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X-Ray and Vacuum-UV Lasers - Current Status and Prognosis

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ABSTRACT

The amplification of x radiation by stimulated emission appears to be nearing the point of being experimentally feasible. This report analyzes the theoretical foundations for an x-ray laser by means of a generalized rate equation approach and determines the general requirements to be placed on the pumping rate, its rate of rise, and on excited-state lifetimes. These requirements give a basis for a selection and optimization of promising excitation (pumping) techniques and possible lasing media. In addition, most of the previous literature relevant to x-ray lasers has been reviewed and evaluated to assess its likely future application in the development of a realistic x-ray laser.

PROBLEM STATUS

This is an interim report on the continuing and expanding problem of developing lasers for the vacuum-ultraviolet and x-ray spectral regions.

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X-RAY AND VACUUM-UV LASERS —
CURRENT STATUS AND PROGNOSIS

INTRODUCTION

Recent advances in laser and related technologies provide strong evidence that x-ray lasers are forthcoming in this decade, perhaps within a few years. Among the most important developments of recent years are the following: Coherent laser emission has been demonstrated at wavelengths as short as 1161Å in the vacuum ultraviolet (VUV). New techniques have been demonstrated and are being developed to generate VUV radiation by the harmonic generation of coherent light. Conversion efficiencies in excess of 20% have been reported for x-ray generation using high-intensity "optical" laser pulses incident on high-Z targets. High-intensity pulses of electrons and x rays, with nanosecond or less risetimes, can be employed for pumping an x-ray laser. The concept of traveling-wave excitation of a laser has been demonstrated. All of these developments and more are discussed in detail later in this report; they all point towards the realization of coherent x rays. The basic ingredients for x-ray laser action are known; however, a great deal of careful work is required to bring these concepts to fruition.

The realization of an x-ray laser offers tremendous potential to many fields of science, and the impact on society would be enormous. Medical technology would have a diagnostic tool that would yield information on a submacroscopic scale; a collimated beam of x-rays would permit tumor therapy with a minimum of damage to surrounding tissue and organs. Also, the ultrashort wavelengths in the x-ray spectrum would make possible the holography of giant molecules, such as proteins. Even further, biologists would have another tool for investigating and altering the RNA and DNA molecules. In the realm of physical sciences the potential is just as great. Atomic and nuclear spectroscopy would clearly benefit from the x-ray laser, and coherent x-ray studies can provide previously inaccessible structural information for crystallographers and metallurgists. For plasma physics and controlled fusion research, short-wavelength lasers will be most valuable for both heating and diagnostics.

The potential for military applications is also very great. The information to be gained in the development of an x-ray laser and in its use as a research tool should greatly enhance the technological base of military devices and systems. The first x-ray laser will almost certainly be a low-power device. However, there is no fundamental reason to believe that a device of this sort could not be scaled up with another generation of research, just as some infrared lasers have grown into high-power devices. If high-power devices are realized, they certainly would permit studies designed to evaluate high-altitude atmospheric effects of an intense x-ray flux from natural or artificial sources.

The purpose of this report is multifold. It will provide an introduction to the basic physical concepts involved in obtaining coherent emission in the x-ray region. Further, it will indicate what has been accomplished in the relevant technologies up to this point in time, describe the fundamental concepts which will most likely provide a basis for an x-ray laser, provide a discussion and delineation of the important experimental approaches to this problem, and, finally, indicate paths of future research and development and their prospects for success. An attempt has been made to include references to most of the published work in the field.

The motivation for this particular report is the initiation at NRL of a research program on x-ray lasers and the need to point this research along relevant paths. The next section of this report gives a background in the fundamental physics involved in this area. This is followed by a discussion in the third section of techniques and approaches, along with a description of past work reported in the literature. The fourth section provides a general discussion of paths for future research. The Appendix contains the general solutions for the rate equations given in the second section.

FUNDAMENTAL PRINCIPLES

Conventional Optical Lasers

A familiar prototype of the optical laser is a uniformly pumped, two-level, atomic medium of length L situated between two parallel, flat, highly reflecting mirrors. If the number of excited atoms N_2 exceeds the number of lower state atoms N_1, spontaneously emitted photons can stimulate additional emission in excess of that lost by absorption. Hence, an amplification path is provided for spontaneous noise radiation in the medium. The effective length of the amplification path is greatest along the axis of the mirrors, and laser radiation essentially consists of photons propagating in this direction. Only certain such axial propagation vectors k are allowed by the resonant cavity, namely, those satisfying the relation $kL = n\pi$ where n is an integer. Since the stimulated emission cross section at a given frequency is proportional to the spontaneous fluorescence intensity at that frequency, it is seen that axial modes near the center frequency of the fluorescence line shape profile are amplified most. This shows up as the monochromaticity of laser radiation.

The gain and the threshold condition for such a laser are simply obtained from the following considerations. Each excited atom in the system spontaneously emits A photons per second. If $S(\nu)$ represents the normalized frequency distribution of this radiation, where $\int S(\nu)d\nu = 1$, with maximum at $\nu = \nu_0$, the number of photons per atom emitted per second in the frequency interval $d\nu$ centered at ν is $AS(\nu)d\nu$. The number of modes in this frequency interval is $p(\nu)d\nu = V8\pi\nu^2 d\nu/c^3$, where V is the volume of the active atomic medium, and c is the phase velocity of light in the medium. A quantum treatment of the radiation field (1) shows that spontaneous emission can be regarded as the emission from excited atoms induced by one photon in each and every mode. If there are $n(\nu)$ photons present in a given mode at frequency ν, they will induce an excited atom to emit $AS(\nu)n(\nu)/p(\nu)$ photons per second into this mode. Consequently, the total net rate of generation of stimulated photons (in excess of the absorption rate) in a mode of frequency ν from all atoms of the medium is given by $[N_2 - (g_2/g_1)N_1]\ AS(\nu)n(\nu)/p(\nu)$ where the g's are level-degeneracies. This must exceed the loss rate $n(\nu)/t_c$ for these photons, where t_c is the lifetime of a photon in the cavity. For a high-Q cavity we have $t_c \approx (L/c)/(1 - R)$, where R is the geometric mean of the reflectance values of the mirrors at opposite ends of the cavity. The preceding threshold condition is

$$\left(N_2 - \frac{g_2}{g_1} N_1\right) \frac{AS(\nu_0)}{p(\nu_0)} \geqslant \frac{1}{t_c}. \tag{1}$$

If we substitute the relations for p and t_c, and assume a Lorentzian line shape, this condition can be written as

$$\alpha(\nu_0)L \geqslant 1 - R \tag{2}$$

where

$$\alpha(\nu) \equiv \frac{c^2}{4\pi^2\nu^2\Delta\nu} A\Delta N. \tag{3}$$

Here $\Delta N \equiv [N_2 - N_1\ (g_2/g_1)]/V$ is the population inversion per unit volume of active medium, and $\Delta\nu$ is defined as the full width at half maximum of the frequency distribution $S(\nu)$. The quantity α is the amplification constant or negative absorption coefficient for Lorentzian line shapes, which are characteristic of lifetime-limiting mechanisms. Indeed, Eq. (2) is the low-gain approximation to the more general threshold requirement (2) given by

$$R \exp(\alpha L) \geqslant 1. \tag{4}$$

For Gaussian line shapes (e.g., as in Doppler broadening in a low-pressure gas) the amplification constant is given by $\alpha_G = \alpha\sqrt{\pi \ln 2} \approx 1.48\alpha$ where α is given (2) by Eq. (3).

Extension to the X-Ray Region

The difficulties of extending conventional optical laser technology to the VUV and x-ray regions of the spectrum can be seen from a consideration of Eqs. (3) and (4). First, Eq. (4) shows that pumping requirements could possibly be relaxed if short-wavelength reflectors were available. Unfortunately, reflectors and windows are notoriously inefficient in the VUV and x-ray spectral regions. Furthermore, the lifetimes of x-ray transitions (A^{-1}) tend to be very short since $A \propto \nu^2 f$. Here f is the oscillator strength of the transition, which tends to be a slowly decreasing function of frequency for a given type of transition. Consequently, the lifetimes of x-ray transitions can easily be so short (e.g., 10^{-15} s at 2Å) that several passes in a practical resonator cannot be made in this lifetime. This basic consideration suggests that early x-ray lasers will be single-pass, superradiant-type devices. However, it is possible that if a metastable transition could be found (f very small), the effective use of a resonator may be practical. In this situation, it is perhaps possible to employ efficient Bragg reflection from crystals at x-ray wavelengths for resonator mirrors (3). Because the Bragg angle is given by $\sin\theta = n\lambda/2d$, where d is the spacing of the Bragg planes, this technique is limited to the hard x-ray region (e.g., $\lambda < 10$Å). The conditions for effective use of such resonators are stringent, and the single-pass device employing synchronized traveling-wave pumping and amplification, as described in the next section of this report, appears more feasible.

The single-pass gain must be high, which typically might require α to be 10^2/cm or higher. This is a very difficult requirement at short wavelengths, as seen by Eq. (3), where ΔN is the population inversion which usually* must be established on the time

*The exception is when the lower laser level is depopulated sufficiently rapidly that the transition is not self-terminating. This is discussed further for specific cases below.

scale A^{-1}. This points up the importance of intense fast-risetime pumps, since lifetimes tend to be short at short wavelengths, and the pump intensity requirement ($\approx h\nu A\,\Delta N$) is proportional to $\nu^3\Delta\nu$. The line width itself usually increases with ν, depending on the mechanism; for example, for Doppler broadening of a gas resonance, the relation is $\Delta\nu \propto \nu(T/M)^{1/2}$, where T and M are the temperature and molecular weight of the gas, respectively. While the above difficulties have hindered the development of lasers at short wavelengths, these difficulties no longer appear insurmountable. The advent of high-intensity fast-risetime pumps, the techniques of traveling-wave pumping, the use of intermediate metastable excited states, and other approaches which appear scientifically and technologically possible today are described in the next section.

Generalized Rate Equation Analyses

A more detailed consideration of the physics of VUV and x-ray lasers provides further insight into the problems involved and indicates means to alleviate them. Essentially all x-ray laser approaches which involve population inversion of an atomic or molecular system can be described in terms of an idealized three-state system, such as shown in Fig. 1. The R's are rate coefficients with, e.g., N_2R_{23} being the rate at which the N_2 atoms (molecules) in state 2 make the radiative laser transition to state 3. Likewise, N_1R_{12} is the rate at which the N_1 atoms in the initial state are pumped into the upper laser level, either directly or via intermediate states. The rate R_{2n} represents the collective effects in the decay of the upper laser state to any other level n except level 3. Similarly, R_{3m} represents the decay of the lower laser level to any other level m, except level 2.

This basic three-level scheme can describe many physical situations. As an example, consider electron or photon pumping by excitation of an atomic or a molecular system, where levels 1, 2, and 3 represent specific bound states. In this case, the rate coefficient is given by $R_{12} = F\sigma_{12}$, where F is the incident electron or photon beam flux density and σ_{12} is the appropriate cross section for excitation.

As another example, consider an atomic system where electron or photon pumping produces inner shell ionization from the K shell, followed by photon emission from the L to K shells of the resulting ions. Here 1 represents the initial ground state atom, 2 the ion with a K-hole, and 3 the ion with an L-hole. Transition R_{12} represents ionization, R_{23} the radiative $K \rightarrow L$ transition for the ionic hole, and R_{2n} and R_{3m} represent other depopulation mechanisms for ion states 2 and 3. The latter include R_{21} and R_{31} recombination transitions from the ion to the atom.

The above examples indicate the generality of the three-level scheme shown in Fig. 1. In this scheme, the pertinent rate equations are

$$\frac{dN_2}{dt} = N_1R_{12} - N_2\,(R_{23} + R_{2n}) \tag{5}$$

and

$$\frac{dN_3}{dt} = N_2R_{23} - N_3R_{3m}. \tag{6}$$

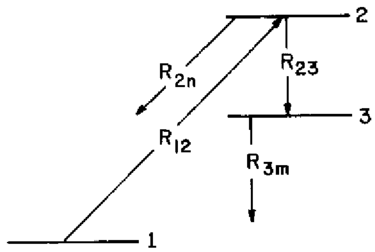

Fig. 1 — Diagram of energy states

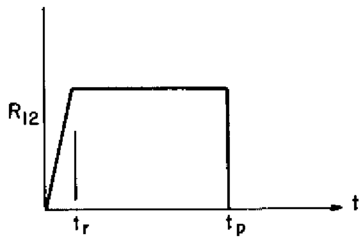

Fig. 2 — Pulse shape for pumping rate coefficient R_{12}

The initial conditions are $N_1(0) \equiv N_0$, and $N_2(0) = N_3(0) = 0$. We assume for simplicity that $N_1 \approx N_0$ throughout, i.e., only a minority of atoms are pumped up to excited states. We also assume the pumping rate coefficient R_{12} to have the idealized time dependence shown in Fig. 2 where the coefficient is linear up to the rise time t_r, and then flat up to the pulse duration t_p where it is truncated. All other rate coefficients are considered constant. We separate two cases for consideration: (a) an upper-laser-level lifetime long compared to the pump pulse rise time t_r, i.e., $(R_{23} + R_{2n})^{-1} \gg t_r$, and (b) the reverse case where $(R_{23} + R_{2n})^{-1} \ll t_r$.

Case A: Fast-Rise Pump $[(R_{23} + R_{2n})^{-1} \gg t_r]$ — The pump pulse time dependence can be regarded as constant from $t = 0$ to $t = t_p$ if t_p is at least comparable to the upper-laser-level lifetime. The solutions to the rate equations are easily obtained for this case. The result is

$$\Delta N = T'(t) N_0 \, R_{12}/(R_{2n} + R_{23}) \tag{7}$$

where the dimensionless function $T'(t)$ contains all the time dependence. The general solution for ΔN is given in the Appendix. In the limit that $R_{3m} \ll R_{2n} + R_{23}$, the lasing is self-terminated and population buildup in the lower laser level ultimately limits the inversion and gain. In this case the solution for $t < t_p$ is

$$\mathrm{T}'(t) = \left(1 - e^{-(R_{2n} + R_{23})t}\right)(1 + b) - b(R_{2n} + R_{23})t \tag{8}$$

where

$$b \equiv g_2 R_{23}/g_3 \; (R_{2n} + R_{23}). \tag{9}$$

The function T′ is plotted versus $(R_2 + R_{23})t$ in Fig. 3 where it is seen that positive gain is limited to a definite time period, and that the maximum $\mathrm{T}'_{\mathrm{max}}$ (and gain) is obtained at a time t_m about 0.5 to 5 times the quantity $(R_{2n} + R_{23})^{-1}$, depending on b. By differentiating Eq. (8) it is found that

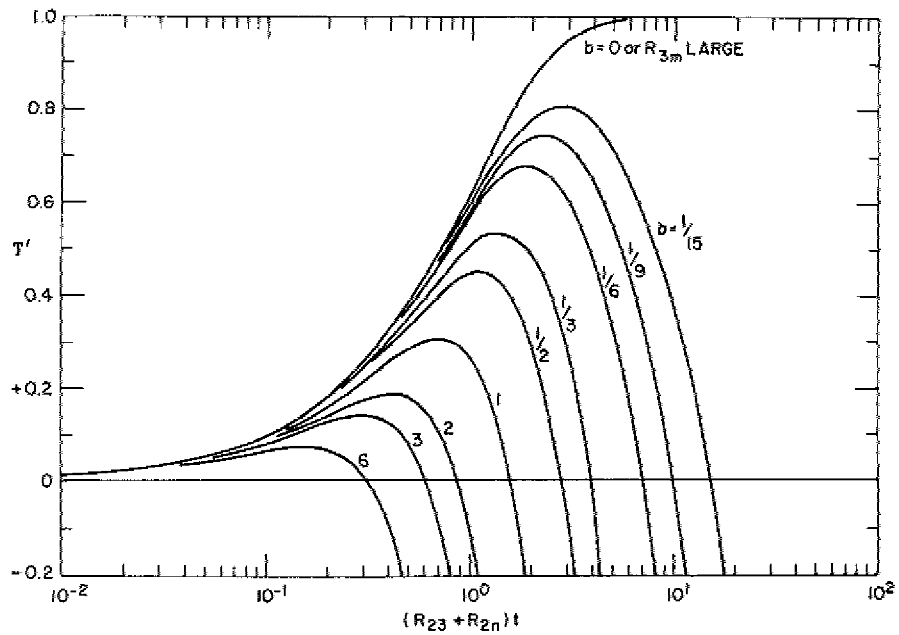

Fig. 3 — Time dependence of inversion parameter T' for the instant-on, fast-rise pumping case (case A). For R_{2n} small, t may be compared to R_{23}^{-1}, the laser transition upper-state lifetime. The $b = 0$ case is also the case for self-depleting final laser states where $R_{3m} >> R_{23} + R_{2n}$.

$$(R_{23} + R_{2n})\, t_{\max} = \ln[(1 + b)/b], \tag{10}$$

and

$$T'_{\max} = 1 - b \ln[(1 + b)/b]. \tag{11}$$

These latter two parameters are plotted versus b in Fig. 4. From the figure it is seen that the selection of a laser transition with a small g_2/g_3 (and hence small b) ratio is advantageous for gain. Raising R_{2n} (and thereby lowering b), however, is *not* desirable [cf. Eq. (7)] since it decreases the overall gain through a loss of pumped states in nonlasing transitions; hence, R_{2n} should always be kept as small as possible.

The factor $T'_{\max}$ in Eq. (11) appears in the formulas for maximum laser gain [Eqs. (3) and (7)] if $t_p > t_{\max}$. If, however, $t_p < t_{\max}$, it is the factor $T'(t_p)$ which appears. In some cases, it is possible to tailor the pump pulse duration without changing its energy content, i.e., a decrease in t_p is accompanied by a corresponding increase of R_{12}. A t_p appreciably less than $(R_{2n} + R_{23})^{-1}$ is advantageous for maximum gain in this case, since the second derivative of $T'(t)$ is negative for all t. In this limit we have from Eqs. (7) and (8)

$$\Delta N \approx N_0 R_{12} t_p, \text{ for } t_r \ll t_p \ll (R_{2n} + R_{23})^{-1}, \tag{12}$$

and it is the energy content of the pump pulse which is the determining factor.

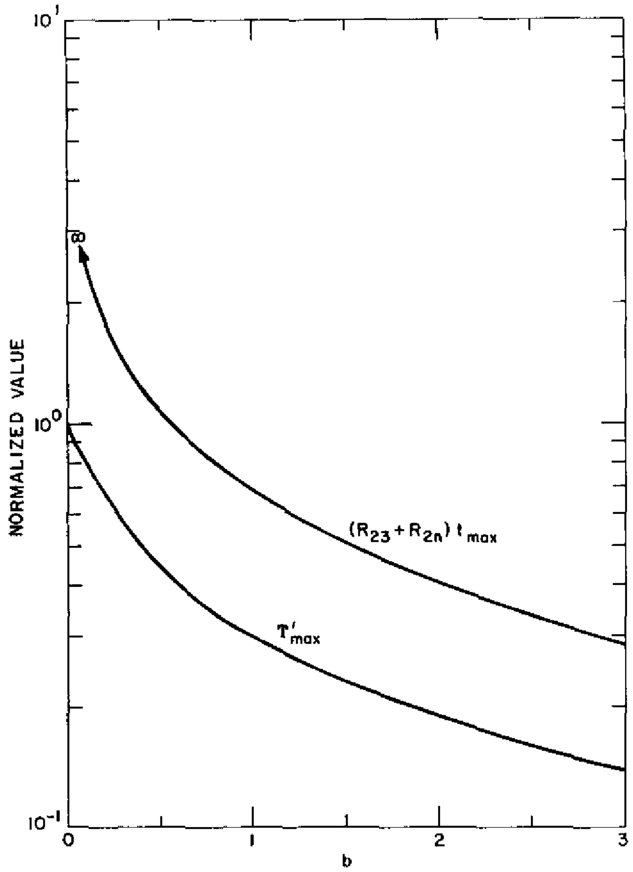

Fig. 4 — Normalized time $(R_{23} + R_{2n})t_{max}$ and amplitude parameter T'_{max} for maximum inversion vs the variable parameter b for the instant-on fast-rise case (case A).

If possible, it would be desirable to introduce a mechanism for selectively depopulating the lower laser level. The effect of increasing R_{3m} can be seen from the limit that $R_{3m} \gg R_{23} + R_{2n}$ when

$$T'(t) \approx 1 - e^{-(R_{2n} + R_{23})t'}. \tag{13}$$

This case, shown in Fig. 3, is the same as the $b \to 0$ limit discussed previously. In this case the gain is maximized and laser action is not self-terminating.

In the case of a Doppler-broadened gas laser transition, where $\Delta\nu_D = 2\nu(2kT(\ln 2)/Mc^2)^{1/2}$ with M being the mass of the radiating particle and T its temperature, the gain becomes [see statement after Eq. (4)]

$$\alpha = \sqrt{\frac{M}{2\pi kT}}\,\frac{c^3}{8\pi\nu^3}\,N_0R_{12}\left(\frac{R_{23}}{R_{2n} + R_{23}}\right)T'. \tag{14}$$

Here T_{max} ($\leqslant 1$) is further enhanced by (a) a careful choice of a laser transition such that the ratio g_2/g_3 (and therefore b) is small, (b) tailoring of the pumping pulse shape to deposit the available energy well within the laser transition lifetime, or (c) introducing a mechanism for selectively depopulating the lower laser level (e.g., a buffer medium which selectively makes charge transfer collisions with the ground state ions).

Case B: Linear-Rise Pump — On the scale of the short lifetimes which normally occur for short-wavelength transitions, the pump pulse may more realistically be assumed to be rising linearly*, i.e., $R_{12} = \dot{R}_{12}t$. Equations (5) and (6) are again easily integrated for $0 < t < t_r$ with the result

$$\Delta N = T(t)\, N_0 \dot{R}_{12}/(R_{2n} + R_{23})^2 \tag{15}$$

where $T(t)$ is a lengthy expression for the time dependence. The general solution for ΔN is given in the Appendix.

In the self-terminated lasing limit (i.e., $R_{3m} \ll R_{2n} + R_{23}$) the time-dependent factor $T(t)$ simplifies to

$$T(t) = \left[(R_{23} + R_{2n})t - \left(1 - e^{-(R_{23} + R_{2n})t}\right)\right](1 + b) - \frac{bt^2}{2}(R_{23} + R_{2n})^2 \tag{16}$$

where b is again given by Eq. (9). In Fig. 5 $T(t)$ is plotted versus $(R_{23} + R_{2n})t$ for several values of b. It is seen that in this case T continues to rise above unity. The maximum (T_{max}) is particularly large for small b values and occurs at a time t_{max} which can be much longer than the upper-level lifetime $(R_{2n} + R_{23})^{-1}$. The time t_{max} is found numerically from the relation

$$1 - e^{-(R_{23} + R_{2n})t_{max}} = \left(\frac{b}{1 + b}\right)(R_{23} + R_{2n})t_{max}, \tag{17}$$

and the corresponding T_{max} is given by

$$T_{max} = (R_{23} + R_{2n})t_{max}\,(1 - b\,(R_{23} + R_{2n})t_{max}/2). \tag{18}$$

Both $(R_{23} + R_{2n})t_{max}$ and T_{max} are plotted versus b in Fig. 6. Figures 5 and 6 show again the desirability of laser transitions with small g_2/g_3 ratios.

As before, a rapid depopulation process for the lower laser level is advantageous. The laser is then not self-terminating if $R_{3m} \gg R_{2n} + R_{23}$. In this limit $T(t)$ becomes

$$T(t) \approx (R_{23} + R_{2n})t - \left[1 - e^{-(R_{23} + R_{2n})t}\right], \tag{19}$$

which is identical with the $b = 0$ limit plotted in Fig. 5. In this situation, inversion is not limited to a set time interval, but the degree of inversion (and gain) is still dependent upon $\dot{R}_{12}$. Also, the appropriate model used (i.e., Cases A or B here, or some intermediate or alternative) still depends on t_r relative to $(R_{23} + R_{2n})^{-1}$.

*Other waveforms, e.g., sinusoidal, are also possible and solvable.

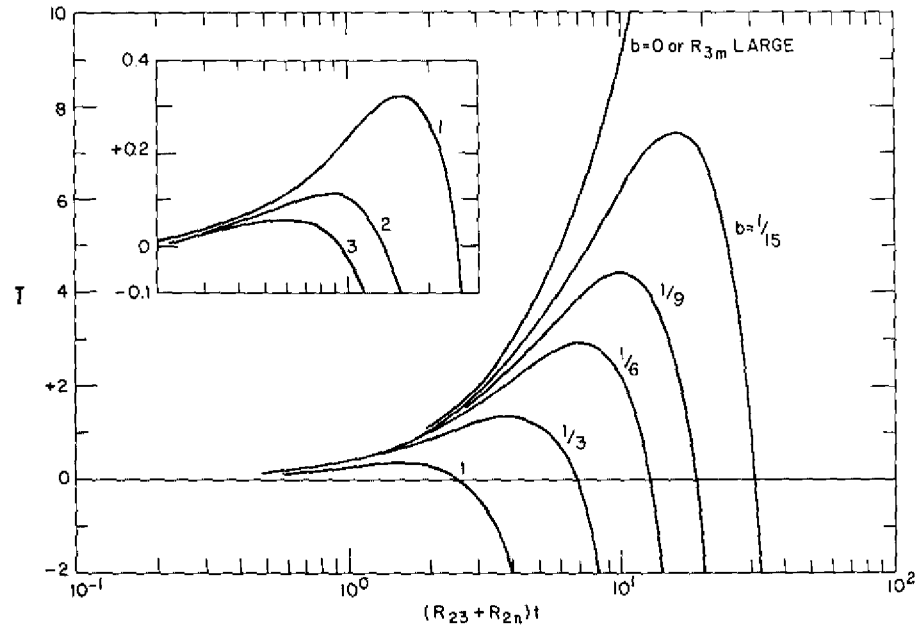

Fig. 5 — Time dependence of the inversion parameter T for the linear-rise (slow) pumping case (case B). For R_{2n} small, t may be compared to R_{23}^{-1}, the laser transition upper-state lifetime. The $b = 0$ case is also the case for self-depleting final laser states where $R_{3m} >> R_{23} + R_{2n}$.

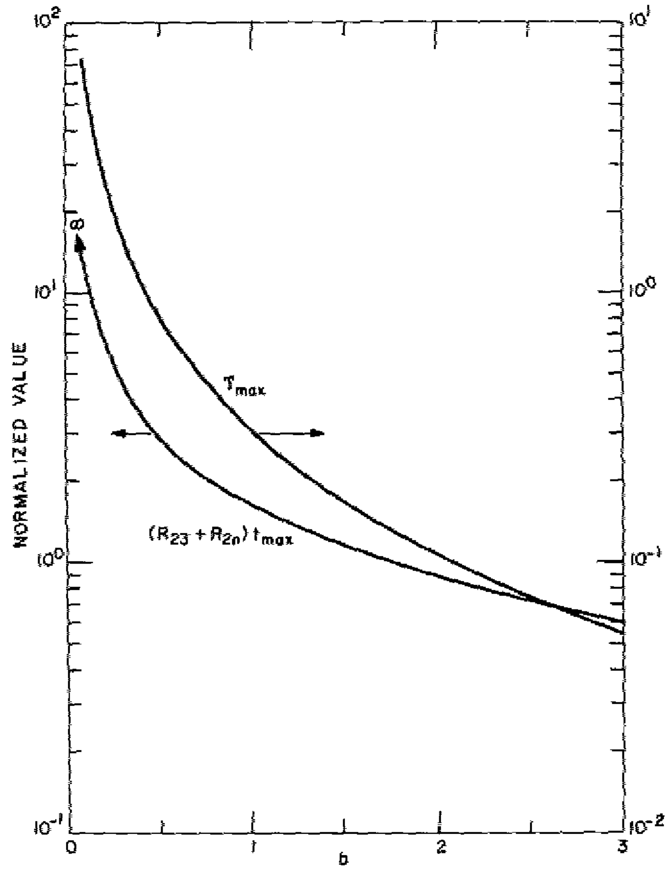

Fig. 6 — Normalized time $(R_{23} + R_{2n})t_{max}$ and amplitude parameter T_{max} for maximum inversion vs the variable parameter b for the linear-rise (slow) pumping case (case B).

The gain for a Doppler-broadened gas transition in the linear-rise pump case is

$$\alpha = \sqrt{\frac{M}{2\pi kT}}\ \frac{c^3}{8\pi\nu^3}\ N_0\dot{R}_{12}\ \frac{R_{23}}{(R_{2n} + R_{23})^2}\ \mathrm{T}(t). \tag{20}$$

In addition to the considerations following Eq. (14), the emphasis here is on $\dot{R}_{12}$, so that pump pulse compression could increase gain.

When $R_{2n} << R_{23}$ it is seen that α_{max} would depend on wavelength as strongly as λ^5, since R_{23} scales approximately as λ^{-2} and T_{max} is independent of R_{23} for self-terminated transitions [cf. Eq. (18)]. At short wavelengths, the gain could thus be severely limited by this dependence. On the other hand, Eq. (19) shows that for a laser which is not self-terminating, when T does not exhibit a maximum during the pump pulse, T is proportional to R_{23} and grows approximately linearly with t for $(R_{23} + R_{2n})t > 1$, along with the pump pulse. In this case α has a λ^3 dependence. This dependence was also found for the fast-rise pump treatment (Case A), which would be more applicable if, e.g., slower "forbidden" transitions were involved.

While we have treated the cases where the lifetime of the laser transition is either much greater or much less than the risetime t_r of the pulse in Fig. 2, the intermediate situation can also be handled simply by combining the solutions for the above cases. For example, the solution for $\Delta N(t_r)$ in the linear-rise pump case would be used as an initial condition in the fast-rise solution in the interval $t_r \leqslant t \leqslant t_p$.

The gain equations derived above can only be evaluated for a particular combination of atom, environment, transition, and pumping mechanism. For example, if pumping is achieved by means of free electrons in a plasma, R_{12} will be given by $Ne\langle\sigma v\rangle$, where N_e is the electron density, σ is the cross section for excitation or ionization, and v is the electron velocity, with the averaging performed over the velocity distribution function (usually Maxwellian). For an electron beam the electron density is replaced by the current density I from $I = N_e ev$, where v becomes the beam velocity. Similar expressions derive for photon beams, where the velocity becomes c and the current density becomes the photon flux.

Before proceeding to a specific example, a further comment about the above analysis is in order. While several approximations have been made to generalize the analysis and keep it amenable to analytical solution, it can also be shown that in most conceivable cases the inclusion of further effects and considerations do not significantly affect the results and do not warrant the necessary numerical routines to solve the nonlinear differential equations that arise. The reason for this is a shortage of information on the basic parameters — such as transition probabilities and cross sections — which are eventually needed. One must estimate and extrapolate to obtain this information, which introduces greater uncertainties than present in the derivations here. It is through further experiments that the x-ray laser will be developed (historically this has generally been the case with lasers), and with presently available information, theoretical estimates alone could even prove misleading.

We may conclude with one particular popular example (4). This is one of the rare cases where reliable cross sections are available. The lasing occurs in ionized sodium following the removal of a 2p electron from the configuration $1s^2 2s^2 2p^6 3s$ by photoionization. Lasing then occurs from the 3s to the 2p level in the neon-like ion and, while the statistical weight ratio is not favorable ($g_2/g_3 = 6/2$), the rate coefficient R_{2n} is low since recombination and autoionization rates are low in this particular case. Inner shell ionization is highly favored here for photoionization, with only a relatively small contribution from R_{13}. Pumping is carried out in a definite photon energy band near 200Å and lasing occurs at 372Å. For an upper-state lifetime R_{23}^{-1} of 0.3 ns and a b value of 3, t_{max} is found in Case B above to be 0.2 ns ($= 0.6\ R_{23}^{-1}$), and $T_{max} = 0.056$. For a power gain factor of $10^2\ cm^{-1}$, the pump flux power coupled into a sodium medium must rise at a rate of about 4 GW/ns/cm^3. A 5-m-long laser of this sort would deliver about 1.5 MW/cm^2 of flux. These numbers are consistent with a frequently quoted figure (5) of 10^{15} W/cm^3 as a required pumping flux density in the x-ray region.

SUMMARY OF PREVIOUS WORK AND PROPOSED APPROACHES

Introduction

The success of Maiman (6) and Javan et al. (7) in extending masers into the optical region caused some workers to immediately speculate about further extension into the x-ray and gamma-ray regions. Perhaps because of the exciting work to be done in the visible and infrared, or because of the obvious difficulties impending at short wavelengths, most of the early laser developers declined to work in the VUV or shorter wavelength regions. During the time since the very first laser appeared, a number of approaches to VUV and x-ray lasers have been suggested. Some of these approaches have been tried and have achieved a measure of success. Others have yet to be tried, and still others await new technological ideas. These approaches are reviewed below, beginning with a fairly comprehensive treatment of VUV molecular lasers and progressing to a discussion of other approaches for VUV and x-ray lasers. Finally, progress on the development of gamma-ray lasers is mentioned.

Vacuum Ultraviolet (VUV) Molecular Lasers

The energy levels of molecules provide potential laser transitions, as evidenced by the early development of the CO_2 laser in which an inversion is produced on bending modes within the first excited vibrational level ($v'' = 1$) and the ground vibrational level ($v'' = 0$). In molecules, the normal vibrational level separation is 1.0 to 0.01 eV, and the rotational separation is 0.001 to 0.00001 eV, all in the near- or far-infrared region. Vacuum ultraviolet transitions originate between electronic energy levels where separations may be as great as tens of electronvolts. A fast-rising high-temperature discharge can populate the vibrational-rotational structure of the upper electronic levels more highly than the competing upper vibrational states of the ground electronic level. In homonuclear diatomic molecules the vibrational and rotational transitions within an electronic level are highly forbidden; hence spontaneous and stimulated emission occurs only between the electronic levels. In most cases, lasing takes place between a number of vibration-rotational states of the two electronic levels.

The molecular nitrogen laser was perhaps the first laser to utilize molecular electronic levels. In this case, lasing takes place between two upper electronic levels, and a number of closely spaced rotational lines have been observed around 3371Å in the near ultraviolet.

The molecular-hydrogen laser was the first to be developed in the VUV region, and it is presently the shortest wavelength laser in existence. The advantage of such small molecules (e.g., diatomic) for short-wavelength lasers is that the population may be inverted on vibrational-rotational levels between electronic states, as exemplified by H_2. In Fig. 7 the ground-state vibrational level (the vibrational quantum number $v'' = 0$) is initially occupied at room temperature, while the $v'' \neq 0$ states are empty. This is due to the large energy separation ($\approx$0.5 eV) between vibrational levels for the ground-state molecule.

If the excited-state potentials were to have minimum coordinates and associated curvatures identical to the ground-state potential, then lasing would not occur. This follows from the fact that the perturbations which induce the pumping (electron or photon) and radiative emission transitions principally act on system electrons, and not nuclei. In the limit that the potentials have identical minima and curvatures, pumping and radiative transition matrix elements are essentially proportional to overlap integrals $\langle v'/v'' \rangle$ between identical excited

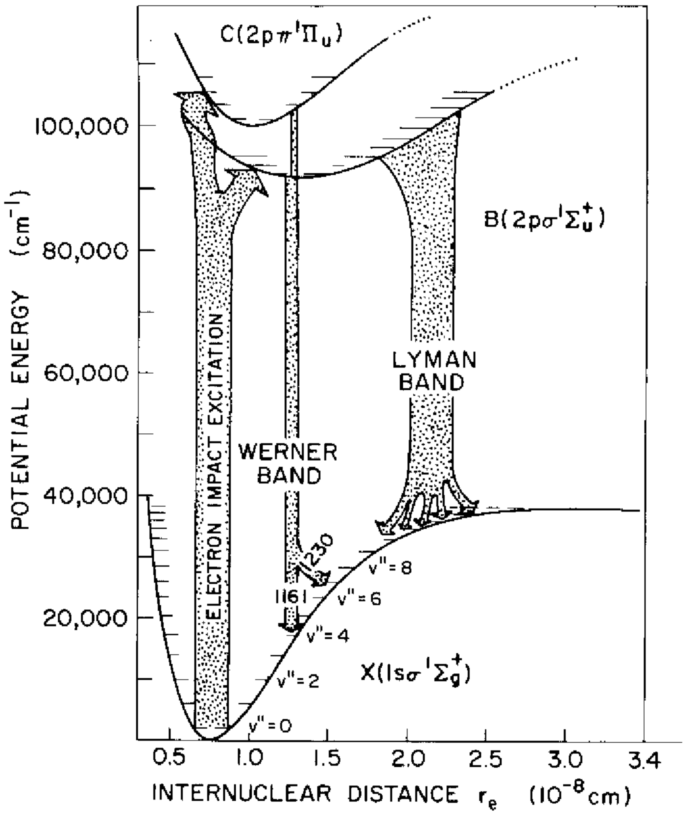

Fig. 7 — Energy level diagram for molecular hydrogen

and ground-state vibrational states, which vanish unless $v' = v''$. Hence, it would be very difficult to effectively pump a population inversion on vibrational states; the requirement is $v'' \rightarrow v'$, where $v' > v'' = 0$. Perhaps some collisional excitation mechanism could accomplish this. Even if this could be done effectively, the radiative emission $v' \rightarrow v'' = v'$ would be self-absorbed by the $v'' \rightarrow v' = v'' = 0$ transition.

In fact, the transition from one electronic state to another in a small molecule is typically accompanied by a change in equilibrium internuclear separation and vibrational force constant. Figure 7 illustrates this for H_2. In the ground electronic $X^1\Sigma_g^+$ state, two electrons fill a bonding orbital formed from 1s orbitals on each H atom. In the $B^1\Sigma_u^+$ state, one electron has been promoted from the $X^1\Sigma_g^+$ state to an antibonding orbital, which is formed principally from 1s, 2s, and 2p orbitals on each H atom, and a larger internuclear equilibrium separation is established. In the $C^1\Pi_u$ state, one electron has been promoted from the $X^1\Sigma_g^+$ state to a bonding orbital formed from the 2pπ orbitals on each H atom, and again a larger internuclear separation is established. The qualitative features of the minima and curvatures of excited state potentials relative to the ground-state potential, shown in Fig. 7 for H_2, appear consistent with the nature of the excited-state orbitals involved. These changes of minima and curvature permit pumping of the excited vibrational states in electronic excitation, as well as radiative emission at longer wavelengths back to excited vibrational states of the ground-state potential. The Franck-Condon principle states that these electronic transitions occur approximately vertically in diagrams such as Fig. 7. Large changes of potential minima or curvature are manifested by a large red shift (or "Stokes shift") of the emission wavelength from the pumping transition wavelength. Since a large Stokes shift also often implies minimal self-absorption losses, it is a valuable asset for lasing. The H_2 laser is effectively a three-level laser, where the upper laser level is directly populated by the pumping transition. The lower laser level is well removed from the initial ground-state level, as seen from Fig. 7.

The first published notion that H_2 (see Fig. 7) might be a suitable choice for a vacuum ultraviolet laser was put forth by Bazhulin, Knyazev, and Petrash (8). This suggestion triggered an effort by Ali and Kolb (9) at NRL to numerically solve the relevant rate equations for the H_2 system. At NRL (10) Shipman's N_2 laser was redesigned and adapted to H_2. Work was also begun on a smaller repetitive H_2 laser.

In early 1970, strong emission in the Lyman band of H_2 (≈1600Å) was noted at NRL in the Shipman traveling-wave system (see Fig. 8), and experiments were begun to verify the gain. In the traveling-wave device the exciting wavefront proceeds down the laser axis ahead of the laser pulse. The pulse is amplified strongly in the direction of travel of the excitation wave, but much less amplification takes place in the opposite direction. Simultaneous with the NRL work, R.T. Hodgson (11) at IBM put together a similar, but nontraveling-wave,

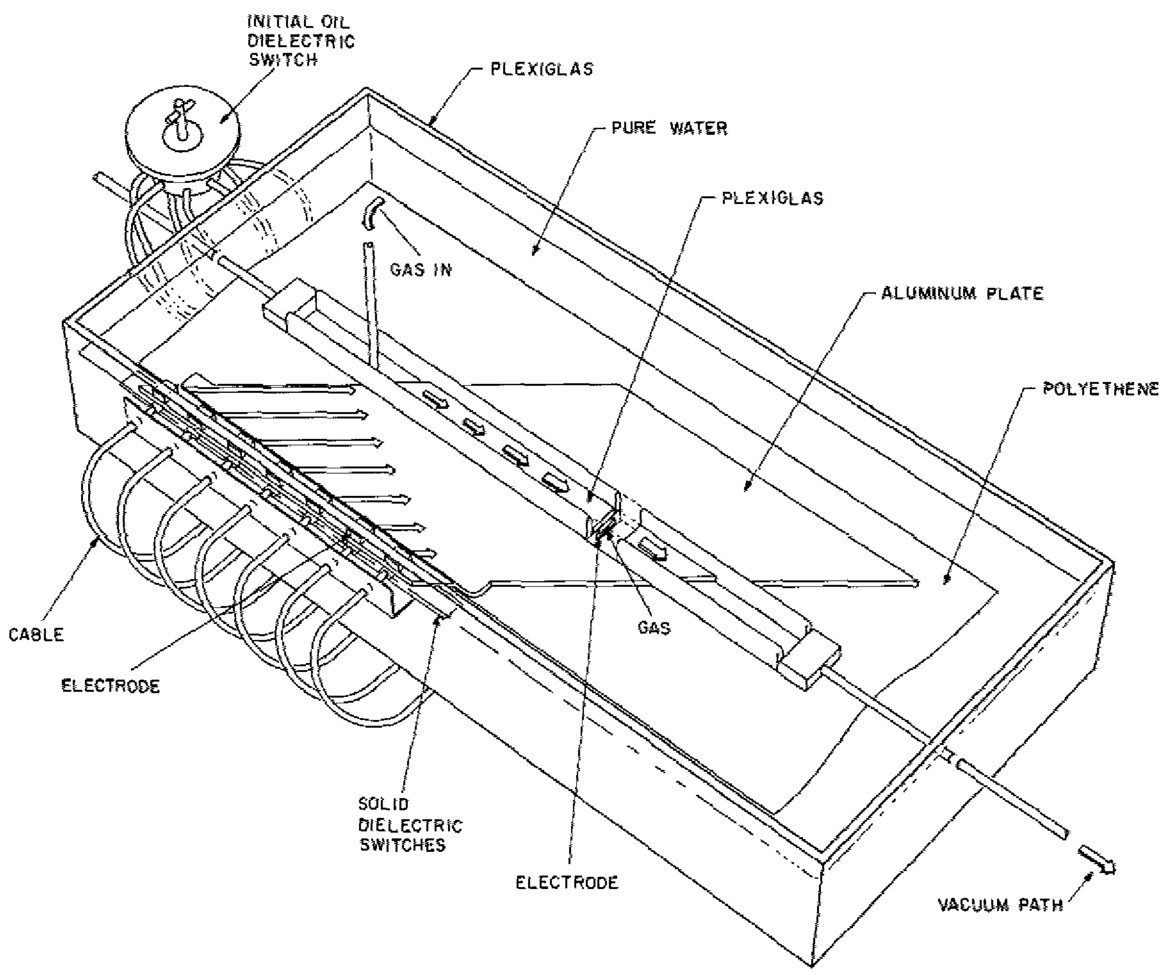

Fig. 8 — Traveling-wave excitation system used by Shipman of NRL to produce lasing in molecular hydrogen

device and noted a lower power (1 kW) in the Lyman band of H_2. At the same time NRL became convinced that the unidirectional emission noted from the traveling-wave system was the required proof of gain and announced (12) high-power (>100 kW) Lyman band lasing. Further immediate work on hydrogen at NRL resulted in even greater power (2 MW) (13), in observation of the "sufficient condition" for confirmation of gain (14), and in the realization of lasing in the Lyman band of deuterium (15). Nearly a year later and lower in power by nearly 100X from that of the Lyman band, prominent Werner band lines were found (14) at 1161Å and 1230Å by the NRL group. The reason for the lower power (≈5 kW) has not yet been fully explained, but may be due to the close competing levels near the upper state or to the filling of the lower states from the Lyman band emission. Hydrogen possesses other shorter wavelength transitions which can possibly be made to lase, but these will likely result in still lower power.

While on the subject of H_2, it should be pointed out that the triplet system (16,17) of H_2 (see Fig. 9), which has an unstable lower level, may not as yet be ruled out as a possible continuous-wave laser tuneable from the vacuum ultraviolet to the visible region. Indirect excitation from the $^1\Sigma_g^+$ ground state to a $^3\Sigma_g^+$ excited state (both are bonding states for the molecule) is followed by broadband emission (e.g., between the points A and B of Fig. 9) to an antibonding repulsive $^3\Sigma_u^+$ state for the molecule. A test for possible gain in such a system was attempted by Palmer (17). His measurements, which showed a loss at 3371Å rather than a gain, were obtained at an extremely low temperature (≈1.2 eV). This temperature may have been too low to sufficiently populate the upper triplet level and produce noticeable gain. In a practical system one must contend with the difficulty of pumping a triplet state efficiently from a singlet ground state. The feature of a rapidly dissociated lower laser level is attractive for the attainment of laser emission from other molecules, such as those of the rare gases, at wavelengths below 1000Å.

Basov and co-workers (19) have demonstrated that the high-energy electronic excitations of rare-gas molecules can be utilized for lasing in the vacuum ultraviolet. Using a high-voltage electron beam pump pulse with a peak current of ≈100 A/cm², they bombarded

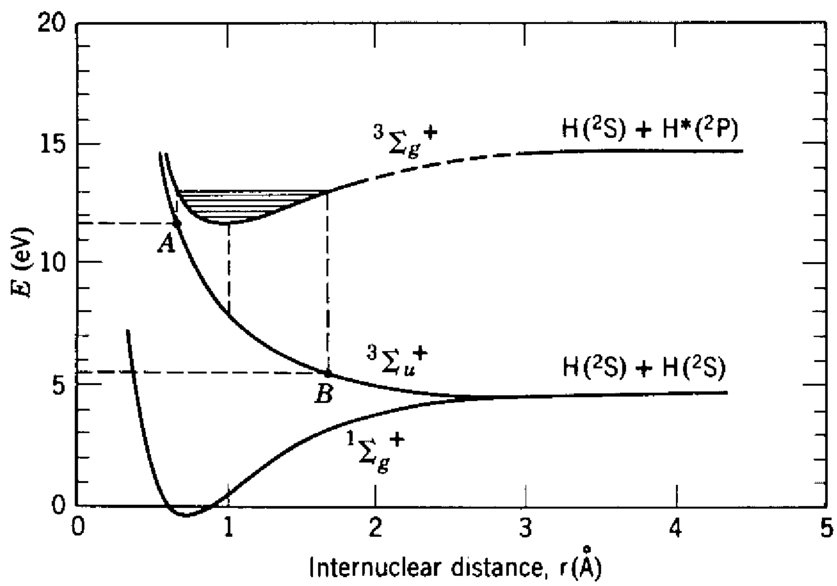

Fig. 9 — Triplet energy levels of hydrogen (from Ref. 18)

about 1 cm^3 of liquid xenon and achieved lasing at 1760Å. An output pulse duration of less than 10^{-6} s was observed, as was an energy density of 200 j/liter and 50% conversion efficiency (from beam power to laser power). For liquid xenon with an outer atomic configuration $5s^2 5p^6$, the steps in the laser process are probably as follows. First, a xenon atom is excited or ionized by a pump pulse to form a Xe^+ ion with a hole in the 5p shell as the final pumping state. In a close encounter with surrounding neutral atoms, the 5p hole on Xe^+ participates in a bond with a xenon atom to form the Xe_2^+ molecule. An electron is then captured by Xe_2^+ to form Xe_2^*, an excited Xe_2 molecule. The excited molecule then relaxes to the lowest metastable excited state, which becomes the upper laser level. The metastable molecule Xe_2^* decays radiatively to the ground state in which the two xenon atoms, having lost the bond, are rapidly repelled from one another. The density of liquid xenon is large enough so that sufficiently high gain can be obtained, even over the limited pumping lengths of the electron pulses. An attractive alternative might be the use of traveling-wave pumping in high-pressure rare gases.

Since other rare-gas atoms should similarly form diatomic molecules in their excited states, one may have the ingredients for extending laser emission far into the vacuum ultraviolet. The lighter rare-gas atoms have higher excitation energies; with He (in which a 1s hole produces the bonding for He_2^+) efficient lasing down to about 600-800Å appears possible. The laser process for He, which resembles that described for xenon, is schematically indicated in Fig. 10. Excitation from ground-state atoms is followed by diatomic molecule formation in the excited state, from which fluorescence to a repulsive ground state takes place.

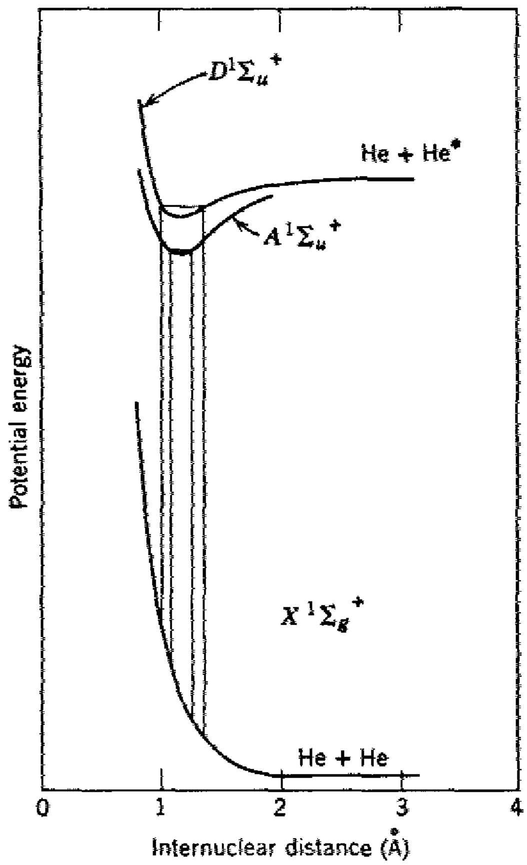

Fig. 10 — Energy level diagram for the helium molecule (from Ref. 18)

It is interesting to speculate on the role of excited states of Xe_2 and He_2 in the laser process. Because of exchange interactions there exist metastable triplet excited states at energies lower than the singlet excited states. Relaxation to these triplet metastables from the excited singlets apparently takes place sufficiently rapidly in liquid xenon so that a relatively long-lived triplet is the upper laser level, which relaxes pumping requirements for lasing. The coupling from singlet to triplet excited states may be assisted by the large spin-orbit interaction in xenon. It is not clear that the coupling from singlet to triplet excited states can take place efficiently in He_2 before radiative decay to the ground state from a singlet excited state occurs. If not, there may be ways to improve this coupling, e.g., through the addition of a buffer medium. These are subjects for future investigations.

Other Approaches for VUV and X-Ray Lasers

For lasing at wavelengths shorter than a few hundred angstroms, one must turn from molecules to atoms and ions for transitions of sufficient energy. Since most high-energy transitions involve inner electrons and terminate in highly populated lower states, the necessary population inversion is difficult to obtain by simple photon or electron collisionally induced excitation in a particular species. Transitions between ionic species which result in new species with innershell vacancies hold more promise. For example, one popular approach often discussed is that of rapidly removing a K-shell electron from an atom by either photon or electron collision, thus producing an inverted population in the resulting ion. This has been evaluated by Duguay and Rentzepis (4) for copper, which would result in lasing at 1.54Å . (This is the same paper in which is analyzed the photon-pumped sodium model for 372Å, used as an example in the above section on Fundamental Principles.) These authors conclude that a copper laser of dimensions 1 μm × 1 μm × 5 mm would require a traveling-wave input pumping power which rises at a rate of 25 GW/fs/μm^2 (1 femtosecond (fs) = 10^{-15} s), clearly beyond the capabilities of present technology. Rozanov (20) suggests, however, that by a careful choice of atoms, photoionization can result in ions in metastable* states. The existence of such metastable states can reduce the risetime requirements on the pumping system by a factor of 10^5 to 10^6, i.e., to a region more easily attainable. Because of the scarcity of information on inner-shell transitions, the location of such metastable states and the verification of laser action will require careful experimental investigations.

It is well to note that the same advantages that have made sodium attractive (i.e., a preferential photoionization of 2p "inner" electrons compared to the single 3s outer electron, the low probability of Auger transitions in the neon-like Na^+ ion created, a low ionization potential, and a low melting point) might be expected to be even more prominent in heavier alkalis such as cesium, with a corresponding slower transition (for easier pumping) in Cs^+ at 927Å, and, more interestingly, in sodium-like ions such as Si V at 118Å, etc., for shorter wavelengths. Such ions are commonly found in high-temperature plasmas where species as high as FE XXVI and Cu XXVIII have been observed (21) in a low-pressure concentrated spark discharge. In the same device is present the necessary fast-rising photon pumping flux, as well as high-energy electrons, most likely associated with turbulent heating in the plasma. Similarly, Mallozzi has reported (22) efficient broadband x-ray generation from a laser-produced plasma which could, for example, be used to pump x-ray laser transitions. Also, stimulated emission was suggested (23) as a possible explanation for a strong forbidden line in Al IV at 117Å, which resulted from a laser-produced plasma, although

*The metastability must include collisional depopulation, for which the rate can become large in high-density plasmas.

measurements with two plasmas in tandem did not confirm this conslusion. It is also possible that the results can be explained by the nearby allowed lines being optically thick, whereas the forbidden line would be expected to be optically thin. Such are the problems encountered in verifying gain, which is essential, and which is unfortunately often an overlooked or underestimated task. There is very little known about the allowed and so-called "forbidden" transitions in highly ionized metallic atoms, but hopefully much useful data can be obtained by isoelectronic extrapolation from lower species, such as sodium in the above case, and from further studies of plasma emission and heating processes.

Both Pantell (24) and Knyazev and Letokhov (25) have proposed two-step pumping processes. In the former, the laser medium can be pumped to a metastable state and then raised from there by a picosecond laser pulse to a higher state from which lasing takes place. Pantell carefully selected a laser material in which the picosecond laser photon energy matched the gap between metastable level and upper lasing level. Knyazev and Letokhov are not specific as to actual transitions and propose generally that a picosecond laser pulse be used to further heat the electrons in a preheated plasma, thereby avoiding the slower gas breakdown phase in the inversion process.

Several other suggested approaches fall into the VUV/x-ray regime. Gudzenko et al. (26) believe that amplification can be obtained on the 1641Å line of the helium ion in a decaying plasma. In the scheme proposed, the population inversion is created by preferential collisional depopulation of the lower state by neutralizing charge-exchange collisions with helium atoms. This type of process may occur in other atomic systems.

Another interesting proposal comes from Presnyakov and Shevel'ko (27) and involves excitation by charge exchange of protons with atoms ($p + A \rightarrow H + A^+$). By bombarding atoms with high-energy protons, it is possible to produce inner-shell ionization resulting in excited ions and a population inversion. Seemingly, the tradeoff involved when a proton beam intersects an atomic beam is that the rate of pumping requirement is superseded by limitations on the size of the interaction region and the velocity of the atomic beam. Calculations indicate a small gain, but the device would operate in a cw mode, with perhaps a few milliwatts of power, using Cs which would emit at 912Å. The most important aspects of this proposal are whether sufficient power can be obtained to make the device useful and whether the principle can be extended to shorter wavelengths. Experiments to determine these aspects should be performed.

Several other suggestions should also be considered. The first is the idea of coherent Cerenkov emission, proposed and pursued by Pantell (24). In this approach a relativistic electron beam is passed through a material and Cerenkov radiation is emitted at the Cerenkov angle $\cos \theta_c = 1/n\beta$, where n is the index of refraction and $\beta = v/c$. Gain appears possible with extremely energetic electrons. The technique seems limited, however, by the lack of materials with an index $n > 1$ which transmit wavelengths below 1000Å. The second suggestion provides an answer to the question concerning lack of good resonators at VUV wavelengths. As pointed out by Kolpakov et al. (28), Bragg reflection from monocrystals can be used in a Cotterill-type resonator. Hence, in low-gain systems with relatively long lifetimes, resonators can be used to increase the radiation field.

The last suggestion has been made by Harris and Miles (29) and involves the use of nonlinear optics to generate VUV radiation. Intense laser radiation at a frequency ω_0 can be used to generate a polarization at the frequency $3\omega_0$ in a medium with a large nonlinear electric susceptibility. This polarization can produce an intense beam of radiation at the frequency $3\omega_0$ if the process is "phase matched." This condition requires that the fundamental wave, which produces the nonlinear polarization, propagate at the same velocity as the radiated third harmonic in order to maintain phase coherence. In the case of Harris and Miles (29), and the experimental work of Young et al. (30) which followed, the nonlinear medium was rubidium vapor. Phase matching for third-harmonic generation requires that the indexes of refraction at ω_0 and $3\omega_0$ be equal. This was accomplished by mixing the rubidium vapor with xenon in the ratio 1:412. In this ratio the anomalous dispersion of the rubidium vapor due to large oscillator strength absorptions in the visible and near infrared completely compensates for the normal dispersion of xenon for ω_0 radiation with a wavelength of 1.06 μm. The experimental work indicates that a 50% conversion efficiency should be possible for 1.06-0.35 μm radiation with an interaction length of 50 cm and 10-MW peak input power.

These experiments demonstrate a potentially promising technique for generating VUV. This third-harmonic technique can conceivably be cascaded many times with fairly high efficiency. Young et al. propose Cd-He for a second stage (3547Å → 1182Å) of their experiment, for example. It may be possible to extend this technique to the 100Å region. However, this will require sophisticated technology.

Gamma-Ray Lasers

The question of stimulated gamma-ray emission was also raised quite soon after the extension of masers into the optical spectrum. A few papers were written at that time, but for nearly ten years little mention has been made of gamma-ray lasers. More than likely, this is due to the extreme lack of information on the energy levels of atomic nuclei. The article by Vali and Vali (31) lays down some of the basic requirements for a gamma-ray laser, but preparation of a material capable of lasing seems very difficult.

Chirikov (32) reiterates the following problems foreseen by others in building a gamma-ray laser: (a) the preparation of a material with a sufficient concentration of excited nuclei, (b) the finding of a sufficiently narrow gamma line so that the cross section for induced radiation is on the order of 10^{-18} cm^2, and (c) the finding of a suitable reflector to form a resonant cavity. Like the Valis, Chirikov has no solution to the problem of preparing the material. The use of Bragg crystals may not be possible because of the strict tolerance imposed on crystal planes. The cross-section problem, however, according to Chirikov might be solved by finding a narrow Mössbauer line. If such a line is used, induced radiation can be obtained from lengths of crystals on the order of 1 mm in length. No estimates of the output characteristics are given.

PROGNOSIS

From the discussion of previous sections, it is clear that in the last ten years a large amount of effort has gone into considering the problems of achieving x-ray laser action. The work and analysis described above indicate that the problems can be reduced to three major areas: (a) the achievement of pumping rates fast enough to compete with the very short spontaneous lifetimes associated with x-ray transitions, (b) the attainment of pumping intensities sufficient to produce a population inversion, and (c) the invention of a resonator or alternative to achieve intense collimated x-ray emission. It is clear that each of these problems is formidable. However, they are remarkably similar to many types of problems researchers had considered even prior to the advent of visible lasers. Ten years ago these problems may have seemed insurmountable, but recently technology in several related areas has made major advances. At this point in time there are very reasonable approaches to each of these problems.

Consider first the problem of pumping rate. There are many ways to pump an x-ray laser, and several mechanisms can be turned on in ultrashort intervals. For example, optical pulses at 1.06 μm exist today with risetime of less that 10^{-12} s. Pulsed x-ray sources exist which have risetimes of less than 10^{-9} s, and pulsed electron beam sources exist with comparable risetimes. Hence, fast-risetime sources exist. The detailed mechanisms by which the sources would couple their energy into a possible laser medium are different and are still not completely understood since, on these time scales, almost everything is transient and far from equilibrium. This is a problem, but not one of impossible magnitude; it is amenable to careful theoretical and experimental investigation. The use of long-lived metastable states as upper lasing levels can aid considerably here.

The problem of pump intensity is also made less significant by modern technological advances. Laser pulses at 1.06 μm are available on subpicosecond time scales with peak powers in the 10^{13} W range. Comparably, energetic x-ray and electron pulses are also available or within reach. This problem can also be diminished, as discussed earlier, through careful use of metastable states which can store energy in the laser medium. There are several possibilities for then extracting this energy as coherent x-ray radiation. These approaches include resonant transfer, double pumping, and stimulated "Raman-type" effects. The detailed physics is not available, but as before there is sufficient comprehension of this problem to indicate solutions in the near future.

The final problem of resonators is associated with the fact that a region of gain in a possible laser medium is very short lived. In all probability a resonator will not be part of early x-ray lasers, since transit times will be comparable to, or longer than, the inversion lifetime. This problem can be overcome by traveling-wave excitation. The basic concept is to have the gain region propagate in the laser medium at the same rate as the coherent laser pulse. This technique has been demonstrated in the VUV, but not with the intense ultrafast risetime pulses necessary for the x-ray region. However, in this case the technological step required is also within sight.

In summary, today's technology has made the fundamental problems associated with an x-ray laser much less formidable. Problems do remain, however. The work to be done includes (a) identification of metastable states in the x-ray region in various materials, (b) development of traveling-wave, high-intensity, ultrashort-risetime, pumping technology, and (c) investigation of the physics of excitation mechanisms during nonequilibrium pumping. At this time the best approach to these problems is probably a step-by-step approach. Starting with the existing VUV laser technology and working toward the x-ray region, all of the problems become increasingly more difficult, but in small enough increments to be solvable.

Finally, from what has been discussed previously it can be concluded that useful x-ray lasers will be a reality in the near future, probably within 5 to 10 years. The major problems seem solvable, and a substantial effort to attain x-ray lasers and unlock their enormous potential seems both timely and well justified.

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Appendix

POPULATION INVERSION FROM RATE EQUATIONS

The general solutions of the rate equations

$$dN_2/dt = N_0 \, R_{12} - N_2 \, (R_{23} + R_{2n}) \tag{A1}$$

and

$$dN_3/dt = N_2 \, R_{23} - N_3 \, R_{3m}, \tag{A2}$$

which correspond to Fig. 1 and are discussed in the second section of the report, are easily obtained for the two cases discussed in the text. We give them here for completeness.

<u>Case A</u>: Fast-Rise Pump ($R_{12}(t)$ = constant)

The population inversion is given by

$$\Delta N = N_2 - (g_2/g_3) \, N_3 \tag{A3}$$

where g_2 and g_3 are the upper and lower laser level degeneracies. For economy of notation, we use the lifetime of the upper laser level, defined by

$$\tau_2 = (R_{2n} + R_{23})^{-1}. \tag{A4}$$

The solutions to the rate equations are

$$N_2(t) = N_0 \, R_{12} \, \tau_2 \, [1 - \exp\,(-t/\tau_2)] \tag{A5}$$

$$\begin{aligned} N_3(t) = N_0 \, R_{12} \, \tau_2 \, \Big\{ & (R_{23}/R_{31}) \, [1 - \exp\,(-R_{3m} \, t)] \\ & - [R_{23} \, \tau_2/(R_{3m} \, \tau_2 - 1)] \, [\exp\,(-t/\tau_2) - \exp\,(-R_{3m} \, t)] \Big\}. \end{aligned} \tag{A6}$$

<u>Case B</u>: Linear-Rise Pump ($R_{12}(t) = \dot{R}_{12} \, t$, where $\dot{R}_{12}$ = constant)

The solutions of the rate equations are, in the notation of Eq. (A4),

$$N_2(t) = N_0 \, \dot{R}_{12} \, \tau_2^2 \, [(t/\tau_2) - 1 + \exp(-t/\tau_2)] \tag{A7}$$

and

$$\begin{aligned} N_3(t) = N_0 \, \dot{R}_{12} \, \tau_2^2 \, \Big\{ & (R_{23}/R_{3m}^2 \, \tau_2) \, [R_{3m} \, t - 1 + \exp(-R_{3m} \, t)] \\ & - (R_{23}/R_{3m}) \, [1 - \exp(-R_{3m} \, t)] \\ & + [R_{23} \, \tau_2/(R_{3m} \, \tau_2 - 1)] \, [\exp(-t/\tau_2) - \exp(-R_{3m} \, t)] \Big\}. \end{aligned} \tag{A8}$$

The population inversion is given by Eq. (A3).