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13. PREFACE <p>The last decade has seen the advent of the He<sup>3</sup>-He<sup>4</sup> dilution refrigerator and the ready availability of high-field superconducting solenoids. These developments have produced a phenomenal increase in the number of research efforts involving the production and use of ultralow temperatures (T &lt; 0.3 K). This research effort has the potential of increasing our understanding of solid state physics and of leading to technological advances both in materials and in techniques.</p> <p>The Office of Naval Research and the Naval Research Laboratory recognized the importance of research at ultralow temperatures and decided to cosponsor a two-day symposium on the production and use of these temperatures. This symposium, held at the Naval Research Laboratory on April 23 and 24, 1970, had as its primary motives (a) to bring together scientists, engineers, and administrative personnel who have an active interest in the area of ultralow temperature research, (b) to provide a basis for the free exchange of ideas and the frank discussion of problems and programs, and (c) to collect the comments and opinions of experts regarding the current status and future of ultralow temperature research.</p> <p>The symposium was divided into four half-day sessions and had nine invited talks covering the history, current status, and future prospectus of the field. These talks were supplemented by short contributed papers at the end of each session. The contributed talks were of a more specific nature involving current research by</p>		

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individuals and groups. One of the most important contributions of this symposium was the providing of ample time for free and uninhibited discourse between the attendees.

Topics for the invited talks and the general planning of the symposium were the responsibility of the organizing committee. Well known speakers from both the United States and Foreign countries were chosen to reflect a large cross section of different ideas, training, and practices. The nine invited speakers were from four universities and three government laboratories, and the contributed papers represented efforts from four universities, one government laboratory, and two industrial laboratories. The attendance at the symposium, numbering approximately 150 people, also represented a diversity of ideas and backgrounds.

**NRL Report 7133**

# **Proceedings of the 1970 Ultralow Temperature Symposium**

*Sponsored by the*  
Office of Naval Research  
*and the*  
Naval Research Laboratory

*Edited by*  
**R. A. HEIN, D. U. GUBSER, AND E. H. TAKKEN**

**April 23-24, 1970**



**NAVAL RESEARCH LABORATORY**  
**Washington, D.C.**



## PREFACE

The last decade has seen the advent of the He<sup>3</sup>-He<sup>4</sup> dilution refrigerator and the ready availability of high-field superconducting solenoids. These developments have produced a phenomenal increase in the number of research efforts involving the production and use of ultralow temperatures ( $T < 0.3\text{K}$ ). This research effort has the potential of increasing our understanding of solid state physics and of leading to technological advances both in materials and in techniques.

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The symposium was divided into four half-day sessions and had nine invited talks covering the history, current status, and future prospectus of the field. These talks were supplemented by short contributed papers at the end of each session. The contributed talks were of a more specific nature involving current research by individuals and groups. One of the most important contributions of this symposium was the providing of ample time for free and uninhibited discourse between the attendees.

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Manuscripts of the talks were supplied by the speakers and were lightly edited and prepared for publication at the Naval Research Laboratory. In these proceedings, the invited papers occur in the order in which they were presented, followed by a separate section composed of the contributed papers.

It is a pleasure to acknowledge the efforts of the following individuals who significantly contributed to the success of this symposium:

Mr. John J. Lister, Mr. Lloyd L. Carter, Mr. John M. Hoggatt, and Mrs. Peggy A. Trollinger, Public Affairs Branch, NRL, for arrangements and smooth operations of the symposium at NRL.

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#### ORGANIZING COMMITTEE

Robert A. Hein, NRL, and Edgar A. Edelsack, ONR

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\*See Phys. Rev. Letters 24, 1181 (1970).

†To be published.

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‡See Phys. Rev. Letters 24, 1061 (1970).

# **Invited Papers**

## **SESSION I**

Thursday Morning, April 23, 1970

Chairman: R. A. Hein  
Naval Research Laboratory



## A History of Paramagnetic Cooling

R. P. Hudson  
Heat Division  
National Bureau of Standards  
Washington, D.C.

In embarking upon a historical survey of *paramagnetic cooling* I deem it highly gratifying and inspiring that in attendance at this Symposium is a man who has himself played a very significant part in that history, namely, Nicholas Kurti. I will also state — with little fear that there be any doubt in your minds — that this first talk is not therefore the keynote address but a warmup for the same, which latter will, to coin a phrase, *follow hard upon*.

The subject of magnetic cooling, as we understand it today, goes back to the early twenties and is associated with W. F. Giauque and P. Debye. But the essential forerunners to them are many and include — to name a few — Curie, Langevin, Weiss, Kamerlingh Onnes, and Pauli.

Langevin showed how one might explain paramagnetism in terms of the random orientation of elementary magnets, developed a very successful magnetic equation of state, and was the first to discuss the magnetocaloric effect. Weiss stimulated much intellectual interest and experimental work by his concepts of the *molecular field* and the *magneton*\*. In Leiden, Kamerlingh Onnes and his associates, employing their unique facilities of high magnetic fields, liquid hydrogen, and liquid helium, made magnetic saturation measurements on a variety of inorganic salts and by so doing laid the groundwork first for the magnetic cooling proposals and ultimately for their successful realization.

You will appreciate that a truly definitive history of any subject is impossible and that a merely thorough one can result only from massive researches and painstaking evaluation. My treatment today is, perforce, rather superficial, although I shall hope to give you the benefit — by judicious distillation — of commentaries by distinguished personages “who were there” or who were, at least, closer to the events than a youngster such as myself.

Let us turn back in time to P. Curie; he postulated the  $1/T$  law for paramagnetism at the end of the 19th century [*J. Phys.* (3) 4, 197 (1895)]. In 1905 Langevin published a

\*Given the “Curie approximation” of the Langevin formula  $M/H = N\mu^2/3kT = cT$  one has, for the Curie constant per mole,  $c_{\text{mole}} = N_{\text{mole}}\mu^2/3k = N_{\text{mole}}^2\mu^2/3R$ , where  $M_{\text{sat}}$  is the saturation moment per mole. Hence  $M_{\text{sat}} = (3Rc_{\text{mole}})^{1/2}$ , and Weiss observed [*Compt. Rend.* 152, 79 and 187 (1911)] that the molar magnetic moment determined from this relation were all multiples of a certain value  $M_{\text{W}} = 1123.5$  gauss-cm, which is now termed the Weiss magneton.

lengthy treatise [*Annal. Chim. Phys.* (8) 5, 70 (1905)] entitled “*Magnetism and the Theory of Electrons*,” which among a host of topics included atomic diamagnetism and a brief excursion into the associated thermal effects of magnetization and furnished the celebrated “Langevin formula”

$$M/N\mu = \coth a - 1/a = L(a), \quad (1)$$

where  $a = \mu H/kT$  and  $M$  is the magnetic moment established by  $H$  in an assembly of  $N$  elementary magnetic moments  $\mu$ . He pointed out that for small values of  $a$  this reduces to

$$M = \frac{N\mu^2 H}{3kT}, \quad (2a)$$

thus accounting for the Curie law.

Beginning in 1908 or so, a strong interaction developed between scientists in Leiden and in Paris, the latter including — in addition to Langevin and Weiss — Mme. Curie, the Bequerels, and the young G. Urbain (who had separated the rare earths in substantial quantities). Shortly after the first successful liquefaction of helium (1908) and the ensuing discovery of superconductivity in 1911, one finds Kamerlingh Onnes measuring the molar susceptibility and studying saturation phenomena in gadolinium sulfate and thereafter in a wide variety of inorganic materials. Although the prime interest was in checking Weiss’ postulate of an atomic unit of magnetic moment termed the *magneton*, Kamerlingh Onnes felt that departures from Curie’s law in paramagnetics and the onset of superconductivity in metals, observable at low temperatures, were not only fruitful for the elucidation of the behavior of electrons in solids but were closely related phenomena. As he expressed it [H. Kamerlingh Onnes and A. Perrier, *Leid. Commun.* 122a. (meeting of May 27, 1911)], with reference to his program of studies on magnetism at low temperatures, “... deviations from Curie’s law which seemed to us to be connected with the problem as to how far the electrons which occasion magnetic phenomena are frozen fast to the atoms when the substance is cooled to very low temperatures.” (As an interesting footnote to history one might here record the remark [*Leid. Commun. Suppl.* 44a (3rd Solvay Conference, Brussels, April 21, 1922)] that a substantial amount of data taken during the years of World War I were lost, apparently by Dr. K. Hof, traveling home to Switzerland [see L.C. Jackson, *Leiden Commun.* 163] in 1916. These were resolutely remeasured when sanity returned — if briefly — to Europe and tranquility to the laboratories.)

As early as 1914 Kamerlingh Onnes and Oosterhuis [*Leiden Commun.* 139e] appreciated and reported upon the concept of magnetic dilution and its importance in the persistence of paramagnetism into the realm of very low temperatures, at a session of the Royal Academy of Sciences, Amsterdam, on the February 28th of that year [English transl., *Proc. Roy. Acad. Sci. Amst.* 16, 917 (1914)]. For me this insight of 50 years ago is one of the jewels in the treasure chest of magnetic cooling’s history.

In 1920 Pauli discussed [*Phys. Zeits.* 21, 615 (1920)] paramagnetic susceptibility in the light of quantum mechanics. He considered the effect of the magnetic moment vector’s constriction to a few discrete spatial orientations but restricted his analysis to the situation for very weak fields. So he confirmed the Curie law. The so-called “old quantum theory,” however, led to the conclusion that the Langevin factor no longer took on the “classical” value of  $1/3$  but varied with quantum number.

Classically, the weak-field susceptibility for  $N$  elementary magnetic moments of magnitude  $\mu$  is given by

$$\chi = \frac{N\mu^2}{3kT} = \frac{M_{\text{sat}}^2}{3RT}. \quad (2b)$$

Following Pauli, for quantum number  $n$  the elementary magnetic moment vector may orient itself with respect to the magnetic field  $H$  at various angles  $\theta_k$  such that

$$\cos \theta_k = k/n, \quad (3)$$

with allowed values for  $k$  of  $\pm 1, \pm 2, \dots, \pm n$ . The different orientations corresponded to states of different energy, namely,

$$E_k = -\mu H \cos \theta_k \quad (4)$$

and thus would be differently occupied, according to the Boltzmann factor  $\exp(-E_k/kT)$ . Then the effective magnetic moment  $\bar{\mu}$  is given by

$$\bar{\mu} = \frac{\sum \mu \cos \theta_k \cdot \exp(\mu H \cos \theta_k/kT)}{\sum \exp(\mu H \cos \theta_k/kT)} \quad (5)$$

where

$$= \frac{\mu^2 H}{kT} \overline{\cos^2 \theta}, \quad \text{for } H \rightarrow 0, \quad (6)$$

$$\overline{\cos^2 \theta} = \frac{1}{3} \frac{(n+1)(2n+1)}{2n^2} \quad (7)$$

Woltjer and Kamerlingh Onnes in 1923 [*Leiden Commun.* 167c (1923)] published the results of careful measurements of the magnetic moment in gadolinium sulfate octahydrate for a temperature range of 1.5 to 4.2 K and in fields up to 22 kilogauss (1 kilogauss =  $10^{-1}T$ ); see Fig. 1. They observed discrepancies with the Langevin function,\* and expressed the opinion that the deviations must be real, i.e., outside experimental error.† Though alluding to Pauli's ideas, they did not attempt a quantitative analysis, and in fact 4 years passed until Giauque [*J. Amer. Chem. Soc.* 49, 1870 (1927)] carried this through and showed that quantum theory gave excellent accord with the data (Fig. 2).

The so-called *Brillouin function*, which replaces the classical Langevin function  $L(a)$  of Eq. (1), is

$$\bar{\mu} = g j \mu_B B_j(g j \mu_B H/kT), \quad (8)$$

where  $\mu_B$  is the Bohr magneton and

$$B_j(x) = \frac{2j+1}{2j} \coth\left(\frac{2j+1}{2j} x\right) - \frac{1}{2j} \coth\left(\frac{x}{2j}\right). \quad (9)$$

This expression follows from a modified version of the summation in Eq. (5): for quantum number  $j$  the projection quantum number  $m$  may take on all values between  $j$  and  $-j$  at unit intervals (Pauli's  $k$  could not be equal to zero). The derivation was first given by Debye‡

\*One notes here the essential requirements of both strong magnetic fields and liquid helium temperatures. The same salt had been studied in 1912 by Kamerlingh Onnes and Oosterhuis [*Leiden Commun.* 129b] up to 17 kilogauss but only at liquid hydrogen temperatures, and there saturation phenomena were still outside the limits of experimental accuracy. With anhydrous ferric sulfate, however, they had been able to detect, for the first time, a dependence of  $\chi$  upon  $H$ .

†There occurs in this paper, too, the suggestion that paramagnetic susceptibility be used as a thermometric parameter; this is the origin of the "magnetic thermometer."

‡Debye used a quantum number  $n$  and used  $\mu$  for the elementary moment rather than the modern  $g j \mu_B$ .

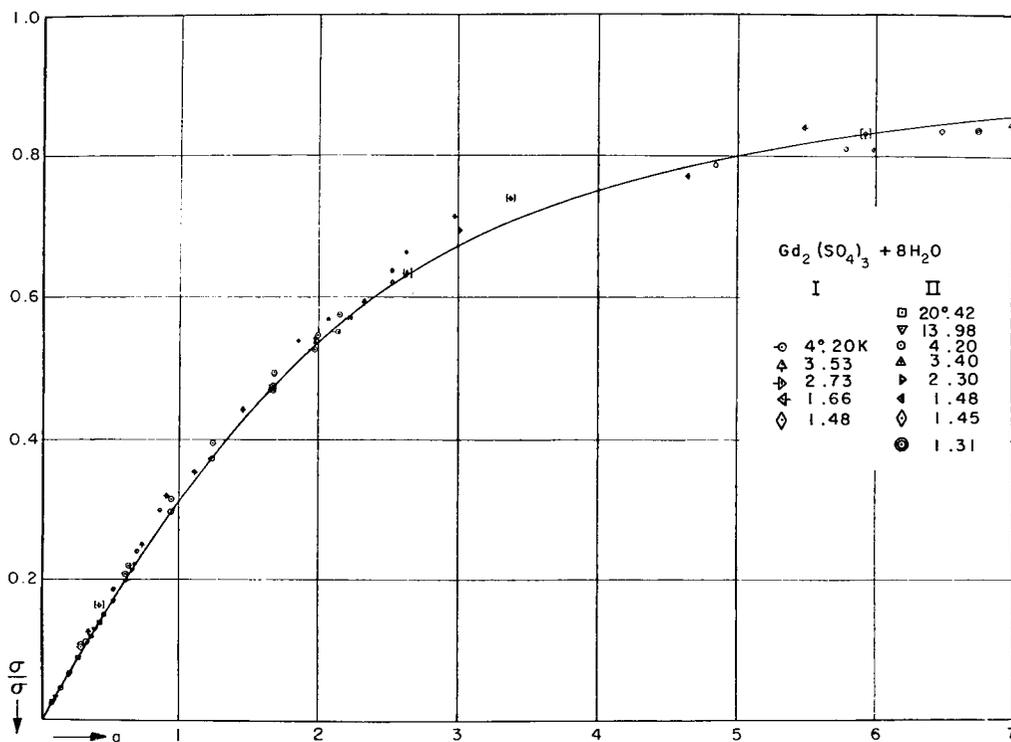


Fig. 1 - Magnetic moment measurements ( $a = \mu H/kT$ ). (From H.R. Woltjer and H. Kamerlingh Onnes, Verslag Kon. Akad. Wetens. Amsterdam 32, 772 (1923); Engl. transl. 26, 633 (1923).)

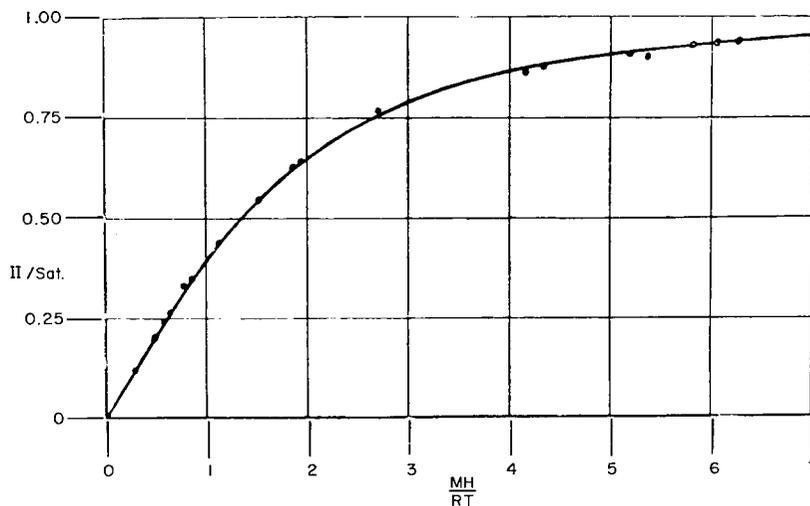


Fig. 2 - Intensity of magnetization of  $Gd_2(SO_4)_3 \cdot 8H_2O$ . The data of Fig. 1 are compared with the theoretical curve, which in no way depends on the measurements. (Reproduction of Fig. 1 from W.F. Giaque, "Proceedings 7th International Congress of Refrigeration," The Hague, June 16-19, 1936, Vol. I, p. 527.)

in 1924 and appears in the 1925 edition of Marx's *Handbuch der Radiologie* [Vol. 6, p. 713]. Two years after this, at the end of 1926, Brillouin submitted a paper "Rotational Moments and Magnetism in Wave Mechanics" to the *Journal de Physique* [*J. phys. radium* 8, 74 (1927)] which included a development of Eq. (5) that went part way to repeating Debye's work. Brillouin obtained the expression

$$\bar{\mu} = \mu_B \left[ \frac{\ell \alpha^{\ell \alpha} + (\ell + 1) e^{-(\ell+1)\alpha}}{\alpha^{\ell \alpha} - e^{-(\ell+1)\alpha}} \right] \frac{1}{\alpha^{\alpha} - 1}, \quad (10)$$

wherein  $\alpha = \mu_B H/kT$  and  $\ell$  is the quantum number of projection varying between  $\ell$  and  $-\ell$ . But he left it at that, manipulating the exponentials no further. [N.B. Stoner quotes Debye's expression in the 1926 edition of his book "Magnetism and Atomic Structure."]

In 1929 Niessen [*Phys. Rev.* 34, 253 (1929)] used the expression *Brillouin function* in referring to Eq. (10). Later, in his famous textbook "The Theory of Electric and Magnetic Susceptibilities," Van Vleck reproduced Debye's expression, Eq. (9), but favored the description *Brillouin function* on the grounds that *Debye function* might lead to confusion with Debye's specific heat function. This recommendation has, by now, the authority of Holy Writ and, like Bunker Hill, which never saw a Battle, is unjustly enshrined in history!

Meanwhile, what of the *magnetocaloric effect*? We have mentioned Langevin's pioneering discussion. The first experimental observations were made by Weiss and Piccard [*J. Phys.* (5) 7, 103 (1917)] in studies of nickel in the region of its Curie point. Their theoretical analysis was given in terms of the molecular field concept originated by Weiss. In a posthumous paper by Kuenen [*Leiden Commun. Suppl.* 47], presented by Ehrenfest before the December 30th, 1922, meeting of the Amsterdam Academy [English transl., *Proc. Roy. Acad. Sci. Amsterdam* 25, 384 (1923)], appears the first *general* thermodynamic treatment, reproducing the results of Weiss and Piccard without recourse to special models.

Combining the first and second laws of thermodynamics, one has

$$dU = T dS + H dI. \quad (11)$$

Changing over to H as independent variable,

$$d(U - HI) = T dS - I dH, \quad (12)$$

whence

$$\left( \frac{\partial T}{\partial H} \right)_S = - \left( \frac{\partial I}{\partial S} \right)_H \quad (13)$$

$$= - \frac{T}{C_H} \left( \frac{\partial I}{\partial T} \right)_H. \quad (14)$$

Noting that the magnetocaloric effect is thus greatly dependent on  $(\partial I / \partial T)_H$ , Kuenen observed that it must become abnormally high near  $T_c$ . (Later, others were to realize that similar arguments, but reinforced by the smallness of  $C_H$ , would apply to the case of paramagnets at very low temperatures.)

And again, starting from Eq. (11), Kuenen derived

$$\left(\frac{\partial T}{\partial I}\right)_S = \left(\frac{\partial H}{\partial S}\right)_I \quad (15)$$

$$= \frac{T}{C_I} \left(\frac{\partial H}{\partial T}\right)_I \quad (16)$$

Thus as, in the paramagnetic region,  $I(T - \theta) = cH$ ,

$$\left(\frac{\partial H}{\partial T}\right)_I = \frac{I}{c}; \quad (17)$$

hence

$$\left(\frac{\partial T}{\partial I}\right)_S = \frac{T}{C_I} \cdot \frac{I}{c}, \quad (18)$$

as found by Weiss and Piccard.

#### THE GIAUQUE-DEBYE PROPOSAL

In the fall semester of 1924 W. F. Giaque was asked to join a colleague in presenting a seminar on magnetism at Berkeley. Giaque was then absorbed in thermodynamics and the third Law in particular but "knew practically nothing about magnetism." He read through the literature on the magnetocaloric effect and was also entranced to find that, according to the Leiden researches, Curie-law behavior persisted in certain paramagnetic substances even at very low temperatures, where heat capacities should be very small. Hence dramatic manifestations of thermomagnetic phenomena — heretofore so miniscule for normal ferromagnets at elevated temperatures — might be observable. Another way of looking at this is: Magnetic fields should be capable of readily removing large amounts of entropy at temperatures so low that (it had been assumed) no appreciable entropy remained. From 1924 on Giaque discussed the concept of magnetic cooling in the environs of Berkeley. At a meeting of the California Section of the American Chemical Society on April 19, 1926, W. M. Latimer gave the first formal, public discussion of Giaque's ideas. Publication before a wider audience was impeded by a phenomenon thoroughly familiar today: referee disdain and opposition! To what must have been his immense chagrin, P. Debye independently published the same ideas late in 1926 [*Annalen der Physik*, 81, 1154 (1926)]; Giaque got his own discussion into print the following year [*J. Amer. Chem. Soc.* 49, 1864 (1927)]. In the light of subsequent developments one must read now with the sympathy born of personal adverse experience his words, "*a helium liquefaction cycle which should soon enable experimental verification is now in process of construction.*" Hydrogen explosions, meager funds, brittle tubing in heat exchangers — to say nothing of the very task of constructing a helium liquefier, its related hydrogen liquefier, electromagnet, and assembling auxiliary equipment — delayed the experimental confirmation for more than 6 years.\*

It is fascinating to realize that (in 1926) Debye saw, as it were, only half of the role played by the "magnetic entropy" in the process, while Giaque (though much closer to

\*Giaque has stated, however (private communication), that he had no interest in merely proving that adiabatic demagnetization worked. He wanted to be able to make accurate magnetocaloric measurements at very low temperatures, employing a large uniform-magnetic-field working volume via a solenoid and shunning the propinquity of iron entirely.

the complete picture) seemed to halt one step away. Both seemed to have thoughts something like "at very low temperatures entropies are *extremely small*, and as the magnetic process is capable of removing *large* amounts of entropy, something dramatic is bound to occur!" Of course, one cannot remove what is not there, and Giauque in fact specified the existence of  $R \ln(2J + 1)$  of magnetic entropy in zero field. Nevertheless each calculated the entropy removable from a suitable paramagnet at liquid helium temperatures by readily achievable magnetic fields and then noted with satisfaction (Debye) that this is 15 times greater than *the lattice entropy* or stated (Giauque), "it does not appear probable that gadolinium sulfate would have any considerable heat capacity in the region about  $1^\circ K$ , but no heat capacity data are available to decide this point."

The fact is of course that the major part of the entropy of the system resides in the *spin system* (to use a modern term) and the lattice entropy is essentially negligible (see Fig. 3). This entropy can be readily removed by a magnetic field at liquid helium temperatures or, in zero field, by thermal excitation at a temperature determined by the magnitude of various indigenous interactions. (And at such a point as the latter heat capacity is very large indeed.) Thus the latter phenomenon determines the temperatures to be reached by adiabatic demagnetization, as most clearly outlined by Kurti and Simon [*Proc. Roy. Soc. A149*, 152 (1935)],\* and in such a process the system moves along an

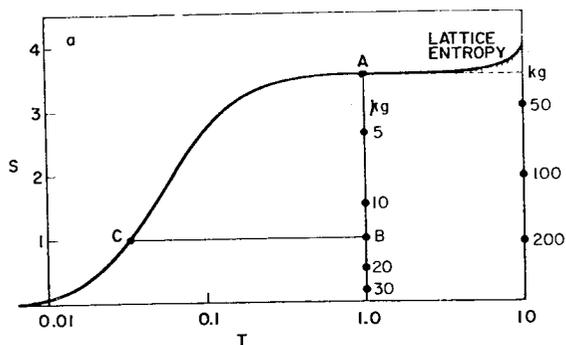
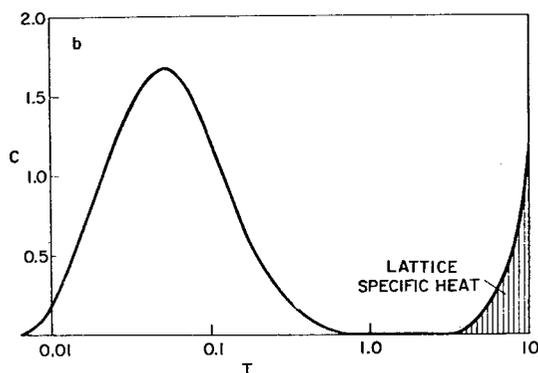


Fig. 3 - Simplified sketch of (a) the entropy and (b) the specific heat of a typical paramagnetic salt. (Reproduction of Fig. 5 from F.E. Simon, "Low Temperature Problems, A General Survey in Low Temperature Physics: Four Lectures," Pergamon Press, London, 1952, p. 15.)



\*The essential features were all laid out by Giauque and Clark in 1932, however [J. Amer. Chem. Soc. 54, 3135, 1932]. In this paper, approximate calculations were made for gadolinium sulfate, treated as an ideal paramagnetic modified only by the presence of lattice entropy. But reference was made to information from F. H. Spedding that spectrographic observations suggested level splittings of the order of  $0.25 \text{ cm}^{-1}$ , and the writers averred that "the calculated reduction in temperature for a given change in field is greater than will be found experimentally."

isentropes from  $B$  to  $C$  in Fig. 3. The entropy diminution for isothermal application of a field  $H$  at temperature  $T$  follows from

$$dG = d(U - TS - HM) = -S dT - M dH, \quad (19)$$

whence

$$\left(\frac{\partial S}{\partial H}\right)_T = \left(\frac{\partial M}{\partial T}\right)_H \quad (20)$$

and

$$[\Delta S]_{H,T} = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH. \quad (21)$$

Use of the *Brillouin function* on the right-hand side of Eq. (21) then leads to the desired result; and there is no problem of infinities, as one encounters using  $L(a)$ .

### EXPERIMENTAL VERIFICATION

Today's generation of experimental physicists knows little of the practical difficulties that loomed so discouragingly large to the cryogenicist of the 1920's. Nor are attitudes, conceptions, and appreciations of the day easy to ascertain — perhaps even for the participants to accurately recall. But we have seen that Giauque, for one, moved steadily and determinedly from his initial proposal toward the goal of realization, the span of 7 years (and more) being determined by the size of the total task relative to available resources. "But what," many ask, "of Leiden?" The Kamerlingh Onnes laboratory was the one place in the world equipped to conduct a test with (few would hesitate to suppose) minimum extra effort. Liquid helium, a powerful and variable magnetic field, and a suitable paramagnetic salt in an evacuable chamber and mounted on a thermal insulator of small heat capacity were required; surely they were readily available. As I probed through the literature, and talked with people who know people who had been there at the time(!), I arrived at the following assessment.

The cryomagnetic facility had been uprooted in 1925 in the cause of expansion. Experimental cryostats were actually an exterior portion of the liquefier, and when the electromagnet was moved away from the vicinity of the latter [Gorter, De Haas, and Van den Handel, *Leiden Commun.* 222d, p. 38], magnetic researches at liquid helium temperatures perforce ceased. When these recommenced in 1932-1933 [Gorter et al., *op. cit.*], a transportable cryostat had to be built. Beyond the fact that, when the 1925 changes were made, much other research was to be tackled, so that magnetism may have seemed to be no shattering loss, I strongly suspect that any magneticist approaching De Haas with a proposal to resume the suspended work by constructing a separately fillable and transportable helium cryostat would have been given very short shrift! (The Collins approach to liquid helium use was a quarter of a century in the future. In cryogenics laboratories of the day, avoidable loss of helium gas was a heinous offence.) Finally, the experiment may have seemed a little wild to almost everybody — witness Giauque's referees!

Also, there was Simon in Berlin; he — expert on entropy and the Third Law *par excellence* — saw immediately the defect in Debye's discussion of the magnetic cooling process. A salt such as gadolinium sulfate *must have a large heat capacity somewhere at very low temperatures*, to match the fall in its (large) entropy which must occur, as required by the Third Law. Early in 1927 Debye had suggested that Simon test his magnetic cooling hypothesis; time passed. In September 1929 Simon gave to Kurti, his new Ph.D. student, Debye's paper to read and set him to look for a heat-capacity anomaly in gadolinium sulfate as part of his thesis program. This latter, completed in the spring of 1931, included evidence that at 3.2 K the heat capacity was indeed rising again, but measurements at still lower temperatures were obviously desirable.

Simon moved to Breslau, and Kurti went with him, setting about rebuilding his heat-capacity apparatus. Simon spent the first half of 1932 as a visiting lecturer in Giauque's laboratory, where he saw emerging the fruits of several years' labor on an impressive installation. Here he received reports from Kurti on the confirmatory experiments on gadolinium sulfate and the suggestion that they should now try magnetic cooling. Simon, however, had no intention of entering into a race with Giauque who, he thought, deserved a clear field. These heat-capacity data [N. Kurti and F. Simon, *Naturwiss.* 21, 178 (1933); details, N. Kurti, *Zeits. Phys. Chem.* 20B, 305 (1933)] showed (Fig. 4) unabiguously the onset of a Schottky anomaly, showed the effect of the crystalline field removing the  $R \ln(2J + 1)$  entropy of angular momentum spatial degeneracy, and permitted an accurate (soon to be confirmed) prediction of the magnetocaloric effect. My late, revered mentor, F. E. Simon, was particularly proud of this piece of work and of both its specific and general importance in the field, and he once took me to task for omitting mention of it in a review article. I stood corrected (and penitent) before the great man and now take this, my first, opportunity to make public — if sadly belated — amends.

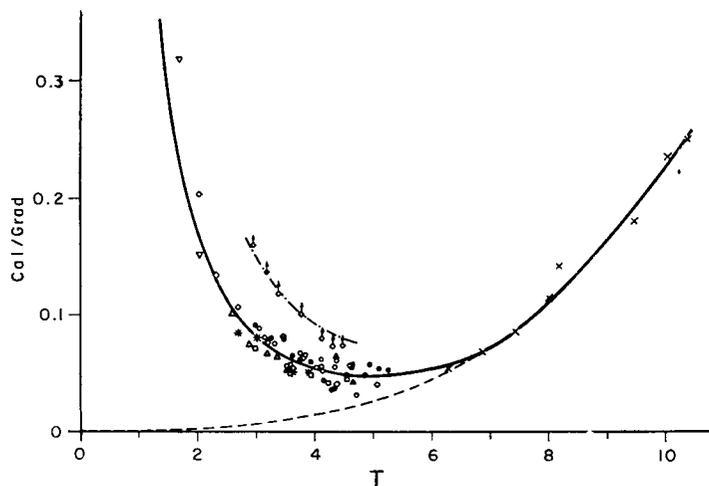


Fig. 4 - Heat capacity data by Kurti in 1933 on 1 gram-ion of gadolinium sulfate. The solid line is the experimental curve for a zero magnetic field, and the dashed curve is the extrapolation with  $T^3$ . The upper curve segment is for a magnetic field of 1800 gauss, corrected in accordance with the experimental curve for zero field. (Reproduction of Fig. 4 from N. Kurti, *Zeits. Phys. Chem.* B20, 305 (1933).)

In 1931 Kramers spent a year in Berkeley, on leave of absence from Leiden. Without doubt he subsequently reported back on the progress made in Giauque's laboratory and the imminence of a test. A decision was made to restore a facility for conducting magnetic studies at liquid helium temperatures — necessitating the said transportable apparatus — and the ensuing race ended almost in a dead heat. On March 19, 1933, Giauque and MacDougall [*Phys. Rev.* 43, 768 (1933)] adiabatically demagnetized gadolinium sulfate from 8 kilogauss at 3.5 K to zero field, reaching a final ("magnetic") temperature of 0.53 K, the salt (within an inductance coil) constituting its own thermometer. Two and a half weeks later, on April 6, 1933, De Haas, Wiersma, and Kramers demagnetized  $\text{CeF}_3$  from 27.6 kilogauss and 1.3 K to 4.5 kilogauss [quoted in *Physica* 1, 1 (1933)], reaching 0.27 K. (Again temperatures were measured via the susceptibility, but here by a force method and so necessitating a sizable measuring field.) In a manifestation of understandable elation the Leiden workers sent out on May 15, 1933,

essentially identical brief notices to *Nature* [131, 719 (1933)], *Naturwissenschaften* [21, 467 (1933)], and *Physica* [13, 175 (1933)]; a fourth delivery was made before the French Academy of Science on June 26 [*Compt. Rend.* 196, 1975 (1933)].

In the very middle of this climactic period, Simon and Kurti uprooted themselves from Germany and reestablished their operations in Oxford. By mid-1934 they were announcing the discovery of a new superconductor (cadmium) through the agency of magnetic cooling [*Nature* 133, 907 (1934)]. Much work followed, up to World War II, in Berkeley, Leiden, and Oxford. From 1948 onward the effort was renewed, and advances blossomed. Today the paramagnetic cooling region spans from 0.001 K to 1 K, and experiments range over thermometry, superconductivity, normal metals,  $^3\text{He}$  and  $^4\text{He}$ , radioactive decay, oriented nuclear targets, paramagnetism, antiferromagnetism, and much more. The perhaps most exotic development, *nuclear cooling*, takes one into the  $\mu\text{K}$  region and will be discussed by Dr. Kurti in the following paper. Two subjects of prime importance — because they sustain all the rest — namely, *thermometry and thermal contact and isolation*, will be dealt with later on by Drs. Abraham and Sarwinski respectively.

### CONCLUDING REMARKS

The foregoing discussion gives only a partial resolution of the “Leiden mystery”: First there were the physical plant changes and intervention of other research interests, as we have seen. Twice in the period 1931-1932, Gorter pressed De Haas to try magnetic cooling but was rebuffed with the statement that there was no hurry. We have seen, too, that the decision *was* reached, in 1932, to build apparatus suitable for the resumption of magnetism research at liquid helium temperatures — and we have concluded that this activity was most probably sparked by Kramers’ reports from Berkeley. But if that be true, how then to explain that the first research using the new apparatus was *not* a test of magnetic cooling?! Could it be that De Haas was obsessed by the desire to maintain the leisurely approach and to present an air of Olympian detachment? Why then the subsequent flooding of the quick-publication journals? It is — to say the least — interesting!

Not only do we today inhabit a different world of physics but also one of different laboratory conditions, external pressures, and extraneous concerns. I cherish one particular glimpse into the world we have lost, which comes to us from a phrase appearing in a followup paper to that describing Leiden’s first successful experiments [W. J. De Haas, E. C. Wiersma, and H. A. Kramers, *Physica* 1, 1 (1933)]. More in sorrow than in anger, or perhaps with a sweet reason engendered by a gentler life, the authors observe, “We investigated the behaviour of  $\text{CeF}_3$ ,  $\text{Dy}(\text{C}_2\text{H}_5\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$ , and  $\text{Ce}(\text{C}_2\text{H}_5\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$ . Then the holidays having arrived, we had to stop work for some months and decided to publish the material now at our disposal.”

### ACKNOWLEDGMENTS

The diagrams used to illustrate this talk are reproductions of figures from original articles whose authors, where possible, and publishers graciously consented to their use. I am also grateful to Professors W. F. Giaque, C. J. Gorter, and N. Kurti for their patient and helpful responses to questions — although the responsibility for any statements, and particularly speculations, herein is entirely mine.

## Historical Survey of Nuclear Cooling

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Van Vleck was probably the first to point out explicitly (in his celebrated "Theory of Electric and Magnetic Susceptibilities") that nuclear paramagnetism, because of the smallness of nuclear magnetic moments, is negligible compared with electron paramagnetism. This fact is important on two counts in a discussion of nuclear cooling, that is, the attainment of very low temperatures by the isentropic (adiabatic and reversible) demagnetization of nuclear spins.

First, as explained by Dr. Hudson in his survey of magnetic (electron-spin) cooling presented in the preceding paper, this method in common with all other cooling processes has a natural limiting temperature below which further cooling is either difficult or inefficient or both. This temperature  $T_{lim}$  is given by the condition  $kT_{lim} = u$ , where  $u$  is an interaction energy between the particle (concerned elementary magnet, atom, or molecule) and its surroundings. Thus for a gas (cooling by adiabatic expansion)  $u$  is the van der Waals energy and  $T_{lim}$  is roughly the boiling point. For spins it may be interaction with the crystalline electric field, or spin-orbit interaction, or hyperfine interaction, and finally - and always present - the magnetic dipole interaction between neighboring spins. Since in this case  $u = \mu^2/r^3$ , where  $\mu$  is the elementary magnetic moment and  $r$  is the distance between elementary magnets, we have

$$T_{lim} \approx \frac{\mu^2}{kr^3}.$$

Clearly therefore  $T_{lim}$  for nuclear spins is about  $10^6$  times lower than for electronic spins. For magnetic cooling  $T_{lim} \approx 10^{-3}$  K; hence for nuclear cooling it will be about  $10^{-9}$  K. This was recognized<sup>1</sup> soon after the first successful magnetic cooling experiments had been carried out, and one is led to ask the obvious question why it has taken so long - compared with magnetic cooling - for nuclear cooling to develop.

The main reason is the smallness of the nuclear magnetic moment. The temperature  $T_f$  that can be reached by demagnetizing to zero field is given in a first approximation by the relation<sup>2</sup>  $T_f = h(T_i/H_i)$ , where  $T_i$  and  $H_i$  are starting temperature and field respectively and  $h = \mu/r^3$  represents the interaction forces; so as far as this goes, small magnetic moment is a help. But the "refrigeration," i.e., the energy that can be absorbed in the temperature region of  $T_f$ , is given roughly by  $T_f \Delta S$ , where  $\Delta S$  is the entropy reduction at  $T_i$  by  $H_i$ . One aims at  $\Delta S \approx R \ln(2I+1)$  or at least  $\Delta S < R \ln(2I+1)$ , where  $I$  is the nuclear spin, and this means that  $\mu H_i/kT_i \approx$  or  $> 1$ , or  $H_i/T_i \approx 10^4$  KG/K. In other words 10 kG at 1 K, which is the initial condition acceptable for magnetic (electron-spin) cooling must be replaced by, say, 100 kG at  $10^{-2}$  K, a condition which in the 1930's, before the advent of superconducting solenoids and <sup>3</sup>He-<sup>4</sup>He dilution refrigerators, was a tough proposition.

Figure 1 shows the disappointingly slow progress of nuclear cooling as compared with magnetic cooling. In this comparison the period 1939 through 1946 was discounted, since

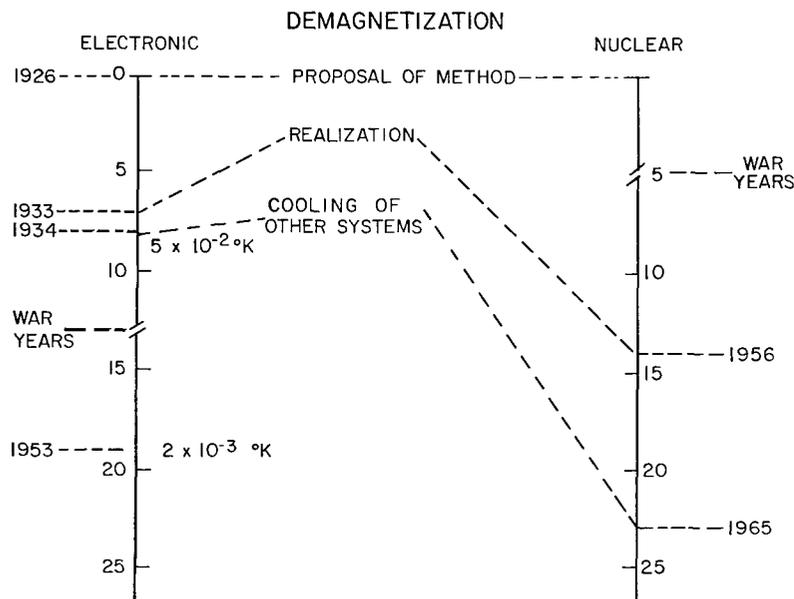


Fig. 1 - Comparison of the developments of magnetic and of nuclear cooling

the war years were not conducive to basic low temperature research. The magnetic cooling was first put into practice in 1933,<sup>3</sup> about 7 years after it had been first proposed<sup>4</sup>; for nuclear cooling the time lag was 14 years. Within 10 years of the first magnetic cooling experiments (always discounting the war years) at least ten laboratories in the world practiced magnetic cooling. For nuclear cooling the corresponding figure today, 14 years after the first experiments<sup>5</sup> in 1956, is four or five laboratories. And whereas magnetic cooling was used within 1 year of its inception for refrigerating other systems, it took 9 years for the same thing to happen in nuclear cooling.

Experimental difficulties are not, however, the sole explanation for the greatly different speeds of development of these two cooling techniques. One of the original incentives for the development of nuclear cooling was the bonus represented by the appreciable nuclear polarization attained under the initial conditions, since polarized nuclei could be used to study directional effects in nuclear reactions. This incentive vanished with the advent of nuclear orientation methods based on hyperfine coupling. Another plausible explanation for the slower development of nuclear cooling is that the temperature range down to about 2 mK, attainable by cerium magnesium nitrate, was still virtually unexplored in the 1950's, and there was little incentive for extending the working temperature range below  $10^{-3}$  K.

It is convenient to divide a review of nuclear cooling into two distinct parts: those experiments in which only the system of nuclear spins is cooled and those in which the nuclear spins are used to refrigerate other systems, either forming part of the same specimen, e.g., conduction electrons in a metal, or a separate specimen, e.g., when cooling liquid  $^3\text{He}$ . We may call the first process simply nuclear cooling and the second process nuclear refrigeration.

## NUCLEAR COOLING

Nuclear cooling is made possible by the fact that the relaxation time  $\tau_1$  for the energy exchange between the nuclear spin system and its surroundings - conduction electrons in a metal and phonons in a dielectric - gets progressively longer at low temperatures. In

metals  $\tau_1$  is governed by the Korringa relation  $\tau_1 T_e = a$ , where  $T_e$  is the temperature of the electrons and  $a$  is the Korringa constant. Expressed in second-kelvin,  $a$  ranges from about  $10^{-3}$  ( $4 \times 10^{-3}$  for Tl) to about  $10^2$  (42 for Li). These are the values in fields above approximately 100 G; for small fields  $a$  drops to about 2/5 of its high-field value. For dielectric crystals, especially in the absence of paramagnetic impurities,  $\tau_1$  becomes too long at  $10^{-2}$  K for the heat of nuclear magnetization to be carried away sufficiently quickly, and for this reason practically all nuclear cooling experiments have hitherto been carried out with metals. However it should be mentioned that the first nuclear adiabatic demagnetization, carried out by Hatton and Rollin<sup>5</sup> in 1949, was done on  $\text{CaF}_2$ . Starting from 12 K and 40 kG the field was reduced to 5 kG, and a nuclear spin temperature - as registered by the spin susceptibility - of 0.3 K was reached.

The first nuclear cooling experiments resulting in microkelvin temperatures were carried out in 1956 by Kurti, Robinson, Simon, and Spohr<sup>6</sup> on Cu. Most other experiments since then, by Fritz, Maria, and Aston,<sup>7</sup> by Dupré, Pitsi, Van Itterbeck, and Vreys,<sup>8</sup> and by Spohr,<sup>9</sup> also used Cu. The lowest temperatures reached were somewhat below  $1 \mu\text{K}$  ( $0.5 \mu\text{K}$  by Spohr<sup>9</sup>) and there was reasonable agreement as to the value of  $h$  (3 G) and the Korringa constant  $a$  ( $0.4 \text{ sec-K}$  in low fields). Walstedt, Hahn, Froidevaux, and Geissler<sup>10</sup> also did experiments on Na, confirmed the Korringa constant measured at higher temperatures, and found  $h = 1.4 \text{ G}$ .

### Temperature Determination

The temperature was determined in all cases by the measurement of susceptibility. The earlier experiments relied on static bulk susceptibility measured by a ballistic galvanometer or an ac bridge. Eddy-current heating even in finely divided metallic specimens can be troublesome, and it is amusing to recall that before the first successful nuclear cooling was performed, several weeks were wasted through using too-high measuring fields; not until the field was reduced to the order of 1 G were signals observed.

A great disadvantage of static susceptibility measurements is that it is not specific. If the sample contains magnetic impurities, they have, especially at low temperatures, temperature-dependent susceptibilities which could mask entirely the nuclear susceptibility. Checks are needed to make sure that the susceptibility-vs-temperature curve flattens out above the usual starting temperature for nuclear cooling, i.e., above 10 mK. Therefore nuclear susceptibilities derived from nuclear magnetic resonance provide a more reliable temperature indication. Both pulse<sup>10</sup> and continuous-wave<sup>11</sup> methods have been used. The continuous-wave method has the advantage of somewhat higher sensitivity and perhaps also greater simplicity. Nuclear free precession on the other hand has the advantage that it can be used in fields of the order of a few gauss and thus can give a more reliable value of  $h$ .

### Irreversible Effects During Nuclear Cooling

One must ask the obvious question: how can nuclear demagnetization experiments which rely on the finite relaxation time  $\tau_1$  for the cooling of the nuclear spin systems give reproducible results? Since the conduction electrons remain substantially at the starting temperature, and since the demagnetization cannot be carried out too rapidly ( $1000 \text{ G/sec}$  is a fairly typical figure if eddy-current effects are to be made negligible), there is a continuous heat exchange between the nuclear spins, the rest of the specimen, and the heat sink. However, as the following calculation<sup>12</sup> shows, this heat exchange has a very small effect on the result in the usual experimental conditions. We may consider two limiting cases: case 1 is perfect thermal contact between the metallic specimen and the heat sink at  $10^{-2}$  K (paramagnetic salt or dilution refrigerator), so that the conduction

electron temperature remains constant, and case 2 is temperature equilibrium between nuclear spins and conduction electrons (short  $\tau_1$ ), with the dominant factor being the heat exchange between the sink and the specimen.

**Case 1** - The energy exchange between conduction electrons and nuclear spins is governed by the relaxation equation

$$\frac{dT}{dt} = \frac{T}{\tau_1} \left( 1 - \frac{T}{T_e} \right) \quad (\text{valid for } \frac{\mu H}{kT} \ll 1)$$

or, for the rate of energy transfer,

$$\frac{dQ}{dt} = \lambda \left( \frac{H}{T} \right)^2 \frac{T}{\tau_1} \left( 1 - \frac{T}{T_e} \right),$$

where  $\lambda$  is the nuclear Curie constant. On the other hand, since the internal energy of an ideal paramagnetic is constant, we have  $\delta Q = -HdM$ , so that, remembering that  $M = \lambda H/T$ , the heat influx to the nuclear spins during demagnetization may be written

$$\frac{dQ}{dt} = -H \frac{dM}{dt} = \lambda \frac{H}{T} \frac{dH}{dt} + \lambda \left( \frac{H}{T} \right)^2 \frac{dT}{dt}.$$

Equating these two expressions for  $dQ/dt$  and assuming a field reduction at constant rate, namely,

$$H = H_i - \beta t,$$

we find by integration between  $t = 0$  and  $t$

$$\frac{T_i}{T} = 1 + \frac{\beta t}{H_i - \beta t} \left[ 1 - \exp\left(-\frac{t}{\tau}\right) \right].$$

For complete demagnetization we write  $H_i - \beta t = h$  and find for the final temperature  $T_f^{(1)}$  reached in these conditions of case 1

$$\frac{T_i}{T_f^{(1)}} = \frac{\beta \tau}{h} \left[ 1 - \exp\left(-\frac{H_i}{\beta \tau}\right) \right].$$

Since in the ideal adiabatic reversible case

$$\frac{T_i}{T_f^{(id)}} = \frac{H_i}{h},$$

the deviation from ideal behavior is given by

$$\frac{T_f^{(1)}}{T_f^{(id)}} = \frac{H_i}{\beta \tau \left[ 1 - \exp(-H_i / \beta \tau) \right]}.$$

**Case 2** - The heat leak to the nuclear spin is governed by the heat influx to the conduction electrons and the lattice. Let us assume this to be a constant  $Q$ . We now have simply

$$Q = -\lambda \frac{H}{T} \frac{dH}{dt} + \lambda \left(\frac{H}{T}\right)^2 \frac{dT}{dt},$$

or, since

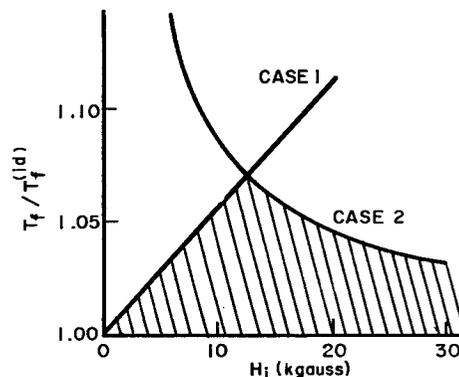
$$\frac{dT}{dt} = -\beta \frac{dT}{dH},$$

we find by integrating between  $H_i$  and  $h$  (between  $T_i$  and  $T_f^{(2)}$ )

$$\frac{T_f^{(2)}}{T_f^{(id)}} = 1 / \left[ 1 - \frac{T_i \dot{Q}}{H_i \beta \lambda} \ln \left( \frac{H_i}{h} \right) \right].$$

**Calculation** - If we now put in the suitable numerical values, we find for 0.3 g-atom of Cu and  $\dot{Q} = 1$  erg/sec the two curves shown in Fig. 2. They give the maximum deviations from ideal behavior, and the actual values of  $T_f/T_f^{(id)}$  must lie below either of these two curves (in the shaded area). We thus see that results obtained under such very unadiabatic conditions differ by less than 8% from the ideal case.

Fig. 2 - The effect of nonadiabatic conditions during demagnetization on the final temperatures. Case 1 is good heat contact at the paramagnetic salt, with the heat influx governed by the relaxation time. Case 2 is short relaxation time, with the heat influx governed by the thermal resistance at the paramagnetic salt.



**Early Experiment** - Figure 3 shows a typical nuclear cooling specimen used in an early experiment. It consists of a bundle of fine (approximately  $3 \times 10^{-2}$  mm in diameter) enameled Cu wires embedded on its upper end in  $\text{CrK alum}$ , which served as a heat sink of about 12 mK. Figure 4 shows the time dependence of the nuclear susceptibility after demagnetization, and Fig. 5 gives  $1/T_f$  as a function of  $(H/T)_i$ . The slope of the straight line gives  $1/h$ .

### Nuclear Cooling In Dielectric Crystals

As was pointed out by Abragam<sup>13</sup> the possibility of dynamic polarization of nuclei in dielectrics makes these substances very attractive for nuclear cooling. About a year ago Cheppellier, Goldman, Hoang Chau, and Abragam<sup>14</sup> carried out such experiments on F nuclei in  $\text{CaF}_2$ . A paramagnetic impurity in the form of  $\text{U}^{3+}$  was introduced into the

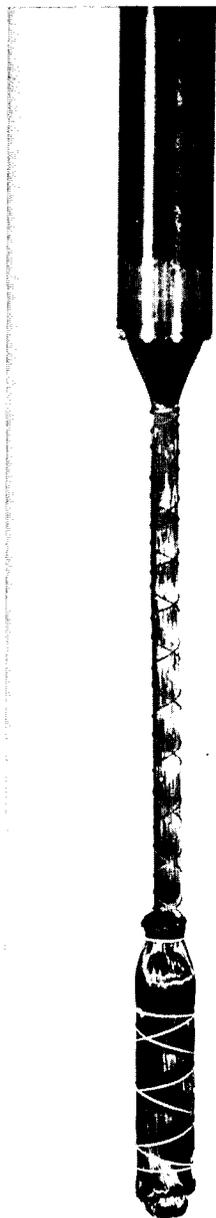


Fig. 3 - Nuclear cooling specimen consisting of a KCr alum heat sink at  $10^{-2}$  K and a bundle of Cu wires

crystal, and the F nuclei were dynamically polarized to up to about 60% in fields of up to 27 kG at a bath temperature of 0.7 K, the corresponding nuclear spin temperature being about 3 mK. The demagnetizations were carried out both to positive and negative spin temperatures. For positive temperatures the behavior of this system was similar to that of Cu ( $T_f$  proportional to  $1/H_i$ ), but for negative temperatures the graph flattened out at polarizations above 30%, indicating the setting in of antiferromagnetic ordering. The Néel point was estimated to be  $0.7 \mu\text{K}$ , and at positive temperatures about  $0.3 \mu\text{K}$  was reached.

More recently Blaisse, Botts, Ottjes, and Koen<sup>15</sup> succeeded in cooling protons in cerium magnesium nitrate by adiabatic demagnetization. The electron spin system to reach a starting temperature of a few millikelvin and the nuclear spin system were in

Fig. 4 - Nuclear magnetic susceptibility, which is proportional to  $1/T$ , as a function of time after demagnetization

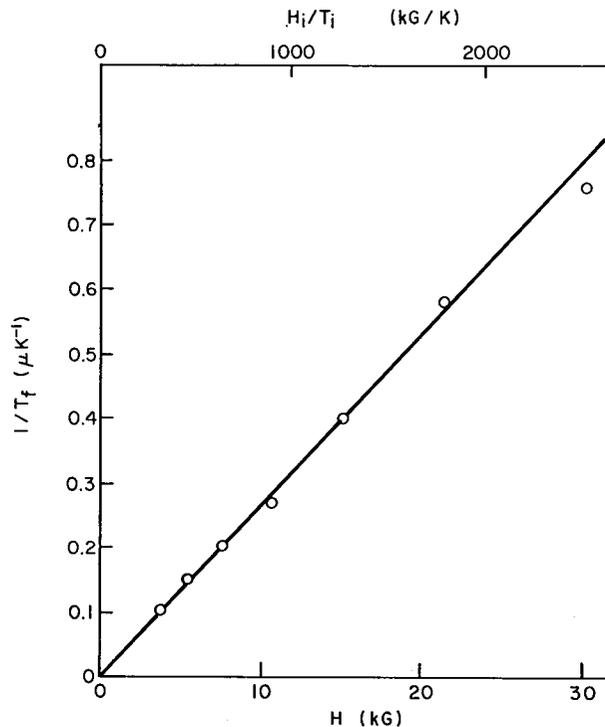
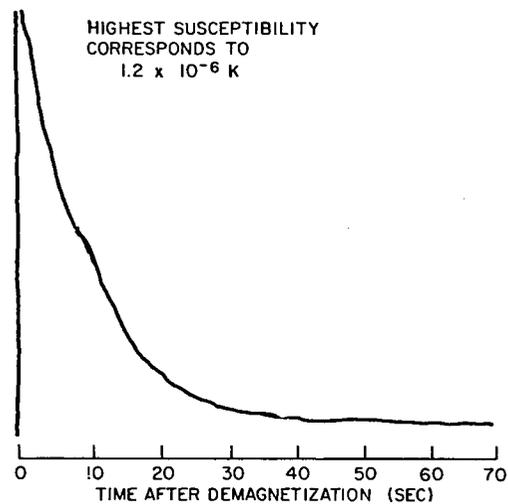


Fig. 5 - Temperatures attained by nuclear demagnetization from 0.012 K and various fields

the same substance. Use was made of the strong magnetic anisotropy of the  $\text{Ce}^{3+}$  ions in this salt, which permits the demagnetization of the electron spins by rotation of the external field. Subsequent reduction of the external field from its initial value of 18 kG to 3 kG resulted in a cooling of the proton to 0.5 mK.

## The Future

One of the two main objects of cooling nuclear spin systems seems to be the determination of the interaction field  $h$  from the relation

$$T_f = h(T_i/H_i).$$

The second main object seems to be the establishment of cooperative nuclear spin ordering and study of its characteristics. So far this has been done only in the Saclay experiment for F in  $CeF_2$ . As was shown in those experiments, and as can be surmised both theoretically and by analogy with electron spin assembly, cooperative ordering cannot be expected unless the entropy  $S/R$  of the spin system is appreciably reduced below  $\ln(2I + 1)$ . In other words one must start with an appreciable spin polarization corresponding to spin entropies of, say,  $0.75 \ln(2I + 1)$ . In dielectric substances this may be achieved by the dynamic polarization method, but in metals the necessary high values of  $(H/T)_i$  must be obtained by brute force.

Table 1 shows the entropies for nuclear spin systems in various metals which have sufficiently long relaxation times  $\tau_1$  to permit reasonably accurate experiments to be carried out with the lattice and the conduction electrons at a temperature not much above 10 mK. The Korringa constants  $a = \tau_1 T_e$  for low fields are somewhat less than half of those given in the table. We see that only for Li can one attain the desired entropy reduction with an  $(H/T)_i \approx 10^4$  kG/K; and it should be noted that the highest  $(H/T)_i$  used so far was about 4000 kG/K. Li has the advantage of a long relaxation time; against it must be set the need of a separate thermal link to the electron spin stage (e.g., and Ag wire) with the corresponding difficulties of good heat transfer between the Li and the link. Since, on the other hand, one can use small nuclear-spin specimens (the warming rate is determined solely by the relaxation time, and the heat capacity is irrelevant), it may be possible to have relatively large thermal contact areas between the Li powder and the link.

Table 1  
Nuclear Entropies  $S/R$  and Korringa Constants  
 $\tau_1 T$  for Some Metals Suitable for  
Nuclear Cooling

H/T (kG/K)	Nuclear Entropy S/R			
	Li	Na	Al	Cu
0	1.386	1.386	1.792	1.386
$5 \times 10^4$	1.293	1.342	1.692	1.339
$10 \times 10^4$	1.071	1.222	1.465	1.210
$20 \times 10^4$	0.62	0.90	0.97	0.87
$50 \times 10^4$	0.09	0.26	0.26	0.24
$a = \tau_1 T$ (sec-K)				
	42	5.0	1.8	1.2



$$\frac{\lambda}{2} \left\{ \left( \frac{H_i}{T_i} \right)^2 - \left[ \frac{h}{T_f^{(1)}} \right]^2 \right\} = \gamma [T_i - T_f^{(1)}],$$

where  $\gamma T$  is the entropy or specific heat of the conduction electrons, and where it is assumed that the nuclear susceptibility follows Curie's law (no saturation effects).

Case 2 is that of a slow rate of energy exchange between nuclear spins and conduction electrons. In the limiting case the nuclear spins cool on demagnetization to  $T_f$  (AB), while the conduction electrons remain at  $T_i$ . The final common temperature  $T_f^{(2)}$  is given by the condition that the energy absorbed by the spins equals the energy given up by the electrons. One sees by inspecting the two shaded areas which must be equal

$$\left( \int_{T_f^{(0)}}^{T_f^{(2)}} T dS_{\text{spins}} = \int_{T_f}^{T_i} T dS_{\text{electrons}} \right)$$

that  $T_f^{(2)}$  can be very much greater than  $T_f^{(1)}$ , especially if  $T_i$  is high or  $\Delta S_m$  is small. Quantitatively

$$\lambda h^2 \left( 1/T_f^{(0)} - 1/T_f^{(2)} \right) = \frac{\gamma}{2} (T_i^2 - T_f^{(2)2}),$$

where  $T_f = h \frac{T_i}{H_i}$ .

Figure 7 shows that there is little chance of cooling conduction electrons *irreversibly* starting from 100 mK or even from 50 mK, and it is not until one gets to  $T_i \approx 10$  mK that the irreversible process gets comparable with the reversible process.

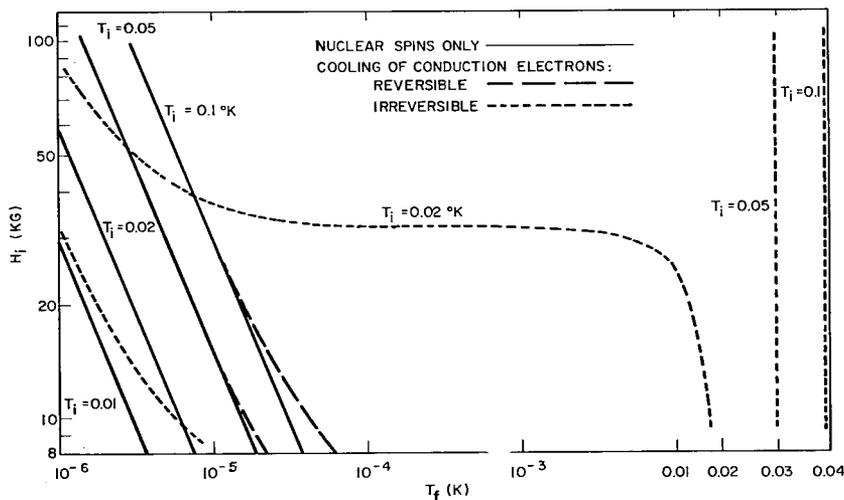


Fig. 7 - Effect of the heat capacity of the conduction electrons on the final temperature reached by adiabatic demagnetization of Cu nuclei. The lines show the fields  $H_i$  required to reach temperatures  $T_f$  for given starting temperatures  $T_i$  under various experimental conditions.

How long does it take for the electrons to follow the nuclear spins after a demagnetization? The long relaxation times ( $\tau_1 \approx 10^6$  sec for Cu at  $1 \mu\text{K}$ ) may make this appear problematic, but, as was shown by Kittel,<sup>16</sup> such a view is due to a faulty argument. While it is true that  $\tau_1$ , which measures the equalization of temperatures with an infinitely large heat reservoir, is very long, the time constant of the process we consider here is short, since the heat capacity of the electrons is small, and even a small rise in nuclear spin temperature suffices to reduce the electron temperature. This is why in nuclear demagnetization spread over, say, 1 minute one has reasonably reversible conditions at the beginning of the demagnetization; by the time the two temperatures begin to diverge, irreversibility ceases to be of importance.

### The Effect of the Heat Influx

It is clear from the foregoing that if there were no heat influx to the specimen, the conduction electrons would follow the nuclear spins with a very small time lag and there would be no difficulty in cooling the whole specimen into the microkelvin temperature range. Let us imagine that, in the absence of heat influx, we have cooled the entire specimen to, say,  $1 \mu\text{K}$ . Any heat influx  $\dot{Q}$  will raise the temperature  $T_e$  of the conduction electrons until  $\dot{Q}$  is balanced by the rate of heat absorption by the nuclear spins. This is determined by the relaxation time  $\tau_1 = a/T_e$  and by the amount of energy that is absorbed by the spin system in each spin flip, which, if demagnetization is to  $H = 0$ , is about  $h/10^6$

The dynamic equilibrium temperature is easily calculated.<sup>2</sup> As we have seen earlier the rate of energy absorption by the spins is

$$\lambda \left( \frac{h}{T} \right)^2 \frac{T}{\tau_1} \left( 1 - \frac{T}{T_e} \right) = \frac{\lambda}{a} h^2 \left( \frac{T_e}{T} - 1 \right).$$

Setting this equal to  $\dot{Q}$  and putting in numerical data for Cu ( $a = 0.4$  sec-K,  $h = 3$  G,  $\lambda = 3.2 \times 10^{-7}$  for 1g-atom) we find

$$\frac{T_e}{T} = 1 + 1.4 \times 10^5 \dot{Q}.$$

Thus if one wants the electron temperature not to exceed the spin temperature by more than 10%,  $\dot{Q}$  would have to be kept below  $10^{-6}$  erg/sec = 0.1 pW, probably unrealizable with the techniques available at present.

The situation becomes more hopeful if one does not aim at such low temperatures but demagnetizes to a finite field  $H$ . The rate of energy transfer then becomes

$$\frac{\lambda}{a} H^2 \left( \frac{T_e}{T} - 1 \right),$$

since in most practical cases  $H^2 \gg h^2$ . Remembering that because of the smallness of the conduction electron entropy

$$H/T = \text{const} = H_i/T_i,$$

we can write

$$\dot{Q} = \frac{\lambda}{a} \frac{H_i}{T_i} H \left( T_e - \frac{T_i}{H_i} H \right)$$

and

$$T_e = \frac{\alpha \dot{Q}}{\lambda(H_i/T_i)H} + \frac{T_i}{H_i} H.$$

The condition for the lowest  $T_e$  for a given heat-influx  $\dot{Q}$  and  $H_i/T_i$  is

$$H = (\alpha \dot{Q}/\lambda)^{1/2}$$

and

$$T_e = 2(T_i/H_i) (\alpha \dot{Q}/\lambda)^{1/2}.$$

Assuming  $\dot{Q} = 10^{-2}$  erg/sec per gram-atom we find for Cu ( $\alpha = 1.2$  sec-K in a field)

$$T_e = 360 T_i/H_i.$$

Thus for  $T_i/H_i = 2 \times 10^{-7}$  k/G (50 kG at  $10^{-2}$  K),  $T_e = 7.2 \times 10^{-5}$  K and  $T = 3.6 \times 10^{-5}$  K. In other words it should be practicable to cool conduction electrons to below 100  $\mu$ K.

It can also be seen that the warming up rate of the specimen should be very slow. We have

$$\dot{Q} = \lambda \left( \frac{H}{T} \right)^2 \frac{dT}{dt},$$

and substituting  $H^2 = \frac{\alpha \dot{Q}}{\lambda}$  we obtain the simple relation

$$\frac{1}{T} \frac{dT}{dt} = \frac{1}{T_e} \frac{dT_e}{dt} = \frac{T}{\alpha}.$$

For the above example  $(1/T_e)(dT_e/dt) = 3.6 \times 10^{-5}$  sec $^{-1}$ , which means that it would take about 1 hour for  $T_e$  to rise by about 10%.

Since for a given heat influx and given  $H_i/T_i$ ,  $T_e$  depends on  $\alpha/\lambda$ , one must have, as far as possible, short relaxation times and large nuclear Curie constants. From Table 2, which gives the relevant constants for Cu, Nb, and Tl, it appears that Tl is by far the most promising substance for nuclear refrigeration. However the difficulty in handling this very toxic substance reduces its usefulness.

Would it be possible to keep the conduction electrons at a *constant* temperature? In principle this is feasible, by continuing to reduce the field after the required temperature has been reached and do so at such a rate that any heat generation is compensated. Calculations<sup>17</sup> have shown that in Tl the temperature of the electrons  $T_e$  reached by demagnetization from 100 kG at  $10^{-2}$  K could be maintained at  $2 \times 10^{-4}$  K for about 1 min with a heat influx of about 10 ergs/sec = 1  $\mu$ W.

## EXPERIMENTAL RESULTS

Unfortunately the experiments carried out so far have not lived up to these expectations. Cooling of conduction electrons in Cu by nuclear demagnetization has so far been

Table 2  
Properties of Some Metals Suitable for Nuclear Refrigeration

Metal	Nuclear Moment, $\mu$ (nuclear magnetons)	Nuclear spin, I	Nuclear Curie Constant, $\lambda$ , for $10 \text{ cm}^3$ (cgs)	Nuclear Entropy Reduction, $\Delta S_m$ , for $H/T = 10^4 \text{ kG/K}$ and $10 \text{ cm}^3$	Korringa Constant, $a = r_1 T$ (sec-K)	$a/\lambda$
Cu	2.3	3/2	$4.5 \times 10^{-7}$	$2.0 \times 10^7$	1.2	$270 \times 10^4$
Nb	6.1	9/2	$13.0 \times 10^{-7}$	$3.9 \times 10^7$	0.36	$28 \times 10^4$
Tl	1.6	1/2	$1.65 \times 10^{-7}$	$0.7 \times 10^7$	0.004	$2.4 \times 10^4$

achieved by March and Symko<sup>18</sup>, by Osgood and Goodkind<sup>11</sup> and by Berglund, Enholm, Gylling, Lounasmaa, and Søvik (whose paper will be presented later in this symposium). The lowest temperature obtained so far in the system of conduction electrons was  $600 \mu\text{K}$  in the recent experiments of Lounasmaa and his colleagues. One can only hope that by improving the experimental conditions it will be possible to reach the predicted temperature and that there is no hitherto unexplained fundamental difficulty.

Instead of demagnetizing to a finite field to improve the energy transfer between electrons and spins one can use a metal with suitable nuclear quadrupole coupling energy, as was shown by Symko<sup>19</sup>, who cooled indium to about  $1.5 \text{ mK}$  by demagnetizing to  $H = 0$ . This method of nuclear refrigeration has the advantage of not requiring a residual field at the end of the demagnetization (although in the case of In a field of a few hundred gauss was retained so as to destroy superconductivity).

Once the technique for experimenting down to about  $100 \mu\text{K}$  has been established, and the accessible temperature range has been extended by a factor of about 20, worthwhile experiments could be carried out on a variety of properties such as size effects in heat capacities, the temperature variation of electrical and thermal conductivities, and superconductivity.

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## Historical Survey of Other Means of Cooling Below 0.3 K

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As far as I am aware the only two practical ways of getting below 0.3 K apart from the magnetic cooling methods discussed in the two previous lectures are the Pomeranchuk refrigerator, in which  $\text{He}^3$  is solidified by compression, and the dilution (or London) refrigerator, in which  $\text{He}^3$  is diluted with liquid  $\text{He}^4$ .

Other methods which, at the moment at least, are not practical include the application of a supercritical magnetic field to a superconducting metal so as to bring it into the normal state. As far as I know the most recent work on this is by M. Yaqub<sup>1</sup>, who tried it as part of his thesis project at Oxford and achieved temperatures of the order of 0.2 K with tin. Another method is to use the mechanocaloric effect in pure liquid  $\text{He}^4$ . The helium II is squirted through a superleak to filter out the entropy carried by the normal component. A practical and continuously operating refrigerator using this effect has been constructed by Staas et al.<sup>2</sup> which operates down to  $\approx 0.7$  K. In both methods the disadvantage is that the "cold" state, the normal metal or the cold helium II, has very little capacity to absorb heat—either to cool other systems or to cope with stray heat influxes. The specific heat per mole in both cases is very small compared to R, and neither method seems useful at the present time for the temperature range below 0.3 K.

Returning to our two practical methods I shall begin with the Pomeranchuk effect, because historically it was suggested first (in 1950) and because it is basically a version of nuclear cooling which Professor Kurti has already discussed.

### THE POMERANCHUK EFFECT

In a 1950 paper Pomeranchuk<sup>3</sup> discussed the probable properties of liquid  $\text{He}^3$ . He postulated that it would be qualitatively similar to an ideal Fermi gas, so that the entropy at low T would be given by  $S/R = (\pi^2/2)(T/T_F)$  with a degeneracy temperature  $T_F$  of the order of  $\approx 5$  K. On the other hand solid  $\text{He}^3$  was expected to retain the full value of the nuclear spin entropy of  $R \ln 2$  down to extremely low temperatures, of the order of  $10^{-7}$  K, which is the order of magnitude of the magnetic dipole interaction energy. This state of affairs is illustrated in Fig. 1. Below  $T_m \approx 0.3$  K,  $S_{\text{solid}} > S_{\text{liquid}}$  and from the Clausius-Clapeyron equation the melting curve has negative slope as shown in Fig. 1b. Below the minimum the latent heat of melting is negative, so that heat is absorbed on solidifying the liquid by compression. As Fig. 1 shows, if the solidification by compression carried out reversibly and completely so that the whole sample is crystallized, the temperature obtained would be the nuclear ordering temperature of the solid, supposed by Pomeranchuk to be  $\approx 10^{-7}$  K but in fact, according to modern measurements of the exchange interaction, of the order of  $10^{-3}$  K. The fact that the melting curve has a minimum leads to a difficulty,

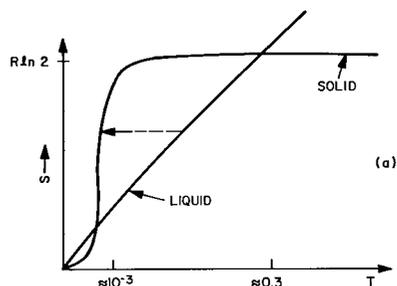
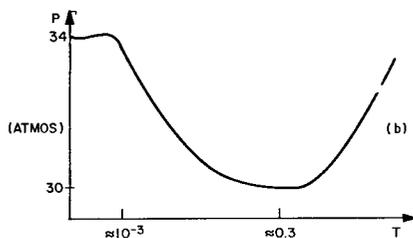


Fig. 1 - Schematic entropy diagram and melting curve for  $\text{He}^3$



both in measuring the melting curve and in using the Pomeranchuk effect for refrigeration. It is impracticable to apply or measure the pressure via a tube leading to higher temperatures. This tube will always block at  $\approx 30$  atmospheres at whatever part of it is at 0.3 K.

In measuring the melting curve<sup>4</sup> at Ohio State in 1960 this difficulty was avoided by having the pressure gage at low temperature. At Los Alamos they<sup>5</sup> used what might be called an intensifier—a spring loaded capsule. Pressures up to 25 atm were applied to the outside of the capsule (a bellows) by liquid  $\text{He}^4$ . The pressure inside was larger than this because of a spring pressing on the bellows. The Los Alamos group used this device to measure the melting curve, but if it had been thermally isolated, it could have been used for cooling experiments.

Curiously enough none of the many laboratories who were interested in measuring the melting curve tried any cooling experiments. I remember there was a general feeling that it would be very difficult, and friction was expected to be a great problem. This feeling depended on the fact that the reversible work to carry out crystallization is large,  $P \Delta V/R \approx 0.5$  deg, so that only 1% of this would heat the sample by 5 mK even assuming the heat capacity of the solid is  $\approx R$ . However, in 1965 Anufriev<sup>6</sup> in Russia published the results of some successful cooling experiments. His apparatus is shown in Fig. 2. The "capsule," which is similar in form to the capsule of an aneroid barometer, is filled with  $\text{He}^4$  to compress the  $\text{He}^3$  which is *outside* in chamber 7. The system was precooled by iron alum (labeled 3) through a tin superconducting switch (labeled 4) connected to a foil immersed in the  $\text{He}^3$ . Another foil was connected to a cerium-magnesium nitrate (CMN) thermometer (labeled 9). Starting at 50 mK he got to  $\approx 17$  mK. The main problem was the poor contact between the CMN and the  $\text{He}^3$  and the fact that the pressure apparently was applied rather jerkily (Fig. 3).

In more recent experiments Wheatley<sup>7</sup> and his collaborators have reached temperatures of about 2 mK. Precooling is by means of a dilution refrigerator, and the thermal switch has been eliminated by using a very large thermal resistance between the dilution refrigerator and the cell. Most important the CMN is immersed in the  $\text{He}^3$ . Some of the earlier results are shown in Fig. 4. Perhaps John Wheatley will tell us what the latest results are during the course of the conference. As far as I know it is not yet possible

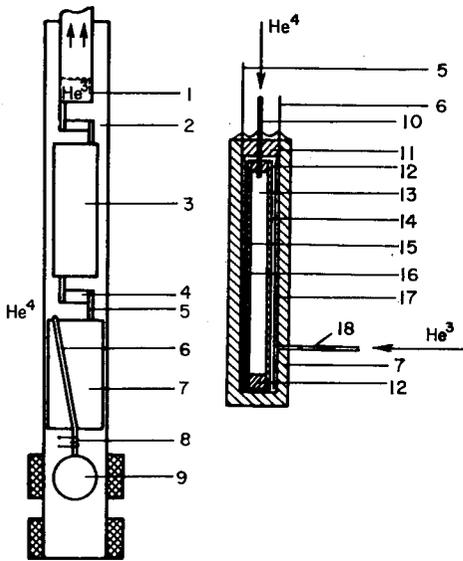


Fig. 2 - Anufriev's apparatus (Fig. 2 from Ref. 6)

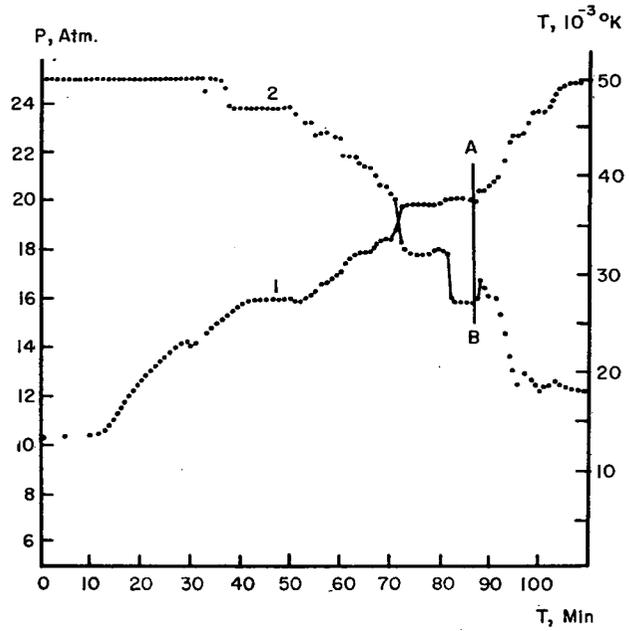


Fig. 3 - Cooling by the Pomeranchuk method (Fig. 3 from Ref. 6)

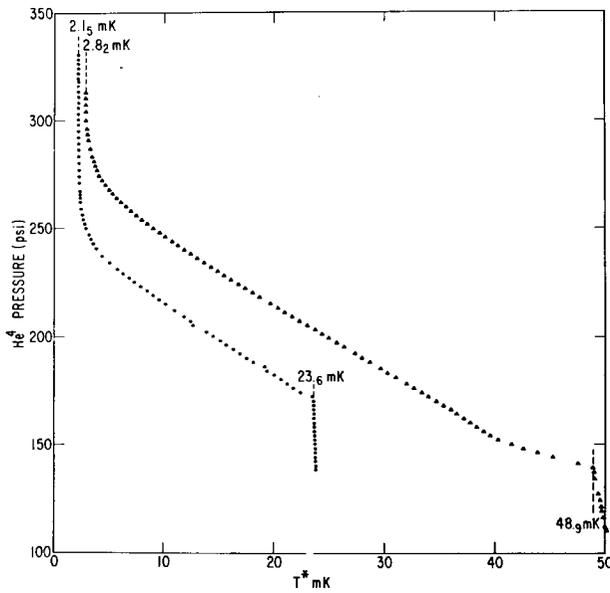


Fig. 4 - Cooling by the Pomeranchuk method (Fig. 1 from Ref. 7)

to say whether the temperatures reached are larger, smaller, or the same as one would expect from theory using empirically determined values of the exchange interaction in the solid and assuming reversibility. The Néel temperature at melting has been estimated<sup>8</sup> at about 1.8 mK with an entropy of  $\approx 0.4R$ . The liquid has this entropy at about 0.1 K, so that temperatures below the Néel temperature,  $\approx 1$  mK perhaps, could be expected if the compression could be carried out to nearly complete solidification. However, there is a great deal of uncertainty in the temperature scale within this region.

In the right circumstances the final temperature *could* be smaller than the strictly reversible crystallization would give. Referring to Fig. 1, if the liquid at some temperature below 0.3 K is crystallized bit by bit and at each step the resulting small amount of solid is removed in some way from the system, eventually the entropy of the last bit of liquid will reach the value at the second crossover point corresponding to the maximum in the melting curve. According to the calculations of Scribner et al.<sup>9</sup> this occurs at about 0.6 mK. (This temperature is in any case the absolute lower limit for cooling by the solidification method. Below this temperature, cooling could be produced by adiabatic melting.) Alternatively, suppose solidification is begun with liquid at temperature  $T_i$  and proceeds with good contact between solid and liquid until some lower temperature  $T_c$  is reached (Fig. 5). Below  $T_c$  thermal contact between liquid and solid might be effectively broken by a large boundary resistance, so that the temperature  $T_f$  of the solid subsequently formed would correspond to reversible crystallization from  $T_c$  rather than  $T_i$ , while any liquid remaining and the solid previously formed would still be at  $T_c$ .

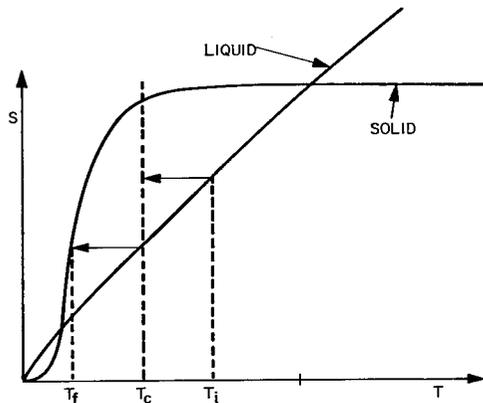


Fig. 5 - Scheme for obtaining a final temperature lower than what strictly reversible crystallization would give

## DILUTION COOLING

In a way it is odd that Pomeranchuk did not also suggest the principle of cooling by the dilution of  $\text{He}^3$  with liquid  $\text{He}^4$ . In 1948 he published a short paper<sup>10</sup> with Landau on the phenomenological theory of dilute solutions of  $\text{He}^3$  in  $\text{He}^4$ . According to this theory the  $\text{He}^3$  in solution acts like a set of quasiparticles with an energy momentum relation  $\epsilon = -E_3 + p^2/2m$ . The thermodynamic properties of the solution at low temperatures therefore resemble those of a Fermi gas with an effective mass  $m$ . Empirically  $m \approx 2.3m_3$ . Since Pomeranchuk was obviously interested in methods of cooling, as shown by his paper on pure  $\text{He}^3$ , it is surprising that he did not point out that this gaslike system of quasiparticles in solutions could be used for cooling in ways similar to those using real gases at high temperatures. In fact this idea was first suggested by Heinz London at the 1951 Oxford Low Temperature Conference in some remarks<sup>11</sup> after a paper by Pryce on nuclear cooling and orientation. Rather than the Landau-Pomeranchuk theory, London quoted the model for  $\text{He}^3$ - $\text{He}^4$  mixtures put forward by Heer and Daunt<sup>12</sup> in 1951. This

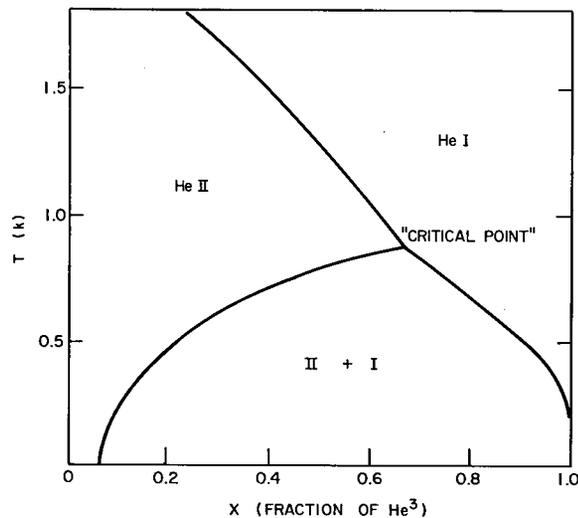


Fig. 6 - Phase diagram of  $\text{He}^3$ - $\text{He}^4$  liquid at saturated vapor pressure

assumes that the liquid is similar to a mixture of ideal Fermi and Bose gases. London suggested that cooling be produced by the reversible adiabatic dilution of the  $\text{He}^3$  with  $\text{He}^4$  via a superleak. Since the entropy of a Fermi gas is simply a function of the ratio  $T/T_F$ , where the Fermi temperature  $T_F \propto n_3^{2/3}$  ( $n_3$  is the  $\text{He}^3$  number density), dilution by a factor of  $10^3$  would reduce the temperature by  $10^2$ .

London did not publish any more about his idea until 1960<sup>13</sup> and 1962<sup>14</sup>. In the meantime the phenomenon of phase separation had been observed for the first time by Walters and Fairbank.<sup>15</sup> London with Clarke and Mendoza in their 1962 paper<sup>14</sup> noticed the resemblance between the phase diagram of  $\text{He}^3$ - $\text{He}^4$  liquid (a modern version of which is shown in Fig. 6), and the phase diagram of a single-component liquid-vapor diagram. The concentrated  $\text{He}^3$  phase is analogous to the liquid, and the  $\text{He}^3$  in dilute solution is analogous to the vapor phase. They suggested a number of refrigerator models which worked by the "evaporation" of  $\text{He}^3$  from the concentrated phase into the dilute, vapor-like phase. One of these designs used a pump at room temperature to circulate the  $\text{He}^3$  continuously, as suggested by H. E. Hall, together with a heat exchanger to precool the returning, concentrated  $\text{He}^3$  and so was identical to the present conventional design of circulating refrigerator. At that time it was not known what the form of the phase-separation curve for low  $X$  would be as  $T \rightarrow 0$ . The fact that the available thermodynamic data indicated that some  $\text{He}^3$  might remain in solution at 0 K was pointed out by Edwards and Daunt<sup>16</sup> in 1961, but this possibility was not experimentally verified<sup>17</sup> until 1965. Under the circumstances it was not possible to make very good estimates of the performance of the refrigerator, and London et al.<sup>14</sup> rather underestimated the effectiveness of the device.

The construction of a working refrigerator was attempted at Harwell, where London was, and then at Manchester by Mendoza in collaboration with London and later by Hall. Another was begun in 1961 at Dubna in Russia for use in nuclear polarization studies, and a third was begun at Leiden. Accounts of these first-generation refrigerators were published in 1964 to 1966. As far as I can ascertain the first "fridge" to operate was the Leiden machine<sup>18</sup>, which reached 0.22 K. Apparently it suffered from convection currents in the dilute side of the circuit. This may happen, as was pointed out by London (reported in Ref. 19), if the hotter, heavier, less-concentrated liquid in the still is above the lighter, more-concentrated dilute solution in the mixing chamber and if the connecting

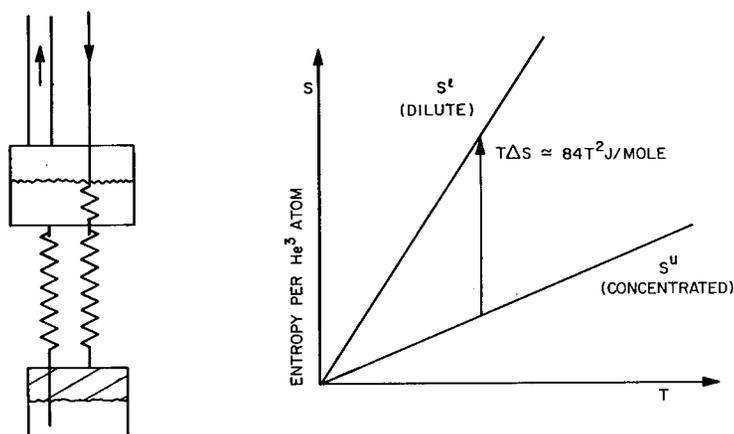


Fig. 7 - Principle of the circulating dilution refrigerator

tube is not narrow enough to inhibit the instability. Both the Hall<sup>19</sup> and Dubna<sup>20</sup> refrigerators worked successfully, but the Dubna machine had the better specifications, reaching 25 mK in its final form.

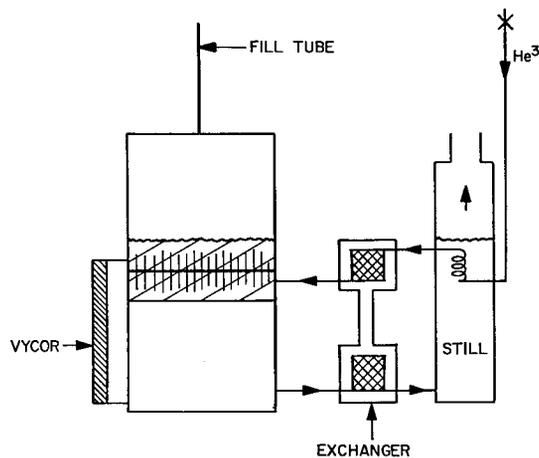
More recent machines, as you all know, have reached 10 mK in continuous operation and of the order of 5 mK in single-cycle operation.<sup>21</sup> Since  $\text{He}^3$  will remain in solution up to a concentration of 6.4%<sup>22</sup>, the final temperature is not limited by an exponentially decreasing concentration in the dilute phase as it would if the London analogy with a liquid-vapor system were exact. In fact I believe that a better analogy at low temperatures is with a thermoelectric refrigerator, the degenerate  $\text{He}^3$  in the upper and lower phases in the mixing chamber being analogous to the electrons in two different metals in contact. The heat of dilution  $T(S_3^l - S_3^u)$ , where  $S_3^l - S_3^u$  is the entropy difference per mole of  $\text{He}^3$  between the lower and upper phases (Fig. 7), is the analog of the Peltier heat. The heat exchangers exploit the "Thomson heat" of the returning  $\text{He}^3$  to precool the incoming  $\text{He}^3$ .

The dilution refrigerator is much more effective than the thermocouple refrigerator for several reasons: As regards circulation rate they are comparable;  $10^{-5}$  moles/sec is equivalent to  $\approx 1$  ampere. Due to the large mass the Fermi temperature of the dilute phase ( $\approx 0.4$  K) is small compared to that found in metals or even in semiconductors, giving a large entropy per particle. Most important the relation between the thermal conduction and "Joule heating" in the "leads" is quite different in the dilution refrigerator. The  $\text{He}^3$  flows in the connecting tubes in hydrodynamic laminar flow in a way which is not known in any electrical conductors. This means that frictional heating can be minimized without incurring too much thermal conduction into the cold region. Nevertheless, as Wheatley, Vilches, and Abel<sup>23</sup> have shown, even with the optimum length of exit tube, giving the best balance between heat conduction and friction, the minimum possible temperature in the mixing chamber is still limited by these effects and is given approximately by

$$T_{\min} = (4 \text{ mK}) [d(\text{mm})]^{-1/3},$$

where  $d$  is the diameter of the exit tube from the chamber. Since the maximum practical value of  $d$  is about 1 mm or so, this gives a practical limit of  $\approx 4$  mK for the circulating or single-stroke refrigerator in which  $\text{He}^3$  is removed through a tube.

Fig. 8 - Cooling by the dilution of  $\text{He}^3$  with  $\text{He}^4$  through a superleak



The original suggestion by London that the  $\text{He}^3$  could be diluted by the reversible addition of  $\text{He}^4$  through a superleak has not yet been thoroughly investigated, although I believe some experiments are being made in Leiden. The difficulty with the heat conduction and friction in the exit tube is avoided in this method. With great care dilution through a superleak might yield much lower temperatures than the conventional refrigerator. Starting with  $1 \text{ cm}^3$  of pure  $\text{He}^3$  precooled by a circulating fridge to  $10 \text{ mK}$ , reversible dilution to  $\approx 12 \text{ cm}^3$  of saturated solution would give  $\approx 2.4 \text{ mK}$ , and further dilution to  $\approx 100 \text{ cm}^3$  would give  $0.6 \text{ mK}$ , the limit for Pomeranchuk cooling. The entropy per mole and the specific heat would both remain constant with temperature at  $\approx 0.03R$ . With a heat leak of  $1 \text{ erg/min}$ , it would take 1 hour for the helium to warm from  $0.6 \text{ mK}$  to  $0.7 \text{ mK}$ .

Figure 8 is offered as a suggestion for an "attachment" to a conventional circulating refrigerator which might allow dilution through a superleak starting at  $\approx 10 \text{ mK}$ . An extra chamber is added above the conventional mixing chamber and connected to it by a superleak made of Vycor glass. It is also thermally connected to the mixing chamber by means of wires or sintered copper in the partition between them, which is the roof of the conventional mixing chamber and the floor of the "dilution" chamber. The dilution chamber is partially filled with a charge of pure  $\text{He}^3$  by means of a filling tube. When the circulating refrigerator has cooled down to its lowest temperature, the valve in the  $\text{He}^3$  return tube is turned off. When all of the concentrated phase has disappeared from the conventional mixing chamber and the liquid there becomes unsaturated, dilution begins in the upper chamber. The removal of  $\text{He}^4$  from the lower chamber through the superleak breaks thermal contact between the chambers automatically.

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## **Invited Papers**

### **SESSION II**

Thursday Afternoon, April 23, 1970

Chairman: J. G. Daunt  
Stevens Institute of Technology



## Thermometry at Ultralow Temperatures

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Although the measurement of temperature is absolutely essential for most experiments, it is no exaggeration to say that its measurement for most experimentalists is an absolute bore. The experimentalist usually seeks to establish the temperature dependence of some physical parameter such as energy absorption, attenuation, or spin diffusion, but more attention is given to the measurement of this quantity than to the temperature itself. Even though experiments are now being performed in the millikelvin temperature range, the temperature scale below 6 mK is yet to be identified with the thermodynamic scale in an unambiguous fashion. Further, the corrections to the common thermometer, cerium magnesium nitrate (CMN), are yet to be agreed upon. In this presentation I shall give a fairly detailed account of the CMN thermometer and then describe briefly various other possibilities with emphasis on the most promising.

When a temperature is assigned to a physical quantity, one refers to a parameter which in some way can be related to the absolute temperature defined by

$$T = \left( \frac{\partial U}{\partial S} \right)_{P,H} = \left( \frac{\partial E}{\partial S} \right)_{V,M} , \quad (1)$$

where  $U$  is the enthalpy and  $E$  the internal energy. In the range where the primary standard, the gas thermometer, cannot be used to establish the temperature, other physical parameters which give a temperature identical to the one defined by Eq. 1 must be found. The susceptibility of a paramagnetic salt was the natural choice, since there is a well defined and well understood magnetic equation of state in the absence of interactions. As long as the experimentalists were preoccupied with the properties of the paramagnetic salts, ellipsoidal single crystals could be used; however, with the extension of experimental investigations to other materials, so that the properties of the salt were a secondary concern, the need for a reliable thermometer became a primary concern.

Since the initial work of Daniels and Robinson,<sup>1</sup> CMN has preempted the role of thermometric substance. It has now been well established, through the original work and subsequently through the work of many others, notably Hudson and coworkers,<sup>2</sup> that CMN obeys Curie's Law to 6 mK. Below 6 mK, where the dipolar interactions begin to play a substantial role, there are deviations from Curie's Law which have not been well established. Not only is the correlation<sup>1,3-5</sup> between the magnetic temperature  $T^*$  and the thermodynamic temperature  $T$  uncertain, but the shape correction for thermometers constructed with powdered CMN in nonellipsoidal shapes still seems to be a subject for debate. Such as it is, the CMN thermometer is still the standard for thermometry below 1 K.

For material which obeys Curie's Law the magnetization per unit volume  $m$  is given by

$$m = cH_1/T, \quad (2)$$

where  $H_1$  is the local field acting on the ions. In the Lorentz field approximation  $H_1$  will be given by

$$H_1 = H + (N - D)m, \quad (3)$$

where  $H$  is the externally applied field. On substituting Eq. 2 into Eq. 3 we find that

$$m/H = \chi_{\text{exp}} = c/[T - (N - D)c], \quad (4)$$

from which we obtain the definition of  $T^*$ :

$$T^* = c/\chi_{\text{exp}} = T - (N - D)c = T - \Delta. \quad (5)$$

If the dipoles are in a cubic array, then  $N = 4\pi/3$ ; and if the specimen is spherical,

$$T^* = T^* = T.$$

For an actual substance  $N \neq 4\pi/3$ , so that we have for a sphere  $T^* + \Theta = T$ , which really expresses the inadequacy of the Lorentz approximation. The correction  $\Delta$  includes the shape correction  $\Delta'$  and the Weiss constant  $\Theta$ . If one considers a powder, then Eq. 4 defines  $\chi$  for each particle, and the effective  $\chi_{\text{exp}}$  is the volume average:

$$\chi_{\text{exp}} = \frac{1}{V} \sum V_i m_i/H = \frac{1}{V} \int_{\phi} \sum \frac{V_i c d\phi}{T - (N - D)c}. \quad (6)$$

It is clear from Eq. 6 that the Curie constant  $c$  in the denominator is the single-crystal value and that for an anisotropic  $g$  value  $c$  is averaged over all angles  $\phi$ . For a powder, then,

$$T^* = \bar{c}/\chi_{\text{exp}} = T - (N - D')\bar{c} = T - \Delta' - \Theta,$$

and the demagnetization correction  $\Delta'$  need not be zero for a sphere of powder. A right circular cylinder of CMN for which the height equals the diameter could have a correction as large +3.5 mK or as little as 0 mK. The magnitude is such that inaccuracies in the  $^3\text{He}$  temperature scale as well as in the calibration procedures preclude the determination of  $\Delta'$  in the normal calibration range of 0.6 mK to 1.6 K.

Illustrated in Fig. 1 is the demagnetization factor<sup>6</sup> for a uniformly magnetized cylinder of varying length-to-diameter ratios  $\gamma$ . For a ratio of 1 we see that the demagnetization factor  $D/4\pi$  (curve  $N_f$ ,  $\chi = 0$ ) is nearly 0.2 and not 0.333, the value for a sphere. The value of  $\Delta'$  for a body whose gross shape is that of a cylinder with an aspect ratio of 1 but is made up of highly anisotropic particles such as CMN granules is clearly an indeterminate quantity. To establish thermal contact with a test body, it is almost essential to use powdered salt. Furthermore, cylinders are much, much easier to fabricate than spheres. If one is interested in working below 100 mK and wants to know the

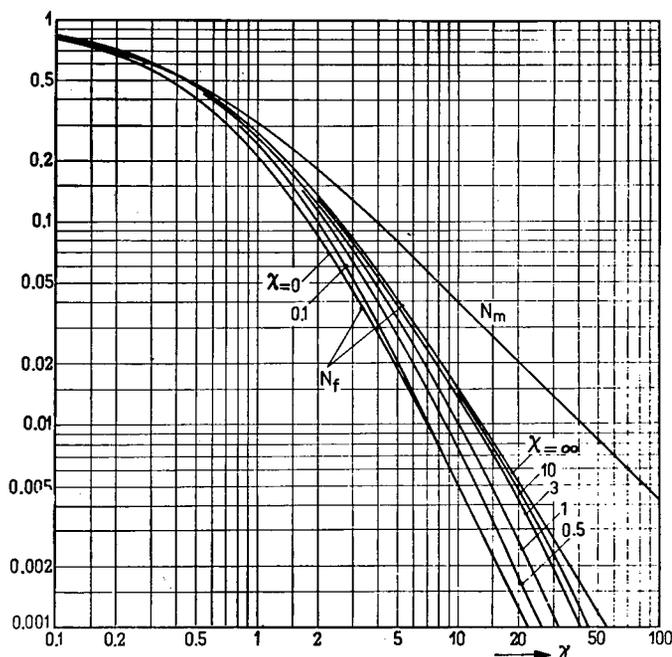


Fig. 1 - Demagnetization factor<sup>6</sup> for a uniformly magnetized cylinder of varying length-to-diameter ratios  $\gamma$

temperature to at least 1%, then the magnitude of  $\Delta'$  becomes very important indeed. The necessity for this correction I consider to be the most serious limitation to the CMN thermometer, much more of a drawback at the moment than the 6-mK lower limit or the large specific heat. There is really no good way to determine  $\Delta'$  *in situ* for a particular thermometer.

The importance of  $\Delta'$  on the CMN temperature scale cannot be overemphasized. To illustrate the effect on interpretation of data, we see in Fig. 2 the specific heat of liquid  $^3\text{He}$  as measured by Wheatley and coworkers.<sup>7,8</sup> Although there are two symbols and two references, the data are the same and they agree with the later measurements of Wheatley *et al.*<sup>9</sup> You will notice that the points lie on a reasonably good straight line except for those below 10 mK and that the line does not go through the origin. Now compare these same data replotted in Fig. 3 with a  $\Delta = 1.7$  mK added to each temperature. The line goes through the origin, and there is an improvement in the fit. I know that some will regard this an unjustifiable tampering, which it may be, but we are using these figures for heuristic purposes only.

The most direct evidence for a lower limit to the value of  $\Delta$  has been obtained by Abraham and Eckstein<sup>10</sup> through measurements of the specific heat of spherically and cylindrically shaped specimens. They found that 0.8 mK had to be added to the temperatures measured with the cylinders to superimpose the specific heats measured with the cylinders on those measured with the spheres. The agreement between the two sorts of data (cylinder and sphere) is illustrated in Fig. 4. These workers tried to go a step further to obtain the thermodynamic temperature by curve fitting; although the result, the correction of the sphere of powder to a single crystal of powder, is reasonable, a less uncertain method would be very desirable. The evidence is mounting that a right circular cylinder of CMN requires a minimum correction of 1.1 mK for the magnetic temperature  $T^*$  to be equal to the thermodynamic temperature  $T$  down to 6 mK. Below 6 mK the scale is still undetermined.

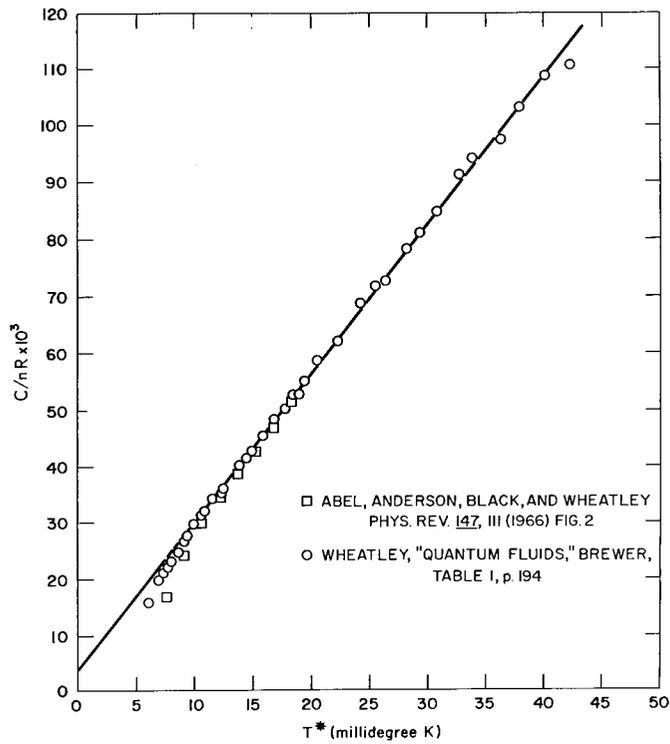


Fig. 2 - Specific heat of  $^3\text{He}$ , <sup>7,8</sup> plotted as  $C/nR$  vs magnetic temperature

Fig. 3 - Same as Fig. 2 except for  $\Delta = 1.7$  mK added to each temperature

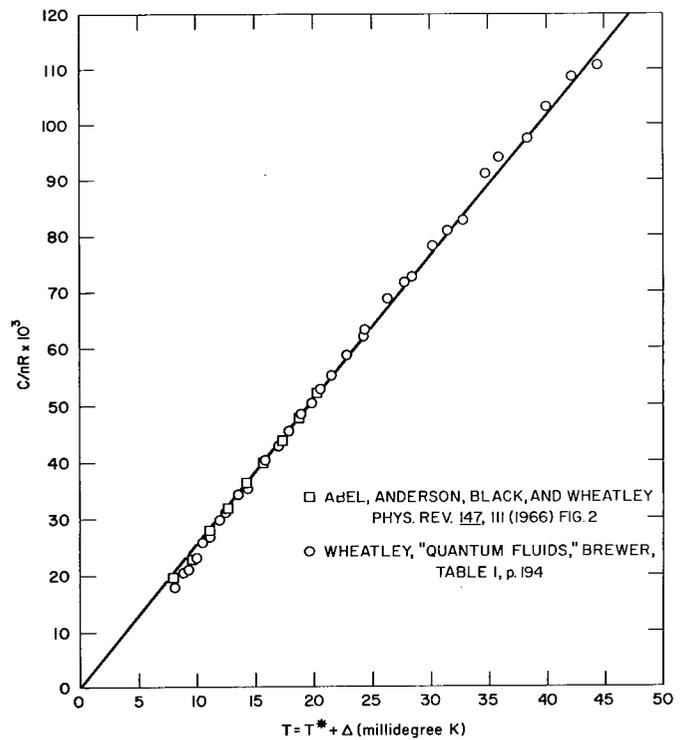
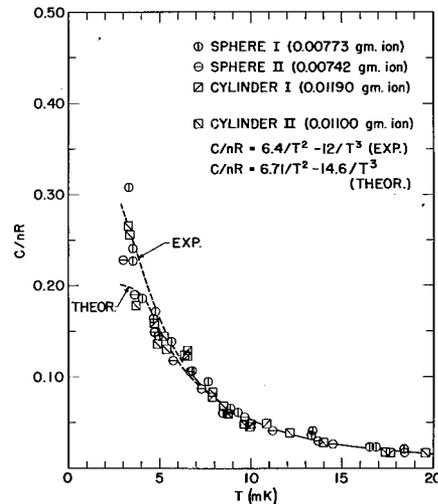


Fig. 4 - Specific heats of spheres and cylinders, with 0.8 mK added to the temperatures measured with the cylinders<sup>10</sup>



In a thermometer used in my laboratory at the Argonne National Laboratory the salt pill is formed around a fan of about 50 silver wires 0.254 mm in diameter. The pill when removed from the die is a right circular cylinder 1.91 cm in diameter composed of approximately 80 volume-percent CMN and 20 volume-percent AgCl. This pill is machined into a cylinder 1.27 cm high by 1.27 cm in diameter or into a sphere 1.27 cm in diameter. It is then inserted into a coil form which has an inner secondary (2000 turns of No. 44 Cu wire) and an outer secondary (approximately 1000 turns of No. 44 wire). The area turns of each is the same; the two coils are balanced to cancel the respective induced emf, and then a turn is taken off the outer coil so that the mutual inductance never goes through zero. Finally a primary (820 turns of No. 38 wire) is wound over the outside. Part of the primary turns are compensating windings to produce a uniform field over the specimen. The primary has a resistance of 2 ohms at 4.2 K; with the calibrating current of 380  $\mu$ amp, 200 ergs/min are introduced into the system. Below 20 mK joule heating is reduced to less than 1 erg/min by reducing the current to 3.8  $\mu$ amp. The maximum field produced by the calibrating current is 0.09 gauss. A completed thermometer has a diameter of 2.29 cm and a length of 3.81 cm. An actual calibration equation is

$$T^* \text{ (kelvin)} = 1353/(M - 19021) ,$$

where  $M$  is the reading on a six-decade ratio transformer. When the maximum excitation is used during calibration, the bridge is sensitive to less than 1 unit, and at the low end when using only 3.8  $\mu$ amp, it is sensitive to 50 units. The fractional temperature sensitivity  $\Delta T^*/T^* = T\Delta M/1353$  is always less than  $10^{-3}$ .

Although the manner of constructing the coils makes them relatively insensitive to external ac fields, the CMN will be affected by stray static fields. The measurement is made at 1700 Hz, so that we always measure  $\chi_{\text{adiabatic}}$ . The quantity of interest is  $\chi_{\text{isothermal}}$ , which is related to the measured quantity by

$$\chi_{\text{ad}} = \frac{\chi_{\text{iso}}}{1 + \frac{cH^2}{T^2 C_p}} ,$$

where  $C$  is Curie's constant,  $C_p$  is the specific heat, and  $H$  is the magnetic field. The magnitude of  $c$  and  $C_p = b'/T^2$  are such that a stray field of 4 gauss will introduce a 1%

error in  $\chi_{ad}$  and consequently a 1% error in the temperature. We therefore keep all ferromagnetic materials at considerable distance, and the superconducting magnet, used for refrigeration, can be lowered 65 cm from the thermometer. Further, the magnet is energized and then deenergized before performing a calibration, so that a standard magnetic state is established. The latter precaution is unnecessary, as we have found that the residual field in the magnet does not influence the measurements.

To sum up the characteristics of the CMN thermometer, we find that thermal contact can be easily established, sensitivity is adequate, and the measured parameter  $\chi$  can be directly related to the thermodynamic temperature. The serious limitations are, most important, the necessity of applying an uncertain shape correction and, second, the inadequacy below 6 mK. If an agreed  $T-T^*$  correlation for CMN could be established, then the scale could be extended to 3 mK. Although there are some claims that the scale is presently known to 3 mK, I must reemphasize that the scale is not known until it can be firmly tied to Eq. 1, and that is far from being accomplished. At present the scale has been established through *ignotum per ignotius*.

From the foregoing it is clear that a thermometer or thermometers are required which do not have the built-in limitations of CMN. A variety of techniques and parameters have been suggested, one of which should, with sufficient perseverance in development, be capable of serving as standard. Among the suggested techniques are measurement of Johnson noise, static and dynamic nuclear susceptibility,  $\gamma$ -ray anisotropy, and the Mössbauer effect. Before going into a description of any one of these I should set up my criteria for a satisfactory thermometer. I hasten to point out that the important technical question of how to establish thermal contact with the thermometer is assumed to be solved. We are concerned only with thermometric systems. The characteristics of an ideal thermometer are these:

1. The measurable parameter can be related to the fundamental definition of temperature. It is not necessary that each experimentalist establish the relationship himself; it is sufficient that someone showed that the thermodynamic temperature is derived from the parameter measured. Someone had to demonstrate that CMN obeys Curie's Law to 6 mK; otherwise it would be useless as a thermometer. I suppose everyone would be willing to accept the Curie Law behavior for nuclei.

2. The internal relaxation time of the thermometer must be short. If  $\tau$  is the relaxation time, then

$$(T_B - T_T) = \tau \dot{T}_B .$$

where  $T_B$  and  $T_T$  refer to the test body and the thermometer respectively. At 10 mK a drift rate of 0.1 mK/min, which one would get with a heat leak of 0.08 erg/sec into 0.01 gram-ion of CMN (7.5 grams), results in an error of 1% in the temperature if  $\tau = 60$  sec. If one were using copper nuclei as the thermometer, for which  $\tau = 1300/T$  sec ( $T$  in mK), the error would be 2%. In general the fractional error would be

$$\frac{\Delta T}{T} = \frac{1300}{T^2} \frac{\dot{Q}}{C} ,$$

where  $\dot{Q}$  is the energy input and  $C$  is the specific heat. If CMN provided the dominant heat capacity, then with constant  $\dot{Q}$  the error would remain constant as the temperature drops; if liquid  $^3\text{He}$  provided the dominant heat capacity, however, the error would increase rapidly as the temperature dropped.

3. The energy required for a measurement should not contribute to the heat leak.

4. The accuracy should be as good as the temperature scale near 1 K, i.e., about 0.1%. Perhaps a more realistic requirement would be 1%.

5. The specific heat should be small, which would reduce the relaxation to the thermometer.

6. The precision or resolution should be at least  $0.001 T$ . This is a *desideratum* but is not essential for the primary thermometer, only for the secondary one. Such a requirement is necessary only for specific heat or thermal conductivity measurements. The conventional manner of making a specific heat measurement is to introduce enough heat to raise the temperature by  $1.1 T$ . To obtain a 2% measurement one must determine the temperature to  $0.001 T$ .

Now that we have set down the essential characteristics of a thermometer, let us examine a few schemes to see how closely one can approximate them. Of the various thermometers I shall describe, the noise thermometer is the only truly absolute thermometer; i.e., no calibration is necessary. The application of such a thermometer was discussed by R. A. Kamper at the Symposium on Superconducting Devices which was held at the University of Virginia in 1967.

If one biases a Josephson junction with a small dc voltage, then one can detect rf radiation from the junction. This radiation will be frequency-modulated by the noise generated in the biasing resistor. The noise voltage generated will be given by Nyquist's formula:

$$V^2 = 4kTRf,$$

where  $R$  is the resistance and  $f$  is the bandwidth of the noise; the other symbols have their conventional meaning. The extent of the modulation  $\Delta f$  is given approximately by

$$\Delta f = \frac{2eV}{h}.$$

If we equate  $\Delta f$  to  $f$ , then we find that the line width  $2\Delta f$  is given by

$$2\Delta f = 32kTRe^2/h^2.$$

On substituting the appropriate values for the constants we find that

$$2\Delta f = 0.3TR \text{ Hz},$$

where  $T$  is in millikelvin and  $R$  is in microohms. With  $R$  equal to 100 microohms one finds the bandwidth at 10 mK to be 30 Hz. With modern sophisticated electronics it should be possible to measure the line width to 0.1 Hz, so in principle one could evaluate the temperature at 10 mK to 0.3% and at 5 mK to 0.6%.

It is apparent from the foregoing that in the temperature range of greatest interest the noise thermometer becomes least sensitive. If it can be developed to the stage of a practical device, it could be used to standardize the CMN thermometer down to about 5 mK, which would be no mean accomplishment.

Probably of greater practical interest and promise is the nuclear resonance thermometer. The nuclear moments in a metal such as platinum follow Curie's Law to at least two orders of magnitude lower temperature than can be considered current working

temperatures. There is therefore no need to perform the laborious and difficult S-Q experiments to establish the validity of Curie's Law; the magnetic temperature calculated from the susceptibility is the thermodynamic temperature. Further, by making the measurement of susceptibility with resonance techniques one need not be concerned about extraneous fields or impurities adversely affecting the measurement.

To improve the accuracy one could make spin echo measurements. In contrast to the noise thermometer a nuclear resonance thermometer becomes more sensitive as the temperature is lowered. The major drawback is the lack of precision. If the amplitude of the echo is taken from an oscilloscope, the maximum precision is 1%. Possibly an improvement can be made with some signal averaging technique such as a boxcar integrator or multichannel analyzer. The overall accuracy will be limited by the constancy of the line shape.

My colleagues, Drs. John Ketterson and Pat Roach, are at present trying to develop the spin echo measurement in platinum as a practical thermometer. Platinum was chosen as the thermometric substance because of the short spin-lattice relaxation time,  $\tau_1 T = 30$  mK-sec, which is to be compared with 1300 mK-sec for copper. Even at 1 mK the relaxation time is only 30 sec, which is probably shorter than other relaxation times in an experimental apparatus.

It is appropriate at this time to mention a most ingenious device used by Professor Wheatley and collaborators<sup>11</sup> to measure the static nuclear susceptibility of copper. A superconducting fluxmeter\* was used to detect the change in nuclear magnetization. The Squid, as the device is called, served the same function as the mutual inductance coils around a pill of CMN. Since the device responds to flux changes, it has the same drawbacks as a conventional susceptibility apparatus. Though the coils may be astatic, insensitive to external fields with no sample present, the magnetization in the sample will change if the external field changes. The device will then register a response. However, that is a minor criticism and is not meant to detract from the utility.

As I understand the operation, a pair of astatic coils, one of which contains a small copper specimen are inserted into an open-ended niobium tube. A persistent current is induced in the tube which generates the magnetic field for nuclear alignment. The astatic pair wound from superconducting wire, are in series with another superconducting coil closely coupled to the Squid. The three coils constitute a superconducting transformer. A change in magnetization of the nuclei induces a persistent current in the superconducting transformer which changes the flux threading the Squid. Appropriate biasing and feedback circuitry keep the Squid locked on a particular oscillation of the V-H curve. The feedback voltage is a measure of the flux change, therefore of the susceptibility. The superconducting quantum interference device Squid is, in my opinion, one of the most promising developments for ultralow temperature thermometry in recent years. Because the Squid is not a resonance device, it suffers from the same interference drawbacks of any static susceptibility; however, if the Squid were used in conjunction with a resonance thermometer, it would make an excellent secondary thermometer.

One cannot terminate a discussion of thermometry without mentioning the two possible radioactive thermometers:  $\gamma$ -ray anisotropy and Mössbauer effect. The Mössbauer thermometer is in principle an absolute thermometer. If one measures the line spacing as well as the intensities, then the ratio of the counting rate for the two lines is  $\exp(-2m_g E/KT)$ , where  $m_g$  is the ground state spin and  $E$  is the separation between two successive levels.

\*Among the contributed papers of this symposium were two papers describing the use of superconducting magnetic flux detectors as thermometers, by R. A. Buhrman, W. P. Halpern, J. D. Reppy, R. C. Richardson, S. W. Schwerterly, and W. W. Webb, and by J. M. Goodkind and D. L. Stofa, submitted to the Review of Scientific Instruments.

The logarithm of the ratio leads to the absolute temperature immediately. In Table 1 are listed several isotopes that could be used for such a thermometer;<sup>12</sup> it can be seen that to cover a wide range of temperatures one would have to use at least two isotopes, with iron being the one for low temperatures. The problem here is the time necessary to develop the lines with reasonable precision. Further, the cryostat design becomes very complicated, since these measurements must be made with the absorber cold. One must then make provision for the radiation from a moving source to impinge on the absorber without first traversing a window. If one wanted to forego the absolute feature and use the Mössbauer effect as a secondary thermometer, very little is gained; the cryostat is still complicated. For either absolute or relative measurements there is an inherent error due to absorber thickness, illustrated in Fig. 5.<sup>12</sup> There seems little chance that this method has utility for low temperature research.

Table 1  
Properties of Isotopes Usable as Mössbauer Absorber  
Thermometers<sup>12</sup>

Isotope	Transition energy (keV)	Ground-state magnetic moment (nm)	Absorber material	Hyperfine field (kOe)	Splitting factor $2\mu_g H_i / k$ (K)	Spectrum
Np <sup>237</sup>	59.6	+2.70	NpAl <sub>2</sub>	3150	0.621	Resolved
Dy <sup>161</sup>	25.7	-0.472	Dy metal	7100	0.240	Resolved
Yb <sup>171</sup>	66.7	+0.493	YbCl <sub>3</sub> · 6H <sub>2</sub> O	2980	0.107	Resolved
Sb <sup>121</sup>	37.2	+3.359	MnSb	353	0.087	Complex
Eu <sup>151</sup>	21.6	+3.463	EuS	332	0.084	Complex
Ir <sup>193</sup>	73.0	+0.159	1% Ir in Fe	1495	0.017	Partly resolved
Sn <sup>119</sup>	23.9	-1.046	Sn ferrite	210	0.016	Resolved
Au <sup>197</sup>	77.3	+0.146	1% Au in Fe	1280	0.013	Partly resolved
Fe <sup>57</sup>	14.4	+0.090	FeF <sub>3</sub>	620	0.004	Resolved

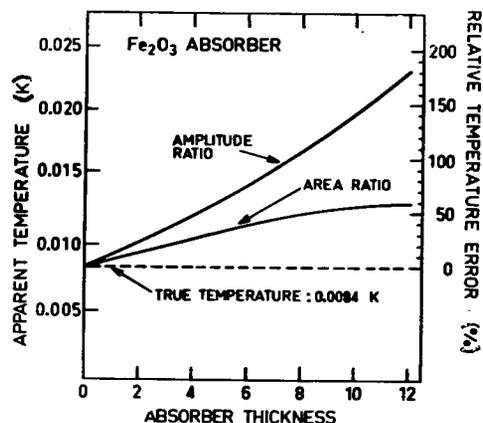


Fig. 5 - Error in a Mössbauer-effect thermometer due to absorber thickness

With regard to measurement of  $\gamma$ -ray anisotropy as a thermometer, the first comparison between magnetic temperature  $T^*$  and thermodynamic temperature  $T$  was made, to my knowledge, by Griffing and Wheatley<sup>13</sup> in 1956. They measured the anisotropy of  $\gamma$  rays from Co<sup>58</sup> nuclei in a copper Tutton salt host and concluded that the anisotropy

thermometer was of questionable validity. Frankel, Shirley, and Stone<sup>4</sup> some 9 years later used the  $\gamma$  rays from  $\text{Cm}^{137}$  to show that the  $T^*-T$  correlation for CMN as given by Daniels and Robinson<sup>1</sup> was in error. Both groups of investigators are probably correct; the former were expecting too great an accuracy for the method.

A detailed analysis of the technique is beyond the scope of this paper; but in brief the measurement is made as follows. A radioactive source, preferably with no  $\beta$  ray, is incorporated in a magnetic host or a paramagnetic host. The nuclei are oriented (magnetic host) or aligned (paramagnetic host) through the hyperfine interaction. If a paramagnetic host is used, then an external field is required. One then counts the  $\gamma$  rays at a fixed angle to the alignment axis. If the decay scheme and the *form* of the spin Hamiltonian are known, then the number of  $\gamma$  rays counted per unit solid angle will be given by

$$W(\theta, T) = 1 + A_2 B_2 P_2(\cos \theta) + A_4 B_4 P_4(\cos \theta),$$

where  $A_2$  and  $A_4$  are geometrical parameters associated with the counter,  $P_2(\cos \theta)$  and  $P_4(\cos \theta)$  are Legendre polynomials, and  $B_2$  and  $B_4$  are the temperature-dependent terms containing the coefficients of the Hamiltonian. The function  $W$  as written here has been normalized to unity at high temperature and  $\theta = 0$ .

Even with better than 1% counting statistics it appears doubtful from the published work<sup>4,13-15</sup> that the temperature can be obtained to better than 10%. Although Frankel *et al.*<sup>4</sup> were able to show that the Daniels and Robinson<sup>1</sup>  $T^*-T$  correlation for CMN was in error, they could not distinguish with certainty between their correlation and that of DeKlerk.<sup>3</sup> As a crude guide the anisotropy thermometer has some value.

To sum up, at the present time experimentalists have a reproducible, precise thermometer in CMN which is good to 6 mK. The major inaccuracy due to the shape can be eliminated by using a spherical geometry;  $\gamma$ -ray anisotropy is not sufficiently accurate to determine the shape correction. As experiments are pushed below 6 mK, the CMN scale is no longer adequate because of the undetermined  $T^*-T$  correction. The most promising standard thermometer is dynamic nuclear susceptibility; static nuclear susceptibility measurements made possible by the development of the fluxmeter has the greatest promise for a general working thermometer.

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## Thermal Contact and Isolation

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As lower temperatures are obtained in the laboratory, problems which were considered solved or negligible at higher temperature become more important. This paper deals with the problems of heat leak into, and thermal contact to, a substance in an experimental cell cooled to temperatures less than 0.3 K. Many of the methods and principles discussed are applicable to cells operating below 5 mK.

We will, in turn, investigate heat leaks due to radio and television emissions, mechanical vibrations, thermal radiation, and exchange gas. We will discuss insulating materials used in the construction of low temperature equipment, heat leaks down leads and fill lines, and thermal switches. Problems with thermal contact at low temperatures will be considered. This usually means a discussion of the Kapitza or boundary resistance. We will then discuss the problems of thermal contact to paramagnetic salts and dilution refrigerators, thermal contact within the experimental cell, and the thermal anchoring of fill lines and leads.

### RADIO-FREQUENCY HEATING

On many experimental cells, heaters, transducers, temperature-measuring resistors, or measuring devices are connected through transmission lines to electrical equipment at room temperature. These transmission lines couple with radiation from nearby radio, television, and FM stations, so that this coupling can lead to a major heat leak<sup>1</sup> into the cell, since the rf field strength in the laboratory can easily be of the order of volts per meter. Shielding the transmission lines with stainless steel tube in the cryostat and with coaxial cables at room temperature is difficult and often has little effect. Some workers<sup>2</sup> suggest using mica capacitors attached to the leads near the cell to provide thermal anchoring and to act as a short for the induced rf. Another method tried is to use lossy ferrite beads in strategic places along the transmission line. Both methods may or may not work for an individual station and at times serve to enhance the heat leak by somehow tuning the line. The usual solution is to take the low temperature data late at night when the offending stations are off the air.

An excellent but expensive solution is to build the whole cryostat in a shielded room. Care must be taken that the room is properly made and maintained. All seams in the room and especially around the door must be electrically closed. Electrical power lines into the room must be well filtered. Vacuum and service lines into the room must be insulated from the room as they pass through a wall. As these connections are made, the integrity of the room should be tested with either a commercial instrument or a portable FM radio. A well-made room provides at least 100 dB of isolation over the important frequency range. Working inside the room with sensitive electronics can be a pleasure, since even 60-Hz pickup is attenuated, and ground loops are easier to control. A shielded room is a good investment.

## VIBRATION

After the shielded room is in place, the problem of mechanical vibration can be tackled. Vibration is known to be a source of heat leak<sup>3</sup>. Some experimental mounts, refrigerators, and cells work better than others, but to predict or design a system guaranteed to give a low vibrational heat leak is difficult. However a few reasonable principles can be provided.

The cryostat should be built on the ground floor of the building, if a choice exists. A massive frame<sup>4</sup> mounted on air springs, steel springs, or even rubber should be made to have as low a natural frequency as possible, since the attenuation of the system varies as the square of the ratio of the natural resonant frequency divided by the transmitted frequency. Adjustable mechanical limits will prevent a "soft" system from rocking. No pumps or vibration sources should be mounted on the frame itself, and all pumping and gas lines should be isolated with rubber tubing, straight bellows, or bellows tees. Kurti once experienced vibration transmitted through the gas in a pumping line and suggests an external ballast when this problem occurs.

The next consideration is the refrigerator system inside the dewar. The pumping lines should be as strong and as rigid as possible. Although both rigid<sup>5</sup> and soft<sup>6</sup> refrigerators have been constructed having low heat leaks (of the order of ergs per minute) it is easier and more sure to make a good rigid system. This applies to both dilution and demagnetization refrigerators.

Although it is difficult to predict how a given cryostat will be affected by vibration, it is not difficult to test.<sup>3</sup> A transducer or variable-speed motor having an unbalanced load can be clamped to the cryostat. With the refrigerator operating, the heat leak can be measured as a function of frequency. Corrective measures could be taken if a large heat leak occurs at a particular frequency. It is a good idea to tune all ultralow temperature cryostats in this manner.

## EXCHANGE GAS

The use of He<sup>4</sup> exchange gas to cool from liquid nitrogen to liquid helium temperatures can cause problems, which experimenters have noted in the literature.<sup>3</sup> For example, Keyston et al.<sup>4</sup> had to worry about a changing heat leak due to adsorption and the heat capacity of the film while doing calorimetry on alloys, as also did Abel et al.<sup>5</sup>, working below 10 mK. Keyston et al. solved their problem by eliminating the exchange gas. They cooled their apparatus to 4 K using a heat exchanger mounted on the refrigerator and through which liquid helium from the bath was pumped. Abel et al. pumped the exchange gas away after calibrating a magnetic thermometer against the bath temperature. After the calibration, with the helium bath low, heaters on the refrigerator warmed it to between 4 and 10 K while the exchange gas was being pumped. The pumping continued until a low reading was obtained on a Veeco MS9A leak detector connected to the system. It is not known how much, if any, of the 2-erg/min heat leak in that experiment was due to using exchange gas. But if one is concerned with much smaller heat leaks, as surely more people will be in the next few years, it is probably better not to use exchange gas at all. A heat exchanger or mechanical thermal switch could be used in cooling. Another possibility is to use hydrogen or neon<sup>7</sup> as exchange gas. The transfer of liquid helium to the bath must be slow enough not to freeze out these gases prematurely. Since the neon will cool the apparatus to only about 20 K, further cooling is accomplished by circulating cold gas through the He<sup>4</sup> subpot, He<sup>3</sup> refrigerator, or dilution refrigerator.

If no exchange gas is used, the temperature calibration, if done to the bath, must be done after the experiment or done using a different type of thermometer such as He<sup>3</sup> vapor bulb or a calibrated semiconducting resistor.

## HEAT SHIELDS

The function of a heat shield is both to stop radiation from reaching the cell and to prevent heat transfer to the cell by "hot"<sup>8</sup> molecules. The shield usually protects the cell from the sides and bottom, protection from the top coming from a guard or some cold part of the refrigerator. It is assumed that the radiation from room temperature down tubes has been blocked by radiation traps. The radiation from surfaces at 4 K is calculated to be about 1 erg/min-cm<sup>2</sup>, so there is a definite need for a colder heat shield. A convenient place to mount the shield may be at the He<sup>3</sup> refrigerator, the still of a dilution refrigerator, or better yet one of the heat exchangers. The place at which the shield is thermally anchored must be capable of not only absorbing the radiation falling on the shield but also the heating produced in the shield by changing magnetic fields from temperature-measuring coils, rf coils, etc. Sometimes another shield is added,<sup>5,9</sup> thermally anchored to some very cold part of the refrigerator. This is a wise precaution, since until measurements are made the effectiveness of a given-design shield is uncertain.

Design of the shield is dictated by what devices act on the cell from outside the shield. An example is that a machined copper shield cannot be used in conjunction with temperature-measuring coils mounted outside the shield. We will discuss two types of shields which can be penetrated by the fields from measuring coils and demagnetizing magnets. D. O. Edwards has used a shield made from a tube of thin-wall stainless steel. The top of the tube is hard-soldered to a copper tube which is thermally anchored. The bottom is made of thin stainless steel and hard-soldered to the tube walls. Flattened copper wires are glued axially to the cylinder and thermally connected to the top copper tube.

Another design is that used by Wheatley<sup>5</sup> and made from coil-foil and epoxy. The shield is constructed by wrapping coil-foil lengthwise over a Teflon cylinder. One end of the coil-foil is cleaned and tied with an overlap to a thin slotted copper cylinder. All of the coil-foil was wrapped with string, and the assembly was mounted on a rotating spit inside an oven. The coil-foil was impregnated with Epibond 100A<sup>10</sup> epoxy. After curing at 150°C for at least 6 hours, the shield was removed from the Teflon form and machined to a wall thickness of about 1.5 mm. The shield and a bottom cap, made from the same material, were painted with a mixture of Dag<sup>11</sup> and varnish. A variation of the above procedure was developed by J. T. Tough to insure light-tight construction. As Epibond was added to the rotating shield, copper was also sprinkled over it. After curing, the shield was machined and painted with Dag.

The shields described above gave little phase shift when used in conjunction with measuring coils and a mutual inductance bridge. A good compromise is to operate a stainless shield at about 1 K and a coil-foil shield at some lower temperature.

## MATERIALS HAVING LOW THERMAL CONDUCTIVITIES

A number of materials having low thermal conductivities are commonly used in constructing a refrigerator. Some of these are listed in Table 1 along with their thermal conductivities and the temperature range over which they were measured. Reasonable approximations of the thermal conductivities at much lower temperatures can be made by extrapolation.

Table 1  
Materials Having Low Thermal Conductivities

Material	Thermal Conductivity (mW/cm-K <sup>power</sup> )	Measured Temperature Range (K)	Ref.
Stainless Steel 304-321-347	$0.45T + 0.14T^2$	1 - 4	12
70-30 Cupro-nickel	$0.93T^{1.23}$	0.3 - 4	13
AGOT Graphite	$0.49 \times 10^{-2} T^{1.86}$	0.3 - 3	14,15
Perspex and Plexiglass	$0.2T$	1 - 4	16
Teflon	$0.038T^{2.4}$	0.3 - 0.7	17
Nylon	$0.026T^{1.75}$	0.2 - 0.8	17
Epibond 100 A	$0.28T^{1.95}$	0.1 - 0.8	17

Thin-wall seamless stainless steel tubing is easily obtained, inexpensive, and available in many sizes. The tubing should be carefully checked for pinhole leaks before construction and after, especially if the tube has been bent or strained. Stainless can be easily hard-soldered and also soft-soldered with the proper flux. Cupro-nickel is more expensive, harder to get, and has a slightly higher thermal conductivity, but it is more easily bent, coiled, and soldered.

AGOT graphite is an extremely good insulator. It is obtained in block form and can easily be threaded and machined. Because of its small thermal contraction from room temperature, care must be taken when bonding it to other materials so that the connections do not loosen to become vibration problems. The same bonding problem exists for Teflon and nylon, except that they have large thermal contractions between room and helium temperatures.

Epoxy resins such as Epibond 100A or Araldites have the advantage that they can be cast into intricate shapes and are easily machined.

Some materials such as stainless steel and Cu-Ni are slightly magnetic and can cause problems when used improperly with sensitive magnetic thermometry. For more information about the magnetic properties of materials, see Salinger et al.<sup>18</sup>

#### HEAT LEAKS THROUGH FILL LINES

Many times the cell will contain liquid He<sup>3</sup>, He<sup>4</sup>, or mixtures of He<sup>3</sup> and He<sup>4</sup> as either a thermal contact agent or as the subject of the experiment. The fill line may be either empty, contain a refluxing film of He<sup>4</sup>, or be filled as when working under pressure.

If the cell contains liquid He<sup>3</sup> at its saturated vapor pressure, the fill line is empty and the heat leak is only that through the tube. When the line is filled with He<sup>3</sup>, conduction through the He<sup>3</sup> must be taken into account. Below 40 mK, the thermal conductivity of He<sup>3</sup> is  $K_3 = (50/T)$  erg/sec-cm<sup>19</sup>. If the cell and fill line is filled with He<sup>4</sup>, the added thermal conductivity of the He<sup>4</sup> can be calculated using the measurements of Fairbank

and Wilkes<sup>20</sup> and the calculations of Bertman and Kitchens.<sup>21</sup> If the cell and fill line contain a liquid mixture of He<sup>3</sup> and He<sup>4</sup>, the calculation for the He<sup>3</sup> fill line will apply at low temperatures.

A problem exists when the fill line is partially filled with He<sup>4</sup>, mixtures of He<sup>3</sup> or He<sup>4</sup>, or the superfluid film alone. The effective conductivity of the fill line can be greatly increased due to refluxing of the superfluid film. Vilches and Wheatley<sup>19</sup> describe a satisfactory solution to the problem. Thermal isolation is attained through the use of long fill tubes wrapped in the form of spirals. One such arrangement for use with a demagnetization system contains an isolation spiral of 0.4-mm-O.D. by 0.08-mm-wall tubing 60 cm long between the demagnetized cell and the guard. Another spiral of the same tubing but 200 cm long isolates the guard from the He<sup>3</sup> refrigerator. A spiral 400 cm long isolates the He<sup>3</sup> refrigerator from the pumped bath. The fill line is thermally anchored to the guard with 40 cm of tubing bonded to coil-foil and to the He<sup>3</sup> refrigerator with 40 cm of tubing soldered to the He<sup>3</sup> pot. The bath anchor is 60 cm of 2.1-mm-O.D. by 0.79-mm-I.D. copper capillary.

The isolation spirals are made from 70-30 Cu-Ni tube and fabricated by winding the tube and a wire together over three strips of cloth laid on and parallel to the axis of a Teflon cylinder. The tubing is tied down, and the spacing wire is removed. The assembly is placed in an oven on a rotating spit. Epibond 100A is sprinkled over the cloth and cured.

The principle of using long enough spirals, thermally anchored where necessary, allows the use of He<sup>3</sup>-He<sup>4</sup> mixtures as a thermal contact agent down to temperatures of the order 2 mK.

#### HEAT LEAKS DOWN LEADS

A number of different materials have been used as leads to the low temperature parts of a system. Manganin, lead-coated manganin or constantan, superconductors, and copper will be discussed.

Manganin has a fairly high electrical resistance from which the thermal conductivity can be calculated to be  $5.5 \times 10^3 T$  erg/sec-cm-K<sup>2</sup>.<sup>17</sup> It is strong and easily soldered. Small-diameter manganin and constantan wire can be obtained with a lead coating. The thermal conductivity is low, and the electrical resistance is zero, at least when the wire is new and has not been handled. After handling, breaks may appear in the lead coating, causing the wire to become unreliable for some applications. Anderson and Bloom<sup>22</sup> have described a method to improve the situation. Two or three strands of cleaned 0.0076-cm-diameter wire is twisted, fluxed, and drawn through a molten bead of 50-50 Pb-Sn solder. Three-wire cables made in this manner can be expected to have a critical current of at least 2 amperes even after severe twisting and bending.

Superconductors such as niobium, Nb-25%Zr, or Nb-48%Ti may be used as leads. They have low thermal conductivities and zero electrical resistance but unfortunately cannot be soft-soldered and are somewhat difficult to plate with copper. An alternative is to buy copper-plated wire and remove the copper in the isolation region with acid. It is found that even commercial copper-plated Nb-Zr has a resistance of about  $10^{-2}$  to  $10^{-3}$  ohms<sup>22</sup> between 1 cm of copper plate and the superconducting wire. Newer types of wire, perhaps Nb-Ti, and better bonding techniques should overcome the problem.

If necessary, small-diameter copper wire can be used as leads having low heat leaks. Insulated copper wire with diameters less than 0.001 mm are available. Even with a high conductivity of about  $2 \times 10^7 T$  erg/cm-sec/K<sup>2</sup>, wires of reasonable length can be used if the necessary thermal anchor points are available. Unfortunately this ultrafine wire is difficult to see, much less work with, and it is easily broken.

Special transmission lines, such as high-voltage capacitor leads or shielded transducer leads may be made in the form of isolation spirals, with the center conductor insulated from the tube with thin-wall Teflon tubing.

### SUPERCONDUCTING THERMAL SWITCHES

Dilution refrigerators and demagnetized-salt refrigerators commonly operate down to between 10 and 20 mK. To reach lower temperatures, another stage of cooling must be added. This stage may be connected to the one above with a superconducting thermal switch in the form of a wire or foil and placed so that the heat flow is along the lines of flux of the activating magnetic field.

Practical switches are made from high-purity (better than 99.999%) material left to anneal at room temperature so as to be reproducible. The normal-state conductivity is field dependent,<sup>23</sup> decreasing with increasing field. Typical switch sizes are of order of 20 mm long, 2 mm wide, and 0.2 mm thick. For pure stock material in low fields  $K_n$  should not vary by more than a factor of 2 from those listed in Table 2. All the materials listed in Table 2 have large switching ratios below 0.1 K and may be used at ultra low temperatures.

Table 2  
Superconducting Switches

Material	$T^2 K_n / K_s$ ( $K^{-2}$ )	$K_n / T$ (W/cm-K)	State	Ref.
Pb	45-50	6.2	Annealed	23,24
In	22	5.5	Annealed	25
Sn	100	3 (est.)	Annealed	9
Zn	6	1	Single crystal	26

### THERMAL CONTACT AT LOW TEMPERATURES

Boundary resistance is the thermal resistance at the interface between two substances. Boundary resistance is usually either measured or forced to fit a  $T^{-3}$  temperature dependence. One exception is the thermal resistance between pure He<sup>3</sup> and powdered cerous magnesium nitrate (CMN), which is very small and proportional to  $T$  at low temperatures.<sup>27</sup> Some useful values of boundary resistance are given in Table 3.

The problem of overcoming the boundary resistance is solved by the brute-force method of increasing the contact area. This means using sintered copper, copper wire brushes and powdered salts and metals.

### CONTACT TO THE SALTS OF A PARAMAGNETIC REFRIGERATOR

Many different methods have been used by experimenters<sup>4,6,19</sup> to make<sup>28,29,30</sup> contact to the salts of a paramagnetic refrigerator, operating down to about 15 mK. Here

Table 3  
Boundary Resistances  $R_B$

Pair of Substances	$R_B T^3 \times 10^6$ (sec-cm <sup>2</sup> -K <sup>4</sup> /erg)	Temperature Range of Measurement (K)	Ref.
He <sup>3</sup> - electropolished copper	20	0.06 - 0.10	19
He <sup>3</sup> -He <sup>4</sup> saturated solution - copper	7	< 0.1	19
He <sup>4</sup> - copper	$4 \times T^{-0.4}$	0.07 - 0.15	19
Coil-foil-CrK crystals using Apiezon "N" grease	4.2	0.03 - 0.2	19
He <sup>3</sup> - Formex-coated copper wires	7	0.025 - 0.04	19
He <sup>3</sup> - Epibond 100A	3	0.015 - 0.1	19
CrK and FeNH <sub>4</sub> single crystals - He <sup>4</sup>	3	0.2 - 0.4	19
Anything and almost anything else without electronic contact	10	< 0.2	-

only a few will be considered. The method to first be considered was used by Wheatley<sup>5</sup> on the guard and first stage of a highly successful two-stage refrigerator. Single crystals of CrK are cut into thin slabs ( $\approx 3$  mm thick) and greased between layers of coil-foil. The sandwich of about six layers with an insulating center plate of machined Epibond 100A is then tied tightly with string. Some coil-foil is brought out the top to make contact to the stage above, but most are brought out the bottom to cool the stage below. The coil-foil serves to transfer heat into and out of the salts with a minimum of eddy current heating on demagnetization. The support may be threaded on each end so as to connect to cylindrical nylon interstage insulators.

Contact between a slurry of crystals in Apiezon "J" oil<sup>28</sup> or glycerol<sup>9</sup> and either a brush of copper wires or coil-foil have also been used. A thermal contact agent used more recently by Vilches and Wheatley<sup>19</sup> is liquid He<sup>4</sup>. Liquid He<sup>4</sup> provides the thermal contact between the small crystals of salt to be demagnetized and a powder of the substance to be cooled.

The methods mentioned above and many others not mentioned have worked well, but the technique of cooling by demagnetization to 15 mK is rapidly being replaced by dilution refrigeration, which brings about some new problems in thermal contact.

#### THERMAL CONTACT TO A DILUTION REFRIGERATOR

Tremendous refrigeration exists in the mixing chamber (MC) of a dilution refrigerator. The problem is to make this refrigeration available to cool an experimental cell. Two options exist: The material to be cooled may be placed either inside the mixing chamber

or external to it. First consider material placed inside the mixing chamber as suggested by Wheatley et al.<sup>31</sup> In his design the MC is mounted on an insulating support some distance from the heat exchangers. The MC is nonmagnetic, cast from Epibond 100A, and contains a CMN thermometer. Experiments on solids, on pure He<sup>3</sup> in the upper phase, and on the 6.4% mixture in the lower phase can easily be performed.

If the cell is external to the refrigerator, some method of thermal contact to the MC must be found. Mota et al.<sup>32</sup> have a suggestion for this type of design. The MC is still Epibond but now contains a number of bundles of copper wires, each bundle being hard-soldered to a 0.51-mm-diameter copper wire to which the experiment may be attached. The bundles are sealed inside the MC and provide about 500 cm<sup>2</sup> of contact area to the liquid inside. The device is operable, but Wheatley suggests that an increase of about a factor of 5 in contact area would be needed to fully utilize the refrigeration of the dilution refrigerator. Sintered copper is purposely not used in this design, since it would cause problems with thermometry and eddy current heating.

Where enough working space exists, a copper MC can be used. For powder less than 44  $\mu$ m in size, 1 cm<sup>3</sup> of sintered copper has a surface area of about 1000 cm<sup>2</sup>, so that thermal contact can easily be made to the mixing chamber with only a few cm<sup>3</sup> of sintered copper. Now the problem becomes one of making contact to the MC. One solution is to use a solderless thermal connection as described by Boughton et al.<sup>33</sup> The MC is machined with a 1-in.-long by 1/4-in.-diameter cylinder on its bottom. The mating female piece is slotted along the axis and has a snug fit to the MC. A cylindrical nylon pressure ring fits over the female. This nylon clamp may be slotted and a bolt used to increase the pressure on the mated copper pieces at room temperature. Both the male and female pieces are gold plated to keep them free of oxide. The thermal contact of this type of system is equivalent to a hard-solder joint. Suomi et al.<sup>28</sup> suggest a simple solution to the contact problem. They found that a copper bolt-nut threaded contact, where the fit is snug enough to clean the oxide, made good thermal contact. So for thermal contact a threaded screw is made as an integral part of the mixing chamber and the nut an integral part of the contact to the cell.

Another method would be to hard-solder or TIG-weld heavy copper wires to the MC and to solder the wires from the cell to it. Solders for thermal contact were investigated by both Steyert<sup>34</sup> and Suomi et al.<sup>28</sup> Steyert found that eutectic Cd-Bi solder in a gap of 70  $\mu$ m and 50-50 Pb-Sn solder in a gap of 10  $\mu$ m obeyed the equation  $\dot{Q}/(\Delta TA) = 4.7 \times 10^6 T^{2.74}$  erg/sec-cm<sup>2</sup>-K for temperatures between 60 and 600 mK. Copper-indium-copper contacts were investigated by Suomi et al.<sup>27</sup> with the following results. Two copper pieces were soldered with a 0.06-cm layer of indium between them. When the indium had not been exposed to magnetic fields so as to trap flux, the contacts exhibited the expected boundary resistance. After the indium was transformed to the normal state, the measured thermal resistance of the contact was due entirely to the copper parts. Other samples were investigated with a much smaller gap. Two machined copper pieces were pressed together, fluxed, and soldered with indium by capillary action. In the virgin state the contacts may have finite resistance, but when enough direct copper-copper contact existed, the resistance would be very small, with the indium serving only to keep the contact areas clean. Once the junction was exposed to a magnetic field, in all cases the thermal resistance became small at all fields.

Experience has shown that soldering near the refrigerator and in screened rooms should be done only when necessary. Since accidents cannot be avoided, at least they can be minimized; it is discouraging to see a nice shiny copper dilution refrigerator turn green or white, depending on the soldering flux used.

## THERMAL CONTACT IN A CELL

With regard to thermal contact in a cell, the first case to be considered is that of the dilution refrigerator or paramagnetic refrigerator in the final stage of cooling. If the sample is bulk metal, contact can be made between it and the refrigerator, or between it and a superconducting switch to the refrigerator, by first cleaning the oxide from the sample and the copper to which it is to be joined, while they are covered with a thin coating of liquid In-Hg solution.<sup>35</sup> The pieces are then mated and held together with thread. It has been found that this method allows electronic thermal contact between the metals. If the sample is not allowed to be treated in the manner mentioned above, perhaps a powdered sample could be used, and thermal contact could be made using liquid helium. Contact between the refrigerator and the liquid helium may be made by using a brush of copper wires hard-soldered to the copper piece which makes contact to the refrigerator. The use of a brush rather than sintered copper is necessary to reduce eddy-current heating, if the cell is in a varying magnetic field. If the sample cannot be powdered to increase its contact area to the helium, it must just be used as it is. Care must now be taken so that no heating inside the metal will take place, since the metal-helium boundary resistance will be large. A powdered CMN thermometer, also in the cell, can be used to measure temperature.

A complete discussion of using liquid helium for thermal contact has been given by Vilches and Wheatley.<sup>19</sup> The liquid helium in the cell may be He<sup>4</sup>, He<sup>3</sup>, or a mixture of He<sup>3</sup> and He<sup>4</sup>. First a system of pure He<sup>4</sup> and CMN powder (<0.2 mm) will be considered. The thermal resistance of a column of liquid He<sup>4</sup> of length  $\ell$ , area A, and diameter d is given by<sup>19</sup>

$$\frac{\Delta T}{\dot{Q}} = \frac{3 \times 10^{-9} \left(\frac{\ell}{d}\right)}{AT^3} \text{ sec-cm}^2\text{-K}^4/\text{erg}$$

when the phonon mean free path is larger than d. Since the CMN heat capacity ( $0.44/T^2$  erg-K/g)<sup>19</sup> varies as the inverse square of the temperature, the column-CMN time constant varies as the inverse fifth power of the temperature. For 1 gram of CMN and a column 5 cm long and 1 cm in diameter the time constant at 10 mK is about 100 sec. With a smaller diameter column and lower temperatures the column becomes a good insulator and may find use as a thermal switch. This method of using He<sup>4</sup> has been used up to about 0.7 K and as low as 13 mK.<sup>19</sup>

Pure He<sup>3</sup> will next be considered as a contact agent. Although the heat capacity of He<sup>4</sup> was neglected, the same certainly cannot be done for He<sup>3</sup>, so that in experiments where a large heat capacity in the cell would cause problems, He<sup>3</sup> may be of limited value. This is especially true where cooling is attained using a paramagnetic refrigerator. He<sup>3</sup> has two main advantages, its low boundary resistance to CMN powder and its high thermal conductivity ( $50/T$  erg/sec-cm) below 40 mK. In fact the conductivities of He<sup>3</sup> and copper are equal at about 2 mK.

Mixtures of He<sup>3</sup> in He<sup>4</sup> at large  $X = n_3/(n_3 + n_4) \approx 6.4\%$  are also good conductors of heat (roughly<sup>31</sup>  $30T$  erg/sec-cm-K<sup>2</sup> at 6.4%, decreasing roughly as X) below 15 mK. The mixtures also exhibit high heat capacities and can be used in a manner similar to He<sup>3</sup>.

When using these liquids as contact agents the important considerations are to have large contact areas to overcome boundary resistance, to be careful in design so as not to have long time constants, and to be careful of thermal gradients caused by heat leaks.

The cell filled with liquid helium, CMN, or the proper metal powder can be connected to the refrigerator through a superconducting switch and demagnetized. The problems

are now not too different, only accentuated. The tolerable heat leaks are smaller, about 1.0 erg/min, when demagnetizing CMN to 2 mK and less than 0.1 erg/min when demagnetizing metals. The demagnetization itself must be slow to minimize eddy-current heating and to be reversible, since all time constants become larger as lower temperatures are reached. One would get old quickly working below 1 mK.

#### THERMAL CONTACT OF FILL LINES AND LEADS

The Cu-Ni fill lines mentioned earlier can be thermally anchored to salts by soldering to coil-foil or copper and then greasing and wrapping with thread to the refrigerator and guard salts. The fill line may be soldered to the He<sup>3</sup> refrigerator pot (a length of about 50 cm) and the He<sup>4</sup> pot or bath (a length of about 100 cm). The lines may be placed inside the He<sup>3</sup> and He<sup>4</sup> pots, but this increases the number of solder connections. The fill lines may be connected to a sintered copper plug and anchored with either a pressure or screw contact. (The dead volume of these pieces may be important in some experiments.) When a dilution refrigerator is used, the same type of thermal anchors may be made to the still, selected heat exchangers, and mixing chamber as were made to the He<sup>3</sup> and He<sup>4</sup> pots mentioned above. Coaxial transmission lines may be treated in the same manner as fill lines except that at the anchor points they should be broken to allow the center conduction to be anchored by gluing.

Leads may be thermally anchored at temperatures above 0.3 K by casting wires to copper with epoxy and greasing, gluing, or bolting the copper to the cold sections. At lower temperatures one must be more careful to put enough wire area in contact with the cold area with only a thin insulating layer of Mylar, paper, or epoxy between. The thermal resistance can be estimated from the boundary resistance data given above. Anderson<sup>36</sup> suggests a technique for anchoring leads. Cigarette paper was glued with GE 7031-toluene to a thin copper foil and dried under a desk lamp. More 7031-toluene was added, and the foil was folded, sandwiching the wires. Then the package was clamped with an electric clip until dry. The foil was either indium-soldered or bolted to a copper heat sink. The thermal resistance was measured to be

$$R = \frac{1.1 \times 10^{-4}}{T^3} + \frac{7 \times 10^{-4}}{T} \left( \frac{\text{K-sec}}{\text{erg}} \right)$$

for a 1-cm length of anchored wire over the temperature range 0.05 to 5 K.

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## **Invited Papers**

### **SESSION III**

Friday Morning, April 24, 1970

Chairman: W. W. Webb  
Cornell University



## Theory of He<sup>3</sup>-He<sup>4</sup> Dilution Refrigerators

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The historical development of the dilution refrigerator was discussed in detail by David Edwards, and so the introduction for this present paper can be kept short. The theory of the dilution process has played a significant role in the evolution of present-day dilution refrigerators, particularly if one recalls that H. London's original proposal<sup>1</sup> for the process in 1951 was based on the theory<sup>2</sup> of Fermi liquids and preceded by 13 years the first refrigerator of Das et al.<sup>3</sup> London's second proposal,<sup>4</sup> which used the discovery<sup>5</sup> a few years earlier of phase separation in He<sup>3</sup>-He<sup>4</sup> solutions, was very explicit regarding the scheme for carrying out the continuous dilution process and led directly to the first refrigerator of Das et al. 2 years later. At the time of the first really successful refrigerators,<sup>6,7</sup> however, the theory was limited to mostly qualitative ideas, and the few quantitative predictions had not taken into account degenerate effects which occur in the dilute He<sup>3</sup> solutions below a few tenths kelvin. The theoretical limitations of the refrigerator were not known.

Prodded by the sudden interest in the refrigerators the theory of the device as well as practical improvements advanced rapidly after 1966. Several publications on the theory have appeared recently in which degenerate effects in the solutions are taken into account. The first of these<sup>8-10</sup> neglected He<sup>3</sup>-He<sup>3</sup> interactions, whereas later papers<sup>11-14</sup> took these weak interactions into account and appear to give quite accurate results for the thermodynamic behavior of the refrigerators. We now can safely say that the general thermodynamic behavior of the refrigerators is well understood, quite unlike the black-magic days a few years ago when the refrigerators were first coming into being. Theoretical advances beyond this point are more for design information than for a basic understanding of the refrigeration cycle.

In this paper we will discuss the theory of the dilution refrigerator that has evolved in the last few years and in addition we will discuss some of the latest results from our laboratory on the theoretical behavior of the heat exchangers. The heat exchanger behavior has been known in principle for some time, but detailed quantitative calculations have never been done to help bring about the design of optimum heat exchangers. This work is still in progress, but we will point out what we have learned to date about the heat exchangers.

### PROPERTIES OF He<sup>3</sup>-He<sup>4</sup> SOLUTIONS

A description of the refrigerator naturally depends on the properties of the working fluid, so first we shall consider some of these properties. Figure 1 is the phase diagram of He<sup>3</sup>-He<sup>4</sup> solutions and shows that phase separation takes place below about 0.86K as first seen by Walters and Fairbanks<sup>5</sup>. If a mixture of He<sup>3</sup> and He<sup>4</sup> containing a molar fraction X of He<sup>3</sup> is cooled below the phase separation line, the solution separates into

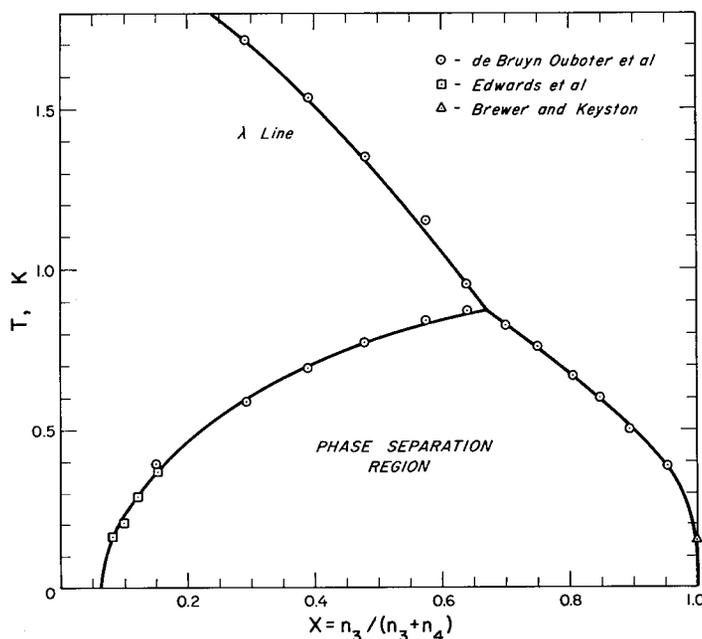


Fig. 1 - Phase diagram of He<sup>3</sup>-He<sup>4</sup> Solutions. The points are from several authors<sup>15,17</sup>, and the behavior at the intersection of the  $\lambda$  and phase-separation lines is from the work of Graf et al.<sup>18</sup>

two phases. The concentrated phase (concentration  $X_u$ ) floats on top of the dilute phase (concentration  $X_\ell$ ). The two phases are in equilibrium with each other just as in the case of a liquid and its vapor. Thus the He<sup>3</sup> chemical potential  $\mu_3$  must be equal in both phases. The He<sup>4</sup> in the dilute phase is superfluid below the  $\lambda$  line, whereas in the concentrated phase it is not. In the dilute phase, then, the relation

$$\nabla\mu_4 = 0 \quad (1)$$

must hold<sup>12</sup> in equilibrium, where  $\mu_4$  is the He<sup>4</sup> chemical potential in solution.

In addition to the data points shown in Fig. 1 there are extensive measurements of the dilute-phase separation line by Ifft et al.<sup>19</sup> using dielectric measurements, and later by Schermer et al.<sup>20</sup> using neutron transmission. Unfortunately the two results are not in perfect agreement. The limiting solubility  $X_0$  of He<sup>3</sup> in He<sup>4</sup> at 0 K is  $0.0640 \pm 0.007$  from the former and  $0.0684 \pm 0.006$  from the latter. Very recently other dielectric measurements<sup>21</sup> and osmotic-pressure measurements<sup>22</sup> give a value of 0.0660 for  $X_0$ . Accurate numerical results for the behavior of the refrigerator rely on a measured value for  $X_0$ . All the results reported here are based on the figure 0.0640, simply because only that figure was available when the calculations<sup>11</sup> were made. A different value for  $X_0$  would alter primarily the heat absorption rate of the refrigerator and will be discussed later.

It is apparent from the phase diagram that at temperatures applicable to the dilution refrigerator dilute He<sup>3</sup> and nearly pure He<sup>3</sup> are the only solutions possible. Properties of pure He<sup>3</sup> necessary for describing the refrigerator have all been measured quite accurately to temperatures as low as a few millikelvin. However, little is known about the properties of concentrated solutions below 0.4 K, i.e., solutions in the range 80 to 95% He<sup>3</sup>.

These are the concentrations usually circulated in actual refrigerators, and the specific heat just below the phase-separation temperature can be considerably higher than pure He<sup>3</sup>. This places an uncertainty in the behavior of the higher temperature part of the refrigerator such as some of the heat exchangers. Several measurements have been made on various properties of dilute He<sup>3</sup> solutions, but, because of the added dimension of concentration, nowhere near enough data exist to completely map out from the data alone the properties as a function of both temperature and concentration.

Fortunately a rather simple and accurate theoretical model exists for the dilute solutions which can map out these properties after a few parameters are determined from the existing data. This theoretical model, first proposed by Landau and Pomeranchuk,<sup>3</sup> is that an atom of He<sup>3</sup> added to superfluid He<sup>4</sup> should behave as an impurity with an ideal-gaslike energy spectrum of

$$E = E_0 + p^2/2m_0^* \quad (2)$$

where  $E_0$  is the binding energy of the He<sup>3</sup> atom to the He<sup>4</sup> bath,  $p$  is the He<sup>3</sup> quasiparticle momentum, and  $m_0^*$  is the effective mass of the quasiparticle. As more He<sup>3</sup> atoms are added to the He<sup>4</sup> bath, the binding energy and the effective mass of each quasiparticle begin to change slightly under the influence of the field set up by the He<sup>3</sup> already present. In addition a term of higher order than  $p^2$  will contribute to the energy in Eq. (2) and is due to exchange scattering of the He<sup>3</sup> atoms. In a somewhat more physical picture what has been done here is to replace each He<sup>3</sup> atom with a fictitious particle known as the He<sup>3</sup> quasiparticle. The He<sup>3</sup> atom actually interacts strongly with the He<sup>4</sup>, whereas the quasiparticle is now a particle interacting only with other He<sup>3</sup> quasiparticles. The effect of the He<sup>3</sup>-He<sup>4</sup> interaction is now contained in  $m_0^*$  and  $E$ , which are different than the bare He<sup>3</sup> atomic mass  $m_3$  and energy. Thus at low temperatures, at which the number of He<sup>4</sup> phonons is extremely small, we can disregard the He<sup>4</sup>. This is an especially attractive model for use with the dilution refrigerator, since in it He<sup>3</sup> atoms diffuse through a nearly stationary He<sup>4</sup> bath. According to the theoretical model, then, we replace the He<sup>3</sup> atoms by He<sup>3</sup> quasiparticles and dispense with the He<sup>4</sup> or replace it by a vacuum or "ether."<sup>12</sup> What we have is simple flow of the gaslike free quasiparticles at a pressure  $\Pi$ , which is actually the osmotic pressure of He<sup>3</sup> in He<sup>4</sup>. A knowledge of the properties of this gas of He<sup>3</sup> quasiparticles would then allow us to analyze the refrigeration cycle.

The earliest attempts to calculate the thermodynamic properties of dilute He<sup>3</sup> solutions were to treat the He<sup>3</sup> quasiparticle gas as an ideal classical gas, which would have a specific heat at constant concentration or constant volume of  $C_v = 3R/2$  and at constant pressure of  $C_p = 5R/2$ . These results are valid only for temperatures much higher than the Fermi temperature, given by

$$T_f = \hbar^2(3\pi^2N_A/V)^{2/3}/2k_B m^* \quad (3)$$

where  $\hbar$  is the reduced Planck's constant,  $N_A$  is Avogadro's number,  $V$  is the molar volume of He<sup>3</sup> in solution, and  $k_B$  is Boltzmann's constant. The behavior of  $T_f$  is roughly as  $X^{2/3}$  and is about 0.38 K at  $X = 6.4\%$ . Obviously, degenerate effects must be taken into account for temperatures reached in the dilution refrigerator. The first approximation would neglect any interactions between He<sup>3</sup> quasiparticles. Such an approximation comes fairly close to experimental results for several properties.

The best accuracy and internally consistent results occur only when weak interactions between He<sup>3</sup> quasiparticles are taken into account. Derivation of the effective mass and total energy of the quasiparticles from theory alone requires a microscopic theory, but such theories to date are not sufficiently accurate. Instead we will use a phenomenological theory by Bardeen, Baym, and Pines<sup>23</sup> (BBP) to predict the variation of these two

quantities with concentration and rely on existing experiments to determine the absolute values. BBP first suggested the interaction potential

$$V(k) = V_0 \cos(\beta k) \quad (4)$$

to fit spin diffusion results<sup>24</sup>, with  $\beta = 3.16 \text{ \AA}$  and  $V_0 = -0.0754 \text{ m}_4 s^2/n_4 = 1.303 \times 10^{-45} \text{ joules-cm}^3$ , where  $m_4$  is the mass of the  $\text{He}^4$  atom,  $s$  is the velocity of first sound in  $\text{He}^4$  at  $T = 0$ , and  $n_4$  is the number density of pure  $\text{He}^4$  at  $T = 0$ . Ebner<sup>25</sup> devised a slightly different potential which should be better for higher concentrations. The form of his potential is much more complicated than Eq. (4) and is not displayed here. All the thermodynamic properties described in this paper, and in much more detail in an earlier work,<sup>11</sup> are based on Ebner's potential. Improvements in  $V(k)$  are still needed to completely explain some of the latest heat-of-mixing<sup>26</sup> and osmotic-pressure<sup>27</sup> measurements. The interaction potential has a reasonably large influence in the low temperature limit on such properties of the  $\text{He}^3$  quasiparticles as the energy and osmotic pressure. It has a much less effect on the effective mass, but it is responsible for the variation of effective mass with concentration. The interaction has no effect on the specific heat and entropy, provided the proper effective mass is used.

Given an interaction potential  $V(k)$  the variation of the specific-heat effective mass with concentration is expressed as<sup>23</sup>

$$\frac{m^*}{m_0^*} = 1 + \frac{1}{3} F_1^s = 1 + \frac{N(0)}{2k_f^2} \int_0^{2k_f} V(k)k \left(1 - \frac{k^2}{2k_f^2}\right) dk, \quad (5)$$

where  $m_0^*$  is the value of  $m^*$  at zero concentration,

$$N(0) = m^*k_f/2\pi^2k^2 \quad (6)$$

is the density of energy states at the Fermi surface for one spin orientation, and  $k_f$  is the magnitude of the Fermi wave vector. The behavior of  $V(k)$  is such that  $F_1^s$  is always positive for concentrations dealt with here. The dependence of  $k_f$  on concentration is shown by the relation

$$k_f = (3\pi^2n_3)^{1/3} = (3\pi^2N_A/v)^{1/3}, \quad (7)$$

where  $n_3$  is the number density of  $\text{He}^3$  atoms in solution, and where  $v$ , the volume of solution containing one mole of  $\text{He}^3$ , is expressed by

$$v = V_m/X = 27.58/X + 7.60 + 1.65X^2 \text{ cm}^3/\text{mole He}^3. \quad (8)$$

In Eq. (8)  $V_m$  is the molar volume of the total solution. Equation (8) fits experimental data<sup>19,28</sup> to within 0.3% for concentrations less than  $X = 0.3$  and all temperatures less than 1.5 K. The solution to Eq. (5) in the form  $m^*/m_3$  is shown in Fig. 2 for both the BBP and Ebner potential. The value of  $m^*/m_3 = 2.34$  was used to give the best fit to the values of  $m^*/m_3$  derived from specific-heat measurements<sup>15,24</sup> and shown in Fig. 2. In addition these specific-heat measurements showed that the specific heat of the dilute solutions was just that calculated by Stoner<sup>29</sup> for an ideal Fermi gas all the way from the highly degenerate ( $C_3 \propto Tm^*/m_3$ ) region to the nondegenerate ( $C_3 = 3R/2$ ) region.

The effect of the  $\text{He}^3\text{-He}^3$  interaction on the energy and osmotic pressure is best evaluated through the chemical potential. We make use of the fact that the concentrated

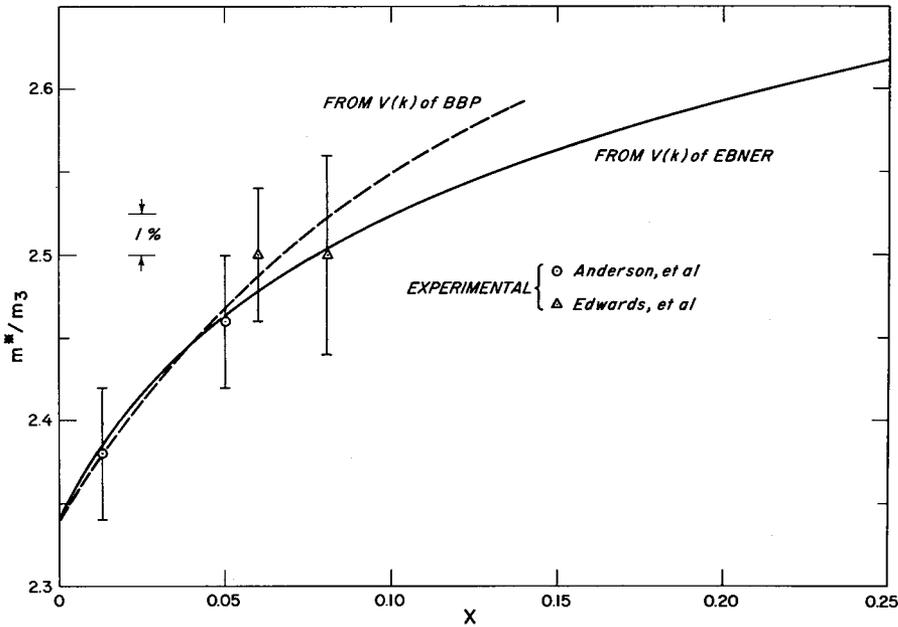


Fig. 2 - Effective mass ratio of He<sup>3</sup> in He<sup>4</sup> as a function of concentration

and dilute phases in a phase-separated solution are in equilibrium with each other, which implies

$$\mu_3(X_\ell, T) = L_3^0 + \mu_3^0(T), \quad (9)$$

where the term on the left is the chemical potential of He<sup>3</sup> along the lower phase-separation line and the terms on the right represent the chemical potential of He<sup>3</sup> in the concentrated phase, assumed to be pure He<sup>3</sup>. The term  $L_3^0$  is simply the heat of vaporization of pure He<sup>3</sup> at  $T = 0$ , and the chemical potential term  $\mu_3^0$  is given by

$$\mu_3^0 = H_3^0 - TS_3^0 = \int_0^T C_3^0 dT - T \int_0^T (C_3^0/T) dT, \quad (10)$$

where  $H_3^0$  and  $S_3^0$  are the enthalpy and the entropy respectively of pure He<sup>3</sup>. The specific heat  $C_3^0$  of pure He<sup>3</sup> has been evaluated<sup>11</sup> from all the experimental data and does not follow the simple Fermi gas model as for dilute He<sup>3</sup> solutions. The first term in a power-series fit of  $C_3^0$  to the low temperature region is 25.3 joules/mole-K, but this expression is good for only the very roughest of approximations at finite temperatures, since even at 50 mK the value of  $C_3^0/T$  has already dropped to 21.6 joules/mole-K<sup>2</sup>. The right side of Eq. (9) is easily evaluated from experimental data, which then gives us experimental results for  $\mu_3$ , but only along the phase separation line. Recent measurements<sup>27</sup> of the osmotic pressure at very low temperatures permit calculation of  $\mu_3$  away from the phase separation line.

However the BBP phenomenological theory has been used here to find the variation of  $\mu_3$  with concentration. We can express  $\mu_3$  as

$$\mu_3(X, T) = \mu_f(X, T) + \mu_3^f(X), \quad (11)$$

where  $\mu_f$  is the chemical potential of an ideal Fermi gas and  $\mu'_3$  is the deviation from this ideal behavior. The theoretical expression given by BBP for  $\mu'_3$  (strictly valid only for  $T = 0$  K but reasonably good for finite temperatures) is

$$\mu'_3(X) = E_0 + (N_A^2/v) V(0) - \frac{N_A}{4\pi^2} \int_0^{2k_f} k^2 \left(1 - \frac{k}{2k_f}\right) V(k) dk \quad (12)$$

where  $E_0$  is the binding energy of a single  $\text{He}^3$  quasiparticle to the superfluid  $\text{He}^4$  bath. The second term on the right is a correction to the binding energy due to the average  $\text{He}^3$ - $\text{He}^4$  quasiparticle interaction energy, and the third term is a result of exchange scattering of particles of like spin. The solution to Eq. (12) in the form of  $(-\mu'_3 - L_3^0)/R$  with Ebner's potential is shown in Fig. 3 along with the Fermi temperature  $T_f$ . The intersection with the  $X = 0$  axis gives  $E_0$  and is adjusted until the theoretical curve best fits the experimental points shown in Fig. 3. Such a fit gives  $(-E_0 - L_3^0)/R = 0.259\text{K}$ . The recent measurements of the heat of mixing<sup>26</sup> and osmotic pressure<sup>27</sup> give an experimental curve which has an intercept of  $(-E_0 - L_3^0)/R = 0.312 \pm 0.007\text{K}$  and blends in with the curve shown in Fig. 3 above the intersection with  $T_f$  at  $X = X_0 = 0.064$ .

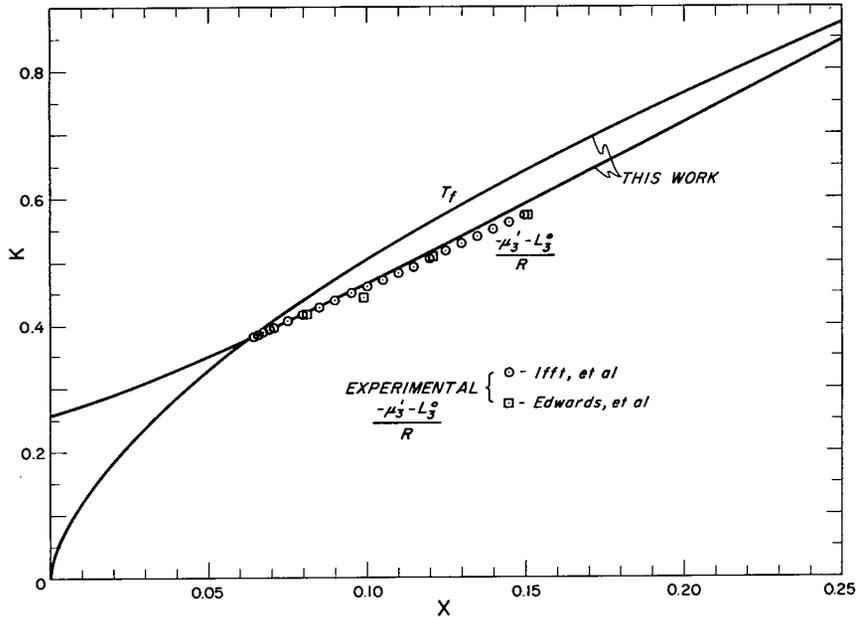


Fig. 3 - Fermi temperature and Deviation of the chemical potential from the ideal Fermi-Dirac value

Now that both the specific heat and the chemical potential of the  $\text{He}^3$  quasiparticle gas have been evaluated, it is easy to calculate other important thermodynamic properties. Two of the quantities necessary for the analysis of the dilution refrigerator are the entropy and enthalpy, given by

$$S_3 = S_f = \int_0^T (C_v/T) dT \quad (13)$$

and

$$H_3 = \mu'_3 + L_3^0 + H_f = \mu'_3 + L_3^0 + RT_f + (5/3) \int_0^T C_v dT. \quad (14)$$

The osmotic pressure of He<sup>3</sup> in He<sup>4</sup>, or the pressure of the quasiparticle gas, is evaluated as follows. Consider two containers connected via a plug permeable only to superfluid He<sup>4</sup>, i.e., a superleak. In one container is a dilute solution of concentration  $X$  and temperature  $T$ , and in the other is pure He<sup>4</sup> at temperature  $T$ . To prevent He<sup>4</sup> from flowing from the container of pure He<sup>4</sup> through the superleak into the dilute solution we must apply a pressure  $\Pi$ , defined as the osmotic pressure, to the dilute solution. The two solutions are then in equilibrium. Because the He<sup>4</sup> in both containers is superfluid, Eq. (1) is valid and results in

$$\mu_4^0(T) = \mu_4(X, T, \Pi), \quad (15)$$

where the term on the left is the chemical potential of pure He<sup>4</sup>. The chemical potential is a point function (a function which is independent of path), so we may then write

$$\mu_4(X, T, \Pi) = \mu_4(X, T, 0) + \int_0^\Pi \left( \frac{\partial \mu_4}{\partial P} \right)_{T, X} dP. \quad (16)$$

The expression  $(\partial \mu_4 / \partial P)_{T, X} = V_4$  follows from the thermodynamic relation  $d\mu_4 = -S_4 dT + V_4 dP$ , where  $V_4 = V_m - X \partial V_m / \partial X = 27.58 - 3.30 X^3$  cm<sup>3</sup>/mole is the partial molar volume of He<sup>4</sup> solution. Equations (15) and (16) can then be combined to yield

$$\Pi V_4 = - [\mu_4(X, T, 0) - \mu_4^0(T)]. \quad (17)$$

The term  $\mu_4$  is related to the quantity  $\mu_3$  through the Gibbs-Duhem equation for constant temperature and pressure:

$$X \frac{d\mu_3}{dX} + (1 - X) \frac{d\mu_4}{dX} = 0. \quad (18)$$

In integral form Eq. (18) becomes

$$\mu_4 - \mu_4^0 = - \int_0^X \frac{X}{1 - X} d\mu_3, \quad (19)$$

which can be evaluated numerically to find  $\Pi$ . The results of this calculation are shown in Fig. 4. Shown for comparison is  $\Pi$  for the noninteracting Fermi gas of  $X = 0.064$ . The value of  $\Pi$  at  $T = 0$  and  $X = X_0 = 0.064$  is 12.2 torr (1 torr = 133.3 N/m<sup>2</sup>), which is somewhat lower than the recently measured values of  $16.8 \pm 0.4$  torr by Landau et al.<sup>27</sup> and  $17.2 \pm 0.5$  torr by London et al.<sup>30</sup> The difference, however, has little effect on the analysis of the dilution refrigerator.

The curve  $p_f$  shown in Fig. 4 is pure He<sup>4</sup> fountain pressure given by

$$p_f V_4^0 = - [\mu_4^0(T) - \mu_4^0(0)]. \quad (20)$$

Comparing this with Eq. (17) we see that the equilibrium condition of  $\mu_4 = \text{constant}$  in the dilute solution also means  $\Pi + p_f = \text{constant}$ ; but for most temperatures  $p_f \ll \Pi$ . If the quasiparticle gas is allowed to warm up at constant  $\Pi$ , as in the case of the gas flowing through a heated tube, then it is evident from Fig. 4 that the concentration or gas density decreases. The specific heat of the He<sup>3</sup> quasiparticles in such a process is given by

$$C_{\mu_4} = T \left( \frac{\partial S_3}{\partial T} \right)_{\mu_4 = \text{const}}. \quad (21)$$

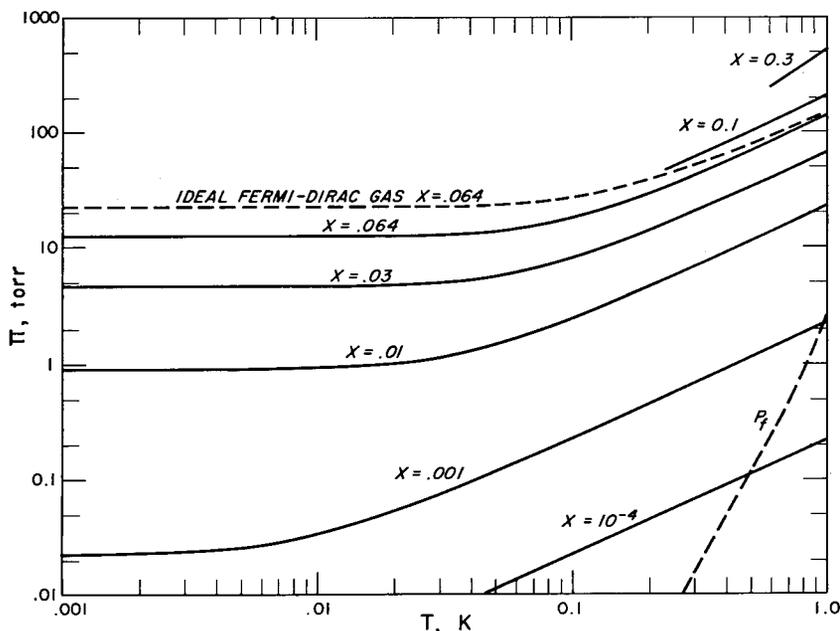


Fig. 4 - Calculated osmotic pressure of  $\text{He}^4$  in liquid  $\text{He}^4$  as a function of temperature for various  $\text{He}^3$  concentrations. Also shown is the fountain pressure  $p_f$  of pure  $\text{He}^3$

#### THE THERMODYNAMICS OF THE DILUTION PROCESS

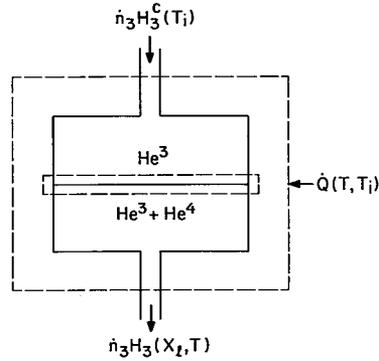
Let us consider the actual process of dilution as takes place in the mixing chamber of a continuously operating dilution refrigerator. A schematic of this process is shown in Fig. 5. The quantity of interest is the heat absorption rate  $\dot{Q}$ . By far the simplest technique for evaluating  $\dot{Q}$  is to treat the mixing chamber as an open thermodynamic system. Such a system is common in engineering but is not well known in physics. Our system of interest, which remains fixed in time, is shown by the outer dashed line completely enclosing the mixer. The system is considered an open system, since mass is allowed to enter and leave. Concentrated  $\text{He}^3$  is entering at a molar flow rate of  $\dot{n}_3$ , and the  $\text{He}^3$  quasiparticle gas is leaving at the same molar flow rate. The process is obviously analogous to the continuous vaporization of a liquid. Since no external work is being done by the system, and since there is no change in the internal energy of the system with time, conservation of energy, or the first law of thermodynamics, for an open system is simply stated as

$$\dot{Q}(T, T_i) = \dot{n}_3 H_3(X_\ell, T) - H_3^0(T_i) \quad (22)$$

For nearly pure  $\text{He}^3$  the last term on the right becomes  $H_3^0$  and for  $T \lesssim 0.04$  K is equal to  $12T^2$  joules/mole  $\text{He}^3$ . The term  $H_3(X_\ell, T)$  can be evaluated from Eq. (14) in principle, but at temperatures below about 0.1 K the first three terms on the right side of Eq. (14) are relatively large and nearly cancel each other. Such a calculation would then be subject to rather large errors. To get around this dilemma we shall consider an alternative calculation of the heat absorption rate. Though exactly equivalent to Eq. (22) this second method gives a way of evaluating  $H_3(X_\ell, T)$  below 0.1 K much more accurately than the direct evaluation from a table of  $H_3$  as a function of  $X$  and  $T$ .

Consider a second open thermodynamic system designated by the inner dashed lines in Fig. 5 just around the phase boundary. Within this limited system, temperature

Fig. 5 - Schematic diagram of the mixing chamber of a continuously operating dilution refrigerator, with concentrated He<sup>3</sup> entering at an inlet temperature T<sub>i</sub> and quasiparticle He<sup>3</sup> leaving at temperature T



equilibrium exists, and the actual crossing of the phase boundary by He<sup>3</sup> is reversible. The total heat absorption rate in this reversible process is simply

$$\dot{Q}_t = \dot{n}_3 T \Delta S = \dot{n}_3 T [S_3(X_l, T) - S_3^0(T)]. \quad (23)$$

But this total heat actually is coming from two sources: the external heat that is being applied and the heat that is being conducted to the interface from the incoming warm He<sup>3</sup>. Thus we write

$$\dot{Q}_t = \dot{Q} + \dot{n}_3 [H_3^0(T_i) - H_3^0(T)], \quad (24)$$

and when combined with Eq. (23) this becomes

$$\dot{Q}(T, T_i) / \dot{n}_3 = T [S_3(X_l, T) - S_3^0(T)] + H_3^0(T) - H_3^0(T_i). \quad (25)$$

If we compare Eq. (25) with Eq. (22), we note that

$$H_3(X_l, T) = H_3^0(T) + T [S_3(X_l, T) - S_3^0(T)] \quad (26)$$

which says that the enthalpy in the dilute (quasiparticle "gas") phase is simply the enthalpy of the quasiparticles in the strongly interacting or pure liquid He<sup>3</sup> phase plus the heat required to "vaporize" the quasiparticles into the "gas" or weakly interacting phase. Equation (26) is used to calculate H<sub>3</sub>(X<sub>l</sub>, T) below 0.1 K, and for T ≲ 0.04 K the heat absorption rate of the dilution refrigerator according to Eq. (22) becomes

$$\dot{Q}(T, T_i) / \dot{n}_3 = 94T^2 - 12T_1^2 \text{ joules/mole He}^3 \quad (27)$$

For mixer temperatures considerably above the minimum the heat exchangers will be quite efficient, and T<sub>i</sub> will be nearly equal to T. In this case the maximum heat absorption rate of  $\dot{Q}(T, T) / \dot{n}_3 = 82T^2$  joules/mole He<sup>3</sup> is achieved. If X<sub>0</sub> were 0.068 instead of 0.064, the heat absorption rate would be 78T<sup>2</sup> joules/mole He<sup>3</sup>.

Figure 6 gives curves of H<sub>3</sub>(X<sub>l</sub>, T) and H<sub>3</sub><sup>0</sup>(T), which can be used in Eq. (22) to find the heat absorption rate at higher temperatures. It is obvious from these curves and from Eq. (27) that the incoming pure He<sup>3</sup> can be at a much higher temperature than the outgoing dilute He<sup>3</sup> and still provide refrigeration. From Eq. (27) we find that the refrigeration

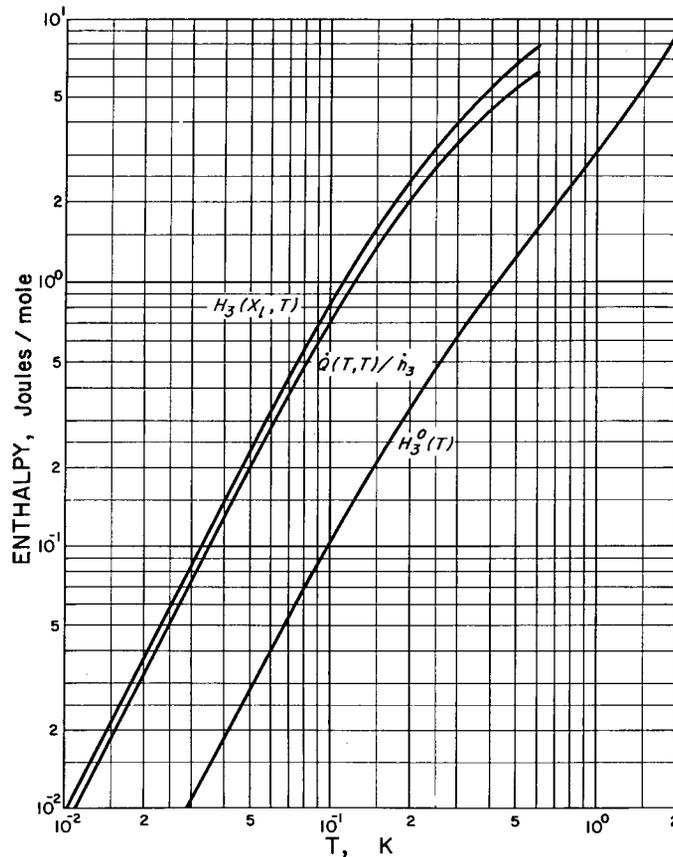
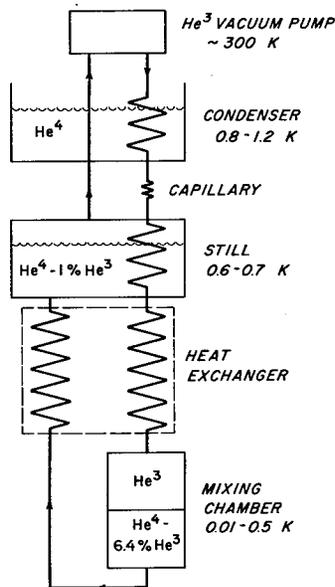


Fig. 6 - Enthalpy and heat absorption curves for calculation of mixer behavior

power goes to zero whenever  $T/T_i = 0.36$ . This ratio decreases at higher temperatures to about 0.25 at  $T = 0.2$  K.

The actual process to bring about the continuous dilution is shown schematically in Fig. 7. The vacuum pump circulates nearly pure  $\text{He}^3$  through the system and compresses the gas to a pressure in the range 25 to 200 torr. At this pressure the gas condenses in a condenser held at about 1 K by a pumped- $\text{He}^4$  bath. The pressure on the liquid is then reduced after passing through the capillary. The initial condensing rate of the empty condenser must be higher than the flow rate through the capillary. When this is so, the condenser begins to fill up, thus reducing the condensing rate due to a loss of condensing surface area. At some point the condensing rate equals the flow rate through the capillary and the condenser is running partially full. If the condensing rate is too small or the capillary impedance too small, then some gas will pass through the capillary and put a large heat load on the still and possibly the warmer heat exchangers. One must also be careful to keep the pressure below the capillary up to at least the vapor pressure of  $\text{He}^3$  at that temperature to prevent gas formation due to vaporization. This is usually done<sup>13</sup> by putting a second capillary below the still. Just the hydrostatic head of the liquid itself is usually sufficient below this second capillary to prevent gas formation there. In addition to causing a heat load on the colder parts the gas formation in the capillary can increase the effective flow impedance tremendously and require rather high pump pressures to get the proper circulation rate.

Fig. 7 - Schematic diagram of the He<sup>3</sup>-He<sup>4</sup> dilution refrigerator



The He<sup>3</sup> liquid continues to cool as it passes through a heat exchanger in the still and then through the main heat exchangers. The liquid exits from the heat exchangers at a temperature  $T_i$ , which is the inlet temperature to the mixing chamber. A small amount of He<sup>4</sup> in the circulated He<sup>3</sup> will not have much direct effect on the behavior in the mixer, but it certainly will in the heat exchangers. The effective specific heat<sup>16</sup> of solutions in the range 80 to 95% He<sup>3</sup> can be at least double that for pure He<sup>3</sup> for temperatures of 0.2 to 0.6 K. Thus He<sup>4</sup> circulation puts a much higher load on the warmer heat exchangers and leads to a higher mixer temperature.

The driving process for dilution in the mixing chamber is brought about by the depletion of He<sup>3</sup> from the dilute solution in equilibrium with the pure He<sup>3</sup> phase. The depletion actually occurs in the still at about 0.7 K, where heat is applied to cause vaporization. Since the vapor pressure of He<sup>3</sup> is much higher than that of He<sup>4</sup> at this temperature, the gas given off is nearly pure He<sup>3</sup>, which returns to the pump. As the He<sup>3</sup> is removed from the dilute solution in the still, He<sup>3</sup> diffuses through the stationary He<sup>4</sup> from the mixer to the still. Ideally the He<sup>3</sup> diffusion, or He<sup>3</sup> quasiparticle gas flow, is at constant osmotic pressure, but any impedance in the heat exchangers causes some pressure drop. The osmotic-pressure curves of Fig. 4 show that for low mixer temperatures the concentration in the still at about 0.7 K will be about 1% He<sup>3</sup>. Any osmotic pressure drop in the heat exchanger will reduce this concentration and cause an increase in the amount of He<sup>4</sup> evaporated in the still.

The flow of cold He<sup>3</sup> quasiparticle gas from the mixer to the still is used by the heat exchanger to cool the incoming pure He<sup>3</sup> liquid. The specific heat being used in the dilute side of the heat exchanger is the specific heat at constant osmotic pressure or, more exactly, at constant  $\mu_4$ . This specific heat, calculated from Eq. (21), is shown in Fig. 8. The various mixer temperatures  $T_m$  represent different osmotic pressures. The limiting low temperature behavior of  $C_{\mu_4} = 107.2T$  joules/mole is just  $C_v$  for a 6.4% solution. The 5R/2 behavior at higher temperatures is just that of an ideal classical gas at constant pressure. The fairly rapid rise in  $C_{\mu_4}$  at the highest temperatures is a result of the rapidly increasing fountain pressure driving down the equilibrium concentration. The high initial values of  $C_{\mu_4}$  for the high mixer temperatures are due to the He<sup>3</sup>-He<sup>3</sup> interactions becoming fairly strong at the high concentrations present at these high mixer temperatures. We are fortunate that  $C_{\mu_4}$  is much higher than  $C_3^0$  at all temperatures. If it were not so, the dilute stream would have little cooling power on the incoming pure He<sup>3</sup>.

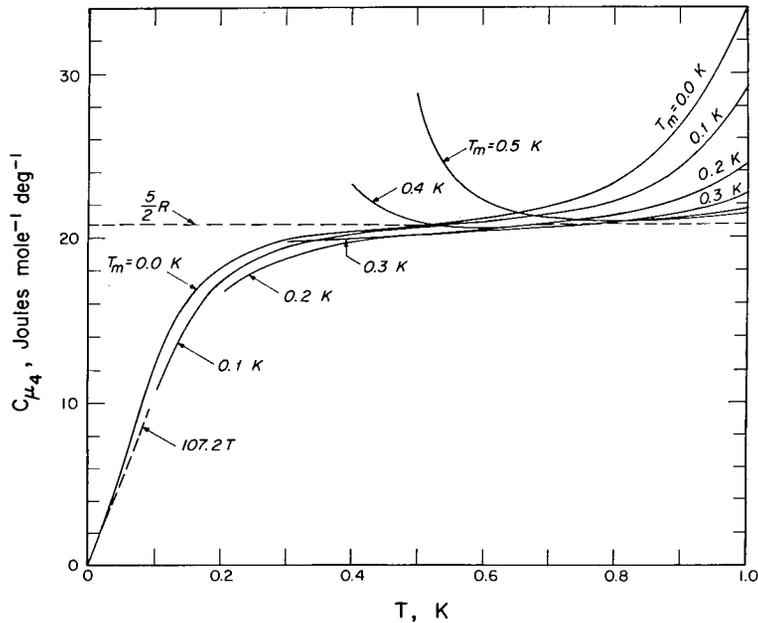


Fig. 8 - Calculated specific heat of  $\text{He}^3$  at constant  $\mu_4$  for various mixer temperatures  $T_m$

#### HEAT EXCHANGER ANALYSIS

The heat exchanger for the refrigerator can be one of two types: (a) continuous and (b) discrete or modular. The discrete heat exchanger can be made to vary between two extreme varieties: (a) long and slender, or length to area ratio  $l/A \gg 1$ , and (b) short and fat, or  $l/A \ll 1$ . It will be shown later that the variety with  $l/A \gg 1$  is the more efficient of the two discrete varieties, if the problem of high impedance can be overcome. In a continuous type, such as a set of coaxial stainless steel or CuNi tubes, the temperature of the heat exchanger body continuously changes with position, whereas in the discrete type the body temperature is uniform throughout. The continuous type was used on some of the first dilution refrigerators and in principle is more efficient than the discrete type. As the temperature is lowered, the Kapitza resistance increases at least as fast as  $1/T^3$ ; so the tube lengths need to be increased considerably to gain enough additional surface area for heat transfer. As the tube is lengthened, the flow impedance increases, resulting in a reduction of  $\text{He}^3$  concentration in the still and increase in the  $\text{He}^4$  circulated.

Wheatley et al.<sup>13</sup> first used the discrete type to gain higher surface areas per unit volume. In this type the total heat exchanger is divided into several separate heat exchangers, each connected by small stainless steel or CuNi tubes to carry the two liquid streams. Each separate heat exchanger is made of a copper block containing two volumes, one for the dilute stream and one for the concentrated stream. These volumes are filled with a sintered copper powder, with the powder particles usually about  $44 \mu\text{m}$  in diameter. To achieve higher efficiencies it is now becoming customary<sup>31</sup> to use a continuous heat exchanger for the higher temperatures followed by a few of the discrete type for the lowest temperatures.

Little information exists on the optimum design of the heat exchangers. Obviously the heat exchangers can be made very large to obtain good heat transfer and reduce flow impedance. The problem with doing this is that large amounts of  $\text{He}^3$  and  $\text{He}^4$  are required

to fill the system. Time is required to condense the liquid into the system and may become rather long for large volumes. Probably even more important is that equilibrium times for a change in mixer temperature are also increased. Optimum heat exchangers would cut down on some of this dead time and make the refrigerator more productive. We are engaged in an extensive theoretical as well as experimental analysis of the heat exchangers, and we present here some of the results to date on the theoretical analysis. So far only the discrete type has been analyzed.

The limiting behavior of a set of discrete heat exchangers is found by letting each heat exchanger be of infinite volume or allowing perfect heat exchange between the liquid streams and each heat exchanger body. The analysis then requires only an enthalpy balance on each heat exchanger. For simplicity we assume that the enthalpy in the concentrated phase is  $H_3^0 = \alpha T^2$  and that the enthalpy in the dilute phase at constant osmotic pressure is  $H_{\mu_4} = \beta T^2$ . An enthalpy balance on  $n$  perfect heat exchangers is then

$$\alpha(T_1^2 - T_1^2) = \beta(T_1^2 - T_2^2), \quad (28a)$$

$$\alpha(T_1^2 - T_2^2) = \beta(T_2^2 - T_3^2), \quad (28b)$$

$$\alpha(T_2^2 - T_3^2) = \beta(T_3^2 - T_4^2), \quad (28c)$$

$$\alpha(T_{n-1}^2 - T_n^2) = \beta(T_n^2 - T_m^2), \quad (28d)$$

where  $T_i$  is the inlet temperature of the pure He<sup>3</sup> to the first heat exchanger and  $T_m$  is the mixer temperature, which we replace by

$$T_m = \gamma T_n. \quad (29)$$

For no heat input to the mixer  $\gamma = 0.36$  for  $T \lesssim 0.04$  K. Equations (28) can be modified to the form

$$\alpha_i T_i^2 = (\alpha_1 + \beta_1) T_1^2 - \beta_2 T_2^2 + 0 + 0 + \dots, \quad (30a)$$

$$0 = -\alpha_1 T_1^2 + (\alpha_2 + \beta_2) T_2^2 - \beta_3 T_3^2 + 0 + \dots, \quad (30b)$$

$$0 = 0 - \alpha_2 T_2^2 + (\alpha_3 + \beta_3) T_3^2 - \beta_4 T_4^2 + \dots; \quad (30c)$$

$$0 = 0 + 0 + 0 + 0 + \dots - \alpha_{n-1} T_{n-1}^2 + [\alpha_n + \beta_n(1 - \gamma_n^2)] T_n^2, \quad (30d)$$

where now the coefficients of  $T^2$  are indexed to permit the coefficient to depend on temperature. The  $n$  simultaneous equations with the variable coefficients are solved by successive approximations. A single value for  $\alpha$ ,  $\beta$ , and  $\gamma$  is chosen, and the equations are solved for each of the  $T^2$  values. Then the coefficient appropriate for that temperature replaces the single value used initially, and the process is repeated until the temperatures are determined to sufficient accuracy.

The mixer temperature resulting from this analysis is shown in Fig. 9 as a function of the number of perfect discrete heat exchangers for various  $T_i$ . As an example

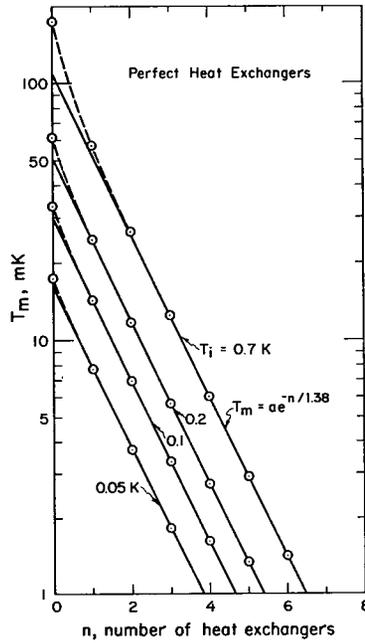


Fig. 9 - Calculated mixer temperatures as a function of the number of perfect discrete heat exchangers for various inlet temperatures

consider the case of  $T_i = 0.7$  K. To reach 10 mK would then require a minimum of four discrete heat exchangers. For low temperatures, where the coefficients  $a$ ,  $\beta$ , and  $\gamma$  remain constant, the mixer temperature is given by

$$T_m = ae^{-n/1.38}, \quad (31)$$

where  $a$  depends on  $T_i$ . The same expression, but with a different  $a$ , also gives the temperature of each of the heat exchangers. The temperature reduction in the  $j$ th heat exchanger implied by an expression of the type in Eq. (31) can be written as

$$T_j = T_{j-1}/\delta \quad (32)$$

For  $T_{j-1} \lesssim 0.05$  K and for perfect discrete heat exchangers  $\delta = \sqrt{\beta/a} = 2.1$ . This number begins to increase at higher temperatures, and at  $T_{j-1} = 0.1$  and  $0.2$  K we have  $\delta = 2.2$  and  $2.4$  respectively. A measure of the efficiency of a practical exchanger is  $(\delta-1)/(\delta-1)_{\text{perfect}}$ . If a 95% solution is circulated instead of pure  $\text{He}^3$ , then the curve in Fig. 9 for  $T_i = 0.7$  K is shifted to the right by about 0.3 of a heat exchanger or upward by 2 mK at 10 mK. There is less effect on the curves with lower  $T_i$ . A similar graph for less than perfect heat exchangers still needs to be done.

The temperature profile which occurs in the liquids in either a continuous or a discrete heat exchanger can be found from a heat balance on a section of the liquid as shown by the equation

$$A \left[ \underbrace{\kappa \frac{d^2 T}{dx^2} + \frac{d\kappa}{dT} \left( \frac{dT}{dx} \right)^2}_{\text{heat conduction}} \right] - \underbrace{\frac{d\sigma}{dx} \int_{T_b}^T \frac{dT}{\rho}}_{\text{kapitza conduction}} + \underbrace{\eta v^2 \dot{n}_3^2 \frac{dz}{dx}}_{\text{viscous heating}} = \underbrace{\dot{n}_3 c_3 \frac{dT}{dx}}_{\text{enthalpy change}}, \quad (33)$$

where  $A$  is the cross-sectional area of the liquid,  $\kappa$  is the liquid thermal conductivity,  $d\sigma/dx$  is the surface area per unit length,  $T_b$  is the heat exchanger body temperature,  $\rho$  is the Kapitza resistivity,  $\eta$  is the viscosity,  $v$  is the  $\text{He}^3$  molar volume,  $\dot{n}_3$  is the molar

flow rate, and  $dz/dx$  is the impedance per unit length. Temperatures are assumed to be uniform in the radial direction. For continuous heat exchanger  $T_b$  will change with axial position, but with a discrete heat exchanger it will be constant. So far we have only solved the equation for the discrete case, and the viscous heating effect has been neglected. If  $\rho$  were proportional to  $1/T^3$ , then the integral would just become the difference in the fourth powers of the stream and body temperatures. The value for  $T_b$ , found by trial and error, must satisfy the heat balance equation in both the dilute and concentrated sides simultaneously. The equations are solved numerically, subject to the proper boundary conditions.

The solution to these equations are shown in Fig. 10 for a rather typical heat exchanger at the very lowest temperatures. The length is 1 cm, and the volume of He<sup>3</sup> on the concentrated side is 0.85 cm<sup>3</sup>, so that  $l/A \approx 1$ . The liquid volume on the dilute side is 2.1 times the concentrated volume. The flow rate is  $2 \times 10^{-5}$  mole/sec, and the surface area per unit volume is 400 cm<sup>-1</sup>. Two sets of curves are shown: one for the case where the full equation except for the viscous term in Eq. (33) is solved and the other for the case where thermal conductivity in the liquid is neglected. This second case actually corresponds to the case of finite thermal conductivity in the very long, small-diameter variety of exchanger with the same volume and surface area. In either case radial temperature gradients are neglected, since in any properly made exchanger these gradients will not exist. The temperature profile in the two cases is drastically different. In comparing the performance of the heat exchanger in the two situations, the mixer temperature has been held constant at 10 mK. For the case of zero conductivity (long, slender exchanger) the concentrated He<sup>3</sup> inlet can be 59.9 mK, whereas for the case where conductivity is considered the He<sup>3</sup> inlet is only 40.5 mK. The long, slender exchanger is able to transfer considerably more heat and thus is a more efficient heat exchanger. As the surface area is increased, both varieties will of course approach the efficiency of a perfect exchanger. In the variety of heat exchanger for  $l/A \ll 1$  and even for  $l/A \approx 1$  the thermal conductivity of the liquid tends to equalize the liquid temperature throughout the exchanger. The initial high  $\Delta T$  between the liquid and the exchanger body possible with  $\kappa = 0$  is lost when  $\kappa \neq 0$ , and the amount of heat that can be transferred is thus reduced. Most of the temperature drop in the concentrated and dilute liquid streams actually occurs in the small inlet tubes just prior to entering the exchanger. In order that heat flow be conserved the ratio of the two slopes in the heat exchanger and in the tube at the inlet to the exchanger is just the inverse ratio of the cross-sectional areas.

In this analysis of the discrete heat exchanger we have taken  $\kappa$  as the thermal conductivity of the bulk liquid, either pure He<sup>3</sup> or a dilute solution. In a sintered-copper discrete heat exchanger the effective  $\kappa$  will actually be somewhat lower, because the path length for heat transfer through the liquid is increased in the sintered material by a factor of approximately  $\pi/2$ . The small channels in the sintered copper can also effect the thermal conductivity whenever the mean-free-path of the dominant heat-carrying excitation is boundary limited. In the case of pure He<sup>3</sup> heat is transported by He<sup>3</sup> quasiparticles for all temperatures of interest. The mean-free-path due to He<sup>3</sup>-He<sup>3</sup> interactions is extremely short, the order of 0.1  $\mu\text{m}$  at 10 mK, and proportional to  $1/T^2$ . Thus even in fine powders the thermal conductivity of the liquid will be the same as the bulk value. The situation is different for the dilute side. The quasiparticle mean-free-path is still small, about 1  $\mu\text{m}$  at 10 mK, but above about 15 mK in a 6.4% solution, heat is also transported by phonons, which dominate at somewhat higher temperatures. The phonon mean-free-path due to collisions with He<sup>3</sup> quasiparticles can be very long<sup>31</sup> (320  $\mu\text{m}$  for a 5% solution at 0.1 K), which leads to boundary-limited mean-free-paths in the small channels of most sintered materials. Deviations of the effective thermal conductivity for a 44- $\mu\text{m}$  sintered powder from bulk values<sup>32</sup> will be largest around 60 or 70 mK and will be about a factor of 4 lower. The heat exchanger analyzed in Fig. 10 is at a low enough temperature that this deviation is negligible. For the warmer heat exchangers this deviation has to be taken into account. The analysis of the warmer exchangers has not been completed yet, but it is evident that the lower thermal conductivity at somewhat higher temperatures

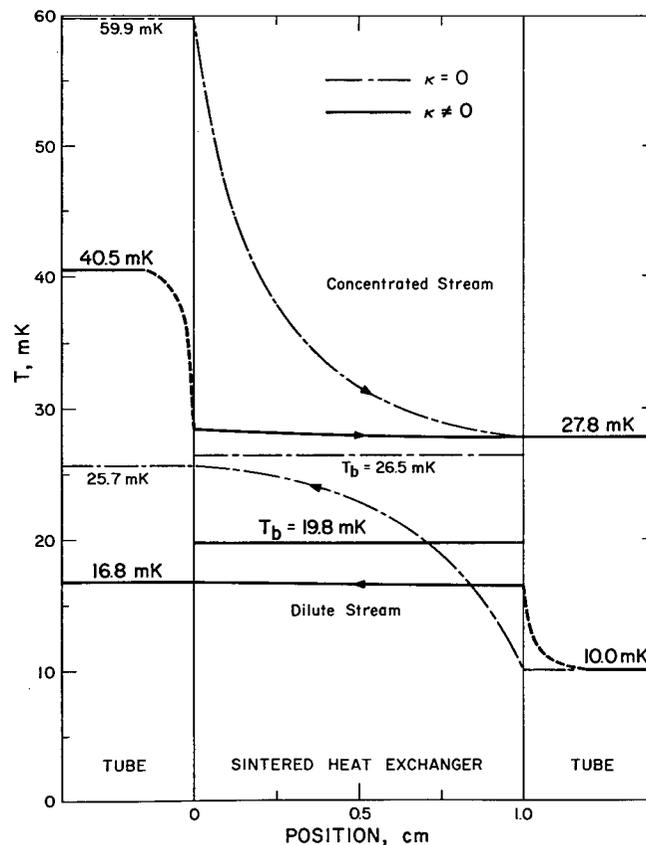


Fig. 10 - Calculated liquid and body temperature profiles within and just outside a sintered, discrete heat exchanger for a mixer temperature of 10 mK

will give rise to higher axial temperature gradients than that shown in Fig. 10 but nowhere near the gradients possible with  $\kappa = 0$ .

The implication from the present analysis of the discrete heat exchangers is that a heat exchanger with a large  $l/A$  value is much more efficient than one with a small  $l/A$  variety for the same liquid volume. However the large- $l/A$  variety would have an extremely high impedance and therefore be undesirable if made by the conventional techniques. To approach the curve for  $\kappa = 0$  would require an increase of at least a factor of 100 for  $l/A$ . A possible way of reducing the impedance in such an exchanger is by means of a large axial hole or holes through the sintered material. Another technique, which we have used, is to make an exchanger with fine powder (approximately  $10 \mu\text{m}$  in diameter in diameter) sintered in the form of a foil 0.018 cm thick. A strip of the foil is rolled in a loose spiral and sintered, or diffusion bonded, to the inside of the copper block. The liquid is able to flow easily through the gaps between each layer of the spiral but at the same time should be in good thermal contact with the liquid trapped in the very fine pores. If one does not wish to go to this trouble, the best technique would be to make the heat exchangers just as short as possible, at least for the lowest temperature exchangers. The curves of Fig. 10 show that a conventional sintered heat exchanger of  $l/A \approx 1$  already is behaving nearly like the case of  $l/A \ll 1$ , in which case no axial temperature gradients would exist for the liquid within the exchanger. To reduce  $l/A$  further would have a negligible effect on the thermal behavior of the exchanger but would cause a significant reduction in flow impedance.

The question of the best distribution of the volumes between the dilute and concentrated sides has also been considered. The total number of moles of  $\text{He}^3$  in both the dilute and concentrated sides of the exchanger was fixed, since this essentially determines the time constant for the refrigerator. The heat exchanger was then analyzed as in Fig. 10 to find the total heat transferred for various ratios of surface areas in the two sides. The mixer temperature was held at 10 mK. Figure 11 indicates how the amount of heat transferred varies with the ratio  $\sigma_d/\sigma_c$ , which in this case is also the same as  $V_d/V_c$ , since the same surface-to-volume ratio is used in the two sides. The optimum ratio is seen to be 2.1 and is the ratio used to obtain the results in Fig. 10. Further progress in analysis of the heat exchanger is being made. Yet to be evaluated is the optimum distribution of sizes for the various heat exchangers at different temperatures. Also the analysis of the continuous heat exchanger is just now beginning. Hopefully we might be able to say at what temperature it is best to shift from the continuous to the discrete type of heat exchanger.

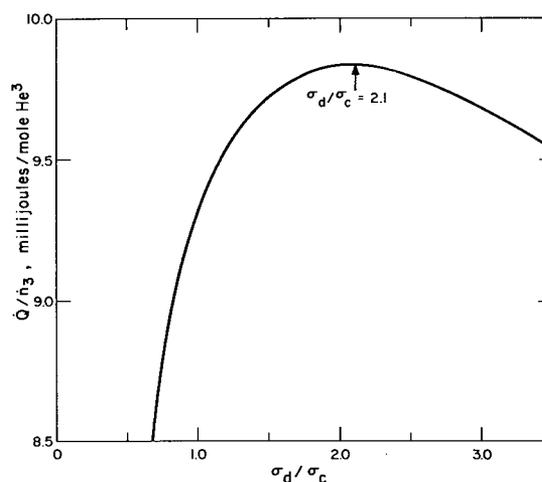


Fig. 11 - Total Heat Transfer Rate in a Discrete Heat Exchanger as in Figure 11 as a Function of the Ratio of Dilute to Concentrated Surface Areas

## CONCLUDING REMARKS

It has not been possible in the length of this paper to discuss several other aspects of the theory of dilution refrigerators. For instance nothing has been said about the various single-cycle or one-shot modes possible. Those interested in the analysis of these modes might care to look at Refs. 11 through 13. In this paper we first discussed the thermodynamics which accurately describe the behavior of the dilution refrigerator. After knowing how the device operates one wants to optimize the behavior. The numerical analysis of the heat exchangers is about the first step in this optimization, and we have discussed the progress we have made so far in these calculations. Future theoretical work on the dilution refrigerator is still mainly on the heat exchangers, and in particular the Kapitza resistance problem needs to be dealt with much more thoroughly. Many uncertainties need to be cleared up about the heat transfer process in the dilute side of the heat exchangers, where heat must pass from the wall through the  $\text{He}^4$  phonons to the  $\text{He}^3$  quasiparticle. We need to know more about the phonon-quasiparticle interaction. At very low temperatures direct heat transfer to the  $\text{He}^3$  quasiparticles may occur. Also the effect of small particles on heat transfer needs to be considered, so that optimum particle size can be used in the sintered heat exchangers. We might end by saying that the desire for improvements in dilution refrigerators still remains as an influence for further theoretical as well as experimental work.

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## Practical Consideration of He<sup>3</sup>-He<sup>4</sup> Refrigerators

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The previous paper has discussed the theory of dilution refrigeration, and the following will consider the application of dilution refrigerators. Thus I have assumed that the present paper should be concerned with the design and construction of practical dilution refrigerators, with emphasis on the word practical. The construction of a *useful* refrigerator is surprisingly simple. This is not to belittle the efforts of early workers in this field. After all, the fact that you or I can now readily build a laser does not reflect on the competence of the pioneers in that field. On the other hand a requirement of *optimum* refrigeration performance necessitates the application of all available theoretical and empirical knowledge, just as with any other device.

Figure 1 shows a schematic flow diagram which will serve to establish the terminology to be used. The *pumps* are at room temperature and serve to circulate the He<sup>3</sup> gas and to raise the pressure of the He<sup>3</sup> gas sufficiently that it can be recondensed. The *traps* serve to remove impurities from the circulating gas, since the refrigerator may be operated for 2 or more weeks at a time. Hence even trace amounts of impurities would tend to plug the small-diameter tubing within the cryostat. The condenser serves to liquify the returning He<sup>3</sup> gas at a temperature of 1 to 2 K. This *consenser* operates at or above the vapor pressure of He<sup>3</sup> at the temperature of the condenser (100 to 1000 torr), whereas the output of the refrigerator operates at a very low pressure. The major fraction of this pressure differential is forced to occur across the *flow impedance*. This impedance is inserted after the condenser, where the temperature is constant during operation of the refrigerator; hence the liquid viscosity and resultant pressure drop also remain constant. In addition the resultant viscous heating is less detrimental in this location. The *heat exchanger* provides the usual function of increasing the efficiency of the refrigerator. The *mixer* is simply a volume where the phase boundary between the He<sup>3</sup>-rich and He<sup>4</sup>-rich liquids is located and where dilution of the He<sup>3</sup> takes place. It is the cold finger of the refrigerator. The *still* serves to separate the circulating He<sup>3</sup> from the He<sup>4</sup>-rich phase by capitalizing on the higher vapor pressure of He<sup>3</sup> versus He<sup>4</sup> at a given temperature, usually  $\approx 0.7$  K.

### A SIMPLE DILUTION REFRIGERATOR

A refrigerator will be described which can be constructed easily and inexpensively, which can be adapted to most cryostats, and yet which provides modest refrigeration to temperatures below 0.05 K. In the next section we will discuss modifications to this simple design which will optimize the refrigeration capacity and/or the ultimate low temperature of the refrigerator. The few dimensions quoted are typical, but it must be understood throughout the discussion that *no* dimension or value is critical.

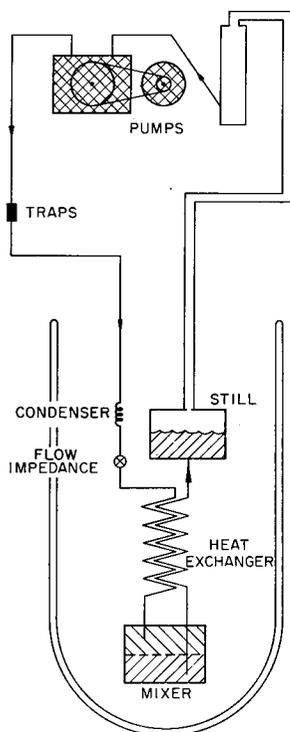


Fig. 1 - A schematic flow diagram for a He<sup>3</sup>-He<sup>4</sup> dilution refrigerator

The cryostat (dewar system) housing the refrigerator serves to provide thermal shielding and to cool the condenser to 1 to 2 K. Hence either a pumped He<sup>4</sup> bath is required or an outer bath at 4.2 K plus an inner pot which may be filled from the outer bath by a needle valve and then pumped to below 2 K is required. We have used both types of cryostats, as shown in Fig. 2, but in the interest of simplicity type II, the one using a pumped He<sup>4</sup> bath, will be discussed first.

The He<sup>3</sup> return line within the cryostat can be a  $\approx 0.16$ -cm-diam thin-wall tube (Cu-Ni or stainless steel). We generally use 5 to 10 cm of  $\approx 0.16$ -cm-I.D. Cu tube as a condenser, but just the return line itself will suffice. The impedance is made by inserting a steel wire in a 0.03-cm-I.D. thin-wall tube and then stretching the tube slightly. The flow impedance is measured at room temperature, and an appropriate length (2 to 3 cm) is cut off. The impedance should pass  $\approx 3 \times 10^{-3}$  cm<sup>3</sup>/sec of N<sub>2</sub> gas (S.T.P.) at room temperature at a pressure difference of  $\approx 200$  torr.

The coaxial, counterflow heat exchanger is made by inserting 0.04-cm-diam thin-wall tube inside a 0.12-cm-diam tube, the total length being  $\approx 100$  cm. The returning He<sup>3</sup> flows down the inner tube. The mixer can be fashioned from a wrought-copper plumber's cap as shown in Fig. 3. The larger tube of the heat exchanger should extend to the bottom of the mixer, so that the level of the interface between the He<sup>3</sup>-rich and He<sup>4</sup>-rich liquids is not critical. The inner tube of the heat exchanger should hook past the larger tube so that returning He<sup>3</sup> will not immediately reenter the larger tube, as shown in detail in Fig. 4. Although the device of Fig. 3 operated to below 0.045 K, it is best to increase the area of contact between the liquid He and the copper to enhance the thermal contact. Hence copper foils, etc., may be soldered into the mixer, or copper powder or lathe clips may be sintered in. Whatever technique is used, the He<sup>3</sup> within the mixer should be allowed to diffuse laterally as well as vertically. The mechanical support tube between the still and mixer can be of any material having a low thermal conductivity.

Fig. 2 - Drawings of two dilution refrigerators with some constructional details omitted to improve clarity. The labels are: C - condenser, S - still, H - heat exchanger, and M - mixer. Device I operates in a 4.2-K He bath and contains a He<sup>4</sup> pot pumped to approximately 1 K, whereas II operates in a 1-to-2-K He bath. Device I has two In O-ring-sealed vacuum jackets, so that during calibration, using a He<sup>3</sup> vapor pressure bulb attached to the inner jacket, the inner jacket alone may be filled with He<sup>3</sup> or He<sup>4</sup> thermal exchanger gas. The ports at the top of each jacket permit entrance of leads and tubing. A typical epoxy seal for electrical leads is shown in I. Note that light traps are placed in the He<sup>3</sup> pumping lines, as well as all other lines. The vacuum jackets are reduced in diameter near the bottom to accept mutual inductance coils for a magnetic thermometer.

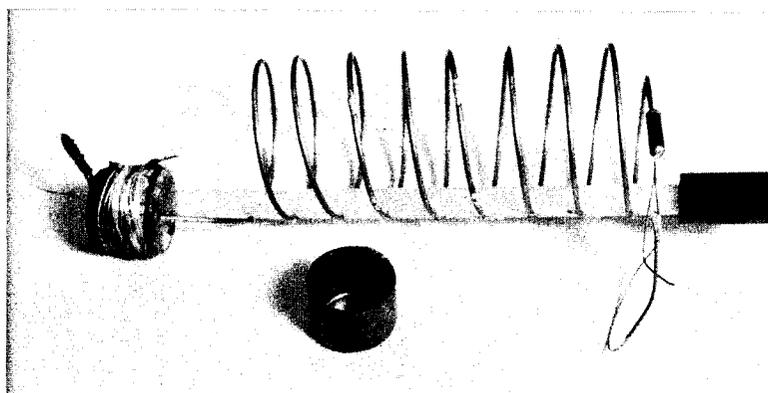
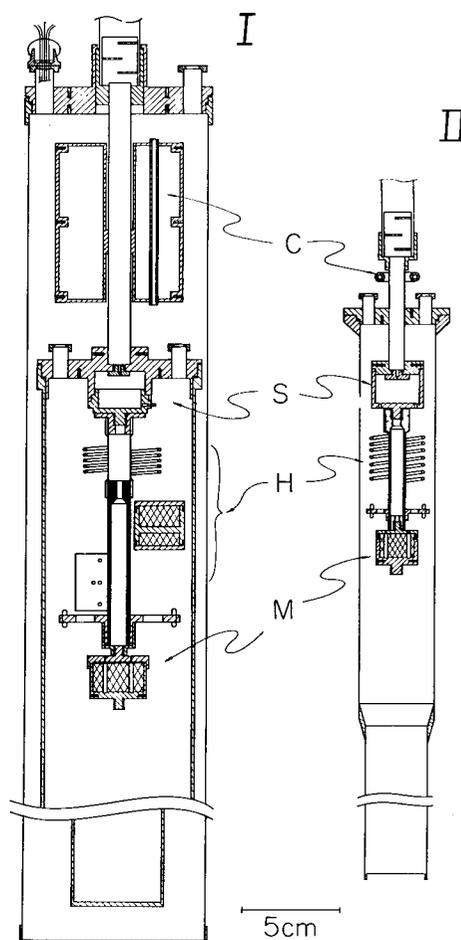


Fig. 3 - A coaxial counterflow heat exchanger and plumber's cap mixer, which was literally assembled from junk and operated on a weekend. The minimum temperature was  $\approx 0.045$  K. An unused 1.5-cm-diam wrought-copper plumber's cap is also shown.

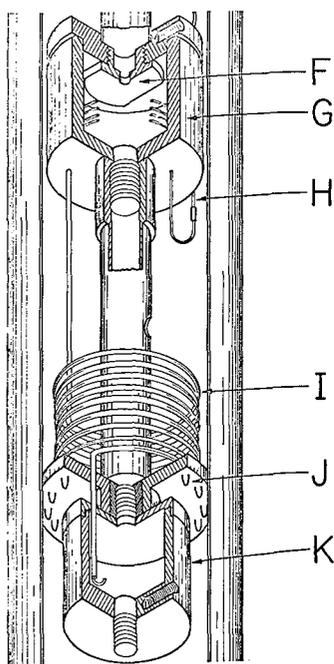


Fig. 4 - A more detailed view of refrigerator II from Fig. 2. The labels are: F - orifice, G - still containing a spiral of thin wall tube for precooling the returning He<sup>3</sup>, H - part of flow impedance, I - counterflow heat exchanger, J - epoxy electrical terminal strip, and K - mixer. A threaded stud is provided below the mixer for attaching experiments, and holes are tapped in the mixer and the still for attaching heaters and thermometers to the outside surfaces.

The still can be an equally simple container but with an orifice diameter of approximately 0.5 to 1 mm to provide a reduced perimeter over which the superfluid He<sup>4</sup> film must flow and thereby reduce the net amount of He<sup>4</sup> circulated with the He<sup>3</sup>. We have always electropolished the orifice, although the refrigerator will certainly operate without this refinement.

Since the still is generally operated at a lower temperature than the liquid He<sup>3</sup> returning from the condenser, it is advisable to precool the He<sup>3</sup> to the still temperature before it reaches the counterflow heat exchanger. This is easily accomplished by continuing  $\approx 50$  cm of the center tube of the heat exchanger into the mixer in the form of a coil, as shown in Fig. 4.

The mechanical pump needs to be completely sealed from the atmosphere. Certain Welch or Edwards models are available for this purpose. The diffusion pump should be designed for use with "heavy" or viscous oils; i.e., the ultimate low pressure capability is sacrificed to gain greater throughput at moderate pressures. Two traps are placed in the He<sup>3</sup> return line, one of loose glass wool immediately at the output of the mechanical pump, the other of a molecular sieve material cooled to liquid-N<sub>2</sub> temperatures. The quantity of He gas in the system is not critical, so long as under all operating conditions the phase boundary is always in the mixer and the upper surface of the He<sup>4</sup>-rich phase is always in the still. For refrigerator II of Fig. 2 the charge is  $\approx 4$  liters (S.T.P.) with a He<sup>3</sup>/He<sup>4</sup> ratio of  $\approx 0.2$ .

The foregoing description has been intentionally vague, since no one dimension is critical or sacred. Every device we have assembled has operated below 0.05 K, some with what later were discovered to be rather serious assembly errors. The low temperature limit of device II in Fig. 2 (with a sintered-Cu core in the mixer) is  $\approx 0.04$  K. Its refrigeration capacity at a He<sup>3</sup> circulation rate of  $\approx 2 \times 10^{-5}$  mole/sec is shown in Fig. 5, curve A, and may be compared with the refrigeration capacity of a typical evaporative type He<sup>3</sup> refrigerator, curve C. The cool-down time to 0.05 K from 4 K is  $\approx 30$  min and to 0.05 K from room temperature is  $\approx 3$  hr.

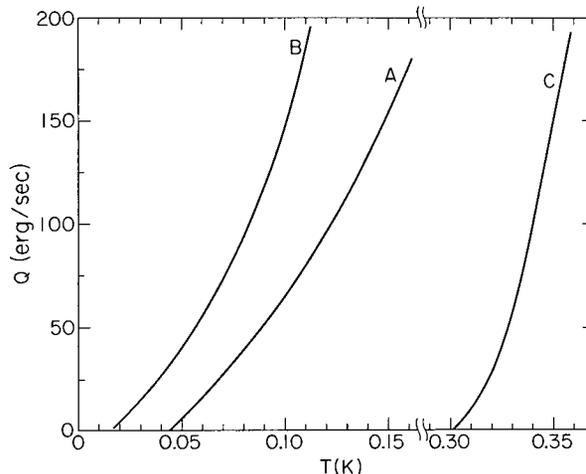


Fig. 5 - Refrigeration capacity versus temperature of: A - refrigerator II of Fig. 2, B - refrigerator I of Fig. 2, C - a typical  $\text{He}^3$  refrigerator. Curves A and B have not been optimized by a judicious choice of still temperatures.

No gravitational or other instability has been observed. However it is worth noting that frequently lower temperatures are reached if the  $\text{He}$  charge is initially liquified in the condenser and *not* in the still by admitting the gas into the low pressure lines. The reason is that the  $\text{He}^4$  superfluid film formed during the later method at higher pressure transports more  $\text{He}^4$ , after commencing operation, than a film formed under very low pressures.

The refrigerator can be operated for any length of time. When the bath runs low, it can be raised to atmospheric pressure, more liquid  $\text{He}^4$  can be transferred, and then it can be pumped back down. Since the bath around the vacuum jacket remains near 2 K, the refrigerator will continue to operate below 0.05 K. Furthermore the bath may be maintained at any temperature between 1 and 2 K. At the higher temperatures little or no extra heat need be supplied to the still to hold it at  $\approx 0.7$  K. In fact the  $\text{He}^3$  pumping tube between the still and the bath is made short intentionally (Fig. 2, II), so that much or all of the still power consists of a heat leak from the bath. Other details of this refrigerator will be published elsewhere.<sup>1</sup>

As was mentioned, this simple refrigerator has also been operated in a cryostat having a pumped  $\text{He}^4$  pot inside an outer bath, as in I of Fig. 2. Hence the overall consumption of liquid  $\text{He}$  is reduced, and smaller  $\text{He}^4$  pumps are required. The pot had a volume of  $\approx 200$   $\text{cm}^3$ . The  $\text{He}$  vapor was pumped through an unpolished 1-mm orifice, giving a holding time of  $\approx 36$  hours when condensing  $\text{He}^3$  at the rate of  $\approx 10^{-5}$  mole/sec. The needle valve (not shown) used for filling the pot from the  $\text{He}^4$  bath had a steel needle (1.5-degree taper) and seat (0.04-cm-diam hole), this combination giving much better results than other combinations of metals or plastics. With this valve the pot may be filled at any rate; i.e., the temperature of the pot may be maintained at any value between 1 and 2 K while filling. A filled pot is signaled by an abrupt rise in temperature or simply by visually observing the quantity of liquid removed from the outer bath. Hence, again, the refrigerator can be operated for any desired length of time.

I would also like to emphasize the use of In 0-rings to seal the flanges, as shown in Fig. 2, although, of course, any type of seal may be used. The flanges in device I use 0.08-cm-diam In wire, an 8-cm-diam flange requiring only eight No. 3-48 ( $\approx 2$ -mm-diam) bolts. We once accidentally caught an electrical lead in the flange, but it sealed anyway.

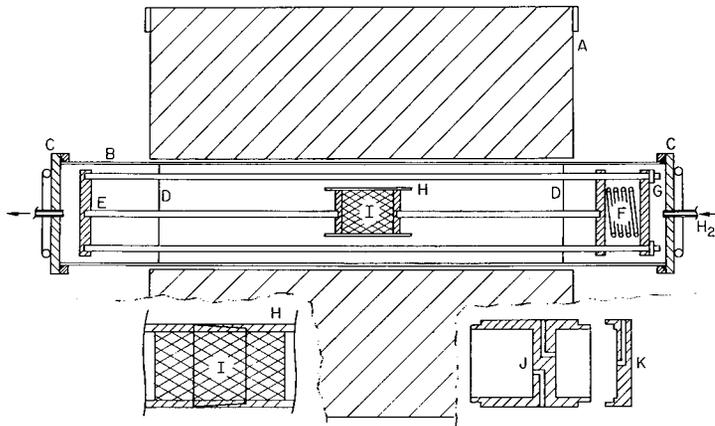


Fig. 6 - Arrangement used for the sintering of copper. The labeled parts are: A - Hoskins furnace, B - fused-quartz tube or other high temperature material, C - O-ring-sealed caps, D - stainless steel radiation shields, E - stainless steel "compactor," F - spring, G - adjustment nuts, H - copper tube, I - copper powder, J - body of heat exchanger, and K - cap for heat exchanger. Since the furnace is also used at temperatures above 1000°C, the end caps C are water cooled. A small flow of H<sub>2</sub> gas is maintained through the furnace while sintering, the flow rate being monitored by allowing the gas to bubble through water before venting. The enlarged insert at the lower left of the drawing indicates how the sintered piece is afterward machined to a taper (with a sharp lathe tool) prior to pressing it into the body of the heat exchanger, shown as an insert at the lower right.

## OPTIMIZING THE REFRIGERATOR

The refrigerator described is a simple and inexpensive device which could, for example, be substituted in an existing He<sup>3</sup> cryostat. Some applications, however, may require greater refrigeration capability and/or a lower ultimate temperature.

The ultimate low temperature is determined primarily by the heat exchanger. At low temperatures greater surface areas are required in the heat exchanger to defeat the Kapitza thermal boundary resistance—hence the popularity of sintered copper.<sup>2,3</sup> Placing two or three sintered-copper heat exchangers between the coaxial-tube exchanger and the mixer described in the preceding Section will result in a temperature of ≈0.016 K on the exterior of the mixer. The interior (the liquid He) cools to ≈0.009 K.

The copper, in the form of powder or chips, may be sintered in an inexpensive Hoskins furnace (Fig. 6) at ≈700°C for ≈1 hr in H<sub>2</sub> atmosphere. The powder is first compacted in a vise or press (≈10<sup>5</sup> g/cm<sup>2</sup> force), so that it is no longer loose. If the powder is maintained under compression while sintering, it will sinter to the copper walls and provide good thermal contact. The outer wall of the sintered plug is machined to a slight taper as shown in the lower left insert of Fig. 6 and then pressed into the body of the heat exchanger shown in the lower right insert to provide excellent thermal contact. End caps are then pressed on and the joints sealed with soft solder. Pressing a sintered core into the copper body permits one to leave a narrow space at each end of the core, thus reducing the overall flow impedance of the component. The impedance of

these exchangers reproduces to  $\approx \pm 10\%$ . The mixer may also be filled by sintering the Cu directly into its cavity, in which case the mixer body should be fashioned from  $O_2$ -free Cu. More detailed prescriptions for sintering have been presented elsewhere,<sup>4,5</sup> but, in terms of the results achieved the technique is not critical. It may be useful to note that copper parts may be "welded" by pressing them lightly together while in the sintering oven. Such "welds" provide excellent low temperature thermal contact.

Large refrigeration capacity means large  $He^3$  circulation rates. Hence the flow impedance of the various components and interconnections must be kept small,<sup>6</sup> including that of the  $He^3$  pumping line between the still and the diffusion pump. As an example of the importance of the pumping lines on refrigeration capacity, curve B of Fig. 5 shows refrigeration versus temperature for device I of Fig. 2, which is similar to that producing curve A except that larger pumping lines were used (and also three sintered-copper heat exchangers, which do not greatly affect the refrigeration capacity). The device appropriate to curve A (device II) had a 1.6-cm-diam pumping tube in the cryostat, becoming 5 cm in diameter at room temperature, and a 5-cm-diam diffusion pump. In comparison the device appropriate to curve B (device I) had a 2.5-cm-diameter pumping tube in the cryostat, becoming 7.6 cm in diameter at room temperature, and a 15-cm-diam diffusion pump. Consideration also should be given to improved transfer of heat in the condenser, heat exchangers, and mixer both by increasing the surface areas and by avoiding poor thermal conductors such as Pb-Sn solder. In particular the poor thermal contact generally provided by soft-solder joints should not be overlooked.

The mixer of the larger refrigerator (device I) of Fig. 2 will operate continuously and stably from  $T < 0.020$  K to  $T > 2$  K, i.e., over 2 decades in temperature. Even higher temperatures can be obtained by placing a thermal impedance between the experiment and the mixer. Sufficient working space is available in this cryostat to include three to five different experiments in each run. Hence some 70 electrical leads have been provided, the vacuum feedthrough being made from Stycast 2850 GT epoxy, as shown in Fig. 2.

These refrigerators can of course be adapted to unusual environments. If a large, time-dependent magnetic field is necessary, for example, the mixer can be fashioned from epoxy using a brush of wires<sup>7,8</sup> to provide thermal contact to the liquid He.

## CONCLUDING REMARKS

In conclusion let me repeat that the lack of numerical detail in this discussion has been intentional, since your imagination should be free to adapt the dilution process to your own research. With modest refrigeration requirements you can expect, with little effort, temperatures of about 0.05 K using a coaxial-tube heat exchanger. Addition of one or two copper exchangers will result in temperatures of 0.03 K and below. Relatively more effort is required to obtain large refrigeration capacity, especially if this capacity is to extend to the lowest possible temperatures.

The dilution refrigerator competes with the technique of adiabatic demagnetization. The relative advantages of the dilution process are lower initial cost, environment free of magnetic fields (of importance, for example, in work on superconductors), and no constraint on the physical size or shape of the cryostat. The advantage of adiabatic demagnetization is that temperatures of about 0.01 K are more easily obtained. Whether the *continuous* refrigeration provided by dilution is an advantage depends on the time-integrated heat load delivered by your experiment. For example, the dilution refrigerator would be more suitable for measurements of thermal transport properties.

Finally, I have avoided reference to single-shot dilution refrigerators<sup>9</sup> simply because I have little experience or interest in or use for these devices.

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## Uses of Dilution Refrigerators

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With the advent of the  $^3\text{He}$ - $^4\text{He}$  dilution refrigerator ultralow temperature research is finding its way into more and more laboratories. It is undergoing an expansion similar to that which took liquid  $^4\text{He}$  from a laboratory curiosity to a common laboratory commodity and to its present use in rockets, accelerator programs, biology research, etc. Some of the reasons for this ultralow temperature expansion lie in the inherent advantages of dilution refrigeration over adiabatic demagnetization. Most of the experiments we will discuss depend on these advantages for their success. Some of these advantages are: a dilution refrigerator can operate in the presence of a magnetic field, no magnetic materials are needed near the experiment, it is simpler to build and operate, no big magnets are required, the total system cost is lower, it provides continuous refrigeration, it provides good thermal contact to the experiment, it is convenient, and it provides long-term temperature stability.

This report will discuss some of the uses to which dilution refrigerators have been put, as well as the techniques for using them in an actual experimental situation.

### SOLID STATE STUDIES

#### Dilute Magnetic Impurities in Nonmagnetic Hosts (The Kondo Effect)

Early experiments using dilution refrigerators as research tools studied the magnetic susceptibility and electron scattering associated with dilute iron impurities in copper.<sup>1</sup> In such systems one sees at high temperatures the paramagnetic, free-spin susceptibility of the iron impurities. As the system is cooled, the susceptibility flattens out and becomes almost temperature independent. The Fe spins in Cu, when measured at 0.04 K have a susceptibility down by about 3 decades from their free-spin value. This small susceptibility is attributed to a compensating cloud of oppositely polarized conduction electrons which condenses around the magnetic impurity at very low temperatures.

The Kondo effect also explains the ancient mystery of the resistivity minima exhibited by so many dilute alloys and even "pure" metals near  $^4\text{He}$  and  $\text{H}_2$  temperatures. Of course, above  $\text{H}_2$  temperature, lattice scattering causes the well understood resistivity increase; at lower temperatures the increase is caused by enhanced scattering due to the spin-compensating cloud. It is found that below a certain temperature ( $\approx 1$  K for Fe in Cu and 0.1 K for Cr in Cu) the impurity scattering is saturated and exactly temperature independent.

Associated with the compensating cloud formation, which occurs over 1 to 3 decades as the temperature is lowered, is a specific heat anomaly, first studied for Cu-Fe by Frank, Manchester, and Martin.<sup>2</sup> Later the dilution refrigerator allowed the cooling of 200-g samples of Cu-Fe and Cu-Cr alloys to 0.04 K.<sup>3</sup> The excellent thermal isolation

afforded by a graphite holder attached to the refrigerator kept heat leaks below 1/2 erg per minute into these large samples. These low heat leaks were necessary, because the 200-g samples had a specific heat which was less than that of  $10^{-3}$  cm<sup>3</sup> of <sup>3</sup>He and because accurate measurements are required, since the impurity specific heat is the difference between the specific heats of the measured dilute alloy and a pure sample.

The susceptibility measurements also relied on the inherent advantages of dilution refrigerators for their success. Only the Cu-Fe alloy, was in the susceptibility coils and no magnetic materials were near the coils (except the Pyrex, whose temperature is held at a constant 1.2 K). Since dilute Cu-Fe alloys have a susceptibility which is down by a factor of  $10^4$  from that of cerium magnesium nitrate (CMN), even a distant salt pill used for cooling could lead to erroneous susceptibility measurements. The cryogenic arrangement of Ref. 4 is shown in Fig. 1.

Both the resistivity and the susceptibility measurements were made with a mutual-inductance bridge designed by Daybell<sup>4</sup> to make possible precise determinations of both the real and imaginary parts of a mutual inductance containing a metallic sample. Alternating-current susceptibility measurements were made on laminated samples. In this way power dissipation in the alloy could be kept below 1 nanowatt.

The absence of leads to the sample from warm temperatures down into the 40-millidegree temperature region was highly desirable. The uncertainties caused by the strains and the solvent oxidation and precipitation introduced in drawing the alloys into the long, fine wires needed in more conventional techniques are avoided. An accuracy of better than 1% can be obtained with this technique, and its precision can be considerably better than 0.3%. A number of samples can be measured on one experimental run.

Kondo systems are subject to measurement error associated with small impurity levels in or on the samples and effects of impurity-impurity interactions. Experimentally and theoretically we expect gross deviations from low-concentration behavior if  $c > 50 T_k$ , where  $T_k$  is the Kondo temperature and is in parts per million.

### Mossbauer Effect

Early experiments studied the Mossbauer effect below 1 K and even developed <sup>57</sup>Co in Fe as a thermometer.<sup>5</sup> This work, done above 0.35 K, used a <sup>3</sup>He evaporation refrigerator for continuous refrigeration. Later work<sup>6</sup> used demagnetization transient cooling to 0.025 K for studies on iron salts. The dilution refrigerator combines the advantages of both methods to provide ultralow temperatures continuously.

Investigators at the Technical University of Helsinki<sup>7</sup> have studied numerous Mossbauer isotopes (<sup>57</sup>Fe, <sup>169</sup>Tm, <sup>197</sup>Au, and <sup>151</sup>Eu) in many chemical environments to study magnetic transitions, hyperfine fields, and  $\beta$ -decay mixing ratios. The problems of thermal contact between the radiation heated source or absorber materials have been handled by this group by locating a very-large-contact-area sample foil or powder in the mixing chamber of a dilution refrigerator. This has allowed work down to about 40 mK. The large area of contact available with the inclusion of a sintered Cu disk in the mixing chamber will, in the future Mossbauer experiments, provide even better contact between the low temperature helium and the sample. A beryllium backing for an absorber foil can provide contact with the Cu and, at the same time, be transparent to  $\gamma$  rays.

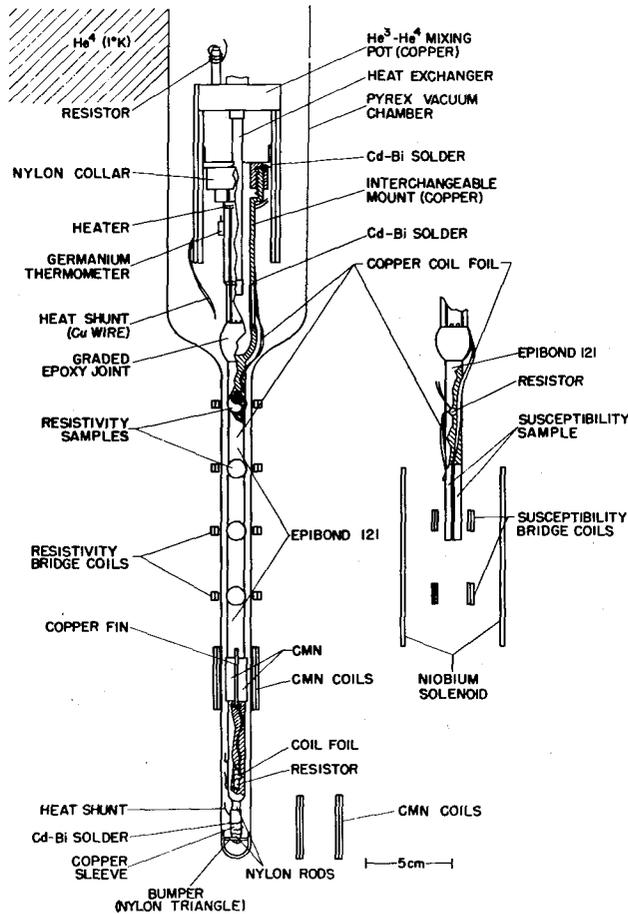


Fig. 1 - Experimental arrangements for resistivity and susceptibility measurements. Thermal contact between the refrigerator and the sample is maintained by the pressure of the nylon collar on the vertically slitted threaded section, then through Cu wires going to the samples. More recent experiments at lower temperatures (14 mK) use spot welds ( $R \leq 10^{-5} \Omega$  at  $N_2$  temperature) in place of the Cd-Bi solder joints for Cu-to-Cu thermal contact. The Speer and germanium resistor thermometers permitted measurements from 0.04 to 40K.

## BOLOMETER APPLICATIONS

Very low temperatures have been used by Daybell, Pratt, and Steyert (unpublished) for the operation of very sensitive infrared detectors. The very thin graphite films were held at about 0.05 K, at which temperature their very low thermal conductivity, combined with a large change in electrical resistance with temperature, results in a very large change in electrical resistance for a very small infrared power input. An additional benefit from ultralow temperature operation of "dielectric" bolometers is the rapid response time associated with their very small specific heat at these temperatures.

These devices, however, presented a problem in that they tended to be transparent and nonabsorbent of the infrared photons of interest. This problem could be overcome by the use of nonbolometric photoconductive devices in which a small bolometer specific heat was not important and a large piece of material was acceptable. In In-Sb detectors a narrow-band frequency response is found<sup>8</sup> associated with transitions between the impurity states located  $10^{-4}$  V below the conduction band and the Landau levels of the conduction band. The resonance frequency is tunable with an applied magnetic field.

## NUCLEAR PHYSICS STUDIES

Experiments aimed at the study of nuclear decay properties are almost as old as ultralow temperatures themselves.<sup>9</sup> The most famous such experiment, and probably the most fundamental low or ultralow temperature experiment, is that of Wu, Ambler, Hayward, Hoppes, and Hudson,<sup>10</sup> which showed the nonconservation of parity in the weak interaction. They found that electrons are emitted preferentially opposite to the direction of nuclear alignment. Positrons are emitted in the opposite direction. Experiments are presently under way to study other symmetry properties. At Los Alamos parity-nonconservation  $\gamma$  decay has been studied, and the  $\beta$  decay of aligned nuclei is being looked at in greater detail.

Other experiments, which we will here call structure experiments, have looked at the magnetic moment,  $\beta$ -decay matrix elements, spin and parity assignments of nuclear levels, and multipole admixtures in  $\gamma$  decay. These experiments reflect primarily the validity of the nuclear models in the description of nuclear structure. They do not directly bear on the poorly understood nature of nuclear forces as do the symmetry experiments.

The structure experiments generally require much lower temperatures than the symmetry experiments; however, these measurements do not require exceptional statistical accuracy and thus can be completed in a short time with a limited number of counts. An accuracy of 1 or 0.5% is generally quite sufficient.

The degree of orientation and hence temperature required can best be understood by considering the form of the angular distribution of a nucleus orientation along a symmetry axis:

$$W(\theta) = 1 + a_1 f_1(\beta) P_1(\cos \theta) + a_2 f_2(\beta) P_2(\cos \theta) + a_3 f_3(\beta) P_3(\cos \theta) \\ + a_4 f_4(\beta) P_4(\cos \theta) \dots$$

Here  $P_k(\cos \theta)$  is the Legendre polynomial of the angle away from the alignment axis, and  $f_k(\beta)$  is the  $k$ th nuclear orientation moment of the oriented parent level. The moment  $f_k(\beta)$  is a simple function of  $\beta$ , namely, the hyperfine splitting  $\mu H/I$  divided by the thermal energy  $k_B T$ ;  $H$  is the effective hyperfine field at the nucleus, generally the 100 to 1000 kOe produced by the atomic electrons of the radioisotope and its chemical environment. The

$a_k$  coefficients contain the nuclear data of interest. For symmetry experiments the  $k = 1$  term is generally of interest, and for structure experiments all the terms are of interest. Note that  $f_1$  becomes large for modest  $\beta$  values; for instance,  $f_1(1)$  is typically 70% of its maximum value,  $f_1(\infty)$ . To get large  $f_4(\beta)$  values, however, much larger  $\beta$  values are required. The value  $\beta = 3$  must be reached to get  $f_4(\beta) / f_4(\infty) \approx 0.7$ ;  $f_4(1) / f_4(\infty)$  is only  $\approx 0.1$ . This can be the difference between a 15 mK dilution experiment for a symmetry measurement and a 5-mK demagnetization for a structure experiment.

### Structure Experiments

To determine nuclear properties the structure experiments measure the  $a_k$  coefficients and  $\mu H$ . If  $H$  is known, we can deduce  $\mu$ , and vice versa. The  $a_k$  coefficients are a function of level spins, decay, and multipolarities (hence level parity). In a given decay if only one spin, or multipolarity, is unknown, we can deduce it from one measured  $a_k$  value. If, however, in a given decay cascade more than one quantity is unknown, we must measure more than one  $a_k$  value to arrive at a unique result. Generally, in studying gammas emitted by aligned nuclei or daughter of aligned nuclei, we measure  $a_2$  and  $a_4$ . This takes measurements at two angles (generally 0 and 90 degrees) at high and low temperatures to determine the two quantities, and requires quite high  $\beta$  values to measure  $a_4$ .

To get sufficiently low temperatures at Los Alamos, CMN is cooled to 0.04 K in a field of 1.5 kOe and adiabatically demagnetized. To provide the required  $10^4 \text{ cm}^2$  area of contact between the coolant, the CMN, and the radioactive source, equal volumes of minus-80-mesh CMN and 1- $\mu\text{m}$  Cu powder are mixed and pressed together under 40,000 psi. Many 99.999% Cu wires are pressed with the metal mixture to provide contact to the source\* (Fig. 2). The temperature is measured to 0.1 mK by  $^{60}\text{Co}$  or  $^{54}\text{Mn}$  in an iron foil.

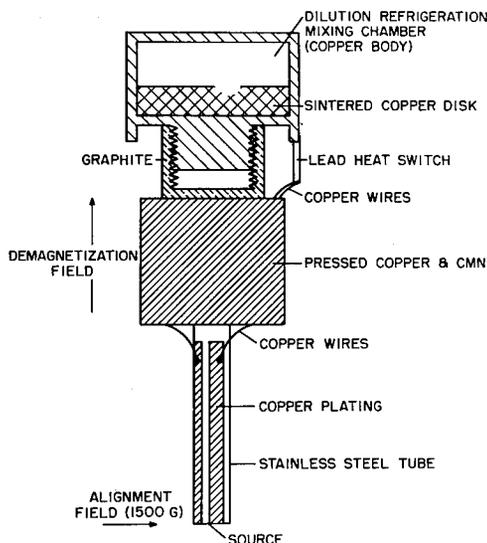


Fig. 2 - Experimental arrangement for the use of the Cu-CMN disk to provide large-area Cu-CMN contact

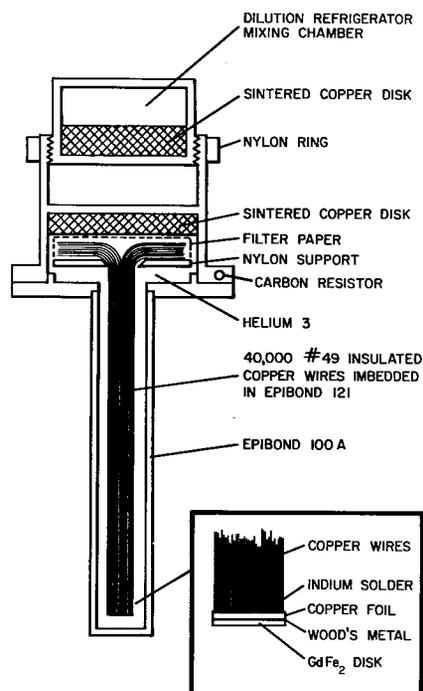
\*This technique, developed by H. Sheinberg, produces a machinable, copperlike disk which has a resistivity of  $\approx 10^{-5} \Omega\text{-cm}$  at room temperature. Preliminary work with the material yields cooling to 8 mK with about 1 erg/sec heat input into the magnetized iron foil. Temperatures below 10 mK are maintained for 3 hours. Much lower temperatures will be available when the heat link between the CMN-Cu and the source copper is improved. Earlier work with a sintered-Cu disk impregnated with a CMN solution was used to study  $^{198}\text{Au}$  aligned in an Fe foil at 5 mK. Temperatures below 10 mK were held for only 1 hour however.<sup>11</sup>

## Symmetry

If parity were conserved, there would be no  $P_k(\cos \theta)$  terms for  $k$  odd. However, Wu et al.<sup>10</sup> have shown for electrons emitted by aligned nuclei  $a_1 = \alpha V/C$ ,  $\alpha \approx 1$ . By looking at  $\beta$  particles with Si(Li) detectors operated at 77 K, it should be possible to measure  $\alpha$  much more accurately. By looking at  $\beta$  particles and  $\gamma$  rays in coincidence, possible time reversal invariance can be measured. Ambler, Hayward, Hoppes, and Hudson made rough checks of time reversal invariance in the  $\beta$  decay of polarized  $^{52}\text{Mn}$  in 1958.<sup>12</sup>

A small parity nonconserving component to  $\gamma$  decay can be seen by the presence of a small  $a_1$  in the angular distribution of  $\gamma$  rays emitted by aligned nuclei.<sup>13</sup> Measurements of this  $a_1$  have been made on a  $^{159}\text{Gd}$ -in- $\text{GdFe}_2$  source (Fig. 3) cooled to 28 mK.<sup>14</sup> Two Ge(Li) counters were held in a fixed position while a 1500-Oe field rotating at 0.01 Hz rotated the  $\text{GdFe}_2$  ferromagnet polarization. The counting rate, measured as a function of polarization direction, yielded  $a_1$ . Measurements at 28 mK were conducted over a continuous 30-hr period to provide a statistical accuracy of about  $2 \times 10^{-4}$  in the coefficient of  $P_1(\cos \theta)$ .

Fig. 3 - Thermal contact for a parity experiment in a rotating magnetic field. Eddy-current-free contact is provided by large area contact through  $^3\text{He}$  bath.



Combining the Kondo effect and nuclear alignment provides a very accurate  $^{54}\text{Mn}$ -in-Cu thermometer for the millikelvin temperature range.<sup>15</sup> Other  $\gamma$ -anisotropy thermometers such as  $^{60}\text{Co}$  or  $^{54}\text{Mn}$  in Fe provide better than 1% absolute accuracy.

## HELIUM STUDIES

Low temperature physicists and theoreticians have always been interested in the ultralow temperature properties of the two helium isotopes and their mixtures. The number of experiments already conducted using dilution refrigerators to study the ultralow

temperature nature of the helium isotopes is too large to enumerate here. The thermal contact problems are not quite as severe as in nuclear studies, in which one must contend with sample self-heating. However, the large specific heat of pure liquid  $^3\text{He}$  requires a large surface area contact to the refrigerator, if rapid  $^3\text{He}$  cooling is desired.

Some of the helium properties measured are the thermal conductivity and susceptibility of solid  $^3\text{He}$ , the phase separation curve of mixtures under pressure,  $^4\text{He}$  film flow, osmotic pressure of mixtures, and the cooling achieved by the adiabatic compression of liquid  $^3\text{He}$  to the solid.

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## **Invited Paper**

### **SESSION IV**

Friday Afternoon, April 24, 1970

Chairman: E. A. Edelsack  
Office of Naval Research



## Experiments of the Future at Ultralow Temperatures (< 0.3 K)

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I certainly do not feel able to foresee the full impact on science in the 1970's of the major scientific and technical developments in ultralow temperature physics which have indeed taken place in the 1960's. Anything I say will be qualified by my own limited knowledge and by my own interests and personal judgment. In this sense a paper like this one cannot help but be rather subjective - more or less a matter of personal opinion. However, with that apology, let us begin.

During the 1960's there were four areas of *technical* advance which stand out in my mind. Two are concerned with cooling using helium, and two are concerned with superconductivity. The first is the He<sup>3</sup>-He<sup>4</sup> dilution refrigerator, invented by Heinz London and developed by a number of workers throughout the world. This refrigerator continuously produce temperatures well below 10 mK (in a recent conference it was reported that B. S. Neganov in Dubna has achieved 5.5 mK in continuous operation) without the use of magnetic fields and only rather minor sensitivity to them. The low temperature is produced in a fluid and hence can be piped around as required by the experiment. The part of the apparatus where the cooling is produced can be rendered entirely electrically nonconductive. Thus rapidly changing magnetic fields of considerable size can also be tolerated. The flow rate of He<sup>3</sup> in the refrigerator is relatively temperature independent, although the cooling power per particle is proportional to T<sup>2</sup>. Thus the refrigeration capability of the dilution refrigerator does approach zero as T approaches zero, but the T<sup>2</sup> dependence is much less rapid than the  $e^{-L_0/kT}$  dependence of the He<sup>3</sup> evaporation refrigerator. A temperature limit of 4 to 5 mK is set on the common type of dilution refrigerator by intrinsic helium properties such as viscosity and thermal conductivity. However, with the possible exception of Neganov's latest result as quoted above, the actual performance of dilution refrigerators, including our recent model which reaches 8 mK, is disappointing compared with the possibilities. Much work remains to be done before the dilution refrigerator reaches its full capabilities, but for now a temperature of 10 mK on a continuous basis should be within the reach of most laboratories.

The second technical advance I have in mind also concerns helium, but its application at this moment is for most laboratories more potential than actual. This is true even though it is, in my opinion, the most powerful means of cooling available in the range of roughly 2 to 50 mK. I refer here to the method of adiabatic compressional cooling of a two-phase mixture of liquid and solid He<sup>3</sup>, first proposed by Pomeranchuk in 1950 and executed by Anufriyev in 1965. It has subsequently been developed extensively in my laboratory and at Cornell University if not elsewhere. Here cooling is achieved by a mechanical motion which converts motionally ordered liquid He<sup>3</sup> into spin disordered solid He<sup>3</sup>. Over a good part of the useful temperature range the associated entropy change is of order R, so the resultant refrigeration capability of about RT per mole converted is certainly about as great as one might expect for any system. Further the end product is highly-spin-ordered solid He<sup>3</sup>, which is itself of considerable interest. There is also an important relation to thermometry, which I will discuss later.

The third technical advance is concerned with the area of weak superconductivity, words which are used in describing the Josephson effects. The impact of this advance on science at ultralow temperatures has not been particularly widespread, even though the possibilities which it has opened are great. In our case it was the development by J. E. Zimmerman of the stable point-contact Squid, which could be preadjusted at room temperature and then operated at 4.2 K, which enabled us to enter the field. Using the so-called RF-biasing scheme, technically simple devices can be operated as magnetometers which are so sensitive that they can be used to measure the low-field static nuclear magnetism of metals. Other extremely weak magnetic effects in dilute alloys have also been measured with precision, and we can look forward to discoveries not only in this area but also wherever the voltage sensitivity of the magnetometer can be used, such as in the measurement of small electrical resistances and thermoelectric power. As stable preadjusted resistive Squids, or their counterpart, are developed, we can expect further applications of importance in ultralow temperatures.

The fourth technical advance is the development of materials which can be used to construct high-field (up to 100 kilogauss and beyond) superconducting solenoids. Multi-filament Nb-Ti conductors in a copper matrix are now available which allow good results to be obtained predictably, and here we refer both to the attainment of high fields when the solenoid is activated and low remanent fields when deactivated. Nb<sub>3</sub>-Sn conductors are still relatively difficult to use, but the technology is advancing, and we look forward to a day when they too can be used on a simple laboratory basis. The high-field solenoids do not entirely replace iron magnets, but they do facilitate electronic and nuclear demagnetization. The high fields also fit in with the field-insensitive process of dilution refrigeration to provide the means for achieving simultaneously and simply both high magnetic fields and ultralow temperatures.

Now that I have cataloged what I regard as the major technical advances in low temperature science in the 1960's, let me try to look ahead to ultralow temperature science in the 1970's. I shall take the realistic position that temperatures down into the millikelvin range and fields to 100 kilogauss can be readily achieved, as can measurements of extremely weak magnetic and electric phenomena. Doubtless I have omitted some other important capabilities, but those mentioned will serve my purpose, which is to show that much interesting and important science lies ahead of us.

For the purpose of this conference an ultralow temperature (ULT) is defined to be one below 0.3 K, that temperature which can be reached readily by a He<sup>3</sup> evaporation refrigerator. The following are some areas which I think are now, or will be in the future, dominant in ULT science.

1. Quantum fluids and solids. Here we are concerned at the least with pure He<sup>3</sup>, a strongly interacting Fermi fluid; solid He<sup>3</sup>, particularly in the range of nuclear ordering; solutions of He<sup>3</sup> in superfluid He<sup>4</sup>, weakly interacting fermions in superfluid Bose liquid; superfluid He<sup>4</sup>; and both He<sup>3</sup> and He<sup>4</sup> in nearly-two-dimensional systems. An important two-dimensional system will be He<sup>3</sup> atoms occupying a surface layer in superfluid He<sup>4</sup> and next to vacuum or pure He<sup>3</sup>. At least in the case of the Fermi systems one must reduce the temperature far below 0.3 K to see effects of statistical degeneracy. The behavior of such systems must be regarded as absolutely basic in the physics of condensed matter.

2. Polarized or oriented nuclei. Definition of the direction of the nuclear spin is of great importance in a variety of scattering, capture, and decay experiments aimed both at cataloging nuclear properties and at studying the basic laws of interaction in both nuclear and particle physics. In particular polarized nuclei will be important in studying nonconservation of parity and time reversal invariance in nuclear physics. Polarized He<sup>3</sup> nuclei will be important, as they provide a source of polarized neutrons, in providing a means in particle physics to test for differences in basic interactions between polarized protons and neutrons.

3. Magnetism. It is argued that if one wants to seek an understanding of magnetic phenomena, then it is best to study systems for which the magnetic ordering interactions are weak and the corresponding critical or characteristic temperature is low. At low enough temperatures other effects beside the magnetic ordering will be smaller and the effective magnetic interactions will be purer. Until recently these arguments have been outweighed by practical considerations, but now (a) the low temperatures are available and (b) means for measuring very weak magnetic phenomena at very low measuring *field* and with high precision are available. Actual examples are certain dilute alloys, solid He<sup>3</sup>, and dilute magnetic crystals. Aside from presently known magnetic systems, we expect the high detection sensitivity and the low temperature will lead to the discovery of a variety of new and interesting weakly magnetic phenomena, many of which are beyond our present knowledge or intuition.

4. Noise. Now that ultralow temperatures are available, we should see if it is not possible to improve meaningfully the sensitivity of systems by reducing Brownian-motionlike thermal noise. This should be important in systems where the desired effect is obtained, as in parametric amplification, by varying the value of some parameter. We suggest as important applications (a) bolometric detection of infrared radiation with application to infrared astronomy and (b) detection of very weak voltages by means of devices based on weak superconductivity. There also exists the possibility of noise thermometry, but this will probably not have the importance of the above. Recently important advances have been made in cryogenic (4 K) amplification using germanium junction FETs. There seems to be no point at present in reducing further the ambient temperature for these devices, but they may serve importantly to allow the reduction in thermal noise mentioned above to be utilized.

5. Electrical phenomena. By use of the devices based on weak superconductivity it will now be possible to measure the electrical resistance and thermoelectric power of metals and alloys at ULT. This has immediate application to studies of dilute magnetic alloys and to low temperature thermometry.

6. The third law of thermodynamics. Here we are concerned with basic questions about the approach to absolute zero. Since no magnetic fields are required with the dilution refrigerator or the adiabatic compression device to produce low temperatures, an extremely low ambient magnetic field can be achieved to provide the necessary conditions for the transition to superconductivity of ultralow temperature superconductors. Thus we may seek answers to basic questions regarding the ultimate state of matter as the temperature approaches zero, whether superconductive, or magnetic, or whatever. Nuclear ordering will also be important scientifically, as will, say, the ultimate state of the neutral Fermi liquids.

7. Thermometry. This subject is fascinating in principle though difficult in practice. It seems that every cooling method has associated with it some means for thermometry. Thus with the magnetic cooling method we have the magnetic thermometer. We also have well-established methods, though difficult to perform, for determining the Kelvin temperature. Here we look forward to the application of Giauque's nonheating method. For the dilution refrigerator we have the analog of the thermocouple, with the analogs of the junctions being the concentrated He<sup>3</sup>-solution interfaces and the analog of the thermoelectric emf being an osmotic pressure difference across a superleak. The adiabatic-compression cooling method, working along the melting curve of He<sup>3</sup>, provides a rather strong hope for a nonmagnetic method to obtain a thermodynamic temperature. Using Clapeyron's equation, rather than the melting curve itself, we can find temperature regions in which our confidence in the value of the solid entropy is as good as our confidence in the entropy of a nearly ideal gas for the vapor pressure analog and in which the liquid entropy is relatively small. The molar volume changes are also relatively well known. The method should apply from roughly 5 to 40 mK. The nuclear magnetism thermometer, though not quite as novel as the above, will come into its own with the application of the new devices

highly sensitive to magnetic flux. And finally, these same devices may bring us right back to room temperature thermometry with the use of thermoelectric emfs for temperature measurement.

8. Thermal contact. This is a subject which is crucial in the application of low temperatures both to cooling other substances and to cooling just the thermometers used to measure the temperatures. Interfacial cooling has been dominated by the Kapitza boundary resistance, but there is at least one case - powdered CMN and pure  $\text{He}^3$  - where this resistance is bypassed. And there is another case - contact to dilute solutions - in which there is probably even more resistance. At the same time both  $\text{He}^3$  and solutions at the lowest temperatures have very high intrinsic thermal conductivities. Other kinds of contact are also important, such as the contact between the electrons and the nuclei in a system where the nuclei are to be polarized, their heat capacity in a magnetic field utilized, or their magnetism measured.

9. Materials. The new extremes of low temperature and magnetic field will have an important effect on various fields of solid state and materials science. Resonance experiments on dilute alloys with low Kondo temperatures, experiments of various types on new materials of extremely high purity, and studies of amorphous solids and in particular high polymers with varying degrees of crystallinity are all examples.

10. Refrigerators, cryostats, and cooling methods. In a certain sense the result of an advancing ULT technology will be to change the definition of the ULT region from temperatures below 0.3 K to temperatures below 0.003 K. Certainly the basic cooling methods are rather far advanced. What is required is to simplify the technology, simplify the cryostats, and make the ULT environment more accessible. In the 1970's substantial progress will be made in this area.

11. Lower temperatures. Up to this point I have stayed within those limits of temperature which are accessible by the new cooling techniques using helium. For lower temperatures still, it will be necessary to return to the methods of adiabatic demagnetization, and in particular to nuclear demagnetization. Great achievements have already been made in this area, but only a few laboratories have contributed thus far. No doubt the  $\text{He}^3$ - $\text{He}^4$  dilution refrigerator and the adiabatic compression method will be instrumental in providing an intermediate temperature stage for nuclear demagnetization. Thermal contact to the demagnetized sample will be a problem, as will the necessity for a large magnetic field.

That completes my list. To a physicist, or at least to this one, lists of past accomplishments or other lists of what might be done in the future, however necessary and important, are rather sterile things. Implicit within the lists are many really beautiful ideas, concepts, and experiments; and I would like to mention some of them in the remainder of this paper.

First, let us talk about helium. It is remarkable that a mixture of some  $\text{He}^3$  atoms in  $\text{He}^4$  at temperatures well below 1 K perhaps may be described in terms of a beautifully simple model. Pure  $\text{He}^4$  we think of as inert, sometimes as a mechanical vacuum. But really "ether" is a better word for it, for when a  $\text{He}^3$  particle moves through  $\text{He}^4$ , its energy depends on the local superfluid velocity and density. On the average,  $\text{He}^3$  atoms in the "ether" attract one another. This can be understood from simple thermodynamics and is rooted in the larger volume occupied by a  $\text{He}^3$  atom than a  $\text{He}^4$  atom. On the basis of an idea originally proposed by Bardeen, Baym, and Pines but worked out more quantitatively by McMillan the  $\text{He}^3$  atoms interact with one another by means of the mutual interaction of their velocity fields, or of their Feynman-Cohen backflows. Some of these ideas are pictured in Fig. 1. The  $\text{He}^3$  atoms are Fermi particles, and at low temperatures strongly degenerate statistical behavior is observed.

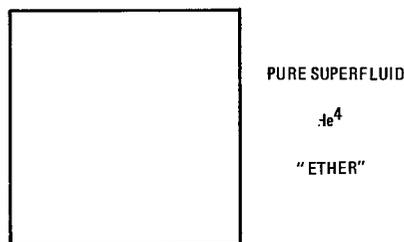
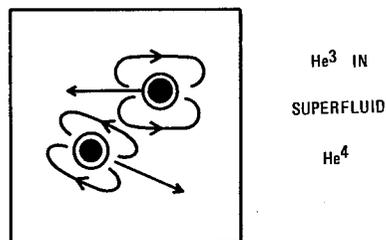


Fig. 1 - Pictorial models of superfluid  $\text{He}^4$  and interacting  $\text{He}^3$  atoms in superfluid  $\text{He}^4$



Pure liquid  $\text{He}^3$  also has many interesting properties at low temperatures, many of them deriving from the strong  $\text{He}^3$  interactions and the ordering of the liquid in translational states. But let us talk about the equilibrium between liquid and solid. This has some remarkable properties, not the least of which is the potential for thermometry. Consider the Clapeyron equation,

$$\frac{dP}{dT} = \frac{\Delta s}{\Delta v},$$

where  $T$  is the Kelvin temperature,  $P$  is the two-phase equilibrium pressure,  $\Delta s$  is the difference in molar entropies of the two phases, and  $\Delta v$  is the difference in their molar volumes. This equation is the basis of vapor-pressure thermometry, a kind of thermometry in which we have a lot of confidence; and the seat of our confidence is that in the equation  $\Delta s$  and  $\Delta v$  are very nearly the entropy and molar volume of an ideal gas, with the corrections for the condensed-phase entropy and volume and for the gas nonideality being relatively small. The ideal-gas entropy is based on concepts in which we have a deep belief. There is a similar situation for the liquid  $\text{He}^3$ -solid  $\text{He}^3$  phase equilibrium above 5 mK and below, say, 30 to 40 mK. The liquid has ordered into translational states and has an entropy decreasing as  $T$ , while all solid disorder has been quenched except that due to the spins. The latter is  $R \ln 2$  on the basis of a  $\text{He}^3$  nuclear spin of  $1/2$ , and this is a computation in which we have a lot of confidence. The situation is shown in Fig. 2. Even though the liquid entropy may be subject to error, the error thrown into the temperature scale obtained by integrating Clapeyron's equation from some known  $T$  is small. This approach can be used to calibrate a thermometer based on the nuclear Curie law and should lead to a rather accurate approximation to the Kelvin temperature.

Let us turn now to a different subject with the same system - liquid and solid  $\text{He}^3$ . The  $\text{He}^3$ - $\text{He}^4$  dilution refrigerator can be used to cool the  $\text{He}^3$  to 10 to 20 mK. At this temperature the liquid is highly ordered, motionally ordered, as may be seen from the low liquid entropy in Fig. 2. Now a mechanical device can be used to convert, without frictional heating, liquid to solid. The temperature will fall, and the end product will be highly ordered, highly spin ordered, solid  $\text{He}^3$ . If this process is carried out in zero field, then an ordered nuclear antiferromagnet is produced whose magnetization, for example, can be measured by the superconducting devices. If the process is carried out

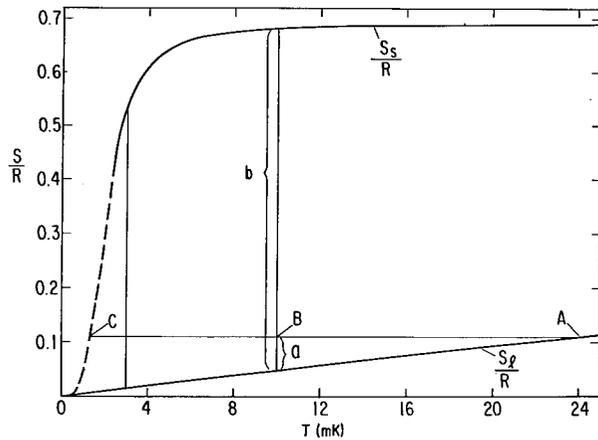


Fig. 2 - Entropy of liquid and solid He<sup>3</sup> as functions of temperature with application to adiabatic compressional cooling

in a large enough field (say 50 to 100 kilogauss), then the He<sup>3</sup> produced will be bulk polarized. This leads to other interesting possibilities.

Turning back to thermometry for a moment, let us consider the thermocouple. An ordinary thermocouple is shown on the left in Fig. 3. If the junctions between metals A and B are at different temperatures  $T_1$  and  $T_2$ , then a thermoelectric emf is produced. The resultant current flow can be nulled by use of a potentiometer. Actually the thermocouple will be an important device at ULT and basic measurements of thermoelectric power at ULT will be possible. If A were Cu(Fe), B were a superconductor, and the potentiometer battery were replaced by some turns of superconducting wire introduced into a superconducting device, then the resultant currents could be measured with precision well into the millikelvin range. A nonconducting helium analog to the thermocouple is shown on the right side of Fig. 3. The analog to a junction is the interface of concentrated He<sup>3</sup> with a saturated dilute solution, the analog of the potentiometer is a superleak, and the analog of the emf is an osmotic pressure difference, here indicated as a height. Owing to the He<sup>4</sup> film covering everything, the superleak shown on the bottom may be unnecessary in practice. This thermocouple should be insensitive to even rather strong magnetic fields.

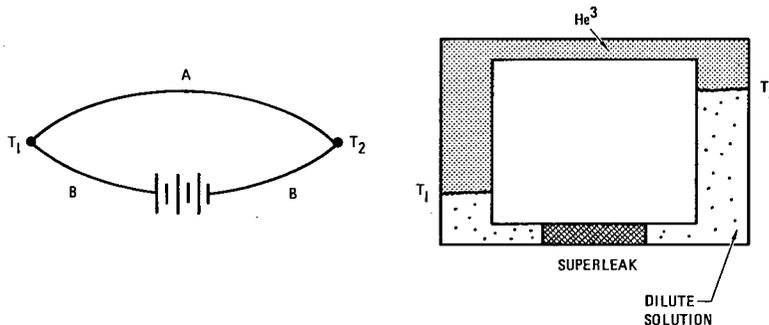
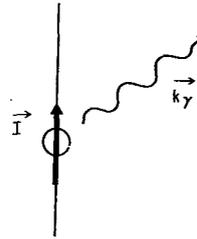


Fig. 3 - Two types of thermocouples

Now let us turn to another subject. I have always preferred to talk about physics at low temperatures rather than low temperature physics. And that explains some of the disorganization of this paper, which has to do with experiments (no qualifications) of the future.

Suppose that I am interested in finding out whether or not the nuclear forces exactly conserve parity. For example suppose that a given nuclear state has an angular momentum of 1 unit and is said to have even parity and that it can decay by emission of a  $\gamma$  ray to a state of zero angular momentum and even parity. Then we expect the  $\gamma$  ray to be characteristic of magnetic dipole radiation. On the other hand, suppose that the nuclear forces do not entirely conserve parity, though they do angular momentum. Then the actual nuclear wavefunction is mainly  $1^+$  but could have an admixture of  $1^-$ . The actual  $\gamma$  radiation would then reflect this admixture by having electric-dipole as well as magnetic-dipole character. To look at this quantitatively we can polarize the nucleus, and here is where low temperatures and high magnetic fields come in. For by measuring the correlation between the direction of emission of the  $\gamma$  ray and the direction of polarization of the nuclear spin (that is, by finding that part of the decay amplitude that depends on  $\vec{I} \cdot \vec{k}_\gamma$ , Fig. 4) we can measure indirectly the relative strength of the parity-violating nuclear force. The effects are small, so very long measuring times and the possibility of frequent, or continuous, change of magnetic field direction are important. These are made possible by the new ULT technology. Other experiments with polarized nuclei relate to the question of time reversal invariance, and still others can measure the spin polarization dependence of various kinds of reactions and decays.

Fig. 4 - Emission of radiation from polarized nuclei



I mentioned the possibility of making bulk polarized solid  $\text{He}^3$  by an isentropic process which converts ordered liquid  $\text{He}^3$  to solid. Suppose that we were interested in studying the nucleon polarization dependence of the pion scattering. Up to now it has been possible to do experiments of this sort only with dynamically polarized proton targets. Polarized  $\text{He}^3$  is a possible source of polarized neutrons. It is possible to think of a polarized  $\text{He}^3$  nucleus as a polarized neutron with an unpolarized proton-proton pair as spectators, and by p- $\text{He}^3$  scattering, say, one can determine the percentage of time the neutron is "free." Thus a polarized  $\text{He}^3$  target will provide means for testing the isotopic spin dependence of the pion-nucleon interaction. It will also make possible scattering experiments not previously done (polarized proton and polarized neutron). However, the polarized  $\text{He}^3$  does not have an indefinite lifetime. It should be straightforward to achieve a 75% polarization by converting only a fraction of the liquid  $\text{He}^3$  to solid  $\text{He}^3$  in a field of 100 kilogauss. Dissipation of energy will decrease the polarization, but calculation shows that if  $5 \times 10^4$  ergs/g are dissipated, the  $\text{He}^3$  polarization is reduced only to 50%. This would correspond to over  $10^{10}$  minimum-ionizing particles per square centimeter. Experiments for which this limitation on the number of incident particles is not serious should give interesting results in the future.

Now let us have another change of pace and talk about a problem of magnetism at low temperatures. For many years the properties of magnetic salts have been of great interest; and from the standpoint of critical points the heat capacity has been a very important quantity. The thermal measurements have been very interesting but may show effects of inhomogeneities. Magnetic measurements will also be important. Near an ordering transition they are usually difficult to make, but by using the new superconducting magnetometer technology we will have essentially a static measurement with very high sensitivity capable of measuring the magnetic moment of samples in low fields. An experimental arrangement is shown in Fig. 5. A suitably low field is trapped in a niobium tube which contains a set of astatic niobium coils as part of a flux transformer circuit which is coupled into a superconducting device. It is possible to use small single-crystal samples and hence to reduce inhomogeneities. It would be tempting to hope for samples which would show a size effect, but these are probably too small to prepare. However, we would hope for interesting new results of some significance in the area of magnetic phenomena near critical points.

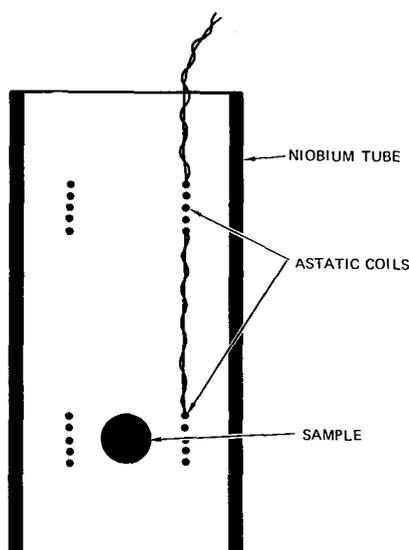
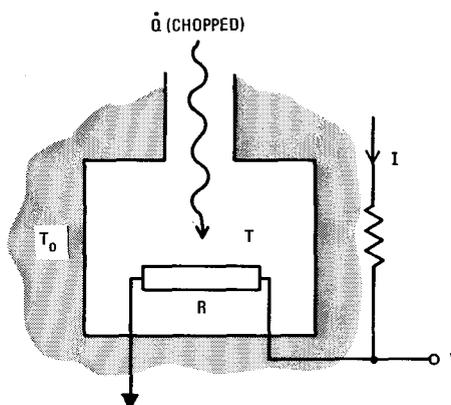


Fig. 5 - Schematic arrangement for magnetic measurements using superconductivity

I will conclude my paper by making some comments about noise. In reducing the temperature from room temperature to 3 K the energy of thermal fluctuations is reduced by 100 times. And on making a further reduction of 10 mK the thermal fluctuation energy is reduced by another 300 times. If thermal noise is a problem, then surely reducing the temperature to the ULT region will play an important role, particularly in parametric amplification. In thinking about this point there comes to mind an outstanding example of how the first factor of 100 (300 to 3 K) was used. Ten years ago infrared astronomy was poorly developed. Now it is providing really important information in astrophysics. The technical advance which made the new measurements possible was the cooling of a semiconducting resistor to helium temperatures. A schematic drawing of the detector is shown in Fig. 6. A direct, constant current dissipates power in the resistance, which then rises in temperature above the ambient. Power absorbed from nonelectrical sources, such as infrared radiation falling on the bolometer, then causes the resistance and hence the voltage across the bolometer to change. Noise is introduced not only from current fluctuations but also from temperature fluctuations resulting from the thermal contact with the environment, described by a thermal conductance  $G$ . If the device is biased properly (not critical), the received power equivalent to noise (NEP) is estimated by  $\sqrt{kT^2 G}$  times

Fig. 6 - Physical arrangement for the detection of radiation



the square root of the effective system bandwidth. If  $G$  were to represent a Kapitza thermal conductance, then the NEP varies as  $T^{5/2}$ . The importance of low temperatures for high sensitivity is clear. At a temperature of 50 mK an NEP of  $10^{-15}$  watt/Hz<sup>1/2</sup> is easily possible. Whether or not such a high sensitivity will be immediately useful is another matter.

The superconducting devices will also be important in the realm of noise suppression. Not only do they maintain a very high sensitivity but also they can be made so that thermal noise is the primary noise source. In the case of the voltage-sensitive superconducting device the noise voltage results in a fluctuating frequency. This may also provide some interesting possibilities. We can look forward to many ingenious suggestions in the future.

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## **Contributed Papers**



## Nuclear Cooling Combined with Dilution Refrigeration

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Although the principle and theory of nuclear cooling have been known for about 35 years,<sup>1,2</sup> this technique has not yet become fully practicable as a method of producing ultralow temperatures.<sup>3-6</sup> The reason is easily understood by examining the necessary initial conditions for a successful nuclear demagnetization: a very low temperature, a very high magnetic field, and a very small external heat leak. The principle of nuclear cooling was thus known long before the method became fully feasible in the laboratory. Recent progress in the development of dilution refrigerators and high-field superconducting magnets has, however, changed this situation, and nuclear demagnetization is now on the verge of becoming a tool for research in the submillikelvin region.

In this paper we shall briefly describe the present, unfinished state of an apparatus in which, for the first time, dilution refrigeration and nuclear demagnetization have been combined. Recently we succeeded in cooling 1500 grams of copper to 16 mK in a field of 46 kG by means of our dilution refrigerator; after demagnetizing to a final field of 727 G, equilibrium between the conduction electrons and the nuclear spins was established at 0.62 mK. The nuclear stage remained below 1 mK for 4 hours.

### THE DILUTION REFRIGERATOR

Figure 1 is a photograph of the cryostat showing the <sup>4</sup>He pot and the dilution refrigerator. The <sup>4</sup>He pot has a volume of 1.8 liters and can be filled through a needle valve from the main bath; refilling is normally necessary after 72 hours. The principal parts of the dilution refrigerator are identified in the figure. The machine was designed for a very high circulation rate, resulting in an exceptionally high cooling power  $\dot{Q}$ ; below 25 mK an optimum is achieved with a <sup>3</sup>He circulation rate of 200  $\mu$ mole/sec. Some measured cooling powers are listed in Table 1. When the machine was operated as a single-cycle refrigerator, the temperature of the mixing chamber could be reduced to 7 mK; and by increasing the quantity of <sup>3</sup>He in the system, still lower temperatures could probably be reached in this mode. We are aware of several improvements that will be made in the refrigerator. It is thus likely that the performance figures given in Table 1 can be considerably improved.

A dilution refrigerator with a large cooling power is essential for the operation of our cryostat; the heat of magnetization which must be removed from the nuclear stage within a reasonable length of time is between 300,000 and 1,000,000 ergs!

Figure 2 shows a cross-sectional view of one of our five sintered-copper heat exchangers. For simultaneously obtaining a large surface area and a low flow impedance thin copper sponges of a rather large diameter were used. The packing factor in the sintered pieces is only 41%. To secure good heat contact between the dilute and concentrated sides many thin copper wires were embedded and sintered into the sponges. The

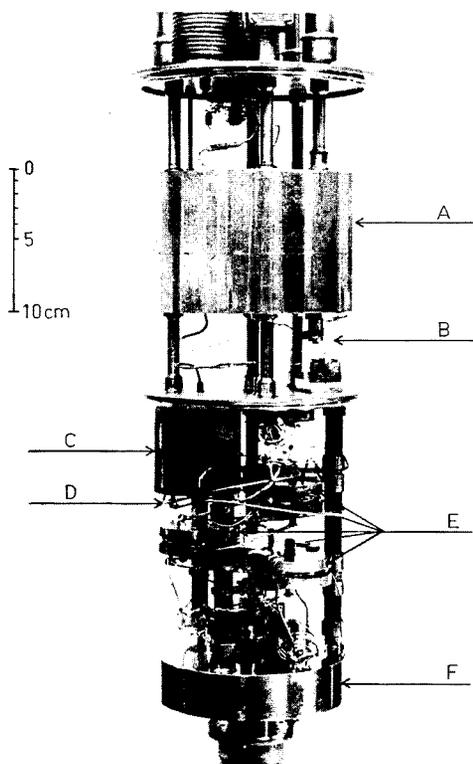


Fig. 1 - The middle part of the cryostat: (A)  $^4\text{He}$  pot; (B) flow-limiting constriction; (C) still; (D) concentric tube heat exchanger; (E) five sintered-copper heat exchangers; (F) mixing chamber

Table 1  
Cooling Power With a  $^3\text{He}$   
Circulation Rate of  $200\ \mu\text{mole/sec}$

T (mK)	$\dot{Q}$ (erg/sec)
11	0
15	10
20	26
30	80
40	155

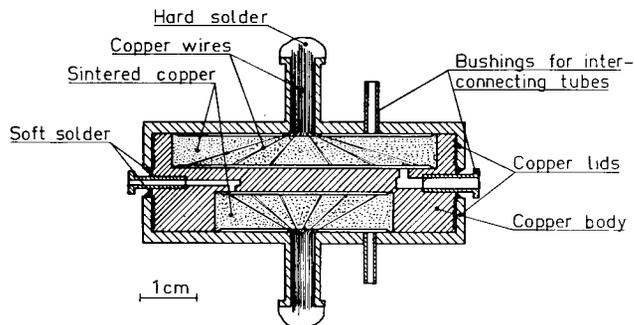


Fig. 2 - Sintered-copper heat exchanger

wires were silver-soldered to the lids which, in turn, were tightly screwed and soft-soldered to the body of the exchanger. The thermal resistance between the sponges on the concentrated and dilute sides could easily become comparable with the Kapitza boundary resistance between liquid helium and the copper surfaces, if these precautions were not taken.

THE NUCLEAR STAGE

Figure 3 is a drawing of the lower part of the cryostat. The nuclear stage is made of 1500 grams of thin copper wire, glued together with Araldite. We chose copper as the cooling agent solely because of its ready availability in the form of thin, insulated wire of reasonable purity. Other more promising metals such as indium and thallium will be tried later.

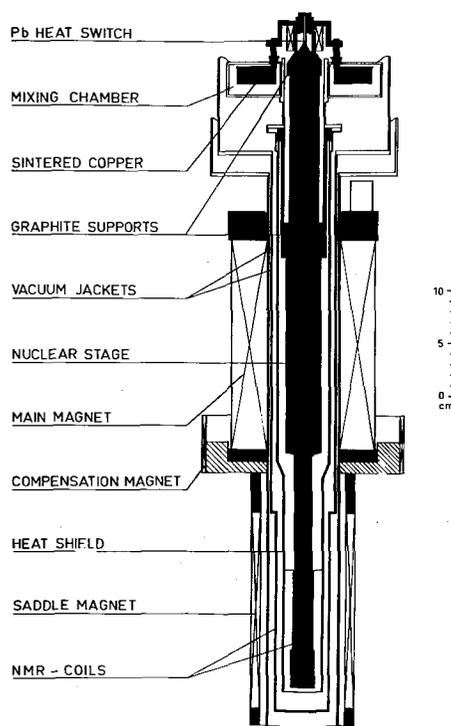


Fig. 3 - Lower part of the cryostat

The copper wires used are 0.05 mm in diameter; about 40% of them proceed straight from top to bottom, and the rest are folded once to form the thicker part of the nuclear stage in the high-field region. The amount of copper inside the magnet is 10.5 moles. At the top the wire bundle is machined to a cone which is coated with indium. A 3-mm-diameter and 15-mm-long lead superconducting heat switch connects the cone to a screw which is thermally anchored to the mixing chamber. The switch is operated by a small superconducting solenoid.

To remove the large heat of magnetization from the nuclear stage, thermal contact to the liquid in the mixing chamber must be good. This was achieved by means of eight sintered copper sectors located inside the mixing chamber and having a total surface area of 8 m<sup>2</sup>.

The nuclear stage is supported at two places by graphite cylinders. The heat shield was made of Epibond, covered with copper coil-foil thermally anchored to the mixing chamber.

The superconducting solenoid used for magnetizing the nuclear stage has an inner diameter of 7 cm and is 20 cm long. The solenoid produces, at present, a maximum field of 57 kG at its center. The magnet is wound with filamentary Nb-Ti wire, is quite free of flux jumps, and has a very small remanent field. In our preliminary experiments the magnet produced a root-mean-square field of 46 kG over the thick part of the nuclear stage.

## MEASUREMENT OF TEMPERATURE

The temperatures reached in the nuclear stage after demagnetization were measured with a pulsed NMR thermometer.<sup>5,7,8</sup> Several of its principal parts can be seen in Fig. 3. A compensating magnet cancels the fringe field of the main magnet over the thermometer coils. The steady homogenous field, in which the copper nuclei precess, is produced in the horizontal plane by a saddle-shaped magnet. The transmitter pulse, also in this plane but at right angles to the steady field, is generated by a second saddle magnet. The signal from the precessing nuclear spins is detected by a receiver coil, located in the vertical direction around the bottom of the nuclear stage.

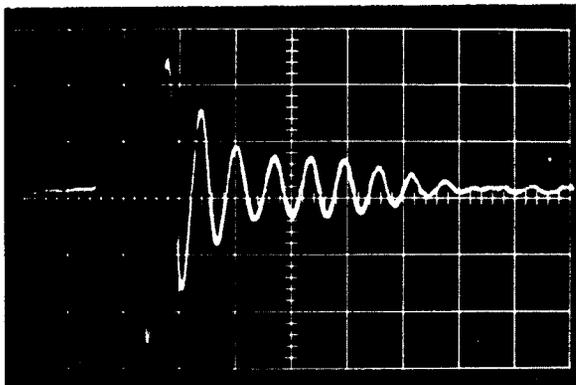


Fig. 4 - Nuclear free-precession signal of copper from the pulsed NMR thermometer. The calculated tipping angle of the nuclear spins, produced by the transmitter pulse, is quite small, only 0.2 degrees, and the total heating caused by the measurement is less than 0.1 erg per pulse. The beat in the decaying signal is caused by the two copper isotopes,  $^{63}\text{Cu}$  and  $^{65}\text{Cu}$ , having slightly different Larmor frequencies. The signal, corresponding to a temperature of 0.62 mK, was photographed from the screen of a Tectronix 549 memory oscilloscope (vertical scale 0.2 V/cm, horizontal scale  $10\ \mu\text{sec/cm}$ ; i.e. the frequency of the signal is about 75 kHz).

Figure 4 shows the signal from the NMR thermometer at 0.62 mK as photographed from the screen of a memory oscilloscope. The thermometer was calibrated between 20 and 100 mK against resistance thermometers which, in turn, had been calibrated against a CMN magnetic thermometer. The estimated accuracy is better than  $\pm 10\%$  in the low temperature region. We have assumed that Curie's law holds for copper nuclei at temperatures relevant to these experiments and thus set  $1/T$  proportional to the measured signal amplitude.

## RESULTS

The warming-up curves after two demagnetizations are plotted in Fig. 5. For runs A and B the initial conditions, 16 mK and 46 kG, and the final field, 727 G, were the same. The only difference between these experiments was that the demagnetization time was increased from 4.5 hours for run A to 7.5 hours for run B. The considerable improvement observed in the results for run B was probably caused by smaller eddy-current

## Superconducting Flux Detector Applied to Thermometry in the 1-mK Range

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In 1967 E. B. Osgood and one of us (JMG) reported the use of nuclear cooling to produce and maintain submillidegree temperatures for as long as 8 hours.<sup>1</sup> Although this work established the utility of nuclear cooling as a laboratory tool, it did not solve many of the difficult problems of making use of that tool. Our motivation for developing it had been to explore the properties of liquid He<sup>3</sup> in the temperature range around 1 mK. However, we were unable to cool it below 4 mK at that time.

One of the problems which had to be solved was that of finding a thermometer which could be immersed in the He<sup>3</sup> so as to measure its temperature but which would not be heated significantly above the temperature of the He<sup>3</sup> by the power dissipation involved in the measurement. In the work of Osgood and Goodkind the He<sup>3</sup> temperature was measured by the nmr signal from a bundle of 0.0005-in.-diameter copper wires. However, even at 4 mK the thermal impedance between the copper nuclei and the He<sup>3</sup> was large enough so that the power dissipation of 0.01 erg/min from our lowest usable rf level was sufficient to warm the thermometer a measurable amount. Since the impedance probably increases as  $T^{-5}$ , the effect would have been intolerably large at 1 mK. We describe here our efforts to solve this problem through the use of a superconducting flux detector used to measure the static nuclear susceptibility in a constant magnetic field. In addition to the low power requirement, we have also attempted to obtain sufficient resolution to measure the temperature to 1 part in  $10^4$  at 1 mK so as to be able to measure the thermal conductivity of He<sup>3</sup> at that temperature.

In the work of Lukens<sup>2</sup> we had learned to make very sensitive superconducting thin-film flux detectors. The sensitivity was sufficient to give us much better than the 1% resolution in the temperature which we could obtain with nmr. In addition, since it would be used to measure the static susceptibility in a constant field, no power would be dissipated in the sample. However, to maintain the accuracy and resolution which we hoped for we felt it necessary to place the flux detector close to the low-temperature sample, so as to eliminate the need for long leads. Thus it was necessary to develop a technique for making a sufficiently weak link so that the device would operate at temperatures far below the transition temperature of the superconductor. Two such techniques are described by Goodkind and Stolfa in an article in the *Review of Scientific Instruments*, June 1970. We explain there also the operation of the device and the circuits with which it can be used.

After the discovery of these techniques, two major problems remained. The first was the response time of the thermometer, and the second was the degradation of the accuracy by stray susceptibilities. By the response time, we mean the time which is required to remove the heat of magnetization of the nuclear spin system through the spin-electron relaxation mechanism and the electron-phonon and phonon-phonon transfer

In these runs a substantial portion of the cooling capacity was wasted, presumably owing to eddy-current heating. This can clearly be observed from Fig. 7, in which we have plotted the heat capacity of the copper nuclear stage before and after demagnetization vs  $\log T$ . Starting from 16 mK and 46 kG (point A), a truly adiabatic process would, after the field has been reduced to 727 G, yield a temperature of 0.25 mK (point B), whereas the observed temperature was 0.62 mK (point C). In Fig. 7 we have also plotted  $\int_{T}^{\infty} T dS$ , which gives the amount of heat the nuclear stage absorbs when it warms from a temperature  $T$  to a high temperature ( $T > 20$  mK) in an external field of 727 G. During run B,  $\int_{T}^{\infty} T dS$  was 3000 ergs (point D), whereas in a truly adiabatic case the integral would have been equal to 7000 ergs (point E). This clearly shows the improvements that can be made, not to mention those resulting from an increased initial field and a lower starting temperature.

As was pointed out earlier the experiments described in this report are quite preliminary. We hope that by the end of the year several important improvements will have been made to the cryostat and that significantly better results can then be obtained, both in terms of the ultimate temperature and in terms of the experimental time available before the cryostat warms up. The next version of the nuclear stage will include provisions for cooling  $^3\text{He}$  and metallic specimens to, hopefully, the submillikelvin region. The fact that we were able to cool the lattice and the conduction electrons, not just the nuclear spins, is essential for the success of these future experiments.

#### ACKNOWLEDGMENTS

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processes to the He<sup>3</sup>. Only in metals will the spin relaxation time be short enough for practical thermometry. For them the relaxation time times the Kelvin temperature is a constant. That constant takes on a wide range of values for different metals but, for example, is 1.27 sec-K for copper and about 0.03 sec-K for platinum. In the temperature range near 5 mK theoretical calculations indicate that the electron-phonon thermal impedance should begin to dominate all other thermal impedances to the He<sup>3</sup>. The measurements of Osgood and Goodkind<sup>1</sup> in this range showed a temperature dependence appropriate to the electron phonon interaction. However, they also showed a large dependence on the pressure between 0 and 20 atmospheres. Since we are not aware of a mechanism by which the electron-phonon interaction could be significantly altered by such low pressures, we are uncertain of the mechanism which dominates or what its temperature dependence will be at 1 mK. The T<sup>-5</sup> dependence of the electron-phonon impedance is likely to be the worst case, so that we assume it in the discussion which follows.

To estimate the response time, we first find the magnetic field which must be used in order that the flux change resulting from a temperature change be large enough to measure the temperature to 1 part in 10<sup>4</sup> at 1 mK. We then find the heat capacity of the nuclear spin system in this field. Next we assume that the thermal impedance is inversely proportional to the surface area (which it would not be if the dominant impedance was the electron-phonon interaction) to calculate the time which would be required to cool a sample of 0.001-in.-diameter wire 0.1 mK with a  $\Delta T$  of 0.1 mK at a temperature of 1 mK. The fields which are required with our present system and the computed times are listed below for several metals:

<u>Metal</u>		<u>H (gauss)</u>	<u>Time</u>
Cu	15		4.7 min
Cd	$7.5 \times 10^3$		40 hr
Mg	860		4.5 hr
Sc	1.7		32 sec
Al	150 (required to quench superconductivity)		4.2 hr

Stray susceptibilities are a problem, since a static measurement technique does not distinguish between different sources of paramagnetism in the way that an nmr measurement does. Spin systems, other than the nuclei of the metallic thermometer material, are likely to exhibit a susceptibility which is not inversely proportional to the Kelvin temperature and/or is dependent on time for the duration of the low-temperature experiment. Thus they would render the susceptibility useless as a primary thermometer. Such spin systems can be present in the thermometer material itself or in the associated circuits and mechanical supports. It is important, therefore, to use for the thermometer material a metal in which the "local moments" due to magnetic impurities are sufficiently weak and few in number so as to contribute an unmeasurable amount to the susceptibility.

After considering many materials with regard to this problem and with regard to the response time problem, we have decided to continue to use copper for our first experiments. We have obtained No. 40 wire drawn from a five-9's-pure sample. We first clean the surface in an acid etch and then anneal it in an oxygen atmosphere at 850°C. This process is known to render the iron impurities nonmagnetic. The resistance ratio which we obtain is then close to its size-limited value.

To eliminate all other materials from the region of the measuring field, we have used toroidal field coils and transformers as shown in Fig. 1.

The toroids are made by winding 0.002-in. Nb wire which is insulated by anodizing to a depth of a few thousand angstroms on a cylindrical copper form. The form, plus

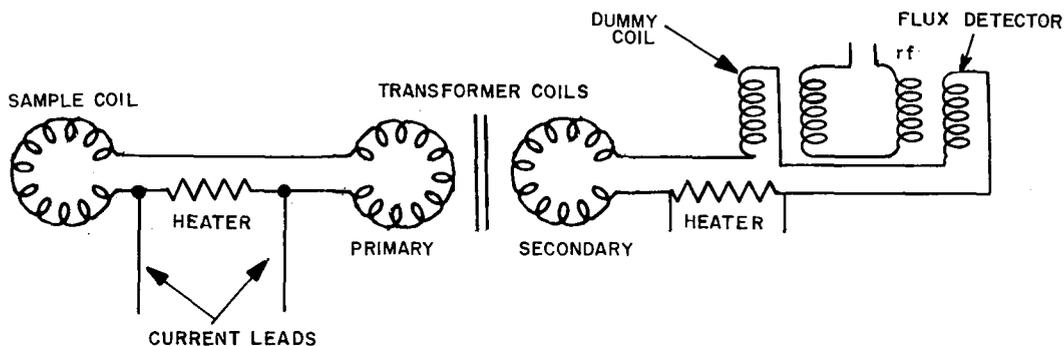


Fig. 1 - Superconducting flux detector

windings, is then bent into a circle to form a toroid and placed inside an anodized Nb shell. For the transformer and sample coils, which have no core, the copper is then dissolved with nitric acid. The heater and current leads of the circuit are used to trap the desired field in the sample toroid and transformer primary and to reduce the field to zero in the transformer secondary and flux detector coils. Thus, in the flux detector coils, where insulated wire is used, no magnetic field is present, so that the susceptibility of the insulation will not be measured. The magnetic field on the apparatus is reduced to 1 milligauss before the superconducting shields are cooled.

The dummy coil serves to prevent any rf field from appearing at the sample, where it could cause heating. The rf current required for operation of the flux detector is passed through two coils in series as shown. These are coupled in opposing senses to the two superconducting coils which carry the dc signal current to the flux detector, so that no rf current flows in the secondary of the transformer.

In our first test of the complete system we were unable to trap a current in the sample toroid. Thus, the system was only in the magnetic field of the earth (about 1/2 gauss at our apparatus), which was not canceled at that time. In that case the temperature-dependent signal down to 20 mK appeared to be primarily the residual temperature dependence of the flux detector itself and was less than 1% of the expected signal from the copper nuclei. Thus we think we have solved the thermometry problem and expect to be using the thermometer in the next few weeks to find out how well we have done with the problem of cooling  $\text{He}^3$  to 1 mK.

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## The Importance of Dipole-Dipole Interactions in Ultralow Temperature Physics

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The magnetic compounds used for thermometry or as working substance for adiabatic demagnetization will eventually "condense" into a magnetically ordered state.<sup>1</sup> The temperatures at which they cease to be paramagnetic are determined by the magnetic interactions between the spins. The interactions are the dipole-dipole and exchange interactions, and the exchange interaction is usually the dominating and hence determining factor. However, materials used in the ultralow temperature region have exchange interactions that are in general small compared to the dipole interactions. Examples are the rare earth magnesium nitrates, chlorides, and ethyl sulphates. We try to obtain the specific heat and susceptibility from the fundamental constants: the lattice parameters, the  $g$  factors, and the interaction. Since the dipole-dipole interaction is known explicitly, contrary to the exchange interaction, which has to be deduced from independent measurement, we are in the position that all necessary information is available. Actually very few theories are available,<sup>2</sup> since most theories require the knowledge of the ground state to initiate the approximation procedures. The exception is the high temperature expansion, which does not rely on any assumptions about the magnetic ordering.<sup>3</sup> On the other hand only the thermodynamic quantities are calculable in the temperature range above  $T_c$ , but by extrapolation one can try to determine  $T_c$  from the radius of convergence. By analytic continuation one can investigate the magnetic ordering below  $T_c$ .

### THE METHOD

To obtain the successive terms in the high temperature expansion we need  $\langle \mathcal{H}^n \rangle$ ,<sup>4</sup> the trace over powers of the hamiltonian:

$$\mathcal{H} = \mathcal{H}_{dd} + \mathcal{H}_{ex} + \mathcal{H}_{Zeeman}.$$

This hamiltonian is bilinear in the spin operators, except for the last term. The trace operation leads to a contraction over vertices, and the resulting expressions are sums over lattice points. These multiple sums can be classified according to their topological structures. The resulting diagrams (Fig. 1) are classified according to the number of bonds (i.e. power in  $1/kT$ ) and according to the number of vertices. The number of vertices  $n_v$  is always equal or lower than the number of bonds  $n_b$ . The diagrams where  $n_v = n_b$  are called the ring diagrams; they are only conditionally convergent and give the largest contribution. We have calculated these diagrams by Fourier transform, which makes it possible to do these summations up to almost any order in  $1/kT$ . However for each order one has an increasingly large number of nonring diagrams. So far we have restricted ourselves to estimating the contribution of these diagrams. The results from some of these Fourier transforms are displayed in Figs. 2a and 2b. These are Fourier transforms of the dipolar sums, without  $g$  factors, for the cerium magnesium nitrate (CMN)

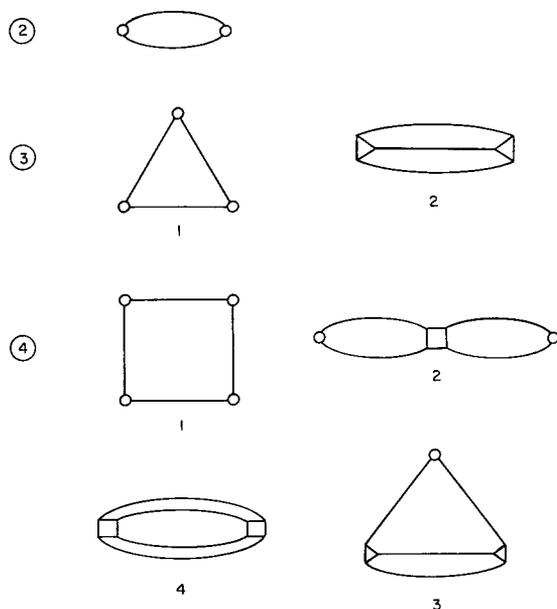


Fig. 1 - Diagrams contributing to the second-, third-, and fourth-order entropy correction. The values are  $\Delta g$  in millikelvin.

lattice. The inserts indicate the direction chosen in the Brillouin zone. The Fourier transform method results in an overcount, since there is no way to prevent the rings from being self-avoiding. The diagrams that are accidentally incorporated are listed in Fig. 3. They have to be computed separately.

Another summation is to be performed. Each tensor coupling can be represented by a matrix, in the same way as Kramers and Wannier used for the Ising model. This step was performed by diagonalization of the tensor matrix and raising the eigenvalues to the power of the order of the temperature correction.

The question has been raised whether the zero-field specific heat could be shape dependent (compare Fig. 4). For CMN it is very simple to prove that this is not so. The only diagrams that could give rise to a shape dependence are the ones that lead to conditionally convergent lattice sums. These are the diagrams that have one or more single bonds between a pair of articulation points. Examples are (5-4)b (6-5)a, and (6-4)b in Ref. 4. It is easy to show that such diagrams always contain at least one pair of odd vertices. Each odd vertex has a trace zero, since CMN has a  $g_{||}$  factor which is almost zero.

The practical bottleneck of the method turns out to be the integration in  $k$ -space over the Brillouin zone. In our first work we used a set of linear inequalities to determine whether the points were inside or outside the Brillouin zone. This method is inflexible, since one has to construct a new set of conditions for each compound. The parallelepiped spanned by the reciprocal lattice vectors contains all equivalent points. Some difficulties were encountered with the numerical three-dimensional integration. There is no universally superior method; every suggestion made in the literature was claimed to be just that! The simple summation used in Ref. 4 was changed to a Simpson rule without too much improvement. (Several options are missing in Fig. 1a, Ref. 4; compare Fig. 5 in this report.) We deduced from our mistakes that the slow convergence to the dense-point-in- $k$ -space value is due to the initial overemphasis on boundary points. The boundary points were originally given half the weight of the integral rule, since they are equal, modulo the reciprocal lattice vector. This distorts the situation, and the network of points in  $k$  space was put on integer-plus-one-half fractions of the parallelepiped. This improved the convergence enormously. It seems moreover that the actual values of the lattice sums for  $k$  values at the boundary give special values due to symmetry. This has to be further investigated.

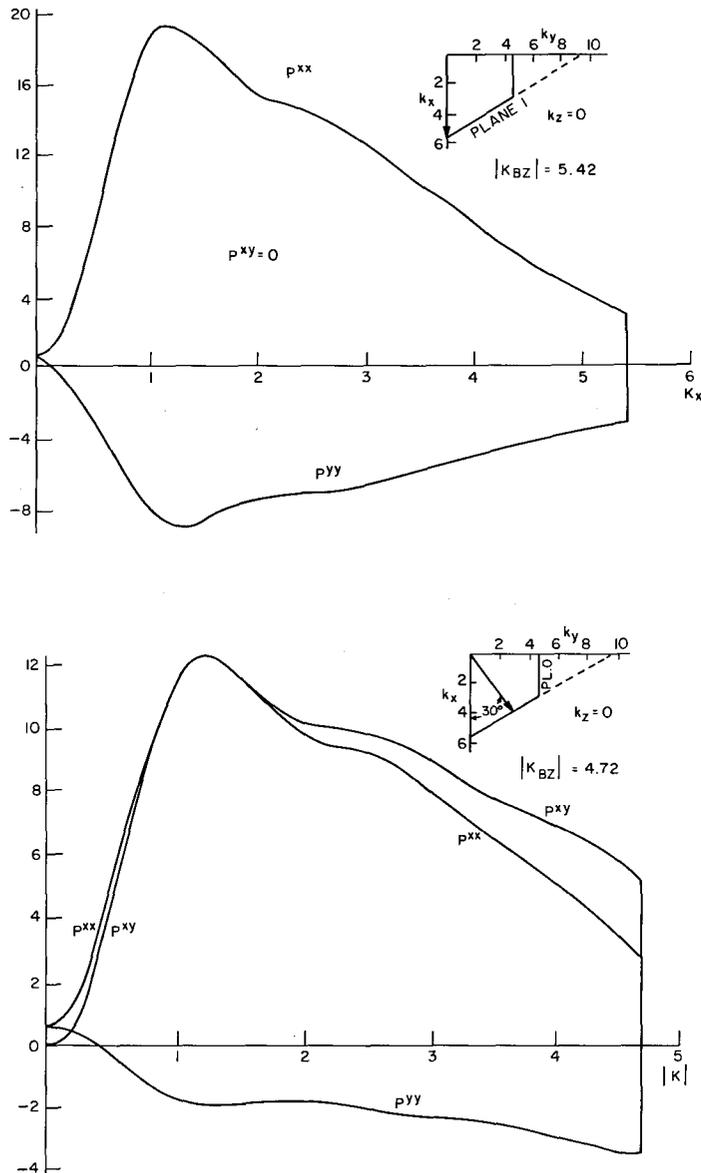
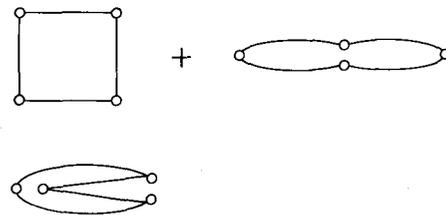


Fig. 2 - A typical  $P(k)$  curve for CMN

Fig. 3 - Diagrams incorporated in the fourth-order ring diagram (collapsed diagram)



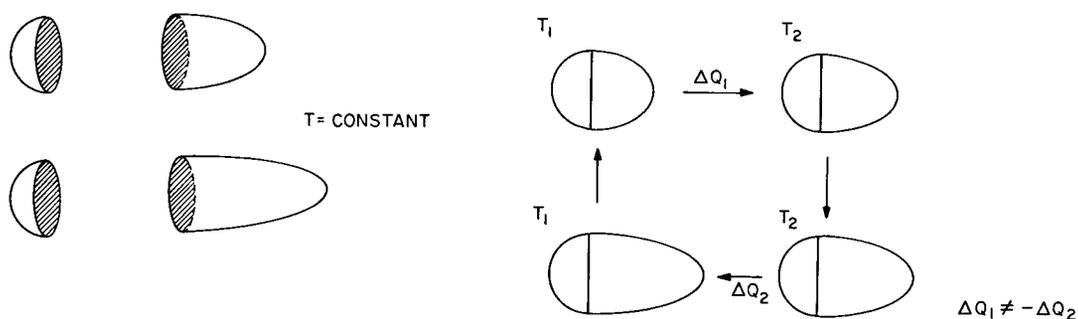


Fig. 4 - Gedanken experiment suggested by Dr. Kurti to change the shape at constant temperature. The half-sphere on the left was originally joined to an half ellipsoid. This half ellipsoid is replaced by another half ellipsoid of different eccentricity, at constant  $T$ . If the specific heat were shape dependent, one could make a heat engine that would subtract heat without doing work.

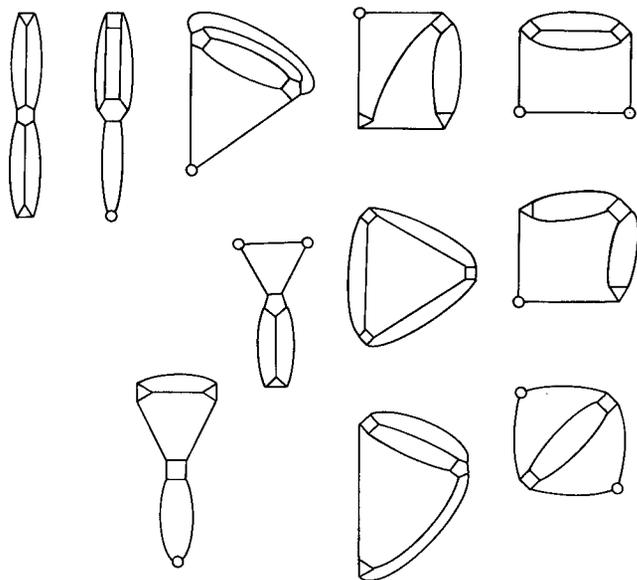


Fig. 5 - Diagrams for the sixth-order entropy correction missing in Ref. 4

## RESULTS

Results obtained are listed in Ref. 4 for CMN and are presented here for CMN and CeTi alum in Table 1 (first and second rows).

To extend the method to so-called lattices with a basis (more than one atom per unit cell) we modified our procedure. In such lattices there is no longer inversion symmetry, and the Fourier transform becomes complex, similar to the so-called structure factor in x-ray analysis. The complexity of the Fourier transform forced us to revise the Kramers-Wannier diagonalization procedure for ring diagrams of arbitrary order.

The previous procedure consisted of the diagonalization of a three-by-three real symmetrical matrix. The successive powers of the eigenvalues  $\lambda_1$ ,  $\lambda_2$ , and  $\lambda_3$  were summed in order to obtain the total contribution to entropy correction. Since this sum

Table 1  
Entropy Corrections and Lattice Sums, Incorporating the g Factors  
The third order is for ring diagrams only.

Compound	R	NPT	$g_{  }$	$g_{\perp}$	c	a	Lattice Sum			$\Delta S$		
							2	3	4	2	3	4
CMN*	4	15	0.032	1.8286	17.034	10.g857	g58.85	13265.	$1.126 \times 10^6$	3.307	7.1653	80.48
CeTi alum†	4.5	11	1.25	1.14	21.079	8.605	166.443	922	$1.887 \times 10^4$	2.4847	922.414	25.268
Nd ethyl sulfate‡	3	7	3.535	2.073	7.07	13.992	111760	$-4.4846 \times 10^6$	$1.2506 \times 10^{10}$	90.29	-274.64	49032

\*The g factors in CMN are different from Ref. 4.

†The ratio c/a in CeTi alum is the fcc lattice ratio:  $\sqrt{6}$ . The length a is  $1/\sqrt{2}$  times the length of the edge of the cube.

‡Ethyl sulfate has two atoms per unit cell; the positions are given by Ketelaar (Physica 4, 619 (1937)).

is equal to a trace, one can avoid the calculation of the individual eigenvalues. The introduction of the complex Fourier transform led to the necessity of the diagonalization of a matrix. Although this matrix is normal and hence can be completely diagonalized, we could not obtain a ready-made numerical procedure to accomplish this. Hence we decided that it was simpler to use a direct procedure. The Fourier transform of the dipole interaction tensor is repeatedly matrix-multiplied by itself, using alternatively the complex conjugate form. The end result needs no diagonalization, since it suffices to take the trace. Although the resulting function of  $\vec{k}$  is complex, the imaginary part is always anti-symmetric in  $\vec{k}$ , hence disappearing after integration over the Brillouin zone. (For results see Table 1, third row.)

Meanwhile improved values of the lattice constants have become available for CMN.<sup>5</sup> This is important, since these values occur to the power  $3n$  in the  $n$ th-order correction.

## CONCLUSION

It is argued that it is feasible to do an "absolute" determination of  $C(T)$  and  $\chi(T)$  (and in the future also of  $C(T,H)$  and  $M(H,T)$ ) for dipole-dipole interacting systems. From this it will be possible to compute  $S = S(\chi)$  for comparison with experiments.

## ACKNOWLEDGMENTS

I thank Dr. Hudson for his stimulation and for his patient waiting till I had eradicated all errors in the computations. A large portion of the actual programming and computation was done by Mr. B. Kessler of the Naval Ordnance Laboratory, White Oak.

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## A Particular Use of a Dilution Refrigerator

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We will describe the use of a dilution refrigerator and the construction of a  $^{59}\text{Co}$  target which has recently been used to study the scattering of polarized neutrons from polarized nuclei. The particular advantage in using a dilution refrigerator over adiabatic demagnetization is that usefully large targets can be cooled and can be kept cold for as long as is desired in quite hostile environments, that is, in the presence of severe vibrations that one is likely to find around Van de Graaff accelerators.

Figure 1 shows a schematic diagram of the refrigerator and cobalt target arrangement. At the top is the still, containing a sintered copper powder sponge, with the return line wrapped around the outside. There are five modular heat exchangers, each containing a pair of sintered copper sponges. The mixing chamber is arranged so that the  $^3\text{He}$  flow is forced through a porous copper plug. This necessarily results in some viscous heating of the liquid, but this feature was needed to fully utilize the surface area of the copper sponge and to cool the cobalt rod efficiently.

The cobalt rod was brazed into a copper tube, which was in turn brazed to the base of the mixing chamber. The rod was mounted in a superconducting solenoid and thermally isolated from this solenoid by means of nylon screws and laminated nylon rings.

The following list summarizes the typical operating conditions of the dilution refrigerator with the  $^{59}\text{Co}$  target attached.

Still power: 2.5 mW,

Still temperature: 0.6 K,

Gas circulation rate:  $9 \times 10^{-5}$  mole/sec  
( $1.7 \times 10^{-4}$  mole/sec max),

$^4\text{He}/^3\text{He}$  ratio: 10%,

Return line pressure: 40 torr,

Final temperature of first heat exchanger: 0.25 K,

Final temperature of fifth heat exchanger: 0.041 K,

Final temperature of mixing chamber: 0.034 K,

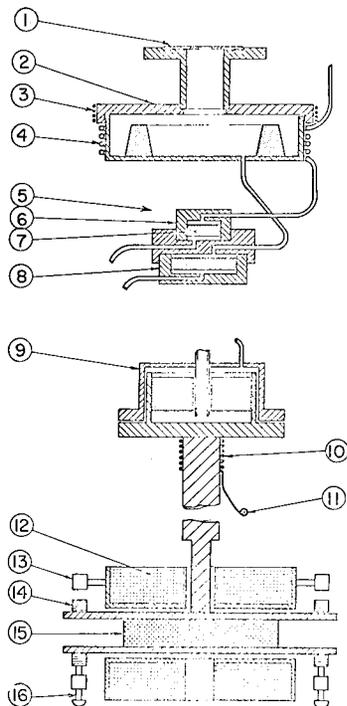


Fig. 1 - Dilution refrigerator and cobalt mounting: (1) electropolished stainless steel aperture, (2) still, (3) No. 38 manganin wire for still heater, (4) return line thermally anchored to still, (5) one of five heat exchangers, (6) concentrated side of the heat exchanger, (7) porous copper plug, (8) dilute side of heat exchanger, (9) mixing chamber, (10) No. 38 manganin wire for mixing chamber heater, (11) copper-foil-wrapped Speer type 1002 resistor used as a thermometer, (12) split-coil superconducting solenoid, (13) clamp ring to keep the cobalt sample rigid with respect to the solenoid, (14) 80 layers of 0.0125-mm-thick Mylar used as a thermal insulator, (15)  $^{59}\text{Co}$  sample, and (16) nylon set screws

Cooling time, 0.5 K to 0.060 K: 1 hr,

Cooling time, 0.060 K to 0.034 K: 2 to 4 hr.

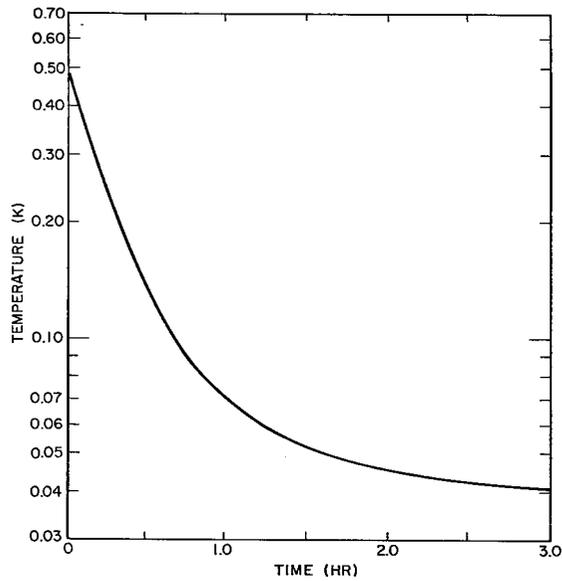
At the typical circulation rate of  $9 \times 10^{-5}$  mole/sec the mixing chamber could dissipate 90 ergs/sec at a temperature of 0.061 K. In the absence of external heat loads the cobalt target could be cooled to 0.034 K. This temperature was measured by observing the anisotropy in the  $^{60}\text{Co}$  gamma radiation emitted by an activated portion of the cobalt rod. At this temperature, and with the solenoid field at 7.5 kOe, the nuclear polarization of the cobalt rod was 40% of saturation, which is quite adequate for nuclear physics experiments.

In practice the dilution refrigerator and cobalt target were mounted in a very hostile environment, on the end of the beam tube of a Van de Graaff accelerator, where there was a great deal of vibration from the belt drive motors and various generators. Under these conditions the cobalt rod did not reach the temperature of 0.034 K achieved under more ideal test conditions. Figure 2 shows a typical cooldown from 0.5 K. After about 3 hours, the target had pretty well leveled out at 0.040 K.

We will describe the experiment of interest. For the past several years there has been some interest in looking for a spin-dependent interaction between nucleons and complex nuclei. One way of observing such an interaction experimentally is by transmitting polarized neutrons through polarized targets and measuring the changes in transmission associated with changes in polarization. Our particular experiment was to look for a spin-spin effect in the total neutron cross section of  $^{59}\text{Co}$ .

The following describes what is meant by the spin-spin effect. With a neutron beam of polarization  $P_n$  obtainable from an appropriate nuclear reaction and a target with polarization  $P_t$ , one can measure the total neutron cross section for the two cases of neutron and target polarization parallel and antiparallel. The difference, normalized to

Fig. 2 - Typical cooldown from 0.5 K



neutron and target polarization, is the spin-spin effect; that is,  $\sigma_{ss} = (\sigma_{\uparrow\uparrow} - \sigma_{\uparrow\downarrow}) / 2P_n P_t$ . This effect has been looked for exhaustively and unsuccessfully in  $^{165}\text{Ho}$ , which has a significant polarization at temperatures obtainable with a  $^3\text{He}$  refrigerator.

Figure 3 shows the success of the search with  $^{59}\text{Co}$ , in that nonzero values of  $\sigma_{ss}$  were found. One point on this graph was measured by a group in Tokyo, using adiabatic demagnetization of a salt for cooling. In this particular case, use of a dilution refrigerator has greatly simplified the experimental procedure and has made possible a much more precise measurement.

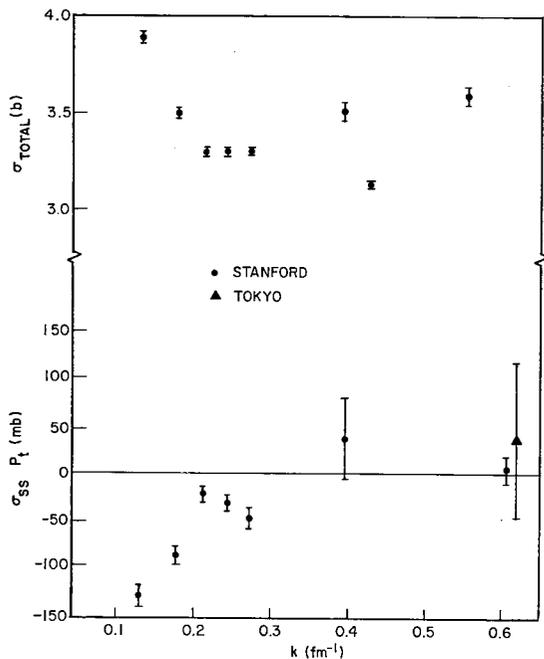


Fig. 3 - Spin-spin effect in the total neutron cross section of  $^{59}\text{Co}$

## Predictions for Helium Film Creep at Ultralow Temperatures

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A moving superfluid vortex line can lose momentum to the normal fluid component of the liquid through the mechanism of quasiparticle scattering by the vortex cores.<sup>1</sup> This viscous drag on the vortex-line cores moving with the superfluid during helium film creep will be shown to have two effects: to enhance the film creep velocity and to push the vortex lines toward the film surface, thus causing their disintegration there for  $T > 0.5$  K. Definitive experiments will be proposed.

One reason for assuming in the conventional way<sup>1-7</sup> that the creation of vorticity is responsible for limiting the film creep velocity is that we can picture a force couple acting on the moving film, as indicated in Fig. 1. Here  $F_g$  and  $F_w$  represent the motivating force of gravity and an effective restraining force from the wall. To aid intuition the wall surface is folded out straight and drawn horizontal. The dotted equipotential lines of the van der Waals plus gravitational fields reach their closest point to the original unfolded wall at the highest point over which the film passes, and, as observed during outflow,<sup>8</sup> an abrupt change in the film profile has been shown at this point. Then the action of  $F_g$  and  $F_w$  in this region must impart angular momentum to the film. Following Donnelly,<sup>6</sup> Anderson's quantum mechanical argument<sup>9</sup> shows that quantized vortex lines should be created in the superfluid at the rate

$$\dot{n} = -gzm/h, \quad (1)$$

where  $g$  is the acceleration of gravity,  $z$  is the difference in heights of the two sides of the film,  $m$  is the helium-atom mass, and  $h$  is Planck's constant.

Whether the vortex lines are created near the wall, near the film surface, or more in the center of the film, one might expect them to be stable as in the bulk liquid, to gain size and energy, and to translate with the moving superfluid through the film, as indicated in Fig. 1. Hence in the formulas that follow, the expression  $e_0 = (\rho_s/4\pi)(h/m)^2 \ln(d/a)$  used for the vortex line energy is that of a unit length of straight vortex line with radius  $d/2$ ,  $d$  being a mean film thickness,  $a$  the core radius, and  $\rho_s$  the superfluid density.

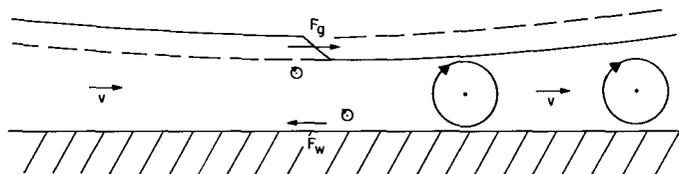


Fig. 1 - Film profile and equipotential lines

We should keep in mind, though, that because the Magnus force  $\rho_s(h/m)u$  moves the vortex lines toward the film surface with velocity  $u$ , this simple expression for  $e_0$  is only an approximation for describing the true physical conditions. Equating the Magnus force and the viscous drag  $2v(m/h)\alpha(T)$ , where  $\alpha(T)$  is defined and measured by Rayfield and Reif,<sup>1</sup> gives

$$u/v = E(d/z), \quad (2)$$

in which

$$E = (2/\rho_s)(Z/d)(m/h)^2 \alpha(T), \quad (3)$$

where  $Z$  is defined as the height of the film. If  $E > 1$  and hence  $u/v > d/Z$ , the vorticity will be forced into the film surface before it can exit into the bulk liquid at the base of the film. Then the assumption of vortex stability and the approximations used for  $e_0$  and  $\alpha(T)$  can easily become invalid.

Instead of using the Landau criterion for the microscopics of the vortex creation processes, we use the condition of energy conservation for the film as a whole and simply add a vortex-line-viscous-dissipation term to Donnelly's energy equation.<sup>6</sup> The intuitive physics goes as follows: Because the angular momentum of the vortex lines is quantized and the torque exerted on the film depends only on the height difference  $z$ , the rate at which vortex lines must be created is purely a function of  $z$  and is independent of the film velocity  $v$ . For the core geometry assumed, the energy of these vortex lines is also quantized, which means that the required supply of energy from the van der Waals and gravitational force fields is also dictated solely by the height difference  $z$  without any dependence on  $v$ . But the rate of energy supply to the film is proportional to its downward velocity  $v$  in the effective gravitational field. Hence  $v$  must be large enough to supply the energy required by Eq. (1), and any increase in energy dissipation necessitates a corresponding increase in  $v$ .<sup>\*</sup> In experiments enhanced flow rates over contaminated or roughened surfaces have, in fact, been observed for many years.<sup>†</sup>

The average number of horizontal vortex lines in the film is roughly

$$n = Z\dot{n}/v \quad (4)$$

or 1/2 of this value, depending on whether we assume that the vortex lines are created all at the top of the film or uniformly throughout the down side of the film.<sup>8</sup> Conservation of energy for a unit width of film requires

$$\rho_s g z d v = e_0 \dot{n} + 2(m/h)\alpha(T) n v^2, \quad (5)$$

which, using Eqs. (1) and (4), becomes

$$v = (1/4\pi d)(h/m) \ln(d/a)/(1 - E), \quad (6)$$

where  $E$ , given by Eq. (3), now takes on the meaning of a velocity enhancement factor due to the viscous drag on the vortex cores.

<sup>\*</sup>The proposed effect is analogous to the tunneling current steps observed when the Josephson frequency  $2eV/\hbar$  is a multiple of the resonant frequency in a Josephson junction.<sup>10</sup> In the case of electron pair tunneling, an increased energy loss at resonance must be compensated for by an increase of dc current flow in the electric field.

<sup>†</sup>Many experiments are summarized on page 217 of Ref. 11; see, however, Ref. 12.

Equation (5) may be compared to a disparity of experimental results.\* The doppler measurements of third sound of Ref. 7 agree well with the formula  $(1/4\pi d)(h/m)$ , which suggests that the numerator of Eq. (6) is about a factor of 5 too large due to the logarithm term. Although the formula used for  $e_0$  can be reduced in magnitude by assuming the vortex lines to be in close proximity to the wall or film surface, such arguments are bound to be qualitative and hard to relate directly to experiment. We turn simply to the possible physical significance of the velocity enhancement term in the denominator.

If the enhancement factor causes Eq. (6) for  $v$  to surpass the value  $(2gz)^{1/2}$ , the assumptions used in deriving this formula must certainly be incorrect. Thus the condition  $E > 1$  implies not only that all the vortex lines are moved to the film surface by the Magnus force but also that they must disintegrate there,† for otherwise the force of gravity could not supply enough energy for first creating and then moving the quantized vorticity through the normal fluid. With  $Z = 1$  cm and the data of Rayfield and Reif,<sup>1</sup> Eq. (3) becomes approximately

$$E = 4.0 \times 10^6 e^{-8.65/T} + 6.6 \times 10^5 n_3 T^{1/2} + 13 T^4, \quad (7)$$

where  $T$  is in kelvin and  $n_3$  is the concentration of  $\text{He}^3$  impurities in  $\text{cm}^{-3}$ . The plot of this expression in Fig. 2 shows that the condition  $E > 1$  occurs when  $T$  is greater than about 0.5 K for the natural density of  $\text{He}^3$  impurity and for progressively lower temperatures when  $n_3$  is increased. Thus, for  $T$  in the upper millidegree range and below, the functional dependence of  $v$  on  $T$ ,  $n_3$ , and  $Z$  is predicted by Eq. (6) to be

$$v = v_0 / (1 - cZn_3T), \quad (8)$$

where  $v_0$  is a constant and  $c = 6 \times 10^5$  cgs units may depend weakly on  $Z$  and on whether the film flow is into or out of the beaker.

Experimental confirmation of Eq. (8) would give considerable support to the often discussed but qualitative concepts concerning the creation of vorticity in moving helium films. Also informative would be experiments in which variable electric forces are applied to vortex lines containing trapped ions.

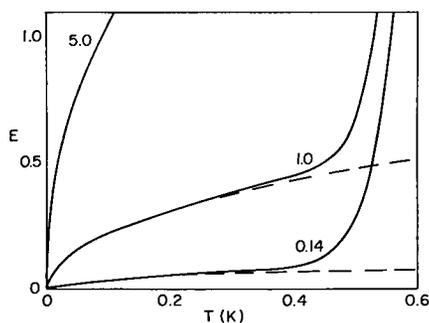


Fig. 2 -  $E$  values for various  $n_3 \times 10^6$ . The dashed lines show the contribution of  $\text{He}^3$  scattering only.

\*See, for instance, Chapter 7 of Ref. 11.

†An alternate but perhaps unlikely possibility is that the viscous drag on the vortex cores could be reduced significantly as they approach the film surface. Then, with  $d \gg a$  and  $E \approx 0$ , Eq. (6) could be made to agree with experiment.

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