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WAVE PROPAGATION IN SUSPENSIONS

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ABSTRACT

This report deals with theoretical methods for determining the effective physical properties of suspensions or statistical mixtures of spherical particles of different isotropic materials. The methods are shown sufficiently general to allow estimates of secondary physical effects such as the average optical rotatory power, the thermoelectric power, and the acoustic attenuation of appropriate suspensions. When particles have sizes comparable with wavelength, the attempted average-property estimates are less satisfactory, and no attempt is made to obtain average-property estimates for anisotropic geometries or particle-materials.

Addition formulas and other useful relations among spherical scalar and vector wave functions are derived for these applications.

PROBLEM STATUS

This is a final report on one phase of the problem. Work continues on other phases.

AUTHORIZATION

NRL Problem R07-02

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PREFACE

This report combines about ten years of the writer's work in wave propagation in random media. Many of the ideas originated in discussions with F. T. Haddock, E. O. Hulburt, and M. Katzin on electromagnetic subjects, and with R. J. Urick on acoustic topics. Valuable help was received from G. Raisbeck in understanding spherical-wave mathematics, and from H. J. Passerini in most of the heavy algebraic work underlying many of the formulas here.

The original writing was done with various aims and at various times; subsequent and spasmodic revisions to present a connected whole have perhaps left irregularities in emphasis and style. Parts of the material, notably Sect. IV and portions of Sect. VII and VIII have been presented orally at technical meetings; partial versions of Sects. I and VIII appear in Naval Research Laboratory Reports Nos. 3238 and 3350. For some years it appeared that the material did not deserve further publication because the spherical wave formulas were without sufficient application, and the physical results were relatively negligible compared with the weight of the necessary mathematics.

When the relatively concise and general theory of Sect. VII was eventually discovered, the theory and a few of the results were submitted to the Physical Review. After some months of perusal, the referee found himself unable to believe the heuristic justification given in the Appendix to Sect. VII, although he must be thanked for clarifying suggestions incorporated elsewhere. As the more formal justification for ignoring interactions among particles of the conceptual small scattering sphere required much of the tedious mathematics of Sect. I and all that of Sect. VI, the writer decided to collect all of the physical applications into a single manuscript, sprawling but connected through consistent concern with problems of wave propagation in suspensions.

Although much of the work is completed, some deficiencies and further developments might be pointed out: Final results for the propagation of sound waves in experimentally realizable suspensions are not deduced, for reasons of algebraic complexity as well as those given in Sect. IX. The theory of the properties of fine suspensions (Sect. VII) can now be extended, concisely though heuristically, to mixtures,[a] through a generalization of an argument of R. Landauer.[b] The extension to suspensions and mixtures of anisotropic materials is also obvious although formal justification along the lines of Sect. VI is lacking. Reciprocity principles are used in Sect. I and especially in Sect. X, and are latent elsewhere. Reciprocity principles[c] can be combined with the self-consistent formalism of Sects. II-V for an approximate answer to the question, latent in Sects. IV and VIII, where does the randomly scattered component of the power flux go? It is hoped that the present material will provide a reasonably solid foundation for such further developments. An extension of Sect. VII to mixtures is made in 'Notes Added in Proof', pp 104-114.

Henry J. Passerini is to be thanked for carrying out the massive algebraic calculations necessary for several of the results presented here, and for eliminating many errors from the typescript.

a) "Physical Properties of Fine Mixtures," Paper Q7 presented at the Washington Meeting of the American Physical Society, May 1957.

b) R. Landauer, J. Appl. Phys. 23, 779 (1952).

c) "Reciprocity and Scattering by Certain Rough Surfaces," paper presented at Session T-10, International Colloquium on Current Problems in Wave Propagation, Paris, Sept. 1956.

CONTENTS

Sect. IV: Frequency dependence of certain artificial dielectrics. (pp 30-36)

Self-consistent calculation of the propagation constant k' of an artificial dielectric formed by randomly suspending small spheres in matrix, k' being given through the first three powers of the sphere's radius. Interpretation of imaginary part of k' in terms of Rayleigh scattering and order imposed by a finite' radius of exclusion'.

Sect. V: Sound waves in suspensions. (pp 36-41)

Self-consistent treatment of acoustic waves in suspensions of spheres small compared with dilatational wavelength, with the principal effects of thermal and shear waves included. Effective propagation constant expressed by factors interpretable as effective density and effective compressibility of the suspension.

Sect. VI; Order of magnitude of interactions for a small sphere of suspension. (pp 41-44)

The self-consistent calculation of the scattering coefficients of a small sphere containing randomly suspended particles is outlined. The average contribution to the sphere's scattering arising from interactions among particles is evaluated and found small compared with average scattering arising from the primary excitation.

Sect. VII: Physical properties of fine suspensions. (pp 44-73)

A. Conceptual experiment for determining properties of a material by measuring scattering coefficients of a small sample sphere. Application of the result of Sect. VI, calculating scattering coefficients of small spheres of a suspension, and hence to determining effective properties for the suspension.

B. Application, illustrative of certain limitations, of the general formulas to artificial dielectrics.

C. Composition laws for properties germane to acoustic waves.

D. Composition laws for Lamé constants. Viscosity of a suspension; comparison with earlier results. Shear and bulk viscosities and elastic constants of special physical suspensions.

E. Reconsideration of the applicability of formal composition laws to the acoustic properties of certain physical suspensions.

Sect. VIII: Approximations Valid for Dilute Suspensions. (pp 74-82)

A. Summary and critique of earlier general methods for obtaining propagation constants and sometimes other properties of suspensions.

B. Interpretation of the imaginary parts of effective propagation constants.

Section IX: Critique of Acoustic Methods and Results. (pp 82-87)

Review of present and some earlier derivations of the acoustic properties of suspensions, emphasizing requirement for exact scattering coefficients. Discussion of some unexplained experimental results. Partial theory of the drift of bubbles in a standing wave system.

Section. X: Optical Activity. (pp 87-103)

Derivation of the optical activity of a suspension of spheres of an optically active material satisfying electromagnetic reciprocity. Comparison with an earlier result. Derivation of the average scattering coefficients of a model, optically active 'molecule'. Discussion of passive vs. electromechanical 'molecules' from the point of view of reciprocity, non-linearity, and types of scattering.

Notes Added in Proof:

Extension of the general formulas of Section VII to include bulk properties of fine mixtures; improvement of those formulas for high concentrations of suspended particles; application to the thermoelectric properties of sintered solids. (pp 104-114)

Section I. Some Formulas Involving Spherical Wave Functions.

The material in this section is an extension of Chapter VII of J. A. Stratton's "Electromagnetic Theory", to which the reader is referred for background. Our aim is to present tables of useful formulas connected with spherical wave functions; applications of the formulas will be found in later sections.

We choose a fixed rectangular coordinate system, with $\vec{i},\vec{j},\vec{k}$ being unit vectors in the positive x,y,x directions, respectively. Then the position vector $\vec{R}$ of a point $\vec{p}$ relative to a point $\vec{q}$ can be expressed in spherical coordinates. R,θ,ϕ through

$$\vec{R} = \vec{p}-\vec{q} = \vec{i}R\sin\theta\cos\phi + \vec{j}R\sin\theta\sin\phi + \vec{k}R\cos\theta. \quad (1.1)$$

Solutions of the scalar wave (Helmholtz) equation

$$\nabla^2\psi + k^2\psi = 0 \quad (1.2)$$

can be expressed as superpositions of the spherical scalar waves

$$f \sim f(R) \sim f_n(k,R) \sim f_{\substack{e\\o}mn}(k,R) = z_n(kR)P_n^m(\cos\theta)\begin{matrix}\cos m\phi\\ \sin m\phi\end{matrix}$$

where z_n is a spherical Bessel function, P_n^m an associated Legendre polynomial, and the shortened notations will be used where the omitted symbols are obvious.

Spherical vector waves satisfying the vector wave equation

$$\nabla\nabla\cdot\vec{C} - \nabla\times\nabla\times\vec{C} + k^2\vec{C} = 0 \quad (1.4)$$

are: $\vec{l} = \nabla f$ (1.5)

$$\vec{m} = \nabla \times (\vec{R} f) = \vec{l} \times \vec{R} = \nabla \times \vec{n}/k \quad (1.6)$$

$$\vec{n} = \nabla \times \vec{m}/k \quad (1.7)$$

where the vector waves $\vec{l}$, $\vec{m}$, $\vec{n}$ are understood to have the same sets of indices as the scalar waves f in their definitions. The vector waves have the further properties:

$$\nabla \times \vec{l} = 0, \nabla\cdot\vec{m} = 0, \nabla\cdot\vec{n} = 0 \quad (1.8)$$

Thus $\vec{l}$ represents a dilatational or longitudinal wave and $\vec{m}$, $\vec{n}$ transverse waves.

The $\vec{i}$, $\vec{j}$, $\vec{k}$ components of solutions of the vector equation (1.4) satisfy the scalar wave equation (1.2). Through mathematical induction using the recursion formulas for spherical Bessel, Legendre, and trigonometric functions, one obtains the general component formulas expressed in Table I. These are in too general a form to be useful in the important case that the n-index is small; we obtain the simpler component formulas of Table II from Table I and the additional identities:

$$f_{\substack{e\\o}}{}_{m,-n} = 0, \quad f_{\substack{e\\o}}{}_{-1,n} = \mp \frac{1}{n(n+1)} f_{\substack{e\\o}}{}_{1n}, \quad P_n^m = 0, \quad m > n. \quad (1.9)$$

In the subsequent $f, \vec{l}, \vec{m}, \vec{n}$ will denote that a spherical Bessel function of the first kind (j_n) appears in the place of z_n in the definitions while $f', \vec{l}', \vec{m}', \vec{n}'$ will denote that the function h_n appears. The functions $j_n(kR)$ are finite at R = 0 (at $\vec{q}$), whereas the functions $h_n \sim h_n(kR) = h_n^{(1)}(kR)$ correspond

Table I - Rectangular components of spherical vector wave functions. All functions have the same arguments.

$$
\begin{aligned}
\frac{2\,(2n+1)}{k}\,\underline{l}_{\substack{e\\o}mn} = \underline{i} & \begin{bmatrix} (n+m)\,(n+m-1)\,f_{\substack{e\\o}m-1\ n-1} \\ +(n-m+1)\,(n-m+2)\,f_{\substack{e\\o}m-1\ n+1} \\ -f_{\substack{e\\o}m+1\ n-1} - f_{\substack{e\\o}m+1\ n+1} \end{bmatrix} \\
(\mp)\,\underline{j} & \begin{bmatrix} (n+m)\,(n+m-1)\,f_{\substack{o\\e}m-1\ n-1} \\ +(n-m+1)\,(n-m+2)\,f_{\substack{o\\e}m-1\ n+1} \\ +f_{\substack{o\\e}m+1\ n-1} + f_{\substack{o\\e}m+1\ n+1} \end{bmatrix} \\
+2\,\underline{k} & \begin{bmatrix} (n+m)\,f_{\substack{e\\o}m\ n-1} \\ -(n-m+1)\,f_{\substack{e\\o}m\ n+1} \end{bmatrix}
\end{aligned}
$$

$$
\begin{aligned}
2\,\underline{m}_{\substack{e\\o}mn} = (\mp)\,\underline{i} & \left[(n+m)\,(n-m+1)\,f_{\substack{o\\e}m-1\ n} + f_{\substack{o\\e}m+1\ n}\right] \\
-\ \underline{j} & \left[(n+m)\,(n-m+1)\,f_{\substack{e\\o}m-1\ n} - f_{\substack{e\\o}m+1\ n}\right] \\
(\pm)\,\underline{k} & \left[2\,m\,f_{\substack{o\\e}mn}\right]
\end{aligned}
$$

$$
\begin{aligned}
2(2n+1)\,\underline{n}_{\substack{e\\o}mn} = \underline{i} & \begin{bmatrix} (n+1)\,(n+m)\,(n+m-1)\,f_{\substack{e\\o}m-1\ n-1} \\ -n(n-m+1)\,(n-m+2)\,f_{\substack{e\\o}m-1\ n+1} \\ -(n+1)\,f_{\substack{e\\o}m+1\ n-1} + n\,f_{\substack{e\\o}m+1\ n+1} \end{bmatrix} \\
(\mp)\,\underline{j} & \begin{bmatrix} (n+1)\,(n+m)\,(n+m-1)\,f_{\substack{o\\e}m-1\ n-1} \\ -n(n-m+1)\,(n-m+2)\,f_{\substack{o\\e}m-1\ n+1} \\ +(n+1)\,f_{\substack{o\\e}m+1\ n-1} - nf_{\substack{o\\e}m+1\ n+1} \end{bmatrix} \\
+2\,\underline{k} & \begin{bmatrix} (n+1)\,(n+m)\,f_{\substack{e\\o}m\ n-1} \\ +n\,(n-m+1)\,f_{\substack{e\\o}m\ n+1} \end{bmatrix}
\end{aligned}
$$

Table II (Part 1)

Rectangular components of $\boldsymbol{\ell}(\rho)/k$. Entries are functions of ρ : e00 = $f_{e00}(\rho)$, etc.

ℓ/k	i			j			k	
e00		$-$ e11			$-$ o11		$-$ e01	
3 e01		$-$ e12			$-$ o12		e00	$-$ 2 e02
3 e11	e00	+ e02	$- \frac{1}{2}$ e22		$- \frac{1}{2}$ o22		$-$ e12	
3 o11		$- \frac{1}{2}$ o22		e00	+ e02	$+ \frac{1}{2}$ e22	$-$ o12	
5 e02	$-$ e11	$-$ e13		$-$ o11	$-$ o13		2 e01	$-$ 3 e03
5 e12	3 e01	+ 3 e03	$- \frac{1}{2}$ e23		$- \frac{1}{2}$ o23		3 e11	$-$ 2 e13
5 e22	6 e11	+ e13	$- \frac{1}{2}$ e33	$-$ 6 o11	$-$ o13	$- \frac{1}{2}$ o33	$-$ e23	
5 o12	$- \frac{1}{2}$ o23			3 e01	+ 3 e03	$+ \frac{1}{2}$ e23	3 o11	$-$ 2 o13
5 o22	6 o11	+ o13	$- \frac{1}{2}$ o33	6 e11	+ e13	$+ \frac{1}{2}$ e33	$-$ o23	
7 e03	$-$ e12	$-$ e14		$-$ o12	$-$ o14		3 e02	$-$ 4 e04
7 e13		6 e02 $- \frac{1}{2}$ e22	+ 6 e04 $- \frac{1}{2}$ e24	$- \frac{1}{2}$ o22	$- \frac{1}{2}$ o24		4 e12	$-$ 3 e14
7 e23	10 e12	+ 3 e14	$- \frac{1}{2}$ e34	$-$10 o12	$-$ 3 o14	$- \frac{1}{2}$ o34	5 e22	$-$ 2 e24
7 e33	15 e22	+ e24	$- \frac{1}{2}$ e44	$-$15 o22	$-$ o24	$- \frac{1}{2}$ o44	$-$ e34	
7 o13	$- \frac{1}{2}$ o22	$- \frac{1}{2}$ o24			6 e02 $+ \frac{1}{2}$ e22	+ 6 e04 $+ \frac{1}{2}$ e24	4 o12	$-$ 3 o14
7 o23	10 o12	+ 3 o14	$- \frac{1}{2}$ o34	10 e12	+ 3 e14	$+ \frac{1}{2}$ e34	5 o22	$-$ 2 o24
7 o33	15 o22	+ o24	$- \frac{1}{2}$ o44	15 e22	+ e24	$+ \frac{1}{2}$ e44	$-$ o34	

Table II (Part 2)

Rectangular components of $\vec{m}(\rho)$, $\vec{n}(\rho)$. Entries are functions of ρ : e00 = $f_{e00}(\rho)$, etc.

m	i			j			k	
e00		0			0		0	
3 e01		− 3 o11			3 e11		0	
3 e11		0			− 3 e01		3 o11	
3 o11		3 e01			0		− 3 e11	
5 e02		− 5 o12			5 e12		0	
5 e12		$-\frac{5}{2}$ o22			−15 e02	$+\frac{5}{2}$ e22	5 o12	
5 e22		−10 o12			−10 e12		10 o22	
5 o12		15 e02	$+\frac{5}{2}$ e22		$\frac{5}{2}$ o22		− 5 e12	
5 o22		10 e12			−10 o12		−10 e22	
n	**i**			**j**			**k**	
e00		0			0		0	
3 e01		e12			o12		2 e00	+ 2 e02
3 e11	2 e00	− e02	$+\frac{1}{2}$ e22		$\frac{1}{2}$ o22		e12	
3 o11		$\frac{1}{2}$ o22		2 e00	− e02	$-\frac{1}{2}$ e22	o12	
5 e02	− 3 e11	+ 2 e13		− 3 o11	+ 2 o13		6 e01	+ 6 e03
5 e12	9 e01	− 6 e03	+ e23		o23		9 e11	+ 4 e13
5 e22	18 e11	− 2 e13	+ e33	−18 o11	+ 2 o13	+ o33	2 e23	
5 o12		o23		9 e01	− 6 e03	− e23	9 o11	+ 4 o13
5 o22	18 o11	− 2 o13	+ o33	18 e11	− 2 e13	− e33	2 o23	

to waves outgoing from $\vec{q}$, the implied time-factor being $\exp(-i\omega t)$ and $\mathrm{Im}(k) \geqslant 0$ in physical applications.

When coherent sources of outgoing spherical waves are uniformly distributed in the $z = 0$ plane, their combined radiation forms two plane waves, one in $z > 0$, traveling according to $\exp(ikz)$, and the other, in $z < 0$, traveling according to $\exp(-ikz)$. One can calculate the amplitude of the scalar plane waves by the stationary-phase principle at large $|z|$; this suffices to determine the amplitude of the wave everywhere, since the asymptotic plane-wave representation of a plane wave is the wave itself. (Induction can also be used, and there are probably other derivations.) From the scalar plane waves, one obtains vector plane waves via Table I; all non-vanishing waves of this type are given in Table III.

It is frequently convenient to expand a spherical wave, originating at $\vec{q}$, in terms of spherical waves for which a second point $\vec{t}$ is origin. If the spherical angles implied in the arguments $\vec{R} = \vec{p} - \vec{q}$, $\vec{\rho} = \vec{q} - \vec{t}$, $\vec{\epsilon} = \vec{p} - \vec{t}$ are referred to the same rectangular axes through relations formally identical with (1.1), then such expansions are relatively easy to obtain. For instance, the known addition formula for $h_0(kR) = f'_{e00}(k,\vec{R})$ can be written (for $\rho > \epsilon$)

$$f'_{e00}(\vec{R}) = \Sigma_{n=0}^{\infty} \Sigma_{m=0}^{n} [m,n] \left\{ f'_{emn}(\vec{\rho}) f_{emn}(\vec{\epsilon}) + f'_{omn}(\vec{\rho}) f_{omn}(\vec{\epsilon}) \right\}$$

where $[m,n] = \left(\frac{1}{4\pi} \int_0^{2\pi} \int_0^{\pi} P_n^m(\cos\theta) \cos^2 m\phi \sin\theta \, d\theta \, d\phi \right)^{-1} =$

Table III
Plane Waves from Planar Distributions of
Sources of Spherical Waves

$$p = (x,y,z), \quad q = (x',y',0), \quad R = p - q$$

$$\int f'_{e0n} \equiv \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dx' \; dy' \; f'_{e0n}\,(R)$$

$$= 2\{\pi(-i)^n\, e^{ikz}/k^2\}, \quad z > z'$$

$$= 2<i^n\pi\, e^{-ikz}/k^2>, \quad z < z'$$

$$\int \ell'_{e0n} = 2ik\{\}k, \quad z > z'; \quad = -2ik<>\,k, \quad z < z'$$

$$\int m'_{e1n} = -n(n+1)\{\}j, \quad z > z'; \quad = -n(n+1)<>j, \quad z < z'$$

$$\int m_{o1n} = n(n+1)\{\}i, \quad z > z'; \quad = n(n+1)<>i, \quad z < z'$$

$$\int n_{e1n} = in(n+1)\{\}i, \quad z > z'; \quad = -in(n+1)<>i, \quad z < z'$$

$$\int n_{o1n} = in(n+1)\{\}j, \quad z > z'; \quad = -in(n+1)<>j, \quad z < z'$$

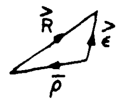
For all other combinations of functions and indices, the integral vanishes.

[Other formula of this type, for spherical and cylindrical wave functions, are given in NRL Report 4747.]

$$= [2 - \delta(m,0)](2n+1)(n-m)!/(n+m)! \qquad (1.10)$$

When $\rho < \epsilon$, the primes on the right appear on functions of $\vec{\rho}$ instead of $\vec{\epsilon}$. To obtain the similar expansion of $f_{e01}(k,\vec{R})$, we take the $\vec{k}$-components of the $\vec{l}$'s given in Tables I and II to regain scalar waves on both sides. The result is of course rather cumbersome and its generalization to higher-order waves on the left is even more so. Furthermore, the process does not lend itself easily to obtaining addition formulas for the transverse vector waves $\vec{m},\vec{n}$. The vector $\vec{\rho}$ is invariant with respect to $\vec{p}$, and although the identity $\nabla_p = -\nabla_q$ (or $\nabla_R = -\nabla_\rho$) could be used, the vector wave functions on the right would have both $\vec{p}$ and $\vec{\epsilon}$ as argument, and it would be tedious to transform the result into the more desirable form in which $\vec{\rho}$ is the argument of scalar waves, $\vec{\epsilon}$ of vector waves. Hence, with physical applications in mind, we simplify the calculations and resulting expressions by confining functions of $\vec{R}$ and $\vec{\epsilon}$ to n-index two or less, carrying functions of $\vec{\rho}$ with whatever indices are necessary. The derivation proceeded tediously as follows: First the addition formula (1.10) was written in tabular form as Row 1 of Table IV. Then $\vec{i},\vec{j},\vec{k}$ components of $\nabla_p = -\nabla_q$ were taken, and entered similarly from Table II to give the partial addition formulas for the $f_1(\vec{R})$. On these the process was then repeated, an $f_{e00}(\vec{R})$ appearing in the

Table IV. Addition formulas for scalar spherical wave functions.
Entries are functions of $\vec{\rho}$; e00 $\doteq f_{e00}(\vec{\rho})$, etc.



$f'(\mathbf{R})$ \ $f(\epsilon)$	$f_{e00}(\epsilon)$	$f_{e01}(\epsilon)$	$f_{e11}(\epsilon)$	$f_{o11}(\epsilon)$	$f_{e02}(\epsilon)$	$f_{e12}(\epsilon)$	$f_{e22}(\epsilon)$	$f_{o12}(\epsilon)$	$f_{o22}(\epsilon)$
$f'_{e00}(\mathbf{R})$	e00	3 e01	3 e11	3 o11	5 e02	$\frac{5}{3}$ e12	$\frac{5}{12}$ e22	$\frac{5}{3}$ o12	$\frac{5}{12}$ o22
$f'_{e01}(\mathbf{R})$	− e01	$\begin{bmatrix} \text{e00} \\ -2\ \text{e02} \end{bmatrix}$	− e12	− o12	$\begin{bmatrix} 2\ \text{e01} \\ -3\ \text{e03} \end{bmatrix}$	$\begin{bmatrix} \text{e11} \\ -\frac{2}{3}\ \text{e13} \end{bmatrix}$	$-\frac{1}{12}$ e23	$\begin{bmatrix} \text{o11} \\ -\frac{2}{3}\ \text{o13} \end{bmatrix}$	$-\frac{1}{12}$ o23
$f'_{e11}(\mathbf{R})$	− e11	− e12	$\begin{bmatrix} \text{e00} \\ +\ \text{e02} \\ -\frac{1}{2}\ \text{e22} \end{bmatrix}$	$-\frac{1}{2}$ o22	− e11 − e13	$\begin{bmatrix} \text{e01} \\ +\ \text{e03} \\ -\frac{1}{6}\ \text{e23} \end{bmatrix}$	$\begin{bmatrix} \frac{1}{2}\ \text{e11} \\ +\frac{1}{12}\ \text{e13} \\ -\frac{1}{24}\ \text{e33} \end{bmatrix}$	$-\frac{1}{6}$ o23	$\begin{bmatrix} \frac{1}{2}\ \text{o11} \\ +\frac{1}{12}\ \text{o13} \\ -\frac{1}{24}\ \text{o33} \end{bmatrix}$
$f'_{o11}(\mathbf{R})$	− o11	− o12	$-\frac{1}{2}$ o22	$\begin{bmatrix} \text{e00} \\ +\ \text{e02} \\ +\frac{1}{2}\ \text{e22} \end{bmatrix}$	$\begin{bmatrix} -\ \text{o11} \\ -\ \text{o13} \end{bmatrix}$	$-\frac{1}{6}$ o23	$\begin{bmatrix} -\frac{1}{2}\ \text{o11} \\ -\frac{1}{12}\ \text{o13} \\ -\frac{1}{24}\ \text{o33} \end{bmatrix}$	$\begin{bmatrix} \text{e01} \\ +\ \text{e03} \\ +\frac{1}{6}\ \text{e23} \end{bmatrix}$	$\begin{bmatrix} \frac{1}{2}\ \text{e11} \\ +\frac{1}{12}\ \text{e13} \\ +\frac{1}{24}\ \text{e33} \end{bmatrix}$
$f'_{e02}(\mathbf{R})$	e02	$\begin{bmatrix} -\frac{6}{5}\ \text{e01} \\ +\frac{9}{5}\ \text{e03} \end{bmatrix}$	$\begin{bmatrix} \frac{3}{5}\ \text{e11} \\ +\frac{3}{5}\ \text{e13} \end{bmatrix}$	$\begin{bmatrix} \frac{3}{5}\ \text{o11} \\ +\frac{3}{5}\ \text{o13} \end{bmatrix}$	$\begin{bmatrix} \text{e00} \\ -\frac{10}{7}\ \text{e02} \\ +\frac{18}{7}\ \text{e04} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{21}\ \text{e12} \\ +\frac{3}{7}\ \text{e14} \end{bmatrix}$	$\begin{bmatrix} \frac{5}{42}\ \text{e22} \\ +\frac{1}{28}\ \text{e24} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{21}\ \text{o12} \\ +\frac{3}{7}\ \text{o14} \end{bmatrix}$	$\begin{bmatrix} \frac{5}{42}\ \text{o22} \\ +\frac{1}{28}\ \text{o24} \end{bmatrix}$
$f'_{e12}(\mathbf{R})$	e12	$\begin{bmatrix} -\frac{9}{5}\ \text{e11} \\ +\frac{6}{5}\ \text{e13} \end{bmatrix}$	$\begin{bmatrix} -\frac{9}{5}\ \text{e01} \\ -\frac{9}{5}\ \text{e03} \\ +\frac{3}{10}\ \text{e23} \end{bmatrix}$	$\frac{3}{10}$ o23	$\begin{bmatrix} -\frac{5}{7}\ \text{e12} \\ +\frac{9}{7}\ \text{e14} \end{bmatrix}$	$\begin{bmatrix} \text{e00} \\ -\frac{5}{7}\ \text{e02} \\ -\frac{12}{7}\ \text{e04} \\ -\frac{5}{14}\ \text{e22} \\ +\frac{1}{7}\ \text{e24} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{14}\ \text{e12} \\ -\frac{3}{28}\ \text{e14} \\ +\frac{1}{56}\ \text{e34} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{14}\ \text{o22} \\ +\frac{1}{7}\ \text{o24} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{14}\ \text{o12} \\ -\frac{3}{28}\ \text{o14} \\ +\frac{1}{56}\ \text{o34} \end{bmatrix}$
$f'_{e22}(\mathbf{R})$	e22	$\frac{3}{5}$ e23	$\begin{bmatrix} -\frac{18}{5}\ \text{e11} \\ -\frac{3}{5}\ \text{e13} \\ +\frac{3}{10}\ \text{e33} \end{bmatrix}$	$\begin{bmatrix} \frac{18}{5}\ \text{o11} \\ +\frac{3}{5}\ \text{o13} \\ +\frac{3}{10}\ \text{o33} \end{bmatrix}$	$\begin{bmatrix} \frac{10}{7}\ \text{e22} \\ +\frac{3}{7}\ \text{e24} \end{bmatrix}$	$\begin{bmatrix} -\frac{10}{7}\ \text{e12} \\ -\frac{3}{7}\ \text{e14} \\ +\frac{1}{14}\ \text{e34} \end{bmatrix}$	$\begin{bmatrix} \text{e00} \\ +\frac{10}{7}\ \text{e02} \\ +\frac{3}{7}\ \text{e04} \\ +\frac{1}{56}\ \text{e44} \end{bmatrix}$	$\begin{bmatrix} \frac{10}{7}\ \text{o12} \\ +\frac{3}{7}\ \text{o14} \\ +\frac{1}{14}\ \text{o34} \end{bmatrix}$	$\frac{1}{56}$ o44
$f'_{o12}(\mathbf{R})$	o12	$\begin{bmatrix} -\frac{9}{5}\ \text{o11} \\ +\frac{6}{5}\ \text{o13} \end{bmatrix}$	$\frac{3}{10}$ o23	$\begin{bmatrix} -\frac{9}{5}\ \text{e01} \\ -\frac{9}{5}\ \text{e03} \\ -\frac{3}{10}\ \text{e23} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{7}\ \text{o12} \\ +\frac{9}{7}\ \text{o14} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{14}\ \text{o22} \\ +\frac{1}{7}\ \text{o24} \end{bmatrix}$	$\begin{bmatrix} \frac{5}{14}\ \text{o12} \\ +\frac{3}{28}\ \text{o14} \\ +\frac{1}{56}\ \text{o34} \end{bmatrix}$	$\begin{bmatrix} \text{e00} \\ -\frac{5}{7}\ \text{e02} \\ -\frac{12}{7}\ \text{e04} \\ +\frac{5}{14}\ \text{e22} \\ -\frac{1}{7}\ \text{e24} \end{bmatrix}$	$\begin{bmatrix} -\frac{5}{14}\ \text{e12} \\ -\frac{3}{28}\ \text{e14} \\ -\frac{1}{56}\ \text{e34} \end{bmatrix}$
$f'_{o22}(\mathbf{R})$	o22	$\frac{3}{5}$ o23	$\begin{bmatrix} -\frac{18}{5}\ \text{o11} \\ -\frac{3}{5}\ \text{o13} \\ +\frac{3}{10}\ \text{o33} \end{bmatrix}$	$\begin{bmatrix} -\frac{18}{5}\ \text{e11} \\ -\frac{3}{5}\ \text{e13} \\ -\frac{3}{10}\ \text{e33} \end{bmatrix}$	$\begin{bmatrix} \frac{10}{7}\ \text{o22} \\ +\frac{3}{7}\ \text{o24} \end{bmatrix}$	$\begin{bmatrix} -\frac{10}{7}\ \text{o12} \\ -\frac{3}{7}\ \text{o14} \\ +\frac{1}{14}\ \text{o34} \end{bmatrix}$	$\frac{1}{56}$ o44	$\begin{bmatrix} -\frac{10}{7}\ \text{e12} \\ -\frac{3}{7}\ \text{e14} \\ -\frac{1}{14}\ \text{e34} \end{bmatrix}$	$\begin{bmatrix} \text{e00} \\ +\frac{10}{7}\ \text{e02} \\ +\frac{3}{7}\ \text{e04} \\ -\frac{1}{56}\ \text{e44} \end{bmatrix}$

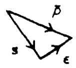
left column was subtracted out by means of the original addition formula, and duplicated entries for the same functions of $\vec{R}$ were compared as a partial check. Table IV presents the results. Here, to get addition formulas for $\vec{l}_n(\vec{R})$, one uses the fact that $\nabla_R = \nabla_\epsilon$, so that the f's in column and row headings are merely replaced with the corresponding $\vec{l}$'s.

A somewhat similar compression in presenting the partial addition formulas for the transverse vector waves $\vec{m},\vec{n}$ is made possible through eqs. (6), (7) and the fact that $\nabla_R = \nabla_\epsilon$; hence a table of expansions of $\vec{m}(\vec{R})$ becomes one for $\vec{n}(\vec{R})$ when $\vec{m}(\vec{\epsilon})$, $\vec{n}(\vec{\epsilon})$ are replaced by the $\vec{n}(\vec{\epsilon})$, $\vec{m}(\vec{\epsilon})$ having the same indices (Table V). To obtain the tabulated entries, we noted first from Table II that with $j_0(k\epsilon) \to 1$ with $\epsilon \to 0$,

$$\vec{n}_{e01}(\vec{\epsilon}) \to 2\vec{k}/3, \; \vec{n}_{e11}(\vec{\epsilon}) \to 2\vec{i}/3, \; \vec{n}_{o11}(\vec{\epsilon}) \to 2\vec{j}/3 \qquad (1.11)$$

whereas all other $\vec{m}$, $\vec{n}$ vanish. Hence the entries in the first three columns are prescribed by the component formulas of Table II. (The analogous property of the $\vec{l}_1$ was then used to check Table IV.) Taking $\nabla_R \times \; = \nabla_\epsilon \times$ in the manner mentioned then allowed the next three columns to be filled in from the $\vec{n}$ - rows of Table II. Then each component of each expanded function was written out through $f_1(\vec{\epsilon})$ terms, from Table IV. The known $\vec{m}(\vec{\epsilon})$ entries could be subtracted away, leaving the remaining entries to be identified as coefficients of $\vec{n}_2(\vec{\epsilon})$'s. The final $\vec{m}_2(\vec{\epsilon})$ entries were found

TABLE V



Expansions of $m(\bar\rho)$ in terms of $m(\epsilon)$, $n(\epsilon)$, and $f(s)$. The usual forms are in terms of $f(R) = f(-s)$; to get these forms, use $f_{\substack{e\\o}mn}(R=-s) = (-1)^n f_{\substack{e\\o}mn}(s)$, i.e., change signs only of f's with odd last index.

	$n_{e01}(\epsilon)$	$n_{e11}(\epsilon)$	$n_{o11}(\epsilon)$	$m_{e01}(\epsilon)$	$m_{e11}(\epsilon)$	$m_{o11}(\epsilon)$	$n_{e02}(\epsilon)$	$n_{e12}(\epsilon)$	$n_{e22}(\epsilon)$	$n_{o12}(\epsilon)$	$n_{o22}(\epsilon)$	$m_{e02}(\epsilon)$	$m_{e12}(\epsilon)$	$m_{e22}(\epsilon)$	$m_{o12}(\epsilon)$	$m_{o22}(\epsilon)$
				e00	$\frac{1}{2}$ e12							− e01	$-\frac{18}{36}$ e11		$-\frac{18}{36}$ o11	
$m(\bar\rho)_{e01}$		$-\frac{3}{2}$ o11	$\frac{3}{2}$ e11	+ e02		$\frac{1}{2}$ o12		$\frac{10}{36}$ o12	$\frac{10}{72}$ o22	$-\frac{10}{36}$ e12	$-\frac{10}{72}$ e22	− e03	$-\frac{8}{36}$ e13	$-\frac{2}{72}$ e23	$-\frac{8}{36}$ o13	$-\frac{2}{72}$ o23
					e00							$\frac{1}{2}$ e11	$-\frac{18}{36}$ e01	$-\frac{18}{72}$ e11		$-\frac{18}{72}$ o11
$m(\bar\rho)_{e11}$	$\frac{3}{2}$ o11		$-\frac{3}{2}$ e01	$\frac{1}{2}$ e12	$-\frac{1}{2}$ e02	$\frac{1}{4}$ o22	$-\frac{10}{12}$ o12	$-\frac{10}{72}$ o22	$-\frac{10}{72}$ o12	$\frac{30}{36}$ e02	$\frac{10}{72}$ e12	$-\frac{1}{3}$ e13	$+\frac{12}{36}$ e03	$+\frac{2}{72}$ e13	$-\frac{2}{36}$ o23	$+\frac{2}{72}$ o13
					$+\frac{1}{4}$ e22					$+\frac{10}{72}$ e22			$-\frac{2}{36}$ e23	$-\frac{1}{72}$ e33		$-\frac{1}{72}$ o33
						e00		$-\frac{10}{12}$ e02				$\frac{1}{2}$ o11		$\frac{18}{72}$ o11	$-\frac{18}{36}$ e01	$-\frac{18}{72}$ e11
$m(\bar\rho)_{o11}$	$-\frac{3}{2}$ e11	$\frac{3}{2}$ e01		$\frac{1}{2}$ o12	$\frac{1}{4}$ o22	$-\frac{1}{2}$ e02	$\frac{10}{12}$ e12	$+\frac{10}{72}$ e22	$-\frac{10}{72}$ e12	$\frac{10}{72}$ o22	$-\frac{10}{72}$ o12	$-\frac{1}{3}$ o13	$-\frac{2}{36}$ o23	$-\frac{2}{72}$ o13	$+\frac{12}{36}$ e03	$+\frac{2}{72}$ e13
						$-\frac{1}{4}$ e22								$-\frac{1}{72}$ o33	$+\frac{2}{36}$ e23	$+\frac{1}{72}$ e33
				$\frac{18}{10}$ e01	$-\frac{9}{10}$ e11	$-\frac{9}{10}$ o11		$-\frac{6}{12}$ o11		$\frac{6}{12}$ e11		e00	$-\frac{10}{84}$ e12	$\frac{8}{84}$ e22	$-\frac{10}{84}$ o12	$\frac{8}{84}$ o22
$m(\bar\rho)_{e02}$		$-\frac{3}{2}$ o12	$\frac{3}{2}$ e12	$+\frac{18}{10}$ e03	$+\frac{6}{10}$ e13	$+\frac{6}{10}$ o13		$+\frac{4}{12}$ o13	$\frac{2}{24}$ o23	$-\frac{4}{12}$ e13	$-\frac{2}{24}$ e23	$-\frac{10}{14}$ e02	$-\frac{24}{84}$ e14	$-\frac{2}{84}$ e24	$-\frac{24}{84}$ o14	$-\frac{2}{84}$ o24
												$-\frac{24}{14}$ e04				
					$\frac{27}{10}$ e01								e00			
			$-\frac{9}{2}$ e02	$\frac{27}{10}$ e11	$-\frac{18}{10}$ e03		$\frac{6}{4}$ o11		$-\frac{6}{24}$ o11	$-\frac{6}{12}$ e01	$\frac{6}{24}$ e11	$-\frac{5}{14}$ e12	$-\frac{30}{84}$ e02	$-\frac{15}{84}$ e12	$-\frac{15}{84}$ o22	$-\frac{15}{84}$ o12
$m(\bar\rho)_{e12}$	$\frac{3}{2}$ o12	$\frac{3}{4}$ o22	$+\frac{3}{4}$ e22	$+\frac{12}{10}$ e13	$+\frac{3}{10}$ e23	$\frac{3}{10}$ o23	$-\frac{4}{4}$ o13		$-\frac{6}{24}$ o13	$+\frac{24}{12}$ e03	$+\frac{6}{24}$ e13	$-\frac{12}{14}$ e14	$+\frac{96}{84}$ e04	$+\frac{6}{84}$ e14	$-\frac{8}{84}$ o24	$+\frac{6}{84}$ o14
									$+\frac{1}{24}$ o33		$-\frac{1}{24}$ e33		$-\frac{15}{84}$ e22	$-\frac{1}{84}$ e34		$-\frac{1}{84}$ o34
													$-\frac{8}{84}$ e24			
					$\frac{54}{10}$ e11	$-\frac{54}{10}$ o11		$\frac{12}{12}$ o11		$\frac{12}{12}$ e11			$-\frac{60}{84}$ e12	e00	$\frac{60}{84}$ o12	
$m(\bar\rho)_{e22}$	3 o22	− 3 o12	− 3 e12	$\frac{6}{10}$ e23	$-\frac{6}{10}$ e13	$+\frac{6}{10}$ o13	$-\frac{4}{4}$ o23	$+\frac{12}{12}$ o13		$+\frac{12}{12}$ e13	$-\frac{24}{24}$ e01	$\frac{10}{14}$ e22	$+\frac{24}{84}$ e14	$+\frac{60}{84}$ e02	$-\frac{24}{84}$ o14	$-\frac{1}{84}$ o44
					$+\frac{3}{10}$ e33	$+\frac{3}{10}$ o33		$-\frac{2}{12}$ o33		$+\frac{2}{12}$ e33	$-\frac{24}{24}$ e03	$-\frac{4}{14}$ e24	$-\frac{4}{84}$ e34	$-\frac{24}{84}$ e04	$-\frac{4}{84}$ o34	
														$-\frac{1}{84}$ e44		
															e00	
		$\frac{9}{2}$ e02		$\frac{27}{10}$ o11		$\frac{27}{10}$ e01	$-\frac{6}{4}$ e11	$\frac{6}{12}$ e01	$-\frac{6}{24}$ e11		$-\frac{6}{24}$ o11	$-\frac{5}{14}$ o12	$-\frac{15}{84}$ o22	$\frac{15}{84}$ o12	$-\frac{30}{84}$ e02	$-\frac{15}{84}$ e12
$m(\bar\rho)_{o12}$	$-\frac{3}{2}$ e12	$+\frac{3}{4}$ e22	$\frac{3}{4}$ o22	$+\frac{12}{10}$ o13	$\frac{3}{10}$ o23	$-\frac{18}{10}$ e03	$+\frac{4}{4}$ e13	$-\frac{24}{12}$ e03	$-\frac{6}{24}$ e13		$-\frac{6}{24}$ o13	$-\frac{12}{14}$ o14	$-\frac{8}{84}$ o24	$-\frac{6}{84}$ o14	$+\frac{96}{84}$ e04	$+\frac{6}{84}$ e14
						$-\frac{3}{10}$ e23			$-\frac{1}{24}$ e33		$-\frac{1}{24}$ o33			$-\frac{1}{84}$ o34	$+\frac{15}{84}$ e22	$+\frac{1}{84}$ e34
															$+\frac{8}{84}$ e24	
																e00
					$\frac{54}{10}$ o11	$\frac{54}{10}$ e11		$-\frac{12}{12}$ e11	$\frac{24}{24}$ e01	$\frac{12}{12}$ o11		$\frac{10}{14}$ o22	$-\frac{60}{84}$ o12		$-\frac{60}{84}$ e12	$+\frac{60}{84}$ e02
$m(\bar\rho)_{o22}$	− 3 e22	3 e12	− 3 o12	$\frac{6}{10}$ o23	$-\frac{6}{10}$ o13	$-\frac{6}{10}$ e13	$\frac{4}{4}$ e23	$-\frac{12}{12}$ e13	$+\frac{24}{24}$ e03	$+\frac{12}{12}$ o13		$-\frac{4}{14}$ o24	$+\frac{24}{84}$ o14	$-\frac{1}{84}$ o44	$+\frac{24}{84}$ e14	$-\frac{24}{84}$ e04
					$+\frac{3}{10}$ o33	$-\frac{3}{10}$ e33		$+\frac{2}{12}$ e33		$+\frac{2}{12}$ o33			$-\frac{4}{84}$ o34		$+\frac{4}{84}$ e34	$+\frac{1}{84}$ e44

similarly, via $\nabla_R X = \nabla_\epsilon X$. Here it was necessary to extend the first four columns of Table IV to include the $f_3(\vec{R})$.

For checking, Table V has been expanded componentwise, using the scalar formulas of Table IV. The functions $\vec{n}_3(\vec{\epsilon})$ have component terms in $f_2(\vec{\epsilon})$. These were found and the $f_2(\vec{\epsilon})$ accounted for among the $\vec{n}_1(\vec{\epsilon})$, $\vec{m}_2(\vec{\epsilon})$, and $\vec{n}_3(\vec{\epsilon})$. The f_2 terms in the component-expansions of $\vec{l}_3$ were then found and used in a similar check of Table IV. The entries $f_4(\rho)$ were not touched in this, so that a scheme of 90^o axis rotations which fairly well scrambled all entries were used. The probably well-known fact was rediscovered that all spherical harmonics with n-index two are representable by superpositions of the harmonics contained in f_{e02}, referred to different axes. This means that in any linear problem the quadrupolar scattering coefficients computed for incident waves f_{e02}, $\vec{l}_{e02}$, $\vec{m}_{e02}$, $\vec{n}_{e02}$ suffice to determine the scattering coefficients for all incident waves f_2, $\vec{l}_2$, $\vec{m}_2$, $\vec{n}_2$, respectively.

We notice, in Table IV, a symmetry about the main diagonal: the entries at the 'intersection' of the $f_{xmn}(\vec{R})$ row, $f_{x'm'n'}(\vec{\epsilon})$ column are identical with those common to the $f_{x'm'n'}(\vec{R})$ row, $f_{xmn}(\vec{\epsilon})$ column, except for a numerical factor which we now discuss from the point of view of a reciprocity principle.

The wave

$$F = \begin{cases} f_{smn}(k,\vec{R})\, h_n(kR'), & R < R' \\ f'_{smn}(k,\vec{R})\, j_n(kR'), & R > R' \end{cases}$$

satisfied (1.2) everywhere except at R = R' (a spherical surface centered on $\vec{q}$) where F, but not its R-derivative, is continuous. The surface R = R' thus contains a source distribution for the wave, which is then 'outgoing' for R > R', finite at R = 0, and satisfies

$$\nabla_\rho^2 F + k^2 F = P_n^m(\cos\theta)\begin{Bmatrix}\cos m\phi \\ \sin m\phi\end{Bmatrix}\frac{i}{kR'^2}\,\delta(R-R')$$

everywhere, the right hand side expressing the sources. Similarly, a wave

$$G = \begin{cases} f_{s'm'n'}(k,\vec{\epsilon})\, h_{n'}(k\epsilon'), & \epsilon < \epsilon' \\ f'_{s'm'n'}(k,\vec{\epsilon})\, j_{n'}(k\epsilon'), & \epsilon > \epsilon' \end{cases}$$

is the solution of

$$\nabla_\epsilon^2 G + k^2 G = P_{n'}^{m'}(\cos\alpha)\begin{Bmatrix}\cos m'\beta \\ \sin m'\beta\end{Bmatrix}\frac{i}{k\epsilon'^2}\,\delta(\epsilon-\epsilon')$$

where the spherical surface $\epsilon = \epsilon'$ is centered on $\vec{t}$. Again we take $\vec{R} = \vec{p} - \vec{q}$, $\vec{\epsilon} = \vec{p} - \vec{t}$, $\vec{\rho} = \vec{q} - \vec{t}$, $\vec{p} = (x,y,x)$ and stipulate that $\rho > |\epsilon' + R'|$, so that the 'sources' do not overlap. By Green's theorem and the orthogonality properties of the spherical harmonics, we then calculate

$$0 = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} dx\, dy\, dz\ \nabla_{\rho}^{2}\,(FG - GF)$$

and find that the $(-\vec{\rho})$-dependent coefficient of $f_{s'm'n'}(\vec{R}')$ in the expansion of $f_{smn}(\vec{R})$ about $\vec{q}$, multiplied by the $[m,n]$ of (1.10) is identically equal the $\vec{\rho}$- dependent coefficient of $f_{s'm'n'}(\vec{\epsilon})$ in the expansion of $f_{smn}(\vec{R})$ about $\vec{t}$, multiplied by $[m',n']$.

Thus the symmetry about the diagonal of Table IV would be complete (except for opposite signs when the n-index of $f_n(\vec{\rho})$ is odd) had the column headings been multiplied by the appropriate $[m,n]$. Similarly, entries in row $X(\vec{R})$, column $Y(\vec{\epsilon})$, Table V, differ from the 'symmetric' entries in row $Y(\vec{R})$, column $X(\vec{\epsilon})$ by factors which are demonstrably required by the electromagnetic reciprocity theorem. These factors are systematically related through 'normalization' factors,

$\frac{n(n+1)}{2n+1} \cdot \frac{(n+m)!}{(n-m)!}$, encountered in verifying the tables according to reciprocity; the sign of the 'symmetric' entries are again the same or different according as the n-indices of the entries are even or odd: $f_n(\vec{\rho}) = (-1)^n f_n(-\vec{\rho})$. (Relabeling the column heading in Table IV, V to display these symmetries would result in more complication in the important 'diagonal' entries and in the subsequent applications.)

We may regard the addition formulas of Tables IV, V as giving the partial excitation of a particle T centered at $\vec{t} = (x'', y'', z'')$ due to waves scattered by some particle at $\vec{q}$, as function of the relative position vector $\vec{\rho} = \vec{q} - \vec{t}$. It will be useful to compute the total excitation of T (through terms with n-index 2) on the premises that the particles Q which scatter waves to T are uniformly distributed through a space Σ, the half space $z > 0$ minus some sphere of radius r centered on $\vec{t}$, and that the wave scattered by the particle at the general point $\vec{q} = (x', y', z')$ in this region varies only through some phase factor $\exp(ik'z')$, We thus evaluate

$$I = \iiint_{\Sigma} \exp(ik'z') f'(k, \vec{\rho})\, d\vec{q} \tag{1.12}$$

Now f' satisfies $\nabla_q^2 f' + k^2 f' = 0$ whereas $E = \exp(ik'z')$ satisfies $\nabla^2 E + k'^2 E = 0$; we therefore have, by Green's Theorem:

$$(k'^2 - k^2) I = \iiint_{\Sigma} (E \nabla^2 f' - f' \nabla^2 E)\, d\,\mathrm{vol}(\vec{q})$$

$$= -\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} dx'dy' \left(\frac{\partial f'}{\partial z'} - ik'f' \right)\Big]_{z'=0} +$$

$$+ \iint_{\rho = r} \left\{ f' \frac{\partial E}{\partial \rho} - E \frac{\partial f'}{\partial \rho} \right\} d\,(\mathrm{area})$$

The first integral on the left may be evaluated by first using the $\vec{k}$-component of $\vec{1}$ from Table I, substituting $f_n(\vec{\rho}) = (-1)^n f_n(-\vec{\rho})$ and referring to Table III. For the second integral, we use the expansion of $e^{ik'z'}$ and the orthogonality relations of the spherical harmonics implied in f, f'. Both integrals vanish unless the m-index of f' vanishes; in this case, with $\sigma = k'/k$,

$$(\sigma^2 - 1)I = i2\pi i^n k^{-3} \cdot \left\{ (\sigma + 1)e^{ikz''} - 2\sigma^n R_n \, e^{ik'z''} \right\} \quad (1.13)$$

Expanded in powers of r through r^3, the $R_n(r)$ have the forms

$$R_0(r) = 1 + \tfrac{1}{2}(1-\sigma^2)k^2r^2 + i\,k^3r^3(1-\sigma^2)/3 + O[k^4r^4(1-\sigma^2)$$

$$R_n(r) = 1 + \tfrac{1}{2}(1-\sigma^2)\,k^2r^2/(2n+1) + O[k^4r^4(1-\sigma^2)], \; n \geqslant 1 \quad (1.14)$$

Lack of a cubic term in R_n, $n \geqslant 1$, is evident from $h_n = j_n + in_n$ and the forms of the power series expansions of the latter functions. It can also be proved by induction that $R_n - 1$ is divisible by $\sigma^2 - 1$, for all n 0.

The symmetry properties of Table IV, discussed earlier, are used in a similar calculation in which the space Σ is replaced by Σ', the interior of a sphere S, of radius r', centered on $\vec{q}$, minus a small sphere s of radius r again centered on $\vec{t}$ and interior to S.

With $\vec{R} = \vec{p} - \vec{q}$, $\vec{\epsilon} = \vec{p} - \vec{t}$, $\vec{\rho} = \vec{q} - \vec{t}$, we calculated

$$J = \int_{\Sigma'} f_{amn}(k',R) f'_{a'm'n'}(k,\vec{\epsilon}) \, d\,\mathrm{vol}\,(\vec{p}). \quad (1.15)$$

Via Green's theorem, the orthonormal properties of the spherical harmonics, and the symmetry exemplified in Table IV, we find that, except for a numerical constant,

$$(\sigma^2 - 1)k^3 J = R_n(r')F(k,\vec{\rho}) - R_{n'}(r)F(k',\vec{\rho}) \tag{1.16}$$

where, if Table IV were sufficiently extended, the F's would represent the $\vec{\rho}$ - dependent coefficient common to the $f'_{amn}(\vec{R})$ row and $f_{a'm'n'}(\vec{\epsilon})$ column except that the spherical Bessel functions (j's in this case) have argument $k\rho$ or $k'\rho$ according as k or k' appears in the F-symbol.

First (on physical grounds) we stipulate that $\mathrm{Re}(k,k') > 0$ so that $k' + k \neq 0$. Then we use the fact that

$$R_n = 1 + \left[\text{terms divisible by } (\sigma^2 - 1) = (k'^2 - k^2)/k^2\right].$$

With the quotient $Q = k\left[F(k,\vec{\rho}) - F(k',\vec{\rho})\right]/(k' - k)$ obviously well behaved at $k' = k$, we conclude that J is analytic in k,k' in all regions of physical interest.

With $r' > \rho$, $r' > r$ we now examine the magnitude of J when $\max\left\{ |kr'| \;, \; |k'r'| \right\} \equiv K \ll 1$. Since the F are weighted sums of various j's, the 'order' of F is K^{ν} $(\nu > 0)$ where ν is the smallest integer subscript of the j's in F; K^{ν} is then the order of J when $\nu \geq 1$. For $\nu = 0$, $F = O(1)$ but $Q = O(K^2)$, as is $\left[R_n(r') - R_{n'}(r)\right]/(1 - \sigma^2)$, so that $J = O(K^2)$. Hence $J \leq O(K)$, equality arising only from $f_1(\vec{\rho})$ term in $F(k,\vec{\rho})$.

Section II: Illustrative Application to an Artifical Dielectric

We apply the formulas developed in Section I to the propagation of waves through suspensions consisting of small particles randomly distributed throughout a fluid of known properties.

The particles will be assumed describable through a set of scattering coefficients, coefficients in the linear relation between waves incident on an isolated particle and the resulting waves scattered into the fluid. We shall assume that a shape S is formed of some of the suspension, and immersed in a bath of the fluid, and illuminated with known waves with (suppressed) time factor $\exp(-i\omega t)$. Then the excitation of any particle in S consists of the known incident wave plus waves scattered by all other particles of the suspension. In principle, our object is to calculate the average wave scattered by S into the ambient fluid; we take this wave as the superposition of waves scattered by all the particles in S, averaged over all arrangements of the particles.

To calculate the particle's excitation, we make the self-consistent assumption: we assume (a) that the particle lies inside a small 'sphere of exclusion', s, into which the remain-particles in S, however arranged, never enter; (b) that the wave incident on the particle is the average wave found in s in the absence of that particle; and finally, (c) that this average incident wave can be determined as an integral over S-s, with the excitation arising from a volume element dv at a point $\vec{q}$ in S - s again an average, namely, Ndv times the average scattering (evaluated in s) of a particle located at $\vec{q}$, where N is the average number of particles in a unit volume of suspension.

The radius of exclusion, r_e (the radius of s) enters this calculation through specifying one boundary surface of this volume integral. Physically, r_e is set by the dimensions of the non-overlapping particles, and perhaps should depend on the size of the particle in s. However, it will be convenient to take a single r_e for all particles. Conclusions about the mode of wave propagation in the suspension depend on the choice of r_e, but for sufficiently small particles, r_e may be taken small compared with wavelength in the fluid, and its influence disappears.

We illustrate the method by calculating the propagation of electromagnetic waves in a suspension of small particles in a fluid of dielectric constant ϵ, permeability μ, both constants being those effective at ω.

We assume that a unit volume of the suspension contains N identical small particles describable, electromagnetically, by dipole scattering coefficients A,B. Taking $\vec{t}$ as the center of s, $\vec{p}$ as a general point in s, and $\vec{\Delta t} = \vec{p} - \vec{t}$, the electric vector of the incident wave (the average wave inside s in the absence of a particle at $\vec{t}$) can be expressed as a superposition of waves of the form $\vec{m}_n = \vec{m}_{\delta mn}(\vec{\Delta t})$, $\vec{n}_n = \vec{n}_{\delta mn}(\vec{\Delta t})$, where the implied propagation constant k is that of the fluid: $k = \omega(\mu\epsilon)^{\frac{1}{2}}$. We assume that the particle responds only to the magnetic and electric dipole excitations $\vec{m}_1$ and $\vec{n}_1$ respectively, so that if the expansion (about $\vec{t}$) of the incident field in s starts with the terms $(u_x\vec{n}_{e11} + u_y\vec{n}_{o11} + u_z\vec{n}_{e01}) + (v_x\vec{m}_{e11} + v_y\vec{m}_{o11} + v_z\vec{m}_{e01})$ then the field scattered from $\vec{t}$ is of the form $A(u_x\vec{n}'_{e11} + u_y\vec{n}'_{o11} + u_z\vec{n}'_{e01}) + B(v_x\vec{m}'_{e11} + v_y\vec{m}'_{o11} + v_z\vec{m}'_{e01})$, where A and B are the appropriate scattering coefficients

For the geometry, we assume the suspension to fill the half-space $S = z > 0$, with fluid filling the space $z < 0$. We assume the wave $\vec{W}_i$ normally incident on S (from $z < 0$) to be plane with electric vector in the x-direction; this wave has an expansion about $\vec{t} = (x'', y'', z'',)$ of the form $\vec{W}_i = \vec{i}\exp(ikz) =$

$$= \exp(ikz'')\ \frac{3}{2}\left[\vec{n}_{e11}(\vec{\Delta t}) + i\vec{m}_{e11}(\vec{\Delta t}) + (\text{terms in } \vec{m}_n,\ \vec{n}_n,\ n > 1)\right]. \quad (2.1)$$

The total excitation $\vec{W}$ of the particle at $\vec{t}$ consists of this plus excitation $\vec{W}_S$ due to the scattering from other particles in $z > 0$: $\vec{W} = \vec{W}_i + \vec{W}_S$.

We now assume that the average wave propagating in the suspension travels according to some propagation constant k', as would certainly be the case had $z > 0$ been filled with a homogeneous medium. In the present plane-wave geometry, we assume, correspondingly, that

$$\vec{W} = e^{ik'z''}\left[U\vec{n}_{e11} + iV\vec{m}_{o11}\right] \equiv e^{ik'z''}\left[U\vec{n} + iV\vec{m}\right]; \tag{2.2}$$

other dipole excitations vanish by symmetry and higher-order excitations are omitted as producing no scattering. To find U,V we use the set of simultaneous equations implied in $\vec{W} = \vec{W}_i + \vec{W}_s$.

Now the scattering into s from a particle Q at $\vec{q} = (x',y',z')$, owing to Q's excitation-term $\exp(ik'z')\, U\vec{n}_{e11}$ is found, from the addition formulas of Table V, to be of the form: (2.3)

$$U\exp(ik'z')\left\{A\left[f'_{e00} - (1/2)f'_{e02} - (1/4)f'_{e22}\right]\vec{n}_{e11}(\vec{\Delta t}) + \right.$$

$$\left. + A(3/2)f'_{e01}\vec{m}_{o11}(\vec{\Delta t}) + \text{(other dipole terms)} + \left(\text{terms in } \vec{\rho} \text{ with n-index} > 0\right)\right\},$$

where the arguments of the function f' is $-\vec{\rho} = (\vec{q} - \vec{t})$. We multiply this by N and volume-integrate on $\vec{q}$ over S - s, to obtain the excitation at $\vec{t}$ due to scattering arising from the excitation term $\exp(ik'z')U\vec{n}_{e11}$.

General integrations of this type are given in (1.13); it is seen from Table V that 'other dipole terms' have coefficients $f(\vec{\rho})$ with positive n-indices and vanish by symmetry on integration. Similarly expanding the excitations of T due to the $V\vec{m}$ term of $\vec{W}$, and applying (1.13), we find:

$$\vec{W}_s(T) = e^{ikz''}\frac{3}{4}(\vec{n} + i\vec{m})(U\alpha + V\beta)/(1-\sigma) - \frac{e^{ik'z''}}{1-\sigma^2}\left\{\begin{array}{l}\vec{n}\left[\alpha U(R_0 + \frac{1}{2}\sigma^2 R_2) + \beta V\frac{3}{2}\sigma R_1\right] + \\ + i\vec{m}\left[\alpha U\frac{3}{2}\sigma R_1 + \beta V(R_0 + R_2\frac{1}{2}\sigma^2)\right]\end{array}\right\} \tag{2.4}$$

where $\alpha = -4\pi Nik^{-3}A$, $\beta = -4\pi Nik^{-3}B$, $\sigma = k'/k$.

From (2.1), (2.2), and (2.4) we see that the formal equation $\vec{W} = \vec{W}_i + \vec{W}_s$ amounts to four simultaneous equations, since $\vec{m}(\vec{\Delta t}) \neq \vec{n}(\vec{\Delta t})$ and $\exp(ikz'') \neq \exp(ik'z'')$ for general $\vec{\Delta t}$, z". From coefficients of $e^{ik'z}$, k' and the ratio U/V are determined, the former through the vanishing of the coefficient determinant, $|D|$, of the system

$$U\left\{1 - \sigma^2 + \alpha(R_0 + R_2\sigma^2/2)\right\} + V\tfrac{3}{2}\sigma R_1\beta = 0,$$

$$U\tfrac{3}{2}\sigma R_1\alpha + V\left\{1 - \sigma^2 + \beta(R_0 + R_2\sigma^2/2)\right\} = 0. \qquad (2.5)$$

The two equations in U,V determined from coefficients of $e^{ikz''}$ are linearly dependent, so that no contradiction arises with the U/V determined by (2.5); from (2.1), (2.2) and (2.6) we have

$$U\alpha + V\beta = -2(1 - \sigma). \qquad (2.6)$$

Now we discuss the suspension's propagation constant k', obtained by solving (2.5) for $\sigma = k'/k$, taking $R_n \equiv 1$ as befitting this case of small particles. The polynomial $|D|$ is now quadratic in σ^2 and factors readily, one factor being $\sigma^2 - 1$. The conclusion k' = k from $(\sigma^2 - 1) = 0$ is rejected, showing no influence of the suspended particles. Equating the other factor to zero yields:

$$\sigma^2 = k'^2/k^2 = \frac{1+\alpha}{1-\frac{1}{2}\alpha} \cdot \frac{1+\beta}{1-\frac{1}{2}\beta} \qquad (2.7)$$

Thus there is a single propagation constant k', the negative root having the usual interpretation as a wave, running in the negative z direction, which is not excited in the present geometry. Writing $k'^2 = \omega^2 \epsilon' \mu'$ we regard the first and second factors on the right of (2.7) as ϵ'/ϵ, μ'/μ, since the factors arise from electric and magnetic dipole scattering, respectively.

The interpretation of (2.6) is reinforced when one calculates (via Table III) the average wave $\vec{i}\, Re^{-ikz}$ scattered back into $z < 0$:

$$R = (1/2)(V\beta - U\alpha)/(1 + \sigma) = (1 - \mathfrak{z})/(1 + \mathfrak{z}) \qquad (2.8)$$

where $\mathfrak{z} = \left[(\epsilon'\mu)/(\mu'\epsilon)\right]^{1/2}$ is the usual relative characteristic impedance. This wave is identical with one reflected at the z = 0 interface when $\vec{W}_i$ is again incident and $z > 0$ is occupied by a homogeneous medium with the above ϵ', μ'.

Operationally, one determines μ', ϵ' by measuring reflected wave amplitudes under a variety of polarizations and obliquities of $\vec{W}_i$. We performed the entire calculation for the case of oblique incidence, using the present method and the formulas mentioned in passing in Section I; the reflected wave remained that obtainable through the present ϵ', μ' and the assignment is 'operationally' valid. Finally, the forms

$$\frac{\epsilon'}{\epsilon} = \frac{1+\alpha}{1-\frac{1}{2}\alpha}\ , \qquad \frac{\mu'}{\mu} = \frac{1+\beta}{1-\frac{1}{2}\beta} \qquad (2.9)$$

are those required by the Clausius-Mossotti law.

We see that the coefficients of $e^{ik'z}$ determine U/V and k', but that the influence of the boundary plane z = 0 appears as a limit in the integrals used to calculate (2.6) and (2.8). We therefore regard (2.6) and (2.8) as establishing the electromagnetic boundary conditions at z = 0. Eq. (2.6) may be regarded as giving the amplitude of the wave transmitted into z > 0, and (2.8) of course establishes that of the wave reflected by z = 0, but in both it is necessary to have determined the effective propagation constant k' (i.e., σ) first.

Section III. Heuristic Theory of Ferromagnetic Exchange Effects at Optical Frequencies

The equations governing electromagnetic wave propagation in a magnetized ferromagnetic metal are Maxwell's equations plus the (simplified) spin-wave equations:

$$\frac{1}{\gamma}\frac{\partial \vec{M}}{\partial t} = \vec{M} \times \left[\vec{H} + (2A/M_S^2)\nabla^2\vec{M}\right] \qquad (3.1)$$

which establishes the relation between the magnetic induction $\vec{B} = \vec{H} + 4\pi\vec{M}$ and the magnetic field $\vec{H}$. The constant A represents the 'exchange integral' and incorporates, for the ferromagnetic continuum, the effect of exchange forces between various atomic spins of the actual metal. The effect of this generally neglected term has been worked out in a special case by

Ament and Rado[1]. They found (following Macdonald[2]) that for propagation normal to an applied d-c magnetic field, the metal was triply refracting, and that at a plane air-metal interface, the new boundary conditions $Ad\vec{M}/d\vec{n} = 0$ had to be used to determine the three relative amplitudes.

We shall treat this problem by the method illustrated in Section II for the reasons that a continuum (i.e., a differential equation such as (3.1)) is not needed, that the exchange integral and the new boundary conditions enter naturally, and finally, that the range of the exchange force enters explicitly and without essential complication of the resulting algebra. (One might expect to include a range effect as a fourth-order derivative term in (3.1); this would give rise to a quintuply refracting medium.) The thin magnetic films described recently are semi-transparent,[3] so that it becomes possible to measure magnetic effects at optical frequencies; the range of exchange forces, comparable with interatomic distances, may no longer be insignificant with respect to electromagnetic wavelengths in the metal, so that the following results may have more than academic interest.

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1) W. S. Ament and G. T. Rado, "Electromagnetic Effects of Spin Wave Resonance in Ferromagnetic Metals", the Physical Review, Vol. 97, No. 6, 1558-1566, March 15, 1955.

2) J. R. Macdonald, Ph.D. Thesis, Oxford, 1950 (Unpublished).

3) C. A. Fowler, Jr. and E. M. Fryer, "Magnetic Domain in Thin Films by the Faraday Effect," Phys Rev, 104, 522, Oct 1956.

We assume that the ferromagnetic metal is a suspension consisting in an isotropic, non-magnetic fluid, of some appropriate metallic conductivity, in which randomly imbedded particles are responsible for the magnetic properties. Again we let the fluid fill $z < 0$, and the suspension fill $z > 0$, and let the plane wave with magnetic vector $\vec{h} = \vec{i}h_o \exp(ikz)$ be incident from $z < 0$. In addition, let the static magnetic field $\vec{j}H_e$ be applied. Now we describe the particle at $\vec{q}_i$ by the vector $\vec{S}(\vec{q}_i)$, and endow $\vec{S}$ with the properties that the magnetic field $\vec{H}(\vec{p})$ at an arbitrary point $\vec{p} = (x,y,z)$ in the fluid is given by

$$\vec{H}(\vec{p}) = \vec{H}_o(\vec{p}) + C \sum_i \vec{S}(\vec{q}_i) \cdot \overset{\Rightarrow}{T}(\vec{p}, \vec{q}_i, \omega) \tag{3.2}$$

and such that

$$\frac{1}{\gamma} \frac{\partial \vec{S}(q_i)}{\partial t} = -\frac{i\omega}{\gamma} \vec{S}(\vec{q}_i) =$$

$$= \vec{S}(\vec{q}_i) \times \vec{H}(\vec{q}_i) + D \sum_j{}' F(|\vec{q}_i - \vec{q}_j|) \vec{S}(\vec{q}_i) \times \vec{S}(\vec{q}_j) \tag{3.3}$$

Here $\overset{\Rightarrow}{T}(\vec{p},\vec{q};\omega)$ represents a 'dyadic Green's function', depending on the frequency ω, so that $\vec{S}$ acts as a radiating magnetic dipole. Eq. (3.3) is the dynamical equation for the spin $\vec{S}$, the first term representing a torque due to the local magnetic field and the second representing the exchange forces, with F containing the range dependence of the forces. The constants C and D are parameters amounting to scattering coefficients.

Now we replace the general vectors $\vec{H}$ and $\vec{S}$ by

$$\vec{H} = \vec{j} H_e + \vec{h} e^{-i\omega t}, \qquad \vec{H}_0 = \vec{j} H_e + \vec{i} h_0 e^{-i\omega t}$$

$$\vec{S} = \vec{j} S + \vec{s} e^{-i\omega t}$$

$$\vec{h} = h_x \vec{i} + h_z \vec{k}, \qquad \vec{s} = s_x \vec{i} + s_z \vec{k},$$

and will linearize the resulting (3.2) and (3.3) by ignoring terms quadratic in the lower-case quantities. Then, assuming a uniformly random distribution of the $\vec{q}_j$ in $z > 0$, we replace the sums by integrals over $z > 0$ minus small spheres centered on $\vec{p}$ and $\vec{q}_i$. Omitting $e^{-i\omega t}$, we than have, with $\vec{r} = \vec{p} - \vec{q}_i$

$$\vec{h}(\vec{p}) = \vec{i} h_0(\vec{p}) + NC \iiint_{\Sigma} [s_x(\vec{q})\, \vec{n}_{e11}(k, \vec{r}) + s_z(\vec{q})\, \vec{n}'_{e01}(k, \vec{r})]\, d\vec{q}$$

$$-\frac{i\omega}{\gamma} \vec{s}(\vec{p}) = S \vec{j} \times \vec{h}(\vec{p}) + \vec{s}(\vec{p}) \times \vec{j} H_e +$$

$$+ ND \iiint_{\Sigma} F(|\vec{p} - \vec{q}|) S \vec{j} \times [\vec{s}(\vec{q}) - \vec{s}(\vec{p})]\, d\vec{q}. \qquad (3.4)$$

In order to apply the results of Section I, it now becomes convenient to let $F(r) = f'_{e00}(k^* r) \equiv \exp(ik^* r)/(ik^* r)$ (where $k^* \equiv i\,|k^*|$ to make F real), and to assume that the wave in $z > 0$ varies as $\exp(ik'z)$, where the propagation constant k' is now to be determined through a secular determinant. From the $\exp(ik'z)$ dependences in the above equations, we obtain four simultaneous homogeneous equations in h_x, h_z, s_x, s_z;

$$h_x \pm c' s_x (2+\sigma^2)/(1-\sigma^2) = 0$$

$$h_z + 2c' s_z = 0$$

$$S h_z + (i\omega/\gamma) s_x + s_z[-H_e + D'\sigma^2/(1-\sigma^2\mu^2)] = 0$$

$$-S h_x + s_x[H_e - D'\sigma^2/(1-\sigma^2\mu^2)] + (i\omega/\gamma) s_z = 0, \tag{3.5}$$

where: $C' = -4\pi N i C/(3k^3)$

$D' = -\mu^2 4\pi N i D/k^3$

$\mu = k/k^*$

and where we have again taken $\sigma = k'/k$, and assumed $R_n = 1$.

Here D' is essentially the conventional exchange integral, and the effect of finite exchange range is incorporated in the μ^2 appearing explicitly in the last two equations.

The expansion of the secular determinant of this system gives a polynomial cubic in σ^2, so that there are three propagation constants $k' = k_1, k_2, k_3$.

To determine the amplitudes of these three waves, we need analogues of boundary conditions applicable at the interface $z = 0$. For this purpose we notice that the integrals on the right of (3.4) imply, from (1.13), that waves can propagate in $z > 0$ according to $\exp(ikz)$ and $\exp(ik^*z)$. The amplitudes of these waves are necessarily zero, since the presence of the $\vec{S}_i$ is ignored in $\exp(ikz)$ and the surrounding 'fluid' is ignored in $\exp(ik^*z)$. Setting the amplitudes of these unphysical waves to zero yields three non-trivial scalar 'boundary condition equations'

$$0 = h_0 + \frac{3}{2} c' [s_{1x}/(1-\sigma_1) + s_{2x}/(1-\sigma_2) + s_{3x}/(1-\sigma_3)] \qquad (3.6)$$

$$0 = k_1 s_{1z}/(1-\mu\sigma_1) + k_2 s_{2z}/(1-\mu\sigma_2) + k_3 s_{3z}/(1-\mu\sigma_3) \qquad (3.7)$$

$$0 = k_1 s_{1x}/(1-\mu\sigma_1) + k_2 s_{2x}/(1-\mu\sigma_2) + k_3 s_{3x}/(1-\mu\sigma_3) \qquad (3.8)$$

from the two vector equations (3.5).

The quantity of experimental interest is the impedance ratio $Z = (h_0 - h_r)/(h_0 + h_r)$, where h_r is the amplitude of the wave reflected back into $z < 0$. From Table III, following the method used toward (2.8), we find that the reflected wave is polarized in the x-direction and has amplitude

$$h_r = -\frac{3}{2} c' [s_{1x}/(1+\sigma_1) + s_{2x}/(1+\sigma_2) + s_{3x}/(1+\sigma_3)] . \qquad (3.9)$$

Equations (3.6) and (3.9) are equivalent to the standard electromagnetic boundary conditions (continuity of tangential components of $\vec{E}$ and $\vec{H}$), in the same manner as were (2.6) and (2.8). The 'new' boundary conditions are (3.7) and (3.8). These are identical with $d\vec{M}/dz = 0$ for $\mu = 0$, but differ slightly as μ becomes finite, i.e., as the range of exchange forces becomes comparable with the skin depth. (More precisely $d\vec{M}/dz = 0$ holds in the approximation that $\mu\sigma = k'/k^*$ becomes small compared with unity, i.e., that exchange is small compared with actual wavelengths in the metal.)

For a given k^*, one can now assign C',D',S, and H_e so that when $\mu = k/k^*$ is ignored, the physical significance of the present equations is identical with those used previously.[1)] Then with $\mu \neq 0$, the calculation of the roots of the secular polynomial and of Z can be carried out, by methods formally identical with those previously[1)] used, to show the effect of $\mu \neq 0$ on the previous results. These identifications and calculations will not be given here. What has been verified is that the previous formal result for $*$ [eq. (2.5), ref. (1)] is obtained when one sets $\mu = 0$ in all equations following (3.6).

Section IV. Frequency Dependence of Certain Artificial Dielectrics.

In this section we treat the problem of Section II for the case that the randomly suspended particles are small identical spheres of some radius r, and will be concerned with the r-dependence of the effective propagation constant, k'. The r-dependence occurs in several ways. The dipole scattering coefficients of the spheres, expanded in powers of $K = kr$, lead to replacement of the α, β of (2.4) by expansions of the form

$$\alpha' = \alpha + \alpha_2 K^2 + i\alpha_3 K^3,$$

$$\beta' = \beta + \beta_2 K^2 + i\beta_3 K^3. \tag{4.1}$$

The constants $\alpha, \alpha_2, \alpha_3, \beta, \beta_2, \beta_3$ depend on the properties of fluid and sphere-material. In the absence of ohmic losses, α_3 is related to α, β_3 to β, as required by Rayleigh scattering (see

Section VIII). Only the leading terms of these expansions are valid in the Clausius-Mossotti formulas (2.9) as derived there. The effect of $K > 0$ will appear as a 'real' effect, $O(K^2)$, and a lossy term, related to Rayleigh scattering, of order K^3.

Thus to obtain an effective loss in k', due to random scattering, we need to carry terms through K^6 in the scattering coefficients of the spheres (through K^3 in α', β'). The quadrupole coefficients a_2, b_2 of the Appendix have expansions starting out $C'K^5 + O(K^7)$, so that their leading terms must also be included. Higher-order coefficients start out K^n, $n \geq 7$, and can be neglected consistently. Finally, the centers of any two spheres are separated by distances $\geq 2r$, so that the 'radius of exclusion' r_e is finite and the R_n of (1.14) must enter the calculation.

We let the suspension fill the half-space $z > 0$ and let the plane wave $\vec{i}\exp(ikz)$ be incident from $z < 0$ as before. For the excitation $\exp(ik'z)\,(A\vec{m}_{o11} + B\vec{n}_{e11} + C\vec{m}_{o12} + D\vec{n}_{e12})$ we get the scattering $\exp(ik'z)\; A\beta\vec{m}'_{o11} + B\alpha'\vec{n}'_{e11} + C\delta\vec{m}'_{o12} + D\gamma\vec{n}'_{e12}$.

Higher order scattering is negligible and other dipole and quadrupole excitation will vanish by symmetry. We then calculate the total excitation on a sphere at $(0,0,z')$, again using the expansion of the plane incident wave and table IV followed by (1.13) From the coefficients of the four linearly independent functions $\exp(ik'z')(\vec{m}_1,\vec{n}_1,\vec{m}_2,\vec{n}_2)$ we find k' determined through the secular determinant $\quad 0 =$

$$= \begin{vmatrix} (\tau_0 + \frac{1}{2}\tau_2)\alpha' + 1 - \sigma^2 & \frac{3}{2} i \tau_1 \beta & i\left(\frac{27}{10}\sigma + \frac{18}{10}\sigma^3\right)\gamma & -\frac{9}{2}\sigma^2\delta \\ -3i\tau_1\alpha'/2 & (\tau_0 + \frac{1}{3}\tau_2)\beta + 1 - \sigma^2 & \frac{9}{2}\sigma^2\gamma & i\left(\frac{27}{10}\sigma + \frac{18}{10}\sigma^3\right)\delta \\ -i(\frac{1}{2}\tau_1 + \frac{1}{3}\tau_3)\alpha' & \frac{5}{6}\tau_2\beta & \left(1 + \frac{5}{14}\sigma^2 + \frac{8}{7}\sigma^4\right)\gamma + 1 - \sigma^2 & i\left(\frac{1}{2}\sigma + 2\sigma^3\right)\delta \\ -\frac{5}{6}\tau_2\alpha' & -i(\frac{1}{2}\tau_1 + \frac{1}{3}\tau_3)\beta & -i(\frac{1}{2}\sigma + 2\sigma^3)\gamma & \left(1 + \frac{5}{14}\sigma^2 + \frac{8}{7}\sigma^4\right)\delta + 1 - \sigma^2 \end{vmatrix} \tag{4.2}$$

where α', β' are as in (4.1), where

$$\gamma = -4\pi N i a_2/k^3, \qquad \delta = -4\pi N i b_2/k^3$$

and where $\tau_n = R_n \sigma^n$, R_n as in (1.14).

When the R_n's are carried to requisite power in $K_e = kr_e$, this appears to give a polynomial of high order in σ^2, but $(\sigma^2 - 1)^3$ factors out and the result is quadratic:

$$0 = 3A'(1-\sigma^2)^2 + B'(1-\sigma^2) + 3C', \quad \text{where}$$

$$A' = 2(\gamma+\delta)(80 - \alpha\beta) + 7K_e^2(\alpha\beta - \alpha - \beta) + 4(\alpha\gamma + \beta\delta) - 80(\alpha\delta + \beta\gamma),$$

$$B' = \left\{ \begin{array}{l} 6(\gamma+\delta)(19\alpha\beta - 185) - 237(\alpha\gamma + \beta\delta) - 300(\alpha\delta + \beta\gamma) + 420 + \\ + 105(\alpha\beta - 2\alpha - 2\beta) + 21K_e^2(\alpha\beta + 11\alpha + 11\beta) - 140 i K_e^3(\alpha\beta - \alpha - \beta) \end{array} \right\}$$

$$C' = \left\{ \begin{array}{l} 2(\gamma+\delta)(175 + 52\alpha\beta) + 215(\alpha\gamma + \beta\delta) + 320(\alpha\delta + \beta\gamma) + \\ + 105(\alpha\beta + 2\alpha + 2\beta) + 126K_e^2\alpha\beta + 140 i K_e^3\alpha\beta \end{array} \right\}$$

Thus there are two propagation constants k'. We examine that k' which is a 'perturbed' version of the $k' = k'_e$ of (2.7); expanding in powers of K and omitting terms of relative order K^4 we get, with $K_e = kr_e$,

$$\frac{k'}{k} = \sigma = \frac{k'_0}{k}\left\{\begin{array}{l} 1 - u(\alpha,\beta)\gamma - u(\beta,\alpha)\delta + P K_e^2 - iQ K_e^3 + \\ + [\alpha_2 K^2 + i\alpha_3 K^3]V(\alpha) + [\beta_2 K^2 + i\beta_3 K^3]V(\beta) \end{array}\right\}, \qquad (4.4)$$

where $u(\alpha,\beta) = \dfrac{(\beta+1)(\alpha+10)^2}{10(2-\beta)(2-\alpha)^2}$,

$$V(\alpha) = \frac{3}{2(2-\alpha)(1+\alpha)}$$

$$3Q = \alpha^2 V(\alpha) + \beta^2 V(\beta)$$

$$P = \frac{3\left\{\begin{array}{l} 4\alpha^3\beta^3 - 13\alpha^2\beta^2(\alpha+\beta) + 14\alpha\beta(\alpha^2+\beta^2) + \\ + 50\alpha^2\beta^2 + 16(\alpha^3+\beta^3) - 26\alpha\beta(\alpha+\beta) - \\ - 44(\alpha^2+\beta^2) - 40\alpha\beta \end{array}\right\}}{20(1+\alpha)(1+\beta)(2-\alpha)^2(2-\beta)^2}$$

Now we interpret the terms of (4.4) for the case of no ohmic losses, where α, β are proportional to the volume fraction f_1 occupied by spheres; for the case of no ohmic losses, they are real numbers lying between -1 and 2. Hence, in the lossless case, it is seen that the coefficients Q and V are non-negative, so that with $\alpha_3 > 0$, the $iVK^3\alpha_3$ is positive imaginary and corresponds to attenuation in a wave propatating as $\exp(ik'z)$, whereas $-iQK_e^3$ represents a <u>negative</u> attenuation. The attenuation due to the V-term seems attributable to Rayleigh scattering by the randomly arrayed spheres, whereas the negative attenuation from the Q-term appears to arise from the <u>decrease</u> in randomness caused by the finite radius of exclusion. This interpretation is supported by the fact that α_3 $V = O(f_1)$, $Q = O(f_1^2)$, for small f_1. For large $f_1 < 1$ and for reasonable values of $r_e/r \simeq 2$, physically realizable α, β are readily found so that the Q-terms exceed the V-terms and the total 'attenuation' $\mathrm{Im}(k')$ is negative. The reason for this breakdown of the theory is not clear to the writer. The U and P terms are real in the lossless case, but can change sign for suitable α, β (i.e., for suitable lossless electromagnetic constants of the fluid and the spheres). These and the K^2V terms then affect only the propagation velocity implied in k'.

The second propagation constant k' is $O(K^{-1})$; the leading term is real, but it would be inconsistent with the present approximation to calculate higher-order terms. The physical significance of this second k' is not clear; perhaps a parallel analysis of a cubic lattice of finite-sized spheres would shed light here. It is believed that (7.4) expresses the kr-dependence of reasonably dilute suspensions of identical small spheres.

Appendix: Scattering Coefficients.

The coefficients here are expansions of the general formulas (10) and (11), page 565 of Stratton's "Electromagnetic Theory." The expansions there suffer from inaccuracy and lack of symmetry between ϵ and μ.

$$a_1 = \frac{2ik^3r^3}{3}\left\{\frac{\mu'-\mu}{\mu'+2\mu}\right\}\left[1 + \frac{3}{10}\frac{\left[\mu'^2+(N^2-6)\mu'\mu+\mu^2\right]}{(\mu'-\mu)(\mu'+2\mu)}k^2r^2 + \frac{i\,2(\mu'-\mu)\,k^3r^3}{3(\mu'+2\mu)} + O(k^4r^4)\right]$$

where primes refer to the material of the sphere, and $N = k'/k$. Coefficient b_1 is obtained by replacing μ, μ' with ϵ, ϵ' ; the same holds for b_2 with respect to a_2:

$$a_2 = \frac{i}{15}\,\frac{\mu'-\mu}{2\mu'+3\mu}\,k^5r^5 + O(k^7r^7).$$

For perfectly conducting spheres we have with $\rho = kr$

$$a_1 = -\frac{i\rho^3}{3} + \frac{i\rho^5}{5} - \frac{\rho^6}{9} + O(\rho^7),$$

$$b_1 = \frac{2i\rho^3}{3} + \frac{i\rho^5}{5} - \frac{4\rho^6}{9} + O(\rho^7),$$

$$a_2 = -\frac{i\rho^5}{45} + O(\rho^7),$$

$$b_2 = \frac{i\rho^5}{30} + O(\rho^7).$$

For intermediate conductivities (finite skin depths) one must proceed from the general formulas.

For lossless media, the scattering coefficients are found, from the boundary condition equations, to have the structure

$$a = \frac{-L[j_n(kr)]}{L[h_n(kr)]}$$

where L is a linear differential operater with real coefficients. From $h_n = j_n + in_n$, the expansions of $j_n(x)$, $n_n(x)$ in powers of x^2, and the asymptotic forms for small x, one finds $|a|^2 = -\mathrm{Re}(a)$; The expansions above have this structure which, for a_1, b_1, is a mathematical equivalent of the theory of Rayleigh scattering. (See Section VIII, 2).

Section V. Sound Waves in Suspensions

To calculate the propagation constant, k', effective for dilatational acoustic waves in a suspension is vastly more difficult than for the electromagnetic case just treated. First, three kinds of waves (at ω) can exist in the fluid; dilatational, thermal, and shear. For only the first two kinds have propagation constants been calculated exactly[4] in terms of fundamental physical properties of the fluid. Only the coefficients for dilatational

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4) C. Truesdell, "Precise Theory of the Absorption and Dispersion of Forced Plane Infinitesimal Waves according to the Navier-Stokes Equations," J. of Rational Mechanics and Analysis, Vol. 2, No. 4, Oct. 1953.

incident and scattered waves of order 0 and 1 (monopole and dipole) are found explicitly in the literature, and these have been calculated with approximate propagation constants and impedances. For a small volume-fraction of spherical particles in the suspension, where interactions are ignorable, the discussion of Section IV shows that coefficients of order 2 (quadrupole scattering coefficients) must be kept to give an academically correct k' in the limit of the fine dispersion. Presumably, those of order 3 are required to give k' correct to order r^2. As dilatational excitation causes thermal and shear scattered waves (and vice-versa), the matrix analogous to (7.2) would be 12x12, except that there are no shear waves of order 0, and thermal waves of orders 1,2,3 may be unimportant. Finally, most suspensions have a variety of particle types and radii.

Consequently we give the general argument in outline only, then make certain simplifying approximations, and close with the specific formulas that have been obtained. We suppose that the average excitation of type i (e.g., monopole-thermal) of a particle of type τ is given by $\alpha_{\tau,i}\vec{W}_i e^{ik'z}$. This results in scattering $\vec{W}'_j$ of type j, according to the appropriate scattering coefficient $A_{\tau,ij}$. The resulting excitation of type k of a particle of type σ [to be calculated through the addition formulas expanding $\vec{W}'_j$ and eq. (1.13)] has the typical term $N_\tau \alpha_{\tau,i} A_{\tau,ij}\ I_{\sigma\tau,jk} e^{ik'z}\vec{W}_k$ where the $I_{\sigma\tau,jk}$ represents the last factor of (1.13) in which the radius of exclusion r_σ, and hence the R of (1.14) depends on both types τ and σ, and where N_γ is the number of type-γ particles per unit volume. Summing over all types of particles and excitations, we have the set of simultaneous equations

$$\alpha_{\sigma,k} - \sum_{\tau,i,j} \alpha_{\tau i} \, N_\tau \, A_{\tau i j} \, I_{\sigma\tau, jk} = 0 \tag{5.1}$$

in which the α's are the unknown amplitudes. The vanishing of the coefficient-determinant of these amplitudes determines the possible values of k'.

To decrease the order of the determinant, the most obvious assumptions are that $r_{\sigma\tau} = r_\sigma$ or r_γ, or r_e = constant; the last approximation, that a single radius of exclusion applies to all particles, produces the great simplification that the excitation W_k and $I_{\sigma\tau, jk}$ are now independent of particle type, so that the relatively simple

$$\det \left| \delta_{ik} - \sum_{\tau,j} N_\tau \, A_{\tau,ij} \, I_{jk} \right| = 0 \tag{5.2}$$

determines the various k'. Here it is seen that one may sum (on τ) over the variety of particles first, so that we may write, for (5.2)

$$\det \left| \delta_{ik} - \sum_j A_{ij} \, I_{jk} \right| = 0 \tag{5.3}$$

where A_{ij} represents the weighted average of the scattering coefficients $A_{\tau, ij}$. Thus the simple generalization of (4.2) to a variety of spherical particles is now obvious.

The A_{ij}, I_{ij} may now be regarded as matrices and the j-sum as matrix multiplication. In the acoustic case the scattering-coefficient matrix (used by us) is

$$\left\| A_{ij} \right\| = \left\| \begin{matrix} A_o & A_o'' & 0 & 0 \\ B_o & B_o'' & 0 & 0 \\ 0 & 0 & A_1 & A_1'' \\ 0 & 0 & C_1 & C_1'' \end{matrix} \right\| \tag{5.4}$$

The scattering (excitation) in row (column) 1,2,3,4, is dilatational, thermal, dilatational, and shear, respectively, and the order of the coefficient (i.e., monopole, dipole) is indicated by the subscript. (Also $A_o = \sum_\tau N_\tau A_{\tau,o}$, etc.) The interaction matrix $I = I_{jk}$ is found via Table IV and the right hand term of (1.13).

$$\left\| I \right\| \doteq \left\| \begin{matrix} \dfrac{\chi}{\sigma^2-1} & 0 & \dfrac{i\sigma\chi}{\sigma^2-1} & 0 \\ 0 & \dfrac{\chi y^3 R_0(b_e)}{y^2\sigma^2-1} & 0 & 0 \\ \dfrac{-3i\sigma\chi}{\sigma^2-1} & 0 & \dfrac{\chi(1+2\sigma^2)}{\sigma^2-1} & 0 \\ 0 & 0 & 0 & \dfrac{\chi z^3[R_0(c_e)-z^2\sigma^2R_2(c_e)]}{z^2\sigma^2-1} \end{matrix} \right\| \tag{5.5}$$

where if k, k_t, k_s, are the fluid's dilatational, thermal, and shear propagation constants, $X = 4\pi/(ik^3)$, $y = k/k_t$, $z = k/k_s$, $b_e = k_t r_e$, $c_e = k_s r_e$, and where $\sigma = k'/k$ as before. The diagonal elements are perhaps obvious; the entry in row 1, column 3 comes from the coefficient of an exciting wave $1_{eC1}(k, \epsilon)$ in the expansion of a scattered wave $1_{e01}(k,R)$. (We have assumed that $kr_e \ll 1$, dilatational wavelengths in fluids being much longer than thermal or shear wavelengths.)

We now perform the matrix multiplication, expand and solve (5.3) for σ. Having made the further simplification (approximate for very small particles) $b_e \ll 1$, $c_e \ll 1$ we obtain

$$\sigma^2 = \left\{ \frac{(1+XA_0)(1+Xy^3B_0'') - X^2y^3A_0''B_0}{1+Xy^3B_0''} \right\} \cdot \left[\frac{(1+XA_1)(1+Xz^3C_1'') - X^2z^3A_1''C_1}{(1-2XA_1)(1+Xz^3C_1'') + 2X^2z^3A_1''C_1} \right] \tag{5.6}$$

Since $k'^2/k^2 = \{m'/m\}[\rho'/\rho]$ we have a natural interpretation for the two complicated factors in (5.6) [m = compressibility, ρ = density]. When the explicit scattering coefficients (7.33)ff and (9.1) are inserted, the interpretation is completely verified if m' and m are adiabatic compressibilities and ρ' is given through the usual additive laws of composition. We have not verified the interpretation by computing a reflected wave. Neither have we considered $b_e, c_e = O(1)$, feeling that the neglect of order-2 scattering and the difficulties discussed earlier would make the result meaningless. The finite-c_e assumption would make no substantial change (a slight modification of z^3) as it appears in (5.6), since it is seen from (1.14) that the numerator of the lower-right element of (5.5) is divisible by the denominator, with the disappearance of σ^2 in this element.

Section VI. Order of Magnitude of Interactions for a Small Sphere Suspension.

Now we take the shape S of Section II to be a sphere of radius r' with center at $\vec{q}$, and will consider the r'-dependence of the average waves scattered by the sphere into the ambient fluid. A wave $\vec{W}_o$ incident on S can be expanded about $\vec{q}$ in terms of spherical waves. The resulting total excitation $\vec{W}$ incident on a typical particle P at $\vec{p}$ in S is then assumed (in the present self-consistent formulation) to vary with $\vec{p}$ in the same way that a physical wave-field depends on $\vec{p}$ when the material of S is homogeneous; i.e., the $\vec{p}$-dependence of the excitation of P is that of solutions of the scalar wave equation with some effective propagation constant k'. (We take k' as representing, in general, any of the possibly several propagation constants of the equivalent homogeneous medium.) Thus the general term in the excitation of P is of the form $f(k',\vec{p} - \vec{q})\vec{W}(k;\vec{\Delta p})$, where $\vec{\Delta p}$ is the position vector, with respect to $\vec{p}$, of a general point in the neighborhood of $\vec{p}$. $\vec{W}(k:\vec{\Delta p})$ in turn causes P to scatter a set of outgoing waves, which excite T at $\vec{t}$, the general term in the expansion of this particular excitation being of the form $Af'(k,\vec{p} - \vec{t})\vec{W}(k,\vec{\Delta t})$ where $\vec{\Delta t}$ is position vector with respect to $\vec{t}$, and A is some scattering coefficient.

Thus the excitation of T due to scattering from P has the general term $A\vec{W}(k,\vec{\Delta t})f(k',\vec{p} - \vec{q})f'(k,\vec{p} - \vec{t})$. To get the average excitation of T, in accord with the self-consistent formulation, we then multiply the above by Ndv and volume-integrate, with respect to $\vec{p}$, over S - s, where s is a small sphere of radius r_e interior to S, and centered on $\vec{t}$. The order of magnitude of this volume integral is discussed in the last section of Section I: when $|kr'| \ll 1$, $|k'r'| \ll 1$, the integral depends on r' as r'^n, $n \geq 2$, except when the excitation of T depends on $\vec{t}$ as $f_1(k,\vec{t} - \vec{q})$ or $f_1(k',\vec{t} - \vec{q})$.

We now consider the magnitude of the effect of T's 'exceptional' excitation which is of the form $k^{-3}NA\vec{W}(k,\vec{\Delta t})f_1(k \text{ or } k', \vec{t} - \vec{q})$. (This is of the order kr' or k'r' because of the f_1 factor.) The general term of the resulting scattering to the observation point $\vec{u}$ outside S is of the form $k^{-3}NAA'\vec{W'}(k,\vec{u} - \vec{t})f_1$, where A' is a scattering coefficient possibly different from A. The total wave, arising from this term, is found by again multiplying it by Ndv and integrating with respect to $\vec{t}$ over S. For this purpose we then expand $\vec{W'}(k, \vec{u} - \vec{t})$ about $\vec{q}$; in this expansion the sole term that will survive the integration is of the form $f_1(k, \vec{t} - \vec{q})\vec{W'}(k, \vec{u} - \vec{q})$.

Therefore the resulting contribution to the average scattering at $\vec{u}$ is different by a numerical factor only from $k^{-3}N^2AA'\vec{W}'(k, \vec{u} - \vec{q}) \cdot \int_0^{r'} j_1(kr)j_1(k''r)r^2dr$ ($k'' = k$ or k'); the integral is $O(k''k^4r^5)$. (6.1)

For comparison, we determine the scattering when T is assumed excited by the incident wave $\vec{W}_o(k, \vec{\Delta t} + \vec{t} - \vec{q})$ only. This wave has an expansion with terms:

$$f_{e00}(k, \vec{t} - \vec{q})\vec{W}_o(k, \vec{\Delta t}) + f_1(k, \vec{t} - \vec{q})\, \vec{W}_1(k, \vec{\Delta t}) + O(kr')^2, \tag{6.2}$$

where $\vec{W}_o(k, \vec{\Delta t})$ is formally identical with the incident $\vec{W}_o$. Replacing the $\vec{W}_o$, $\vec{W}_1$ by the appropriate scattered waves, expanding them about $\vec{q}$, multiplying by Ndv and integrating on $\vec{t}$ over S, we find that the first term of (6.2) yields $[1 + O(k^2r'^2)]$ times ($Nv' = N4\pi r'^3/3$) times the average scattering to $\vec{u}$ of particles placed individually at $\vec{q}$ and illuminated by $\vec{W}_o$, being of 'order' $Nv'A$. By the analysis leading to (6.1), the $f_1\vec{W}_1$ term of (6.2) yields scattering of order $NA(k^2r'^2)v'$ and the remaining terms of (6.2) clearly give contributions of this order or less.

Gathering results together, we find that, for r' small, and for a fixed incident wave in the ambient fluid, the sphere S of suspension scatters, on the average, a wave equal to $(4\pi Nr'^3/3)$ times the average wave scattered by the individual, isolated particles, placed one at a time at $\vec{q}$, the locus of the center of S. This statement is in error by terms of relative order $(kr')^2$ or $(k'r')^2$ where k' is any one of the propagation constants effective for the suspension. Consequences of these conclusions are explored in Section VII.

Section VII. Physical Properties of Fine Suspensions

A. General Theory

To measure the properties of some substance, one would form a simple shape of the substance, immerse the shape in a fluid of known properties, illuminate the shape with a known wave of frequency ω, and measure the field scattered by the shape. Then, in the linear relations (scattering coefficients) between the incident and scattered fields given in terms of the properties of the fluid and of the substance, one substitutes the measured scattering coefficients and solves for the properties of the substance. One theoretical procedure is to compute the average scattering coefficients for a simple shape formed of the suspension, in terms of the scattering coefficients of the suspended particles; these average scattering coefficients then play the role of those measured experimentally in the inference of unknown (effective, or average) properties of the suspension.

For computational purposes, the simplest shape, formed of the suspension, is a small sphere; this sample-sphere is immersed conceptually in a bath of the fluid constituent, so as to avoid introduction of an irrelevant set of physical properties. We are dealing with randomly distributed, small particles, so that the radius r' of the sample sphere can be quite small also ($r'/L \ll 1$). The excitation of a typical particle in the sample sphere consists of the original incident wave plus scattering from all the other particles in the sample. Since the sample is small, the number of other particles is small, and heuristically, the scattered excitation appears negligible (of order r'/L or r'^2/L^2, say) compared with that of the incident wave. (In Section VI, it was shown, through a self-consistent formulation and the use of addition formulas for spherical waves, that the scattered excitation is indeed relatively negligible for small particles. We give in the Appendix an alternative heuristic demonstration that scattered excitation can be ignored; the argument also holds for anistropic media in which spherical waves are inapplicable.)

All particles in the sample are close to its center (which is at $\vec{p}$, say), and the scattering from any particle (to the 'measuring probe' at some fixed distance from the sample) can be regarded as having originated at $\vec{p}$, with a small relative error of order (r'/L). It follows that, with small error, the total wave scattered by the sample sphere is the sum of waves scattered by the individual particles of the sample, where the wave scattered by each particle may be 'measured' by placing the particle by itself at $\vec{p}$ and illuminating it with the common incident wave. From the finite radius r' of the sample-sphere arises the small error (of maximum order r'/L, heuristically) owing to interactions and eccentricity. The magnitude of these errors is discussed in Appendix

We may express these conclusions quantitatively. Let N_t be the number, per unit volume, or particles of type t in the suspension, let A_t, B_t, ... be scattering coefficients, effective at ω, for the particle of type t relative to the fluid, and let A', B', ... be analogous scattering coefficients for the sample sphere relative to the fluid. Then

$$A' = (4\pi r'^3/3)\Sigma_t N_t A_t \left[1 + O(r'/L)\right] \tag{7.1}$$

Equation (7.1) expresses the scattering coefficients $A', B', \ldots$ of the sample sphere of radius r', in terms of corresponding scattering coefficients A_t, B_t for the particles. To deduce the bulk physical properties $(a', b', \ldots)$ of the suspension, we use the formulas for the scattering coefficients in terms of r', of $a', b', \ldots$ and of a, b, ..., the corresponding properties of the fluid. For small spheres, the dominant scattering coefficients are generally volume proportional.

$$A' = (4\pi r'^3/3)\ G_A(a,b,\ldots)F_A(a,b,\ldots;a',\ b',\ldots) \tag{7.2}$$

where the subscript A denotes that the factors G, F are those appropriate for scattering of the type to which the coefficients A', A_t pertain. (The error in (7.2) will be ignored; it is of order $(r'/L)^2$. See Appendix .) From (1) we therefore obtain

$$F_A(a,b,\ldots;a'b'\ldots) = (1/G_A)\Sigma_t N_t A_t. \tag{7.3}$$

If the particle of type t is a sphere of radius r_t and physical properties $a_t, b_t, \ldots$, then its scattering coefficient A_t (relative to the ambient fluid) is of the form (2), with subscripts t replacing the primes. The volume fraction f_t occupied by the spherical particles of type t is then $N_t(4\pi r_t^3/3)$; in this case we therefore have

$$F_A(a,b,\ldots;\ a',b',\ldots) = \Sigma_t f_t F_A(a,b,\ldots;\ a_t,b_t,\ldots) \qquad (7.4a)$$

Thus we have deduced for each volume proportional scattering coefficient A,B,..., a relation connecting the bulk properties $a', b', \ldots$ of the suspension of the fluid, and with the analogous properties and volume-fractions of the various kinds of spherical suspended particles.

If all particles are of the same material, with constants $a_1, b_1, \ldots$, then the right side is independent of the distribution of radii and we have

$$F(a,b,\ldots;\ a',b',\ldots) = f_1\, F(a,b,\ldots;\ a_1,b_1,\ldots) \qquad (7.4b)$$

Each type of volume-proportional scattering gives a relation of the form (7.3) or (7.4) among the relevant constants of the suspension, the fluid, and the spherical particles. A full set of such relations then provides a number of simultaneous equations which may be solved for the constants of the suspension. Similarly, one can solve a set of relations of the form (7.3) for the constants of the suspension in terms of the scattering coefficients of the particles, if the point-particles scatter only those waves for which

A' is volume-proportional; otherwise the sphere's volume $(4\pi r'^3/3)$ cannot be factored out of (7.2) to obtain (7.3). This fact appears to exclude the assignment of effective constants to the suspension when the particles produce higher-order scattering.

The formulas (7.2), (7.3), (7.4) are too general for fruitful discussion of such composition laws, and we therefore turn to particular cases, considering first the case of electromagnetic waves.

B. The Electromagnetic Case

In the electromagnetic case, we consider, as the fluid, any homogeneous medium described by the complex dielectric constant, ϵ, and the complex permeability, μ, both constants being those effective at angular frequency ω. Then plane waves propagate in the pure fluid according to $\exp(ikx - i\omega t)$ where the propagation constant k is given by $k^2 = \omega^2\mu\epsilon$. Let the suspension have the similar constants ϵ' and μ', and let some of the suspension be formed into a small sphere of radius r. Let B_s be the scattering coefficient, of the small sphere, connecting the scattered electric dipole wave with an incident electric dipole excitation, and let A_s be the similar coefficient for magnetic dipole waves. Let B_t, A_t be similar scattering coefficients for the particles. The two types of waves are linearly independent, and we find[5)] that

5) J. A. Stratton: "Electromagnetic Theory" (McGraw-Hill 1941), Ch.7, and Sect. 9.25. In Formula (39), p. 571, the right side of the first equation should be multiplied by 2.

$$B_s = \frac{2ik^3r^3}{3} \cdot \frac{\epsilon' - \epsilon}{\epsilon' + 2\epsilon}; \quad A_s = \frac{2ik^3r^3}{3} \cdot \frac{\mu' - \mu}{\mu' + 2\mu} \qquad (7.5)$$

Hence, from (2) we have

$$\frac{\epsilon' - \epsilon}{\epsilon' + 2\epsilon} = \frac{-2ik^{-3}\pi}{3} \Sigma_t N_t B_t; \quad \frac{\mu' - \mu}{\mu' + 2\mu} = \frac{-2ik^{-3}\pi}{3} \Sigma_t N_t A_t \qquad (7.6)$$

If, in turn the scattering particle of type t is a sphere of radius r_t, constants ϵ_t, μ_t, then we have, from (7.3),

$$\frac{\epsilon' - \epsilon}{\epsilon' + 2\epsilon} = \Sigma_t f_t \frac{\epsilon_t - \epsilon}{\epsilon_t + 2\epsilon}; \quad \frac{\mu' - \mu}{\mu' + 2\mu} = \Sigma_t f_t \frac{\mu_t - \mu}{\mu_t + 2\mu}, \qquad (7.7)$$

where $f_t = 4\pi r_t^3 N/3$ is the volume-fraction occupied by the particles of type t.

The 'composition laws' (7.7) will be recognized as the Clausius-Mossotti laws for dielectric constant and permeability, rearranged for convenience in handling differences among the properties of the suspended spherical particles. If the particles are all of a single type, these laws become rearrangements of the formulas of Lewin[6]. (The same may be said of (7.6), if allowances for differences in notations are made. Lewin arrays his particles in a cubical lattice but replaces sums over the lattice with integrals; this replacement removes the lattice spacing from the

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6) L. Lewin, "The Electrical Constants of a Material Loaded with Spherical Particles," Inst. of Elec. Engr. Journal, Part III, Vol. 94, pp 65-68, 1947.

resulting formulas, and is equivalent to an assumption that the particles are randomly arrayed.)

The laws (7.7) must be interpreted as relations among these ϵ's and μ's which are effective at angular frequency ω. To use them for information about the statically measured dielectric constant and premeability leads to incorrect inferences. To illustrate this point, let the materials have finite conductivities, σ and σ_1. The fluid's dielectric constant, effective at ω, is then (in MKS units) $\epsilon_0 + i\sigma/\omega$ where ϵ_0 is some dielectric constant necessarily measured under the time-varying conditions owing to the non-vanishing relaxation time σ/ϵ_0. (In a steady-state measurement, one necessarily measures σ.) Relation (7.7) now becomes

$$\frac{\omega(\epsilon_0' - \epsilon_0) + i(\sigma' - \sigma)}{\omega(\epsilon_0' + 2\epsilon_0) + i(\sigma' + 2\sigma)} = f_1 \frac{\omega(\epsilon_{01} - \epsilon_0) + i(\sigma_1 - \sigma)}{\omega(\epsilon_{01} + 2\epsilon_0) + i(\sigma_1 + 2\sigma)} \tag{7.8}$$

When conductivities are finite, a steady voltage gradient applied to the material results in the flow of current of amount determined by the effective conductivity, σ', obtained from (7.8) by setting $\omega = 0$. Setting $\omega = \infty$ yields an expression for ϵ'_0 in terms of ϵ_{01}. But the equality in (7.8) fails for general ω if these values of σ' and ϵ_c' are inserted. Hence we conclude that the general formulas (7.3) and (7.4) apply only among the effective constants.

With highly conductive particles, it is possible that $ka \ll 1$ for particle radius a commensurable with wavelength in the particles. Here the scattering coefficients depend on the particle radii[5], and the application of (7.3) and (7.5) leads to

$$\frac{\epsilon' - \epsilon}{\epsilon' + 2\epsilon} = \sum_a f(a) \frac{2\epsilon_1 j_1(k_1 a) - \epsilon\left[(k_1 a)\ j_1(k_1 a)\right]'}{2\epsilon_1 j_1(k_1 a) + 2\epsilon\left[(k_1 a)\ j_1(k_1 a)\right]'} \tag{7.9}$$

$$\frac{\mu' - \mu}{\mu' + 2\mu} = \sum_a f(a) \frac{2\mu_1 j_1(k_1 a) - \mu\left[(k_1 a)\ j_1(k_1 a)\right]'}{2\mu_1 j_1(k_1 a) + 2\mu\left[(k_1 a)\ j_1(k_1 a)\right]'}$$

where $f(a)$ is the volume concentration of the particles of radius a, and $\sum_a f(a) = f_1$. Formulas (7.9) are required in computing the losses arising from a skin depth $(\omega\sigma_1\mu_1)^{-\frac{1}{2}}$ comparable with particle radius. In the limit of $\sigma_1 \to \infty$, $|k_1 a| \to \infty$, we get

$$\frac{\epsilon' - \epsilon}{\epsilon' + 2\epsilon} = f_1, \quad \frac{\mu' - \mu}{\mu' + 2\mu} = -\tfrac{1}{2} f_1. \tag{7.10}$$

Heuristically, the two expressions of (7.10) differ in form because of the fact that electric lines of force end in the perfectly conducting particles, whereas magnetic lines of force must pass around the particles. [One sets $\epsilon_t = \infty$, but $\mu_t = 0$ in (7.7) to obtain (7.10)]. A similar visualization applies to the conductivity relation obtained by setting $\omega = 0$ in (7.8):

$$\frac{\sigma' - \sigma}{\sigma' + 2\sigma} = f_1 \frac{\sigma_1 - \sigma}{\sigma_1 + 2\sigma} \tag{7.11}$$

which yields

$$\frac{\sigma' - \sigma}{\sigma' + 2\sigma} = \begin{cases} f_1, & \sigma_1 = \infty \\ -\frac{1}{2} f_1, & \sigma_1 = 0 \end{cases} \tag{7.12}$$

When $\sigma_1 = \infty$, lines of current-flow end in the particles, but when $\sigma = 0$, current flows around the particles. Finally when $\sigma_1/\sigma \gg 1$, (11) may be rewritten as $\sigma' = \sigma(1 + 2f_1)(1 - f_1)$ so that $\sigma' \to 0$ when $\sigma \to 0$, a result required by the fact that, for current to flow through the suspension, it must somewhere flow through the fluid.

C. Acoustic Case

We turn now to physical properties important in the propagation of small-amplitude acoustic waves. The scattering coefficients to be used here are those found or implied in the recent paper of Epstein and Carhart[7]. In this work, the relevant physical parameters appear to be density (ρ), shear viscosity (η), bulk viscosity (μ) thermal conductivity (τ), the usual specific heats (Cp,Cv), and α, the temperature coefficient of volume expansion at constant pressure.

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7) P.S. Epstein, and R.R. Carhart, "The Absorption of Sound in Suspensions and Emulsions, I. Water Fog in Air," J. Acoust. Soc. Amer., Vol. 25, pp 553-565, 1953.

In the usual treatment, one takes $\mu = 0$ a priori, and the thermal effects are ignored or lumped phenomenologically in a compressibility, $m = \rho^{-1}(\partial\rho/\partial p)$, where p = pressure. For isothermal conditions τ can be neglected and we may deduce an $m_T = \rho^{-1}(\partial\rho/\partial p)_T = T_o\alpha^2/(\rho C_p - \rho C_v)$. ($T_o$* = mean, or 'undisturbed' temperature.) Likewise, under adiabatic conditions, τ can have no effect; here we deduce the compressibility $m_A = m_T Cv/Cp \equiv m_T/\gamma$.

Epstein and Carhart deal primarily with the propagation of dilatational waves in suspensions; for this purpose they compute two scattering coefficients, A_o and A_1. The coefficient A_1 relates dilatational dipole scattering with dilatational dipole excitation. Some of these scattering coefficients are given in Section IX. Here the particle moves relatively to the viscous fluid, so that shear waves are also scattered; the scattering coeffieicnt is C_1. One may also assume a shear-wave dipole excitation, and compute the scattering coefficients A_1" and C_1" for the resulting scattered dilatational and shear waves. All four of these coefficients are volume-proportional and arise from essentially the same set of four simultaneous boundary condition equations. One might reasonably expect that the application of (7.4) would lead to the determination of the composition laws for four of the physical parameters, or at least for ρ and η, the two parameters most relevant for the dipole scattering. The only constant of the particle material which appears explicitly in the leading, volume-proportional, term in each of the four coefficients is ρ_1, and the application of (7.4) leads to

$$\rho' = f_1\rho_1 + f\rho \tag{7.13}$$

in each of the four cases, without contradiction or further information.

From the four simultaneous equations expressing the boundary conditions for monopole (spherically symmetric) waves, one can compute the volume-propertional coefficients A_o, B_o, A_o'', B_o''. The first two pertain respectively to the radial dilatational and thermal waves scattered when a purely radial dilatational wave is incident, and the last two have similar meanings for radially incident thermal waves. By systematically applying the approximations of the Epstein-Carhart paper, carrying only terms of lowest order in the particles's radius, one obtains the four monopole coefficients in reasonably compact form. Applying (7.4) first to B_o'', one obtains

$$\rho' Cp' = f\rho Cp + f_1\rho_1 Cp_1. \tag{7.14}$$

This result simplified the application of (7.4) to A_o and A_o''; both coefficients yield

$$\alpha' = f\alpha + f_1\alpha_1. \tag{7.15}$$

After using both (7.14) and (7.15) to simplify the application of (7.4) to A_o one obtains

$$m_T' = fm_T + f_1 m_{T1} \tag{7.16}$$

We gain further information by calculating the scattering coefficients appropriate to incident thermal waves of a dipole character. Here the coefficient for scattered thermal dipole waves yields the composition law for thermal conductivity.

$$\frac{\tau' - \tau}{\tau' + 2\tau} = f_1 \frac{\tau_1 \quad \tau}{\tau_1 + 2\tau} \tag{7.17}$$

The coefficients for scattered shear and dilatational waves yield no new laws, being combinations of (7.17) and (7.13).

The angular frequency appears explicitly in the dielectric law (7.8); to obtain analogous acoustic expressions would require carrying the relevent propagation constants without approximation. Acoustic analogues of the 'skin-depth' formulas (7.9) may also be obtained at the expense of algebra; this case, however, is not so interesting as the case where the suspended particles are relatively rigid and of sizes comparable with thermal and shear wavelengths in the field. [See Section VII, E.]

The truth and significance of the four "additive" composition laws (7.13) through (7.16) are obvious. The association of the density composition law (7.13) with dipole scattering coefficients seems to arise from the fact that except in the 'thermal' case the dipole scattering coefficients are volume-proportional only if $|\rho_1 - \rho| > 0$. For the force tending to move the particle relative to the fluid in the oscillatory acceleration-field is of the nature of a buoyancy proportional to the mass of the particle minus the mass of the displaced fluid, a force vanishing if $\rho_1 = \rho$. The composition law

(7.14) essentially gives the heat necessary to raise a unit mass of suspension a unit temperature at constant pressure; the law seems an appropriate deduction from the coefficient connecting between thermal incident and scattered waves. Analogously, the α's, connected through the law (7.15), are physical constants relating a thermal cause and a mechanical effect, in keeping with the derivation of (7.15) from A_o'' and B_o. Both of these coefficients relate thermal with dilatational waves, and the fact that both lead to the same composition law suggests some underlying reciprocity principle. Only mechanical effects are associated with A_o and the resulting law (7.16). The reason this law concerns m_T rather than m_A seems to be that the particles have been assumed small enough so that they are in local thermal equilibrium with the fluid.

Finally, since steady-state electrical and thermal conduction are entirely similar, the formal identity of the Clausius-Mossotti laws (7.17) and (7.11) is not surprising. It is interesting to note that (7.17) is derived from the scattering coefficient connecting thermal <u>dilatational</u> incident and scattered dipole waves, whereas (7.11) was derived indirectly from a scattering coefficient for electric, transverse, dipole waves.

For acoustic waves in homogeneous substances, the dilatational propagation constant k is given by $k = \omega (m\rho)^{\frac{1}{2}}$ to an accuracy improving with $\omega \to 0$. If one ignores thermal waves (by setting $\tau = 0$), the dipole scattering coefficients A_1, C_1, A_1'', C_1'' remain practically unaltered, while the sole monopole coefficient is Ao. In the small-radius limit, this A_o would lead to the composition law $m'_A = fm_A + f_1 m_{A1}$. The law (7.16) is the pertinent one. The thermal equilibrium is reached by the flow of heat between particles and fluid. On thermodynamic grounds, this flow of heat should lead to an energy loss from the wave propagating through the suspension. But the implicit assumption has been that the suspension is passing through a series of reversible equilibriums, so that no heat losses are entailed.

For a strictly rotational incident velocity field, the particle's moment of inertia, rather than its mass, determines the magnitude of the scattering, so that the scattering coefficient is of order r^5. For a shear velocity field, the scattering coefficient is volume-proportional and leads to a composition law for viscosity. This law is derived in Sub-section VII, D.

D. Composition Laws for Elastic Constants and Viscosities

For the spherical scatterers, we have so far encountered only two formally different composition laws; Those for ϵ, μ, σ, and τ, of which the laws (7.7) may be taken as typical examples, and those for ρ and m, of which (7.13) may be taken as typical. The first law (7.7) is simply the Clausius-Mossotti law, rearranged algebraically in a manner convient for taking into account suspended particles of differing dielectric constants. The additive law (7.13) states simply that the mass of a unit volume of suspensions is the sum of the masses of the component materials in the unit volume, a law probably well known to Archimedes. If we visualize non-oscillatory measurements ($\omega \simeq 0$) of the various properties, the Clausius-Mossotti composition law appears when in the visualized measurements, the particles affect a vector field of force or steady flow, whereas the additive law appears when they affect some scalar property, such as a pressure-volume relation in the case of m. (It might be objected that a density measurement requires an acceleration-field, but the dense particles do not disturb the field in any way, so that their effect can be construed as scalar)

When the viscosity or shear modulus of the suspension is being measured, the particles affect tensor relations and the corresponding composition laws are not expected to follow the additive or the Clausius-Mossotti form. The scattering coefficients required in finding these laws by the present method do not seem to be given explicity in the literature, and will now be derived.

We now compute the volume-proportional scattering coefficients pertaining to scattering by a solid sphere embedded in a second solid, thermal effects being ignored. From these coefficients, we deduce the composition laws for density, ρ, and for the Lamé constants, λ and μ. From the law for the shear modulus, μ, we deduce the composition law for fluid viscosity, η; the particular reason for this roundabout derivation will be given subsequently.

For axially symmetric motions, the dilatational and shear-wave scattering coefficients (A_n and C_n, respectively) are to be computed through four simultaneous boundary-condition equations[8]:

$$aj_n'(a) + A_n a h_n'(a) - n(n+1)C_n h_n(c) = A_n' a' j_n'(a') - n(n+1)C_n' j_n(c') \tag{7.18}$$

$$j_n(a) = A_n h_n(a) - C_n[h_n(c) + c h_n'(c)] = A_n' j_n(a') - C_n'[j_n(c') + c' j_n'(c')] \tag{7.19}$$

$$\mu\{a j_n'(a) - j_n(a) + A_n[a h_n'(a) - h_n(a)] - (1/2)C_n[c^2 h_n''(c) + (n^2+n-2)h_n(c)]\}$$
$$= \mu'\{A_n'[a' j_n'(a') - j_n(a')] - (1/2)C_n'[c'^2 j_n''(c') + (n^2+n-2)j_n(c')]\} \tag{7.20}$$

$$a^2[\lambda j_n(a) - 2\mu j_n''(a)] + a^2 A_n[\lambda h_n(a) - 2\mu h_n''(a)] + \mu 2n(n+1)C_n[c h_n'(c) - h_n(c)]$$
$$= a'^2 A_n'[\lambda' j_n(a') - 2\mu' j_n''(a')] + \mu' 2n(n+1)C_n'[c' j_n'(c') - j_n(c')]. \tag{7.21}$$

The first two express continuity of (radial and tangential) velocity at the surface of a sphere of small radius r and of substance described by ρ', λ', μ', and the last two express the balance of (tangential and radial) forces across the boundary. Here the outgoing and spherical Bessel functions $h_n(x) \equiv h_n^{(1)}(x)$ and $j_n(x)$ both satisfy

$$j_n = x[j_{n-1} + j_{n+1}]/(2n+1)$$
$$x j_n' = x[n j_{n-1} - (n+1)j_{n+1}]/(2n+1) \tag{7.22}$$

$$x^2 j_n'' + 2x j_n' + x^2 j_n - n(n+1)j_n = 0 \tag{7.23}$$

8) P. Epstein, "On the Absorptions of Sound Waves in Suspensions and Emulsions," (Eq. 25), Theodore von Karman Anniversary Volume, 1941.

Also, for $|x| \ll 1$,

$$j_n(x) \simeq \frac{x^n n!\, 2^n}{(2n+1)!} \quad , \quad h_n(x) \simeq \frac{-i}{x} \cdot \frac{(2n)!}{n!\, 2^n x^n} \qquad (7.24)$$

Finally $a^2 = \omega^2 r^2 \rho/(\lambda + 2\mu)$, $c^2 = \omega^2 r^2 \rho/\mu$, (7.25)

with similar definitions for a', c'.

We are interested here only in the limiting case $|a| \ll 1, |c| \ll 1$. For the spherically symmetric case n = 0, only dilatational waves are involved, and the 'tangential' equations (7.19) and (7.20) do not apply; setting n = 0 in (7.18) and (7.21) and applying the identities (7.22), (7.23) we obtain

$$j_0(a) + j_2(a) + A_0[h_0(a) + h_2(a)] = A_0'(a'^2/a^2)\,[j_0(a') + j_2(a')]$$

$$3c^2 j_0(a) - 4a^2[j_0(a) + j_2(a)] + A_0\{3c^2 h_0(a) - 4a^2[h_0(a) + h_2(a)]\}$$
$$= A_0'(\mu'/\mu)\,\{3c'^2 j_0(a') - 4a'^2[j_0(a') + j_2(a')]\}$$

respectively. Applying the expansions (7.24), we have

$$1 - 3ia^{-3} A_0 \doteq (a'^2/a^2)\, A_0', \qquad (7.26)$$

$$(3c^2 - 4a^2) + 4a^2\, 3ia^{-3} A_0 = (\mu'/\mu)\,[3c'^2 - 4a'^2]\, A_0' \qquad (7.27)$$

Solving for A_0, and applying (7.25), we obtain

$$3iA_0 a^{-3} = \frac{(3\lambda' + 2\mu') - (3\lambda + 2\mu)}{3\lambda' + 2\mu' + 4\mu} \qquad (7.28)$$

For $n \geq 1$, we find that, with the identities (7.22), the combinations (18) + (n + 1)(19) and (18) - n(19) yield, respectively, the 'new' velocity equations

$$aj_{n-1}(a) + A_n ah_{n-1}(a) - (n+1)C_n ch_{n-1}(c) = A_n'a'j_{n-1}(a') -$$

$$-(n+1)C_n'c'j_{n-1}(c') \tag{7.29}$$

$$aj_{n+1}(a) + A_n ah_{n+1}(a) + nC_n ch_{n+1}(c) = A_n'a'j_{n+1}(a') + nC_n'c'j_{n+1}(c'). \tag{7.30}$$

In the force equations, we first eliminate the second derivatives through (7.23), then in (7.21), use (7.25) to eliminate the explicit appearance of λ. Then (7.20) - 2(n+1)(7.21) and (7.20) +n2(7.21) result in the simplified force equations

$$\mu\left\{c^2 j_n(a) - 2(n-1)aj_{n-1}(a) + A_n\left[c^2 h_n(a) - 2(n-1)ah_{n-1}(a)\right] - C_n(n+1)\left[c^2 h_n(c) - (n-1)ch_{n-1}(c)\right]\right\} =$$

$$\mu'\left\{A_n'\left[c^2 j_n(a') - 2(n-1)a'j_{n-1}(a')\right] - C_n'(n+1)\left[c'^2 j_n(c') - 2(n-1)c'j_{n-1}(c')\right]\right\} \tag{7.31}$$

$$\mu\left\{c^2 j_n(a) - 2(n+2)aj_{n+1}(a) + A_n\left[c^2 h_n(a) - 2(n+2)ah_{n+1}(a)\right] + nC_n\left[c^2 h_n(c) - 2(n+2)ch_{n+1}(c)\right]\right\} =$$

$$\mu'\left\{A_n'\left[c^2 j_n(a') - 2(n+2)a'j_{n+1}(a')\right] + nC_n'\left[c^2 j_n(c) - 2(n+2)c'j_{n+1}(c')\right]\right\} \tag{7.32}$$

For a general solution, it might be convenient to subtract $\left[(2n+1)c^2 - (2n+2)\right]$(7.30) from (7.32); in the present small-radius case, after this step and the substitution of the expansions (7.24), one obtains a consistent set of four independent equations in which A,C,A',C' are of order a^3, c^3, 1, 1 respectively, for n = 1,2. This last equation is not required, however, since (to the present order of approximation) the right-hand side of (7.30) vanishes, and those of (7.29) and (7.31) differ by a constant factor. To obtain A,C we then need only the first three equations, with (say) the A' terms eliminated.

For n = 1, in (7.31) we then factor out μc^2, then apply (7.22) and substract (7.26), and then substitute $\mu' c'^2/(\mu c^2) = \rho'/\rho$. With the expansions (7.24), equations (7.29), (7.30), and (7.31) become

$$0 - 3ia^{-2}A_1 - 3ic^{-2}C_1 = 0$$

$$a - iA_1 + 2iC_1 = -2c'C_1' \text{ (or } a = -2c'C_1')$$

$$0 - 3ia^{-2}A_1 + 6ic^{-2}C_1 = [\rho'/\rho - 1]\,(-2c')C_1'.$$

Solving, we have $A_1 \simeq ia^3(\rho' - \rho)/(9\rho)$, $C_1 = -iac^2(\rho' - \rho)/(9\rho)$. (7.33)

For completeness, we compute the scattering coefficients A_1'' and C_1'' for the case that the incident excitation is the axially symmetric shear wave of order n = 1. As may be obvious, the last set of simultaneous equations is changed only by the appearance of -2c in place of $\underline{a}$ as the leading term of the second equation, so that $A_1'' = -2cA_1/a$, $C_1'' = -2cC_1/a$.

For n = 2, we again omit (7.32) and obtain from (7.29), (7.30), (7.31) respectively

$$a^2/3 - iA_2a^{-1} + 3iC_2c^{-1} = -C_2'c'^2 \ ; \ 0 + A_2a^{-3} + 2C_2c^{-3} = 0;$$

$$- 2a^2/3 - iA_2[3c^2 - 2a^2]a^{-3} + 3iC_2c^{-1} = 2C'_2(\mu'/\mu)c'^2$$

Including the case of shear excitation (again with doubly primed coefficients) we have $A_2'' = (-3c^2/a^2)A_2$, $C_2'' = (-3c^2/a^2)C_2$, $C_2 = -(1/2)A_2$,

$$A_2 = \frac{-ia^3 4\mu(\mu' - \mu)/3}{3(\lambda + 2\mu)\cdot(2\mu' + 3\mu) + 4(\mu' - \mu)} \qquad (7.34)$$

Now we apply the theorem (7.4) (using for simplicity, particles of constants ρ_1, λ_1, μ_1, with $\Sigma_t f_t = f_1$) to obtain the composition laws

$$\frac{(3\lambda' + 2\mu') - (3\lambda + 2\mu)}{(3\lambda' + 2\mu') + 4\mu} = f_1 \frac{(3\lambda_1 + 2\mu_1) - (3\lambda + 2\mu)}{(3\lambda_1 + 2\mu_1) + 4\mu} \qquad (7.35)$$

$$\rho' - \rho = f_1(\rho_1 - \rho) \qquad (7.36)$$

$$\frac{\mu' - \mu}{3(\lambda+2\mu)(2\mu'+3\mu) + 4\mu(\mu'-\mu)} = f_1 \frac{\mu_1 - \mu}{3(\lambda+2\mu)(2\mu_1+3\mu) + 4\mu(\mu_1-\mu)} \qquad (7.37)$$

from coefficients of order n = 0, 1, 2, respectively. The law (7.36) is the familiar density law (7.13).

The conversion of these laws into composition laws for emulsions of viscous fluids (in which thermal effects are to be neglected) proceeds via the scheme $3/(3\lambda + 2\mu) \rightarrow m$ (m = compressibility), $\mu \rightarrow -i\omega\eta$ (η = viscosity), $\rho \rightarrow \rho$ (ρ = density). Here $\omega\eta \ll 1/m$ for practical substances and frequencies (i.e. $a^2 \ll c^2$) and we may simplify the composition laws through ignoring terms of order $a^2/c^2 \sim m\omega\eta$:

$$m' - m = f_1(m_1 - m) \tag{7.38}$$

$$\frac{\eta' - \eta}{2\eta' + 3\eta} = f_1 \frac{\eta_1 - \eta}{2\eta_1 + 3\eta} \tag{7.39}$$

The law (7.38) is identical with (7.16); (7.39) is the sought-for law of combination of viscosities.

The viscosity law (39) arose from the fact that the 'quadrupole' scattering coefficients A_2, C_2, A_2'', C_2'' are volume proportional. In the simultaneous boundary-condition equations for the fluid sphere, Epstein's[9] analogue of the present (7.21) differs from the corresponding equation of Epstein and Carhart[10] by terms of relative order a^2/c^2. In neither case does the analogue of the italicized step leading to (7.31) and (7.32) appear possible;

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9) See footnote 8 on p. 59

10) See footnote 7 on p. 52

the consequence is that A_2, etc., are not volume proportional unless further judicious use of $a^2/c^2 \simeq 0$ is made. Hence the present roundabout derivation of (7.39) via the 'elastic' laws.

Properties of composite media containing completely rigid suspended spheres are to be found by setting $\lambda_1 = \infty$, $\mu_1 = \infty$ ($m_1 = 0$) in (7.35) through (7.39). For bulk properties when the suspended particles are completely soft, one sets $\lambda_1 = 0$, $\mu_1 = 0$ ($m_1 = \infty$) on the right sides of these equations. In particular, the viscosity η' of a suspension of rigid spheres may now be written as

$$\eta' = \eta \frac{1 + (3/2)f_1}{1 - f_1} \simeq \eta\left[1 + (5/2)f_1 + (5/2)f_1^{\,2}\right] \qquad (7.40)$$

Comparison should be made between this viscosity law and the Einstein-Hughes[11)] law

$$\eta' = \eta \frac{1 + (1/2)f_1}{1 - 2f_1} \simeq \eta\left[1 + (5/2)f_1 + 5f_1^{\,2}\right] \qquad (7.41)$$

The assumptions of the present derivation include that of negligible physical contact between particles, a situation approximated in viscous flows only for small volume-fractions of particles. Thus the difference between the composition laws is probably academic.

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11) A. J. Hughes, "The Einstein Relation between Relative Viscosity and Volume Concentration of Suspensions of Spheres," Nature, Vol. 173, pp 1089-1090, June 5, 1954.

The same is true of a preference for the denominator $1 - f_1$ of (7.40) to the $1 - 2f_1$ of the Einstein-Hughes formula; the latter denominator yields perfect rigidity as 50% concentration, and negative viscosities for higher concentrations, which are achievable without particle contact only on paper. The measurements cited by Hughes compare about equally well with both (7.40) and (7.41):

$100f_1$	Observed	(7.40)	(7.41)
0.88	1.022	1.022	1.022
1.76	1.042	1.045	1.045
3.5	1.087	1.091	1.094
7.0	1.195	1.188	1.204
14.0	1.512	1.407	1.486

The writer believes that solid suspensions would be more convenient for experimental verification of the analogous formula (7.37); at least, the problem of colliding particles would be avoided. A prediction for μ' should also be derivable along lines closely paralleling the Einstein-Hughes derivation of η'.

Mackenzie[12)] has computed the elastic constants for a solid containing spherical holes. We set λ_1, μ_1 to zero in (7.35) and (7.37). The former yields Mackenzie's result (7.7) if his holes had been of uniform radius, while the latter agrees with eq. (7.19) when our f_1 is small and $\mu' \simeq \mu$.

12) J. K. Mackenzie, Proc. Phys. Soc. Lond., Vol. 63B, p. 1, 1950

To obtain the viscosity of a suspension of fluid spheres which retain spherical shape owing to surface tension, we equate each side of (7.18) to zero, (19) and (20), but ignore the radial force equation (7.21). After some care with the approximations $a \ll c \ll 1$, $a' \ll c' \ll 1$, we obtain an A_2 which, via (34), (4) and $\lambda \gg \mu \to -i\omega\eta$, yields

$$\frac{\eta' - \eta}{2\eta' + 3\eta} = f_1 \frac{5\eta_1 + 2\eta}{10(\eta_1 + \eta)} . \qquad (7.42)$$

For small f, where $\eta' \simeq \eta$, this becomes the result (Mackenzie's eq. (26)) derived by G. I. Taylor.[13)]

When a fluid has a finite coefficient of bulk viscosity, ζ, the value of $q(\omega) \equiv 1/m(\omega)$ is effectively complex: for small vibrations according to $e^{-i\omega t}$, $q(\omega) = q(0) - i\omega\zeta$. We have taken $\zeta = 0$ previously, so that q, q_1 are independent of ω. But an imaginary term may be deduced for q' when $\mu \to -i\omega\eta$ is retained in (7.35), which now becomes $(q' - q)/(q' - 4i\omega\eta/3) = f_1(q_1 - q)/(q_1 - 4i\omega\eta/3)$. From this we obtain

$$\mathrm{Im}(q') = -i\omega\zeta' = \frac{-i\omega\{4\eta f_1(1-y^2)\}}{3f\,(1 + f_1 y/f)^2}\left[1 + O\left(\left[\frac{m\, m_1 \omega\eta}{m'}\right]^2\right)\right], \qquad (7.43)$$

where $y = q/q_1 = m_1/m$, $f + f_1 = 1$ and where (7.38) gives m'.

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13) G. I. Taylor, Proc. Roy. Soc. A. 138, 41, 1932.

The formula becomes identical with the result of Taylor and Davies[14)], when $y \gg 1$, $f_1 \ll 1$, the error term is small, and when m_1 is construed to include surface tension effects. Our $\rho' \to 0$ when $y \to 1$, in consonance with the idea that ρ' depends on η and must arise from shear flow in the ambient fluid surrounding the particle: the flow should be strictly dilatational when $m_1 = m$.

Even when computed exactly, our $\rho' \to \infty$ when $m = 0$ and $f_1 \to 0$, as does Ta ylor's. It therefore seems reasonable to explore the attenuating effect on waves propagating through the suspension. To get the propagation constant $k' = k(\omega) = \omega(m(\omega)\,\rho')^{\frac{1}{2}}$, we therefore compute $m(\omega) = 1/q(\omega)$ exactly, obtaining

$$m(\omega) = \frac{m' + iWff_1(m_1 - m)^2 + W^2 m\, m_1(m_1 + m - m')}{1 + W^2(m_1 + m - m')^2} \tag{7.44}$$

where m' is as in (38), $W = 4\omega\eta/3$. Hence

$$k(\omega) \simeq \omega\sqrt{m'\rho'}\left[1 + iWff_1(m_1 - m)^2/(2m') + O(W^2)\right] \tag{7.45}$$

so that the attenuation per centimenter $(2\mathrm{Im}(k))$ is proportional to $\omega^2 f_1$ for small f_1, and $m > 0$. When $m = 0$, the attenuation per cm. goes as $\omega^2 f_1^{3/2}$. Intuitively, for small f_1, we expect $\mathrm{Im}(k)$ to be

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14) Proc. Roy. Soc. 226A, pp 34-39, 1954

proportional to the number of scatterers per cubic centimeter, i.e., to f_1; thus the case $m = 0$ is either unsatisfactory, or it is meaningless to talk of sound waves in an incompressible fluid.

For suspensions of very small spheres in fluids of finite compressibility, we have obtained a reasonable, ω-dependent acoustic attenuation which arises from the fluid's viscosity and vanishes when all compressibilities are the same. It should be pointed out that equally important thermal effects have been ignored.

E. Acoustic Scattering by Particles of Finite Size

With regard to obtaining an ω-dependence in some of the acoustic composition laws, one original purpose of this paper was to obtain a viscous loss term in the effective density of a suspension of rigid spheres of radius a. It is well known that the viscous loss depends on the parameter $\beta a = a(\omega\rho/2\eta)^{(1/2)}$ so that (unless one included the small effect probably obtained when all propagation constants are carried without approximation) scattering coefficients showing viscous losses cannot be strictly volume-proportional, and the present method is inapplicable. If however, we treat a as an effective radius of a point particle, the coefficient A_1 leads to

$$\rho' = (f\rho + f_1\rho_1) + i\omega(\rho_1 - \rho)^2 \cdot (9\eta)^{-1} \cdot f \sum_a a^2 f(a) + O(\beta a)^3 \quad (7.46)$$

where $f(a) = \sum_a N_a 4\pi a^3/3$, and $f_1 = \sum_a f(a)$, $f = 1 - f_1$ (Similar derivations of ρ' from A_1'', C_1, C_1'' differ from (7.46) and mutually by terms of order βa, unless further assumptions are made regarding $\underline{a}$).

The writer has a number of reservations concerning the applicability of the previously derived[15,16] formula (7.46). The loss term (on the right) is significant only when $\beta a = O(1)$, in which case the interactions among the (finite) particles of the suspension are not negligible, as assumed in the present derivation. A proper self-consistent calculation of the interaction requires the assumption of some radius of exclusion $a_e \simeq 2a$, a minimum distance between interacting particles. No such radius of exclusion appears in this or previous derivations. (In deriving (7.46) the assumption of point-particles also entailed the assumption $\beta a_e \ll 1$.)

Finally, it is clear from the Epstein-Carhart paper[17] that thermal waves and viscous shear waves (in fluids) are of commensurable wavelength, so that thermal losses should be commensurable with viscous losses; thus the acoustic attenuation in suspensions is not predictable through a 'lossy' ρ' alone.

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15. R. J. Urick and W. S. Ament, J. Acoust. Soc. Am., <u>21</u>, p. 115 1949

16. W. S. Ament, J. Acoust. Soc. Am., <u>25</u>, pp 638-641, (1953)

17. See footnote No. 7

APPENDIX

We assume that an average unit volume of the suspension consists of the fluid plus total of N point particles, defined through scattering coefficients. We then select at random a number of small spherical samples of the suspension, insert the samples one at a time in a bath of the fluid, measure a given scattering coefficient of a similar sphere of some 'equivalent homogeneous material' having physical properties equivalent to those of the suspension. With randomly located point particles, the Poisson statistics apply, so that the probability P(n) that n particles will be found in the volume $v' = 4\pi r'^3/3$ of the sample-spheres is given by $P(n) = (Nv')^n e^{-Nv'}/n!$. Then the average scattering S is determined through $S = \sum_n S(n)P(n)$, where S(n) is the scattering, on the average of those sample-spheres containing precisely n particles. Since the samples containing no particles are continuous with the surrounding fluid, they produce no scattering: $S(0) = 0$. With r' at our disposal, we take it additionally small so that $Nv' \ll 1$; we then have

$$S = (Nv')\, S(1) \tag{7A.1}$$

(If r' is now taken so small that the scattering from any particle may be considered as arising from the sample's center, to relative order (r'/L), eq. (7A.1) is recognized as substantially identical with eq. (7.1) of the text.) The error in (1) is seen to be of order $(Nv')^2(2)$. The relative error thus hinges on the magnitude of $S(2)/S(1)$; i.e., on the average magnitude of the net scattering by two interacting particles separated by a distance less than $2r'$, compared with the scattering by one of the particles. With small interaction $S(2)/S(1) \simeq 2$; with interaction producing some sort of resonance in the two-particle scattering, one might have $S(2)/S(1) = O(1/r')$; but $S(2)/S(1) = O(1/r'^2)$ still leads to a negligible error, since $v' = O(r'^3)$. With $P(n) = O(r'^{3n})$ the $n > 2$ terms are seen to contribute to the average S even more negligibly.

For finite particles, we can choose some fixed point, say the center of gravity, of the particle, and say that the particle is or is not in the sample sphere according as the fixed point is or is not in the sphere. Then the probability $P(n)$ that exactly n finite-sized particles are in the sample is obviously smaller than $P(n)$ for point particles, so that the foregoing argument again leads to $S \simeq S(1)P(1) \simeq (Nv')S(1)$.

The foregoing argument is heuristic, but is simple and to the point. We have shown in Section VI that if a self-consistent theory applies to propagation in the suspension, the error in (1) is of relative order $(k''r')^2$, where k'' is the propagation constant of greatest magnitude effective for the fluid or for the suspension. This error estimate includes both interactions within the sample-sphere and the possible eccentricities of particles there.

It follows that any scattering coefficient A for the sample sphere has a power series development in powers of r' starting out like

$$A = S_o r'^3 (1 + cr'^2) \tag{7A.2}$$

where c is a constant and where S_o is a constant for a 'dominant' volume-proportional scattering coefficients, but should be taken as zero for the higher-order scattering because the physical particles must be assumed small and therefore incapable of producing 'higher order' scattering. It can be shown that (7A.2) is also the form of the dominant scattering coefficients of a <u>homogeneous</u> sphere (again of radius r' and immersed in the fluid), so that the error estimate in (7A.2) is of the order of magnitude required for an equivalence of the suspension's properties with those of an equivalent homogeneous material.

(Suppose one chooses sample-volumes having fixed ellipsoidal shape and orientation. The present argument would suggest that small ellipsoids made of a homogeneous substance have volume-proportional scattering coefficients which are independent of eccentricity and orientation. But the scattering of a small dielectric ellipsoid depends on its shape and orientation. Hence the present argument is suspect, as is the Section's main argument which would appear to lead also to shape-independent scattering. This matter is treated more constructively in the 'Notes Added in Proof'.)

Section VIII - Approximations Valid for Dilute Suspensions

A. Dilute Suspensions of Small Particles.

Following Rayleigh,[18] we assume the suspension to fill a slab between the $z = 0$ and the $z = d$ planes, with $kd << 1$, and with the remainder of space filled with the fluid. A plane wave transmitted normally through the slab emerges with a relative amplitude equal to unity (as if there were no particles in the slab) plus a plane wave formed from the scattering of the particles. If the particles of the slab interact negligibly (as is the case in the dilute limit), the excitation of each particle is that from the expansion of the incident wave Ee^{ikz} about the particle. From the scattering coefficients and Table III we then compute the total plane wave Ete^{ikz} in $z>d$. Assuming no reflections at the slab's boundaries and a wave inside traveling as $e^{ik'z}$, we set $ETe^{ikd} = Ee^{ik'd}$, expand in powers of d, and obtain from the coefficients of d^1

$$\sigma = k'/k = 1 - iN\pi k^{-3}\sum_{n=1} (2n + 1)(a_n + b_n) \qquad (8.1)$$

$$\sigma = k'/k = 1 - 2\pi iNk^{-3}\sum_{n=0} (2n + 1)A_n \qquad (8.2)$$

for the electromagnetic and acoustic cases, respectively. Here N = number of (identical) particles per unit volume of suspension, the scattering coefficients of (8.1) are given in Stratton[1], those

18) Rayleigh, Scientific papers, Cambridge Univ. press, V. 4, p. 397-405 (1892-1920); also in Phil. Mag. 47, 375-384, 1899.

those of (8.2) in Epstein and Carhart[7], and one can generalize to a variety of particle types by replacing NA_i by $\Sigma_\tau N_\tau A_{i,\tau}$, etc. The writer noticed [9, 19] that the superimposed scattering of particles of the slab should also form in $z < 0$ a reflected plane wave traveling as ERe^{-ikz}, where R (like T) may be calculated through Table III. He then chose the effective physical properties of the suspension as those of an equally thick slab of homogeneous material which would have produced the same R and T. By expanding in powers of d and equating coefficients of d^1, he obtained

$$\frac{\mu'}{\mu}, \frac{\epsilon'}{\epsilon} = 1 - i2\pi N k^{-3}\left\{\Sigma_{\text{odd } n, \text{ even } n}\,(2n+1)a_n + \right. \\ \left. + \Sigma_{\text{even } n, \text{ odd } n}\,(2n+1)b_n\right\}, \tag{8.3}$$

$$\frac{m'}{m}, \frac{\rho'}{\rho} = 1 - i4\pi N k^{-3}\,\Sigma_{\text{even } n, \text{ odd } n}\,(2n+1)A_n \tag{8.4}$$

for the electromagnetic ($k^2 = \omega^2\mu\epsilon$) and dilatational acoustic ($k^2 = \omega^2 m\rho$) cases, respectively.

Darwin[20] and Twersky[21] apply a self-consistent calculation to 'suspensions' filling $z > 0$. In place of the present 'sphere of

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19. NRL Report No. R-3238
20. C. G. Darwin, Trans. Comb. Phil Soc. 23, 137-167, 1924
21. Verbal Communication

exclusion' they assume (essentially) that a particle at (x,y,z') receives no excitation from any other particle in $z' - d' < z < z' + d'$ $(kd' \ll 1)$. Thus the particle's total excitation is deducible from the expansions of forward and backward-traveling plane waves. For normal incidence, the most general results of these assumptions are identical with those derivable by the "method" just described. In the electromagnetic case, the results (8.3) do not express the Clausius-Mossotti laws, but give the leading terms of these laws when they are developed in powers of N.

The acoustic case is more interesting. We note that the theory of Section V gave k'/k as the product of two factors which were verifiably m'_A/m_A and ρ'/ρ. The adiabatic compressibility of the suspension m'_A was computed as if fluid and particles in a small region of the suspension were at the same temperature, a condition valid under our assumption that the particles were extremely small. This result was obtained by substituting the scattering coefficients of (9.1) into (5.6). When the same coefficients [i.e., A_0 of (9.1)] are used in the low-concentration formula (8.14), we find that the m' cannot be interpreted either as an adiabatic or as an isothermal compressibility for the suspension, owing to the complicated dependence of A_0 on the physical constants. If we had included the volume-proportional A_2 in (8.4) the m' would not have been improved as A_2 involves the viscosities explicitly. On the other hand, terms suggestive of the 'bulk viscosity' found in (7.43) might have appeared.

A better deduction of m' by the 'thin slab' method leading to would have been to compute the R and T of the slab for a plane thermal incident wave as well as for the dilatational wave. Then the m' would be computed as one of the properties of an equally thick slab of homogeneous material producing the same thermal as well as dilatational R's and T's. Shear waves, which are transverse, would not be encountered for normal incidence. (This seems to be the reason that ρ' of (8.4) is correct.) We have not performed this thermal wave calculation and therefore cannot say that the 'thin slab' method gives a wrong m'. On the other hand, the acoustic k' of (8.2), calculated by Rayleigh's method, is also incorrect when regarded as an expansion of the 'correct' k' in powers of N. The error again arises in the compressibility and is due to the structure of A_0. But this k' has generally been used in former theories of propagation in suspensions. The writer feels that theories of acoustic waves in suspensions in which all coefficients of order 0, 1, 2 are not taken account of are oversimplified, although in many cases practical results may agree closely with experiment.

B. Extinction and the Meaning of k'.

The 'average' plane wave in the present suspension travels according to $e^{ik'z}$ where k' is a complex-valued effective propagation constant. To determine k' by the simple theory of part A, one must measure a slab-thickness d and a plane-wave transmission coefficient, T. For the necessarily interferometric measurement of the complex number T, we imagine an interferometer (immersed in the fluid) with which one measures the transmission coefficient T' of a sample-slab (of thickness d) of some homogeneous substance of properties approximating those of the fluid. The measurement is also possible when stray radiation from random, fluctuating sources is added to the transmitted field. Forming a set of sample slabs of suspension, we similarly measure a complex T_s for each sample, and obtain T as the average of T_s over the set. Alternatively, we visualize the particles in the sample slab as swirling randomly about in the sample slab and measure T for the time-varying transmitted field just as T' was measured in the presence of stray radiation. This power counts as so much receiver noise. (Similar conceptual interferometric measurements of average reflection coefficients R and scattering coefficients A_i are obvious and will not be described.)

Thus $e^{ik'z}$ describes the propagation of an average coherent (interferometrically measurable) plane wave in the suspension. In the electromagnetic case, the power carried by this average coherent waves is attenuated according to $\exp(-2z\mathrm{Im}(k'))$; from (8.1) we have (for k real)

$$NQ_t = 2\,\mathrm{Im}(k') = N\left\{-2\pi k^{-2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}(a_n+b_n)\right\}$$

where Q_t is the 'extenction cross-section', or relative power removed per particle from the average coherent plane wave. Part of the 'power removed' is converted into randomly scattered power such as the 'stray radiation' of the interferometric measurement. Computing in the usual way relative power randomly scattered per particle (when the exciting wave is plane and k is real) we have: $Q_s = 2\pi k^{-2}\sum_{n=1}^{\infty} (2n + 1)(|a_n|^2 + |b_n|^2)$. When there are no ohmic losses all 'power removed' appears as randomly scattered power and we must have $Q_s = Q_t$; by replacing h_n by $j_n + in_n$ in the general electromagnetic scattering coefficient, one finds $|a_n|^2, |b_n|^2 = -\mathrm{Re}(a_n, b_n)$ so that $Q_s = Q_t$ follows as a methematical identity.

(We suppose the particles in the slab of note (1) are opaque and large compared with wavelength, and each has projected area A on the z = 0 plane, and finally that NAd << 1 so that shadowing of one by another can be ignored. Computed by Huygens' principle, ray theory, or otherwise, T = 1 - NAd so that $k'/k = 1 + iNA/k$ and $Q_t = 2A$, the well-known result that the extinction cross-section of a large opaque object is twice the area of its geometric shadow. About half of the 'power removed' by the opaque particle is scattered in a cone subtending k^2A steradians, with center parallel to the positive z-axis in the positive z direction. The angular width is calculable by antenna theory (physical optics) and the phase is such that the incident plane wave is canceled immediately behind the particle, to form its shadow. If the particle were completely absorbing ('black', or 'matched to space') the forward radiation remains, and we have an analogy to the fact that a resistor connected across a constant-voltage generator dissipates maximum power when equal power is lost in the generator's internal resistance. We conceptually measure T and hence Q_t interferometrically to discriminate scattered power flowing at finite angles with the z direction; this power has random phase if the particles swirled, and would count as stray light. Van de Hulst[22] discriminates against this stray light by measuring power remaining in the coherent transmitted plane wave with a properly trained telescope of aperture A'; the stray light's fractional contribution to the measured power is made arbitrarily small by taking A'/A sufficiently large. The large A' is obtainable as the area of a hole in a mask in the $z = d$ plane, with the discrimination A'/A being realized when the fixed-aperture power-measuring device is placed at sufficient distance along the ray through the hole's center; the results of Sinclair and LaMer,[23] obtained with substantially this geometry, confirm $Q_t = 2A$. Finally a relative power flux $1 - NAd$ flows (the principal portion rectilinearly, by ray theory) through the slab, of which $1 - 2NAd$ retains phase-coherence with the originally incident plane wave. If the particles were reflectionless half-wave plates (parallel to $z = 0$)

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22) H. C. Van de Hulst, "On the Attenuation of Plane Waves by Obstacles of Arbitrary Size and Form," Physica XV, No. 8-9, Sept. 1949.

23) Sinclair, D., Light scattering by spherical particles, J. Optical Soc. Am. 37, 475-480.

the relative power flux is unity, the coherent portion is 1 - 4NAd, and $Q_t = 4A$; the forward-scattered radiation has the same phase but twice the amplitude as that producing the shadow of the opaque particle. It is hoped that misunderstandings of Van de Hulst's exposition will be clarified by these supplementary remarks.)

We derive Q_t as the imaginary part of an effective propagation constant k'. At least in the electromagnetic case, the writer believes that an appropriate version of the Kramers-Kronig relation should apply to $k' = k'(\omega)$, so that $Re(k')$ should be deductible from $Im(k')$ and vice versa, for all particle concentrations. For dilute suspensions, Q_t therefore should imply a perturbation (linear in N) in propagation velocity.

The cross-sections Q_t, Q_s do not apply for high concentrations where interactions are important. The area $Q_t - Q_s$ might be termed the 'cross-section for heat generation' and is the cross section required for calculating the decay of energy in a reverberation chamber filled with a dilute suspension. For the acoustic case, only dilatational waves are measured (or propagate across the chamber) and we have

(power attenuation exponent) =

$$= N(Q_t - Q_s) = \frac{-4\pi N}{k^3} \sum_{n=0}^{\infty} (2n+1)\left[Re(A_n) + |A_n|^2 \right]$$

(which has the same significance as eq. (13.3), ref. 7). With small particles of radius r, the dominant scattering coefficients A_0, A_1, A_2

are of order r^3 and their real parts are also of this order, owing to shear and thermal losses. Therefore, as observed by Epstein and Carhart, $Q_s = O(r^6)$ (Rayleigh scattering) is relatively neglible.

Section IX - Critique of Acoustic Methods and Results

As our methods and results in acoustic propagation problems are incomplete and scattered through the text, we summarize them here, point out the remaining difficulties, and close with remarks on related problems.

The chief block toward more complete results in the acoustic cases is the lack of accurate, sufficiently general scattering coefficients. In the notation of Section VII C, Henry Passerini and the writer have found the following coefficients;

$$3A_0 = \frac{-ia^3}{3}\left\{1 - \frac{m'_A}{m_A} - (\gamma-1)\left[\frac{\rho' C'_p}{\rho C_p} - \frac{2\alpha'}{\alpha} + \frac{(\gamma'-1)m'_A}{(\gamma-1)m_A}\right]\right\},$$

$$3B_0 = \frac{-ia^2 b}{3}(\gamma-1)\left[\frac{\rho' C'_p}{\rho C_p} - \frac{\alpha'}{\alpha}\right],$$

$$3B_0'' = \frac{-ib^3}{3}\left[1 - \frac{\rho' C'_p}{\rho C_p}\right], \tag{9.1}$$

$$3A_0'' = \frac{-ib^2 a}{3}\left[\frac{\rho' C'_p}{\rho C_p} - \frac{\alpha'}{\alpha}\right],$$

$$3B_1'' = -ib^3\left[(\tau'-\tau)/(\tau'+2\tau)\right],$$

where A_0, A_0'', B_0, B_0'' are as in (5.4), where a (b) is sphere radius times dilatational (thermal) propagation constant, and where use has been made of the thermodynamic relation $(\gamma - 1) m_A = \alpha^2/(\rho C_p)$. The coefficients of orders 1 and 2 were found essentially as in Section VII D, thermal effects being ignored in the calculations. Some efforts were made to develop the coefficients of order 0 and 1 in powers of radius; the results have been remarked on in Section VII E but are not reported, both because of lurking doubts as to the accuracy of the results and because, as mentioned in Section V, one needs similarly developed order-2 coefficients, and probably the leading terms of the order-3, for consistency in applications. (The derivations were made routinely from equations [9.1] of the Epstein-Carhart paper[7]; in addition to difficulties with this paper mentioned after (7.39) and the use there of approximate propagation constants, mentioned implicitly in Section V, Passerini's calculation differed from their result [10.2] in the sign of the last term. We have used Passerini's sign, which is the minus before the last term in curly brackets in the form of A_0, above; this choice of signs is responsible for the clean interpretation of (5.6) in terms of adiabatic compressibilities.)

We have used available scattering coefficients here in three theoretical approaches to acoustic propagation in suspensions. The first, in Section V, should be regarded as illustrating the self-consistent method since we have not included the volume-proportional coefficients of order 2 in the formulation, nor have we properly treated the variety of exclusion radii in practical suspensions. Furthermore, the formulation should have produced the suspension's thermal propagation constant. No doubt this would have occured had the thermal dipole scattering (coefficient B_1'' of (9.1)) been considered in (5.4), as now seems proper; what effect this might have had on (5.6) is hard to imagine. The second method uses the general theory of Section VII to achieve the results of VII C and VII D. The latter contain some novelties, but one feels that explicit ω-dependencies would be found from exact scattering coefficients. The third simple 'thin slab' method of Section VIII is valid for dilute suspensions; even here the results could be made more satisfactory by separating thermal and dilatational effects, a separation ultimately worthwhile only if exact coefficients are used.

Now we discuss the present theories against the background of four incompletely explained experimental results. First, observations at 1 Mc/s of the attenuation in a kaolin-in-water suspension versus kaolin concentration, showed maximum attenuation at roughly 20% as compared with a theoretical 38% concentration.[24), 25)] The feeling

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24) R. J. Urick, "The Absorption of Sound in Suspensions of Irregular Particles", J. Acous. Soc. Am. Vol. 20, No. 3, 283-389, May 1948

25) W. S. Ament, "Sound Propagation in Gross Mixtures", J Acous. Soc. Am., Vol. 25, No. 4, 638-641, July 1953

is that the losses are due to the kaolin particles' slipping with respect to the water, and that thermal effects are relatively small. Physically, attenuation would be reduced if, with increasing concentration, some mechanism made the variously sized particles move in unison, which would happen if the viscosity of the water increased. We altered the 'old' theory (apparently properly) by assuming that the fluid's resistance D to motion a rigid particle of radius r was, instead of $D = 6\pi\eta r$, $D = 6\pi\eta' r$, where η is the viscosity of water, and η' the viscosity of the suspension given by (7.40). The albebraically convenient approximations $\rho_1 = 2.5$, $m_1 = 0$ and $r^2\omega\rho'/\eta \ll 1$ then led to an attenuation maximum at 20.5% by the modification as compared with 36.5% by the 'old' theory. If the above modification $\eta \rightarrow \eta'$ is valid, then the present theories must include the order-2 coefficients which led to (7.40), as these theories (in Section VII E in particular) at best give substantially the same prediction as the 'old' theory.

The second experimental result[24] is an observation that the attenuation of a kaolin suspension did not change substantially as the water's viscosity was increased over a 100-to-1 range by addition of methyl cellulose. The strange negative result might be explained by proper account of thermal losses; but then our explanations of the first experiment are questionable.

In the third experiment,[26] acoustic heat losses due to fogs

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26) Knudsen, Wilson, and Anderson, J. Acoust. Soc. Am. 20, 849 (1948)

were measured in a reverberation chamber. (This is partially discussed in Section VIII). Observed sound absorbtions were higher than theoretical.[7] Here it is possible that surface tension in the droplets played some role; again an exact calculation of the revelant scattering coefficients is called for.

Surface tension plays a definite part in the fourth empirical observation,[27), 28)] that bubbles move to preferred positions in standing sound waves in fluids. Here the problem of radiation pressure is foreign, but the 'thin-slab' method of Section VIII gives a heuristic explanation. We take N identical bubbles per unit volume of 'thin-slab', of thickness d, placed parallel to the wave fronts of the standing plane wave system. It is easy enough to calculate the radiation pressure F per unit area of slab from its effective density ρ' and compressibility m': the result is, with particle velocity = $V\cos(\omega t)\cos(kx)$

$$F = \frac{1}{4}\rho V^2 k d\left[\frac{m'}{m} - \frac{\rho'}{\rho}\right]\sin(2kx)$$

where m, ρ represent properties of the fluid as usual. Then we use the approximate equations (8.4), N being arbitrarily small, and divide by Nd to obtain the force on one bubble. More simply, for vanishingly

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27) D. E. Goldman and G. R. Ringo, "Determination of Pressure Nodes in Liquids," J. Acoust. Soc. A., Vol. 21, No. 3, p. 270, May 1949

28) F. G. Blake, Jr. "Bjerknes Forces in Stationary Sound Fields," J. Acoust. Soc. Am. Vol. 21, No. 5, p. 551, September 1949

small bubbles, the additive laws apply for ρ', m' and $m'/m - \rho'/\rho = f_1[m_1/m - \rho_1/\rho]$, with f_1 being the volume fraction of air in the slab, and with ρ_1, m_1 the properties of air. The result is that the bubble migrates toward a pressure node when $m_1 = 0$ or is effectively negative. Through a combination of surface tension and high compressibility, bubbles resonate when much smaller than wavelength in the fluid. The sign of $\mathrm{Im}(A_0)$ changes across resonance so that larger, more visible, bubbles have effectively negative compressibilities and are driven to pressure nodes, in agreement with observation.

This approach to radiation pressure is based on coherent waves; the energy carried in random scattering is not used. The error is unevaluable in the absence of a rigorous treatment of radiation pressure on the bubble. Rigorous calculation would again require rigorous scattering coefficients.

In sum, theories of acoustic propagation in suspensions where particle sizes are comparable with shear and thermal wavelengths remain in an unsatisfactory state. According to the writer, the best hope of improvement lies in a rigorous application of the methods of Section V; the chief obstacle here is lack of satisfactory scattering coefficients including surface tension effects.

Section X - Optical Activity

Now we introduce a volume-proportional effect which follows no previous law of composition. This effect pertains to the isotropic optical activity for electromagnetic waves. To introduce the new constant, β , we first require some general statements about optical activity.

A solution of sucrose in water is optically active, by which is meant that a beam of linearly polarized light traveling through this solution will have its plane of polarization rotated. This optical activity is isotropic because of the rotation, per centimenter of travel, is independent of the directions of propagation and of original polarization. The effect may be explained by postulating that the sugar molecules have certain peculiar scattering properties, and are numerous, small, and randomly oriented.

We now consider simple model molecules which, in cumulative effect, would produce optical activity. To produce activity, the models must scatter, in the forward direction of a plane-polarized incident wave, waves with polarization at some finite angle with that of the incident wave. We first choose optically active particles composed of isotropic materials shaped in such a way as to be characterizable as right or left-handed. A turn of a right-handed copper helix serves as an example. These shaped particles are suspended in an isotropic but inactive ambient medium. For any fixed arrangement of the particles, the suspension is everywhere describable through the isotropic scalar electromagnetic constants $\epsilon(x,y,z)$, $\mu(x,y,z)$ (effective at angular frequency ω). For waves propagating an inhomogeneous medium with space-varying scalar constants, there holds a reciprocal relation

$$\int \vec{E}_1 \cdot \vec{J}_2 \, d(vol) = \int \vec{E}_2 \cdot \vec{J}_1 \, d(vol), \qquad (10.1)$$

where $\vec{E}_1$ and $\vec{E}_2$ are the electric fields produced by the respective current distributions $\vec{J}_1$ and $\vec{J}_2$, and the volume integrals extend over all space. If $\vec{J}_1$ and $\vec{J}_2$ are fixed, but the shaped particles are arbitrarily rearranged, so that new fields $\vec{E}_1'$, $\vec{E}_2'$, are produced, then $\int \vec{E}_1' \cdot \vec{J}_2, = \int \vec{E}_2' \cdot \vec{J}_1$. For any particle rearrangement (and fixed $\vec{J}_1$, $\vec{J}_2$) the resulting fields $\vec{E}_1$ and $\vec{E}_2$ must likewise satisfy (10.1). Hence the electric fields, averaged over all arrangements of the particles, must satisfy (10.1). We now regard these average fields as those that would have been produced by the homogeneous, isotropic, optically active medium to which the suspension (suspension I) is equivalent, when the fixed source-currents are $\vec{J}_1$, $\vec{J}_2$.

Electromagnetic wave propagation thus takes place in the homogeneous medium, I, equivalent to suspension I, according to a version of Maxwell's equations in which there are new constants producing optical activity, and for which the reciprocity relation (10.1) holds. At angular frequency ω the equations

$$\begin{aligned} &\nabla \times \vec{E} - i\omega\mu\vec{H} - \omega\beta\vec{E} = 0 \\ &\nabla \times \vec{H} + i\omega\epsilon\vec{E} - \omega\beta''\vec{H} = 0 \end{aligned} \qquad (10.2)$$

contain added isotropic constants β, β'' and have plane wave solutions with rotating polarization. But (10.1) holds if and only if $\beta'' = \beta$. (The proof of this statement for $\beta = \beta(x,y,z)$,

$\mu = \mu(x,y,z)$, $\beta''(x,y,z) = \beta(x,y,z)$ is a slight generalization of the proof in the case of $\beta'' = \beta = 0$ and will not be given.) Hence medium I satisfies the Maxwell's equations

$$\begin{aligned} \nabla \times \vec{E} - i\omega\mu_1 \vec{H} - \omega\beta_1 \vec{E} &= 0 \\ \nabla \times \vec{H} + i\omega\epsilon_1 \vec{E} - \omega\beta_1 \vec{E} &= 0 \end{aligned} \tag{10.3}$$

where the time-factor $\exp(-i\omega t)$ is again suppressed. These equations are satisfied by the two circularly polarized plane waves

$$\left. \begin{aligned} E &= \mu_1^{\frac{1}{2}}(\vec{i} + i\vec{j}) \exp(ik_+ z) \\ H &= -i\epsilon_1^{\frac{1}{2}}(\vec{i} + i\vec{j}) \exp(ik_+ z) \end{aligned} \right\} \quad k_+ = \omega[(\epsilon_1\mu_1)^{\frac{1}{2}} + \beta_1]$$

and

$$\left. \begin{aligned} E &= \mu_1^{-\frac{1}{2}}(\vec{i} - i\vec{j}) \exp(ik_- z) \\ H &= i\epsilon_1^{\frac{1}{2}}(\vec{i} - i\vec{j}) \exp(ik_- z) \end{aligned} \right\} \quad k_- = \omega[(\epsilon_1\mu_1)^{\frac{1}{2}} - \beta_1]$$

Thus the constant β_1 measures the optical activity of the medium I, since it appears with opposite signs in the two propagation constants; $\omega\beta_1$ measures the rotation, in radians per unit distance of travel, of the plane of polarization of a linearly polarized plane wave.

If $\vec{m}$ and $\vec{n}$ are spherical vector wave functions (See Stratton, loc. cit.) having the same indices, it is readily verified that the fields

$$\vec{E} = \mu_1^{\frac{1}{2}}(\vec{m}_+ + \vec{n}_+) \; ; \quad \vec{H} = -i\epsilon_1^{\frac{1}{2}}(\vec{n}_+ + \vec{m}_+)$$

$$\vec{E} = \mu_1^{\frac{1}{2}}(\vec{m}_- - \vec{n}_-) \; ; \quad \vec{H} = i\epsilon_1^{\frac{1}{2}}(\vec{n}_- - \vec{m}_-)$$

satisfy the Maxwell's equations (10.3) where the subscript + (-) denotes that k+ (k-) is the propagation constant appearing in the spherical Bessel function. (From the foregoing equations, it is perhaps apparent that for $\beta \neq 0$, electric modes of oscillation of a spherical cavity are accompanied by waves of magnetic type, and vice versa; this fact suggests a method of measuring β at microwave frequencies.)

Now we form some of the substance into a small sphere of radius $\underline{a}$, immerse the sphere in an optically inactive fluid with constants ϵ, μ, $k \equiv \omega\sqrt{\epsilon\mu}$, and let the waves $\vec{E} = \vec{m}_{e11}$ and $\vec{E} = \vec{n}_{e11}$ be incident. Applying the usual boundary conditions of continuity of $\vec{R}\times\vec{E}$ and $\vec{R}\times\vec{H}$ across $|R| = a$, we find the scattering coefficients A, P, B, Q of the resulting outgoing waves $A\vec{m}'_{e11} + P\vec{n}'_{e11}$ and $B\vec{n}'_{e11} + Q\vec{m}'_{e11}$, respectively. The results are

$$A = \left[(\mu_1-\mu)(\epsilon_1+2\epsilon) - \beta_1^2\right]\cdot\left\{\frac{2ik^3a^3}{3(\mu_1+2\mu)(\epsilon_1+2\epsilon)-3\beta_1^2}\right\}$$

$$P = Q = (\mu\epsilon)^{\frac{1}{2}}\beta_1\cdot\{\ \} \tag{10.4}$$

$$B = \left[(\epsilon_1-\epsilon)(\mu_1+2\mu) - \beta_1^2\right]\cdot\{\ \}$$

Now we form medium I into a number of small spheres of radius $\underline{a}$, and suspend these spheres in the fluid in such a way that an average unit volume of the new suspension II contains an average of N of the spheres of medium I. Letting $f_1 = 4\pi Na^3/3$, we apply

(7.4) and (10.4) to find the constants ϵ', μ', β' of the suspension II, obtaining, after some algebra

$$\frac{(\epsilon'-\epsilon)(\mu'+2\mu)-\beta'^2}{\langle(\epsilon'+2\epsilon)(\mu'+2\mu)-\beta'^2\rangle} = f_1\,\frac{(\epsilon_1-\epsilon)(\mu_1+2\mu)-\beta_1^2}{\{(\epsilon_1+2\epsilon)(\mu_1+2\mu)-\beta_1^2\}} \tag{10.5}$$

$$\frac{(\mu'-\mu)(\epsilon'+2\epsilon)-\beta'^2}{\langle\ \rangle} = f_1\,\frac{(\mu_1-\mu)(\epsilon_1+2\epsilon)-\beta_1^2}{\{\ \}} \tag{10.6}$$

$$\frac{\beta'}{\langle\ \rangle} = \frac{f_1\beta_1}{\{\ \}} \tag{10.7}$$

These simultaneous expressions can be readily solved for ϵ', μ', β', but the resulting expressions are complicated, relatively unsymmetrical, and seem to contain no further physical information. When $\beta_1 = 0$, $\beta' = 0$, and equations (10.5) and (10.6) becomes identical with (7.7) if $\Sigma_t f_t = f_1$ in (7.7), so that when $\mu_1 = \mu$, then $\mu' = \mu$. But when $\mu_1 = \mu$, and $1 \gg |\beta_1^2/(\epsilon\mu)| > 0$, $\mu' \simeq \mu - f_1\beta_1^2/(3\epsilon)$, a conclusion perhaps obvious from (10.6). Thus β_1 acts as a property coupling electric with magnetic effects, and the composition laws (10.6), (10.7) are qualitatively different from the special 'Clausius-Mossotti' laws (7.7).

Hoek[29] has obtained a formula for the 'non-specific' (non-chemical) effect of concentration of active molecules on the rotation per centimeter: $\beta' = Cf_1 (n^2 + 2)$, where n is the average index of refraction of the solution at molar concentration f_1, and C is a constant. To obtain a similar formula, we solve (10.7) for β';

$$\beta' \simeq \left\{ \left[1 + C_1^2 f_1^2 (\mu' + 2\mu)(\epsilon' + 2\epsilon) \right]^{\frac{1}{2}} - 1 \right\} \div (C_1 f_1) \qquad (10.8)$$

For small $C_1 f_1$, this gives essentially $\beta' = Cf_1(\mu' + 2\mu) \cdot (\epsilon' + 2\epsilon)/(3\mu\epsilon)$ which is identical with Hoek's result provided that the dissolved molecules produce no appreciable changes in the permeability of the solution (i.e., $\mu' = \mu$), and that his n is interpreted as the refractive index of the solution relative to that of the pure solvent, or fluid.

As pointed out in Hoek's paper, chemical (or specific) effects of increasing concentration are highly important in determining how the observed value of β' varies with concentration (with N or f_1, in the present case). Such effects are apparent in the fact that, for an optically active solution of sucrose in water, the density of the solution is a non-linear function of either the molar or the molal concentration

29) H. Hoek, "General Theory of the Rotatory Power of Isotropic Media", Physica VIII, No. 2, pp. 209-225, Feb. 1941

of sucrose. The specific, or chemical effects would be minimal in an optically active gas, if one could be synthesized. For the purpose of experimentally verifying the present formulas one could construct an artificial dielectric consisting of paraffin (say) containing randomly distributed and oriented sections of right-handed wire helices of diameter and length small compared with wavelength.

Hoek's last paragraph deals with reasons why one can neglect the quadrupole moment of the active molecules. We now rejustify this neglect. With the present assumption that the active 'molecules' of suspension II are small spheres of radius $\underline{a}$ (and composed of medium I) the dipole scattering coefficients A,B,P,Q are volume proportional, i.e., proportional to a^3. It can be verified that the quadrupole scattering coefficients are of the negligible order a^5, as is the case when medium I is inactive. For the 'molecules' of suspension I, which are composed of isotropic, inactive materials and produce rotation by virtue of their shape, the molecules (or scatterers) must have some finite dimension d to produce the optical rotation, and the finite dimension also implies a quadrupole moment. In the example of the Appendix we show that the scattering coefficients corresponding to P and Q (of(10.4)) are of the order d^4, whereas quadrupole scattering is of order d^5. We may regard the 'volume fraction' f_1 occupied by the active scatterers as proportional to Nd^3; increasing N and decreasing d^3 proportionately (so that f_1 remains constant) decreases the optical activity of suspension I according to $N^{-3/4}$ but decreases the effect of the quadrupole moment at the greater rate $N^{-(2/3)}$. Thus a 'quadrupole effect' can be made negligibly small compared with the optical activity.

APPENDIX

In deriving (10.5) through (10.8) it was mathematically convenient to suppose that the scatterers producing the optical acticity of suspension II were small spheres composed of some isotropic, optically active material, called suspension I (or medium I). Suspension I, in turn consisted in an inactive fluid in which were suspended scatterers of some isotropic, inactive material, having structures such as to produce, on the average over all orientations, scattering similar to that of the spheres. In this Appendix, we describe a physically realisable scatterer having the requisite properties. In terms of a characteristic scatterer dimension d, this scatterer will be shown to have a scattering coefficient comparable to the B of (10.4) of order $(kd)^3$, those comparable to A, P and Q or order $(kd)^4$, all other scattering coefficients being of order $(kd)^5$ or higher.

The 'volume-proportional' coefficients, those of order $(kd)^3$, produce no optical activity. But the analogues of P and Q vanish more slowly with $d \to 0$ than any non-dipole scattering coefficients, so that with a fixed 'volume-fraction' of sufficiently small particles, suspension I will have some optical activity (β) of order kd. Due to neglect of quadrupole and higher order scattering, the error made in ascribing bulk constants to the suspension is of order $(kd)^2$. Relative to the optical activity, however, this error is now of order kd; thus the fractional error made in neglecting particle size is far more serious in the computation of β than in the computation of ϵ or μ, where the fractional error is of relative order $(kd)^2$

In order to describe the scatterer we assume two rectangular coordinate systems, with common origins, one with the unit vectors $\vec{x}_i$, $i = 1,2,3$, the other with unit vectors $\vec{x}'_i = a_{ij}\vec{x}_j$. (The Einstein summation convention is used here.) The scatterer consists in two rigidly connected identical passive dipoles, $\vec{P}_1$, $\vec{P}_2$, $\vec{P}_1$ being parallel to $\vec{x}'_1$ at the position $\vec{\rho}' = \rho_i\vec{x}'_i$, $\vec{P}_2$ parallel to $\vec{x}'_2$ at $\vec{\rho}' + \vec{d}' = \rho_i x_i' + d\alpha_i x_i'$ $(\Sigma\alpha_i^2 = 1)$. (Call this scatterer right-handed; we obtain a left-handed scatterer by changing any sign in the coefficient of d.)

The vector $\vec{\rho}'$ is the position vector of the dipole $\vec{P}_1$ with respect to the origin of coordinates; the average scattered field should be independent of the choice of the origin and hence independent of $\vec{\rho}'$. If there is a preferred origin, however, it has some fixed geometrical relation with the scatterer, so that we may take $\rho'_i = \mu_i d$. We may take the dipoles as copper wires of length ad, diameter bd, with a and b/a being sufficiently small so that at distances of order d, the scattered field produced by each wire may be regarded, within any preassigned fractional error, as that of a point dipole. The electric field incident on this scatterer has the sufficiently general form $E_o\vec{x}_1 + \vec{\vec{F}}_{ij}r_j + O(kd)^2$

where $\overleftrightarrow{F}$ is a dyadic fixed in the unprimed system and $\vec{r}$ measures distance from the origin.

The volume of each wire being proportional to d^3, the dipole moment of each is proportional to $(kd)^3$; each dipole causes a field parallel to the other proportional to the common dipole moment $p(kd)^3$ times a distance-dependence be having as $(kd)^{-3}$. Hence the 'Interaction' is independent of kd for $|kd| \ll 1$ and may be incorporated as a constant Q. Then, if e_1, e_2 are the electric field components along $\vec{P}_1$ and $\vec{P}_2$ respectively, we have

$$e_1 = E a_{11} + d a_{1i} F_{ij} \mu_k a_{kj} + Q e_2 + O(d^2),$$

$$e_2 = E a_{21} + d a_{2i} F_{ij} (\mu_k + \alpha_k) a_{kj} + Q e_1 + O(d^2).$$

Solving, we obtain

$$(1-Q^2)e_1 = (a_{11} + Q a_{21})E + d(a_{1i} + Q a_{2i}) F_{ij} \mu_k a_{kj} + \\ + d Q a_{2i} F_{ij} \alpha_k a_{kj} + O(d^2),$$

$$(1-Q^2)e_2 = (a_{21} + Q a_{22})E + d(a_{2j} + Q a_{1j}) F_{ij} \mu_k a_{kj} + \\ + d Q a_{1i} F_{ij} \alpha_k a_{kj} + O(d^2).$$

Multiplying e_1 by $pd^3\vec{x}'_1$, and e_2 by $pd^3\vec{x}'_2$ yields the two dipole moments $\vec{p}_1$ and $\vec{p}_2$.

We shall now compute the average scattered waves rather than the scatterer's multipole moments. The magnetic field $\vec{M}(\vec{R})$ at position $\vec{R} = R\sigma_i\vec{x}_i$ $(\sigma_i^2 = 1)$ arising from an oscillating electric dipole $\vec{p}$ at position $\vec{r}$ is given asymptotically ($kR \gg 1$) by

$$\vec{M} = M_0 \frac{e^{ikR}}{R^2} (\vec{p} \times \vec{R})\, e^{-\left\{\frac{ik\vec{r}\cdot\vec{R}}{R}\right\}} \cdot [1 + O(r/R)], \text{ or}$$

$$M_1, M_2, M_3 = M_0 \frac{e^{ikR}}{R} [p_2\sigma_3 - p_3\sigma_2, p_3\sigma_1 - p_1\sigma_3, p_1\sigma_2 - p_2\sigma_1] \cdot$$

$$\cdot [1 + ik(r_i\sigma_i) + O(r^2)]\{1 + O(r/R)\}$$

Hence we wish to compute the average components $\langle m_i \rangle$ of the scattered magnetic field $M(R)$: $m = m_1, m_2, m_3 =$

$$= \left\{\frac{M_0 e^{ikR} d^3 P}{R(1-Q^2)}\right\} \cdot \left[(a_{12}\sigma_3 - a_{13}\sigma_2), (a_{13}\sigma_1 - a_{11}\sigma_3), (a_{11}\sigma_2 - a_{12}\sigma_1)\right] \cdot$$

$$\cdot [1 - ikd\,\sigma_\ell \mu_m a_{m\ell}] \cdot$$

$$\cdot [(a_{11} + Qa_{21})E + d(a_{1i} + Qa_{2i})F_{ij}\mu_k a_{kj} + dQa_{2i}F_{ij}\alpha_k a_{kj}] +$$

$$+ \{\quad\} \cdot [(a_{22}\sigma_3 - a_{23}\sigma_2), (a_{23}\sigma_1 - a_{21}\sigma_3), (a_{21}\sigma_2 - a_{22}\sigma_1)] \cdot$$

$$\cdot [1 - ikd(\sigma_\ell \mu_m a_{m\ell}) - ikd(\sigma_\ell \alpha_m a_{m\ell})] \cdot$$

$$\cdot [(a_{21} + Qa_{11})E + d(a_{2i} + Qa_{1i})F_{ij}\mu_k a_{kj} + da_{2i}F_{ij}\alpha_k a_{kj}] \;.$$

for (large) R and arbitrary direction cosines σ_i, where the average is to be taken over all orientations of the primed coordinate system, the common origin remaining fixed.

We now require averages of products of the a_{ij}. Through terms of order $(kd)^4$ in the expansion of $\langle\vec{m}\rangle$, averaged products contain no more than three factors. Here the only non-vanishing averages are $\langle 1\rangle = 1$, $\langle a_{ij}{}^2\rangle = 1/3$, $\langle a_{ij} \cdot a_{k\ell} \cdot a_{mn}\rangle = +1/6$ or $-1/6$ according as the product appears in the formal expansion of $\mathrm{Det}(a_{ij}) (\equiv 1)$ with positive or negative sign. Applying these averages, we obtain,

$$\{\}^{-1} 6\langle m_1\rangle = 0 - ikdQE\alpha_3(\sigma_3^2+\sigma_2^2) + dQ\alpha_3[\sigma_2(F_{21}-F_{12})+\sigma_3(F_{31}-F_{13})]$$

$$\{\}^{-1} 6\langle m_2\rangle = -2\sigma_3 E + ikdQE\alpha_3\sigma_1\sigma_2 + dQ\alpha_3[\sigma_3(F_{32}-F_{23})+\sigma_1(F_{12}-F_{21})]$$

$$\{\}^{-1} 6\langle m_3\rangle = 2\sigma_2 E + ikdQE\alpha_3\sigma_1\sigma_3 + dQ\alpha_3[\sigma_1(F_{13}-F_{31})+\sigma_2(F_{23}-F_{32})]$$

or: $\langle\vec{m}\rangle = \vec{m}_o + \vec{m}_k + \vec{m}_D$

We have now calculated the leading terms of the average scattered magnetic field $\langle\vec{m}\rangle$ at a point many wavelengths away in a direction $\vec{\sigma}$ from the scatterer. These are the only terms through order $(kd)^4$ which do not vanish on the average. Since the μ_i do not appear, the choice of origin is unimportant.

The partial field $\vec{m}_o$, composed of the three terms in $(kd)^3$, is clearly the magnetic field of an electric dipole oriented along $\vec{x}_1$. The partial field $\vec{m}_k$, having components quadratic in the direction cosines, might appear to be 'quadrupole', but it is actually the field of a magnetic dipole parallel to $\vec{x}_1$. To see this, we let $\sigma_1 = \cos\theta$, $\sigma_2 = \sin\theta\cos\phi$, $\sigma_3 = \sin\theta\sin\phi$ and compare the result with a dipole field evaluated at $|kR| \gg 1$. As function of direction $\vec{\sigma}$, the field $\vec{m}_D$ has the form $\vec{\sigma} \times \vec{P}/R$ and hence is the magnetic field of an electric dipole determined by $\vec{\vec{F}}_{ij}$, i.e. by the incident magnetic field. If the fluid is slightly conductive, then a steady current flowing through suspension I will have a non-vanishing curl, owing to the right- or left-handed character of the two-wire scatterers. (The present discussion in terms of dipole moments does not bring out this fact.) Generalizing, one suspects that a steady current through a real optically active solution may produce a parallel magnetic field, or that a varying magnetic flux may produce a parallel component of electromotive force.

All the scattering through terms of order $(kd)^4$ is of dipole character. We further note that the magnetic dipole field $\vec{m}_k$ is parallel to the incident electric vector and does not depend on the propagation direction of the incident wave (on $\vec{\vec{F}}_{ij}$). Finally, if we take the incident field as a plane wave travelling with propagation vector $\vec{x}_2$, electric vector $E\vec{x}_1$, we have $F_{12} = ikE$ and other F-components vanish. Then, in the direction $\sigma_2 = -1$, $\sigma_1 = \sigma_3 = 0$, the $\vec{m}_k$ and the $\vec{m}_D$ fields cancel; the electric polarization of the remaining scattered field $\vec{m}_o$ is parallel to $\vec{x}_1$. This result is a necessary consequence of the reciprocity relation (10.1).

The term $\vec{m}_k$ or $\vec{m}_D$ relates incident electric or magnetic dipole excitation with magnetic or electric dipole scattering, respectively. Each has amplitude proportional to $Q\alpha_3$. In the present case, the factor $Q\alpha_3$ is, for fixed d, proportional to $\alpha_1 \cdot \alpha_2 \cdot \alpha_3$, and maximum when the three direction cosines are equal.

These results are in disagreement with the statements of Volkenshtein[7] to the effect that there must be quadrupole scattering of the same order of magnitude as that producing optical activity, and that the magnetic incident field produces no electric dipole scattering, except to a degree quadratic in the amplitude of the incident field. This last fact suggests that Volkenshtein's scatterer is of an electromechanical nature, with scattering therefore not strictly comparable with that of the present motionless scatterer.

The writer conjectures that all electromechanical scatterers producing optical activity will be found to depend quadratically on the field amplitude. A model consisting in an electret, which serves as axis for a rotating massive charged disk, produces optical activity through precession; here the required interaction of various parts of the model is of a mechanical nature. This model has some steady precession at a rate proportional to the square of

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[7] M. V. Volkenshtein, "on the Theory of Natural Optical Activity" Journal of Experimental and Theoretical Physics, USSR, Vol. 20, No. 4, pp. 342-6 (1950). Naval Research Laboratory Translation No. 423.

the amplitude of the oscillatory incident field. As with the present passive scatter, the activity of Volkenshtein's general model appears to depend (at least partially) on electromagnetic interaction between various parts. For any interactions depending on distances between moving parts, the motions would alter these distances and hence perturb the interactions. The amplitudes of the motions are approximately proportional to the incident field's amplitude, as are the interactions; thus perturbations of the motions would give rise to quadratic terms in the interaction. (For incident waves of angular frequency ω, these effects would be found at angular frequency 2ω or 0.)

Analogous quadratic effects are found in the phenomenon of ionospheric cross-modulation. Non-linear phenomena connected with acoustic waves, such as hydrodynamic streaming, are also somewhat analogous. Numerous observations of non-linear effects of magnetic precession have been made in recent years in experiments with ferrites at high microwave power levels. Probably all 'natural' interactions producting activity are of an electromechanical nature; The artificial dielectric described in Section X shows, however, that activity may be produced by strictly electromagnetic interactions. There the requirements of reciprocity produced the simplifying restriction $\beta'' = \beta$ in (10.2). It is not clear to the writer that the necessary reciprocity relation (10.1) is satisfied for electromechanical interactions, even if the conjectured non-linearities are neglected.

Notes added in proof

We discuss matters relating to the extension of Section VII to statistical mixtures. The physical properties $a', b', \ldots$ of a suspension formed by volume fractions f_i of small spherical particles p_i with volumes v_i and properties $a_i, b_i, \ldots$, in a fluid with properties $a, b, \ldots$, were given implicitly through the set of equations

$$F_j(a,b,\ldots;a',b',\ldots) = \sum_i f_i F_j(a,b,\ldots;a_i,b_i,\ldots) \tag{7.4a}$$

where $A_j = v_i\, F_j(a,b,\ldots;\, a_i,b_i,\ldots)$ are volume-proportional scattering coefficients of p_i isolated in the fluid.

For a homogeneous mixture, such as a two-phase alloy, there is no ambient fluid; the problem is to find some substitute for the fluid so that the method of (7.4a) applies. Here Landauer[1] argues that each granule may be regarded as immersed in a 'fluid' having the average properties of the mixture. Alternatively, we regard all particles of the mixture as spheres of graded sizes; the space between particles of any finite size is packed with particles of the next smaller size, with the same volume fraction as that obtaining in the final mixture. Thus any finite spherical particle is surrounded by a fluid having the average properties of the mixture.

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1) Rolf Landauer, "The Electrical Resistance of Binary Metallic Mixtures", J. Applied Physics, Vol. 23, No. 7, 779-784, July 1952.

Thus in (7.4a) above, one sets $a, b, \ldots$ equal to $a', b', \ldots$. The F_j on the left then vanish because a particle physically identical with its surroundings produces no scattering. Hence, with $\sum_i f_i = 1$ for the mixture, we have

$$0 = \sum_i f_i F_j (a', b', \ldots; a_i, b_i, \ldots) \tag{N1}$$

as a set of formulas implicitly determining the bulk properties $a', b', \ldots$ of the mixture.

But here we have an inconsistency: Let the property b be dielectric constant, permeability, electrical or thermal conductivity. Then for two materials of volume fractions f_1, f_2 in a fluid, (7.4a) gives

$$\frac{b' - b}{b' + 2b} = f_1 \frac{b_1 - b}{b_1 + 2b} + f_2 \frac{b_2 - b}{b_2 + 2b} . \tag{N2}$$

When $f_1 + f_2$ tends to unity, the suspension should approach a mixture and, by (N1), we should have

$$0 = f_1 \frac{b_1 - b'}{b_1 + 2b'} + \frac{b_2 - b'}{b_2 + 2b'}$$

in the limit; the properties of the fluid being squeezed out of the formula as the fluid is squeezed out of the suspension. But the b' of (N2) depends on b in the no-fluid limit. This casts suspicion on the validity of (7.4a) for high particle concentrations, and hence on the validity of its derivation.

In deriving (7.4a) for dielectric constant, ϵ', in the static case, we would have a sample sphere S (of volume V) embedded in fluid, the typical particle P in S being in a small sphere s containing no other particles. Then a potential Bz applied to S would result in a scattered potential

Az/r^3, A to be computed as if S had some ϵ'_1 and some r to be measured from the center of s. It was then assumed that Bz is the potential incident on each particle, so that the ith particle scatters $A_i z/r^3$. Since

$$\iiint_{S-s} \left\{ \frac{\vec{k}\cdot(\vec{p}-\vec{q})}{|\vec{p}-\vec{q}|^3} \right\} d\vec{q} \qquad = \text{const.}$$

$\vec{p}$ inside s, the average potential scattered by other particles in S has vanishing gradient at $\vec{p}$ and the assumption is justified. Furthermore,

$$\iiint_{S} \left\{ \quad \right\} d\vec{q} \qquad = VZ/r^3 \text{ for } \vec{p} \text{ outside S,}$$

so that the average scattered potential is $\Sigma_i A_i z/r^3$. Equating A with ΣA_i yields (7.4a) (for ϵ') when the dependence of A, A_i on ϵ, ϵ', ϵ_i and V, v_i is taken into account.

Here it was implicitly assumed that P scatters as if it were isolated in the fluid, so that no waves are rescattered by other particles back to P to form part of P's excitation. But if a wave incident on S 'sees' an ϵ', should not waves leaving s also see ϵ' at the boundary of s and be reflected? To calculate this reflection and the consequent excitation of P caused by its own scattering appears difficult but we may accomplish the same end by borrowing Landauer's idea that P is to be regarded as surrounded by the average-material.

General boundary condition equations at the surface of S have the form $Bb + A(a/V) = B'b'$, where b is a two element row vector with elements functions o the fluid's properties and for standing, or exciting waves; a is a similar vector for outgoing, or scattered waves and b' is for standing waves but with elements depending on the properties of S; B, A, and B' are wave amplitudes, with A proportional to V. In the previous example, it was proved that the waves exciting P were independent of P's position (except perhaps for P's self-excitation); we similarly assume that the internal wave of amplitude B' is the exciting wave so that $B'b' + A_i^*(a_i'/v_i) = B_i b_i$ are the boundary condition equations, determining the fields inside P. But in the fluid actually surrounding P we must have $B^* b + A_i(a/v_i) = B_i b_i$, where B^* is an undetermined exciting wave. Superposing the waves scattered into the fluid as before, we have $\Sigma A_i = A$. With $|ba|$ representing the determinant formed from the two indicated row-vectors, the three boundary condition equations become

$$\frac{VB}{|ab'|} = \frac{VB'}{|ab|} = \frac{A}{|b'b|} ,$$

$$\frac{v_i B'}{|a' b_i|} = \frac{v_i B_i}{|a' b'|} = \frac{A_i^*}{|b_i b'|} ,$$

$$\frac{v_i B^*}{|a\, b_i|} = \frac{v_i B_i}{|a\, b|} = \frac{A_i}{|b_i\, b|} .$$

Now we ignore the undetermined starred terms and among the others, solve for A_i and A in terms of B; $A = \Sigma A_i$ holds if

$$\frac{|b\, b'|}{|a' b'|} = \sum f_i \frac{|b\, b_i|}{|a' b_i|} , \qquad \text{(N3)}$$

where $f_i = N_i v_i$ is the volume fraction for type i, as before. The previous assumption was that $B^* = B$; using this and ignoring the middle equation, we get $\Sigma A_i = A$ holding when

$$\frac{|bb'|}{|ab'|} = \sum f_i \frac{|bb_i|}{|ab_i|} \; ; \qquad \text{(N4)}$$

this is equivalent to (7.4a).

At low concentrations, $\sum f_i \ll 1$, $a', b' \sim a, b$, and the two formulas are nearly equivalent. When $\sum_i f_i = 1$ and there is no fluid, the ad hoc substitution of a',b' for a, b in (N4) to represent the "average fluid" surrounding each particle results in

$$0 = \sum f_i \frac{|b'b_i|}{|a'b_i|} \, , \qquad \text{(N5); (N1)}$$

but this follows identically from (N3) and an inherent improvement has been made.

For dielectric constant, $b = (1, \epsilon)$, $a = (1, -2\epsilon)$ and (N3) becomes

$$\frac{\epsilon' - \epsilon}{3\epsilon'} = \sum f_i \frac{\epsilon_i - \epsilon}{2\epsilon' + \epsilon_i} \qquad \text{(N6)}$$

This is a slight generalization of Boettcher's result,[2] derived by essentially the same reasoning about fields inside the particles.

When two kinds of waves are required in computing scattering coefficients, the equation-sets are of the form

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2) C. J. F. Boettcher, "The Dielectric Constant of Crystalline Powders", Rec. Trav. Chim. Pays-Bas 64 P. 47,1945.

$$(Bb \text{ or } Cc) + Aa/V + Dd/V = B'b' + C'c',$$

$$(B'b' \text{ or } C'c') + A_i^* a'/\mathcal{N}_i + D_i^* d'/\mathcal{N}_i = B_i b_i + C_i c_i,$$

$$(B_i b_i \text{ or } C_i c_i) = B^* b + C^* c + A_i a/\mathcal{N}_i + D_i d/\mathcal{N}_i;$$

here C and D are amplitudes of the second type of incident and scattered wave, respectively, and a,b,... are now four-element vectors. Setting $C = 0$ and solving for A, D, A_i, D_i in terms of B, we find that $A = \Sigma A_i$, $D = \Sigma D_i$ when

$$|b'dbc| = \sum f_i \left\{ |b_i dbc|\,|a'd'b'c_i| + |c_i dbc|\,|a'd'b_i b'| \right\} / |a'd'b_i c_i|$$

$$|ab'bc| = \sum f_i \left\{ |ab_i bc|\,|a'd'b'c_i| + |ac_i bc|\,|a'd'b_i b'| \right\} / |a'd'b_i c_i|.$$

When $B = 0$, $C \neq 0$, similar steps yield

$$|c'dbc| = \sum f_i \left\{ |b_i dbc|\,|a'd'c'c_i| + |c_i dbc|\,|a'd'b_i c'| \right\} / |a'd'b_i c_i|$$

$$|ac'bc| = \sum f_i \left\{ |ab_i bc|\,|a'd'c'c_i| + |ac_i bc|\,|a'd'b_i c'| \right\} / |a'd'b_i c_i| \tag{N7}$$

Ignoring the middle equation and setting $B^*, C^* = B, C$ yields

$$\begin{Bmatrix} |bdb'c'| \\ |abb'c'| \\ |cdb'c'| \\ |acb'c'| \end{Bmatrix} \div |adb'c'| = \sum f_i \begin{Bmatrix} |bdb_i c_i| \\ |abb_i c_i| \\ |cdb_i c_i| \\ |acb_i c_i| \end{Bmatrix} \div |adb_i c_i| \tag{7.4a}$$

When $\Sigma f_i = 1$, we have

$$0 = \sum f_i \begin{Bmatrix} |b'd'b_i c_i| \\ |a'b'b_i c_i| \\ |c'd'b_i c_i| \\ |a'c'b_i c_i| \end{Bmatrix} \div |a'd'b_i c_i| \tag{N8}$$

as an identity from (N7) or by ad hoc replacement of a,b,c,d by a',b',c',d' in (7.4a).

Landauer's method for mixtures seems to have been developed by Odelevskii[3] and to have been applied to the thermoelectric properties of two-component sintered materials by Airapetiants[4]. We now apply the present (N8) as the natural generalizations of Odelevskii and Landauer's method to the thermoelectric problem.

In the standard treatment[5] of thermoelectricity, a material is described by coefficients X,Y,Z, such that electric current $\vec{j}$ and heat current $\vec{Q}$ are given by

$$-\vec{j} = (X/T)\nabla\mu + Y\nabla(1/T)$$

$$\vec{Q} = (Y/T)\nabla\mu + Z\nabla(1/T)$$

where μ is the electrochemical potential and T is absolute temperature. For steady state conditions it is necessary that $\nabla\cdot\vec{j}$ and $\nabla\cdot\vec{Q}$ vanish everywhere; assuming small potential and temperature gradients, we apply $\nabla\cdot$ to the above, write $1/T = 1/T_0 + \theta$, and ignore terms of second order in μ and θ as small to obtain an equation linear in $\vec{j}$, $\vec{Q}$, μ, θ in which μ and θ satisfy $\nabla^2\mu = 0$, $\nabla^2\theta = 0$. At boundaries between different materials, μ,

3) V.I. Odelevskii, J. Tech. Phys. (USSR) 21, 678 (1951).

4) C.V. Airapetiants, "Thermal Electromotive Force and Additional Thermal Conductivity of Statistical Mixtures", Soviet Phys. (Tech. Phys.) 2, 429 (1957)

5) Encyclopedia of Physics (Springer 1956) Volume XIX, page 271-274.

θ and the normal components of $\vec{j}$ and $\vec{Q}$ are continuous. Thus we solve for the amplitudes of scattered μ, θ fields of the form z/r^3 when waves in the X,Y,Z medium are incident on a sphere of volume V of an X',Y',Z' medium, and form the equivalent of (N8):

$$0 = \Sigma f_i \frac{[(X'-X_i)(2Z'+Z_i)-(Y-Y_i)(2Y'+Y_i)]}{\{(2X'+X_i)(2Z'+Z_i)-(2Y+Y_i)^2\}},$$

$$0 = \Sigma f_i [(Y'-Y_i)(2Z'+Z_i)-(Z'-Z_i)(2Y'+Y_i)]/\{\ \},$$

$$0 = \Sigma f_i [(Y'-Y_i)(2X'+X_i)-(X'-X_i)(2Y'+Y_i)]/\{\ \},$$

$$0 = \Sigma f_i [(Z'-Z_i)(2X'+X_i)-(Y'-Y_i)(2Y'+Y_i)]/\{\ \}. \qquad \text{(N9)}$$

These are four equations for three unknowns X',Y',Z' but they prove to be linearly dependent so that no inconsistency occurs.

For more direct comparison with Airapetiants' work, we convert these via $X = T_0\,\sigma/e^2$, $Y = -T_0 e\alpha X$, $Z = T_0^2/\tau + Y^2/X$, where σ and τ are the usual electrical and thermal conductivities, α is thermoelectric power, and e is the electronic change, to

$$1 = \Sigma \Gamma_i \sigma_i (2\tau'+\tau_i)/\tau',$$

$$1 = \Sigma \Gamma_i (2\sigma+\sigma_i),$$

$$\alpha' = \frac{\Sigma \Gamma_i \sigma_i \alpha_i}{1-2\sigma\Sigma\Gamma_i}, \qquad \text{(N10)}$$

where $\Gamma_i = 3\tau' f_i \div \{(2\sigma'+\sigma_i)(2\tau'+\tau_i)+2T_0\sigma'\sigma_i(\alpha'-\alpha_i)^2\}$.

For all α_i the same, $(=\alpha)$, there are no thermoelectric currents and the first two equations are equivalent to the Odelevskii-Landauer result,

$$0 = \Sigma f_i (\sigma_0' - \sigma_i)/(2\sigma_0' + \sigma_i), \quad 0 = \Sigma f_i (\tau_0' - \tau_i)/(2\tau_0' + \tau_i).$$

For general α_i, $\tau' > \tau_0'$ and $\sigma' < \sigma_0'$ in accord with the intuitive idea that thermoelectric currents provide extra channels for carrying heat and therefore increase thermal conductivity, whereas heat generated by the thermoelectric effect at particle boundaries is dissipated through heat flow and therefore increases the work done by electric currents traversing the mixture, hence decreasing electrical conductivity. Airapetiants predicts a different increase of thermal conductivity owing to α_i and apparently ignores the resulting decrease in σ' from σ_0'.

Airapetiants compares his prediction with measured values of α' for various volume-fractions of a two-component sintered material. (his Fig. 2). The experimental points appear to lie about midway between his prediction and the present α' calculated for his data, his prediction lying closest to the line $\alpha' = f_1\alpha_1 + f_2\alpha_2$.

The calculation of the present scattering coefficients is different from Airapetiants', so that it is not clear that he was not attempting to use the present general method; our expression (N10) for α' is formally identical with that of his eq (12).

For the boundary condition equations pertaining to the Lamé constants, two types of waves are involved (7.18) and (7.21) (i.e., dilatational and shear) with two types of physical constants. Our tedious algebra (if correct) gives, with $z = (3\lambda + 2\mu) \simeq 1/(\text{compressibility})$,

$$z' - z = \Sigma_i f_i \frac{z' + 4\mu'}{z_i + 4\mu'}(z_i - z) \quad , \tag{N11}$$

$$\mu' - \mu = \Sigma_i f_i \frac{(\mu_i - \mu) 15(\lambda' + 2\mu')\mu'}{3(\lambda' + 2\mu')(2\mu_i + 3\mu') + 4\mu'(\mu_i - \mu')} \, . \tag{N12}$$

These should apply to solid aggregates like concrete or granite. The small-f_i agreement with (7.35), (7.37) is almost obvious. When $\Sigma_i f_i = 1$,

$$\Sigma f_i \frac{z_i - z'}{z_i + 4\mu'} = 0 = \Sigma f_i \frac{\mu_i - \mu'}{3(\lambda' + 2\mu')(2\mu_i + 3\mu') + 4\mu'(\mu_i - \mu)}$$

When $z >> \mu$ as in a liquid, (N11) becomes the additive law (7.38, 7.16) for compressibility and (N12) yields

$$\frac{\eta' - \eta}{5\eta'} = \Sigma f_i \frac{\eta_i - \eta}{2\eta_i + 3\eta'}$$

for the viscosity η'. This again has the right limiting forms. In particular, for concentration f_1 of rigid particles, we have $\eta' = \eta / \{1 - \frac{5}{2} f_1\}$, to be compared with (7.40) and (7.41), where the irrelevance of η' for high concentrations was already discussed.

In summary, we have developed, in (N3) and (N7), general formulas for the properties of suspensions which (to judge from the examples) agree with the result (7.4a) at low concentrations and tend to the pure-mixture laws (N1) for high concentrations. [Thus the results of Section VII, stemming mainly from (7.4a), are to be replaced for high concentrations with those from (N3), (N7).] A further application would be to a mixture of materials X and Y, in which the interfaces tended to be convex on the X-side. Then the role of fluid would be played by some subfraction of X; how to assign this subfraction in terms of the convexity of the Y particles is unclear. Finally, the present reasoning would introduct an effective viscosity into the acoustic attenuation calculation of p 95 in a natural way; but the real problem there is that the particle sizes are comparable with shear wavelength and the present formulas do not apply.

Odelevskii's argument properly includes the assumption that the granules of the mixed substances are of comparable sizes; if the X granules are systematically smaller than the Y, the resulting bulk properties would appear affected as if the particles were of about the same sizes, and surface effects caused the Y particles to be the more nearly spherical, as in the foregoing discussion.

Recent orally presented papers of V. Twersky deal with propagation in suspensions of particles of dimension comparable with wavelength. The typical particle is also considered surrounded by an average medium, but Twersky's subsequent argument differs interestingly from the present one. For future comparisons, it should be easier to apply his method to our small-particle geometry than to try to extend our argument to his large-particle problems.