Approaches to the development of gamma-ray lasers

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The interdisciplinary theoretical and practical problems involved in the development of devices analogous to lasers for generation of coherent radiation in the 6- to 120-keV photon energy range by stimulated emission of recoilless radiation from nuclear isomers are discussed in depth with a comprehensive bibliography.

CONTENTS

I. INTRODUCTION

A. Scope of this article

This paper reviews efforts to develop stimulated emission devices, using the Mössbauer effect in nuclear transitions, for generating coherent electromagnetic radiation in the 6- to 120-keV photon energy range. These devices are commonly called "grasers." Earlier general reviews of this subject include: Molchanov, 1972; Baldwin, 1974a, 1974b; Cohen, 1974; Baldwin and Khokhlov, 1975; Bushuev and Kuz'min, 1974; Svoren, 1975; Baldwin, Pettit, and Schwenn, 1975; Waynant and Elton, 1976; Baldwin, 1977a; Kane, 1977; Kuz'min, 1978; Gol'danskii, Kuz'min, and Namiot, 1981.

The difficulty of graser development, together with the perceived rewards of success, make it a major scientific and technological challenge. It is our hope that, by documenting what has been proposed and explaining why each proposal has fallen short of success, we will encourage others to contribute new ideas, to seek essential information now lacking, and to avoid duplication of past efforts, so that the elusive goal can be reached. Although most of this article deals with possibilities and with proposals, rather than with accomplished fact, we believe it can serve a useful purpose.

The problems of the graser are interdisciplinary. In addition to quantum electronics, they involve nuclear and optical spectroscopy, chemistry, solid-state physics, metallurgy, and in the majority of proposals—that would employ neutrons as the primary pump—the generation, moderation, and interaction of neutrons. The subject blends both basic science and engineering technology.

A review that is to cover all relevant facets of this subject cannot be addressed to specialists in any one of these fields alone. Each has its own special concepts and terminology, and few readers can be expected to be familiar with all of them. In order to spare the reader an undue amount of literature perusal outside his own specialty, as well as to provide a consistent set of symbols, nomenclature, and basic relationships, we have provided a number of appendices, each presenting relationships from a contributing discipline that are essential to an understanding of the main text. With this structure we can discuss the history, nature, and difficulties attending each of the graser concepts without digression.

Definitions of those symbols that are used consistently throughout the text, and units employed for dimensional quantities and conversion factors are given in Appendix A.

B. The challenge of short wavelength

The laser has revolutionized optics. Many other fields of science and technology, previously unrelated, have benefited from the use of coherent radiation in the infrared, visible, and ultraviolet regions of the spectrum. The development of lasers that would generate radiation at still shorter wavelengths, comparable with atomic dimensions, can likewise be expected to lead to a vast array of new scientific and technological developments. The unique characteristics of x and gamma radiation (viz., penetration, ionizing ability, short wavelength, interaction with electrons in inner shells of atoms and with nuclei) would be greatly enhanced by the properties (coherence, intensity, monochromaticity, directionality, time and frequency dependence, etc.) that distinguish radiations generated by stimulated emission.

Despite these incentives and numerous proposals, attempts extended over two decades have not yet succeeded in developing such a laser. More than one study, in fact, concluded that the goal is unattainable. Nevertheless, new proposals continue to appear. Figure 1 charts the 150-year history of development

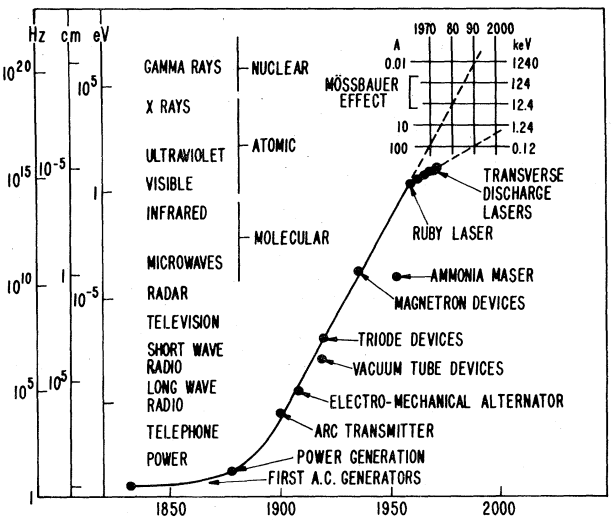

FIG. 1. Development of sources of coherent electromagnetic radiation. Chronology, illustrating the trend toward shorter wavelengths. From Nagel (1974), with addition.

of coherent sources for electromagnetic radiation (Nagel, 1974), characterized by an inexorable trend toward ever-increasing frequencies. This is actually an envelope of many distinct sigmoid curves, describing the history of a great diversity of techniques. Electric alternators, vacuum tube oscillators, magnetrons, transistors, and gas lasers have little in common. Each type of device is limited to a small spectral range and its history therefore shows saturation. New techniques, sometimes entirely new principles, each appropriate to the next spectral range, have nevertheless contributed steady progress toward ever-higher frequency exhibited by this curve.

The family of lasers based on Fabry-Perot resonators indeed appears to show saturation. A new principle must be found if further advances are to be made toward higher frequencies. Figure 1 suggests that the Mössbauer effect (see, for example, Frauenfelder, 1962; also Appendix D) might provide one.

Einstein's prediction that induced transitions must occur in quantum systems was based on a purely thermodynamic argument, of general application (Einstein, 1917). His derivation of

$$A/B = 8\pi h/\Lambda^3 , \tag{1}$$

the ratio of coefficients of probability for spontaneous (A) and induced (B) transitions between a pair of energy states, makes no reference to the type of system, other than that it can interact with electromagnetic radiation of the wavelength Λ. This ratio, however, increases with the third power of the frequency (transition energy), so that spontaneous emission becomes an ever-stronger competitor to stimulated emission as the wavelength is reduced. Although it is more difficult to achieve at shorter wavelengths, amplification by stimulated emission is possible in principle at any wavelength for which photon losses are not too great, and it has been shown (Baldwin, 1973b) that this condition is met in material media for all wavelengths longer than about 1 pm (photon energy less than about 1 MeV). In practice, the shortest wavelength generated by laser devices, approximately 18 nm (68 eV), is far short of this limit (Hull University group, 1980). Optical-frequency-multiplication devices also have reached only about 38 nm (33 eV) (Reintjes, She, Eckhardt, Karangelen, Andrews, and Elton, 1977).

C. General features of lasers

It is helpful, first, to enumerate the essential features of existing lasers and laserlike devices, and then to consider analogous features of the alternatives that are proposed for generating very-short-wavelength coherent radiation.

Three features common to all lasers are (1) an active medium, (2) a host, and (3) a pump. The active medium is an ensemble of identical quantum systems (e.g., atoms, ions, or molecules) that undergo the lasing transition. The host serves to confine the active medium to a definite volume and geometry. Examples include CO_2 gas in a glass tube, Cr^{+++} in a sapphire crystal, and Nd^{+++} in a glass rod. The pump supplies energy to excite the active medium to an inverted condition (e.g., a flash lamp for Nd-glass and ruby lasers, an electron gun for CO_2 lasers, and the energy of chemical reaction for HF lasers). Often, but not invariably, two additional features are provided: (1) a resonator and (2) a Q-switch. The resonator reduces the number of possible modes of oscillation and improves the efficiency with which energy is extracted from the active medium. The Q-switch is a means for holding off the onset of oscillation until the pump has created a high degree of excitation, in order to achieve short, intense pulses.

The active medium is selected on the basis of (1) transition energy; (2) excited-state lifetime; (3) availability of a mechanism by which it can be excited into an amplifying state; (4) ease of stimulation; (5) transparency at the operating frequency; (6) compatibility with the host; and (7) availability of a suitable pump.

The excitation requirements for a laser become increasingly difficult to satisfy as one goes to shorter wavelengths. Not only do the states to be pumped have higher energy, but their lifetimes tend to decrease (for a given transition type) and the ability to absorb an exciting radiation usually diminishes, reducing the efficiency of pumping. Because of spontaneous deexcitation, the pumping power needed to maintain inversion must be increased at least as fast as the product of transition energy by spontaneous decay rate, i.e., as the fourth power of frequency. The temperature rise accompanying an intense pump and the recoil associated with both emission and absorption of energetic photons can further reduce the stimulation cross section, and therefore exacerbate the excitation requirement for short-wavelength lasers.

II. ALTERNATIVE APPROACHES TO SHORT-WAVELENGTH LASERS

A. Possibilities

For the spectral range under consideration in this article, four basic approaches conceivably may lead to the development of sources for coherent electromagnetic radiation. They are distinguished by the nature of the quantum system that constitutes the elementary oscillator. We designate them as (1) bound electronic, (2) unbound electronic, (3) unbound nuclear, and (4) bound nuclear.

B. Electronic transitions

1. Bound-electron lasers

This category is based upon electronic transitions between states, at least one of which is a bound state of an atom. These would differ from existing lasers only in the magnitudes of the binding energies of the terminal laser state. The latter could include those states that in being filled give rise to characteristic x radiation. Their lifetimes are so extremely short—ranging from femtoseconds to picoseconds—that maintaining a density of excitation adequate for lasing is a major experimental obstacle.

The lifetime T of the upper laser state is the most critical consideration, since, for a given transition energy, it determines the order of magnitude of the minimum pumping power required to establish and maintain an above-inversion density of excitation. For

a pair of levels, separated by E keV, with total number density N cm^{-3}, the pumping power density for reaching inversion is at least

$$P \geq NeE/2\eta T(1-\rho), \tag{2}$$

where η is that fraction of the pumping energy that excites states from which the upper state of the laser transition can form, and ρ is the reflection coefficient for the pumping radiation. The quantum energy of the latter necessarily is higher than E, that of the transition to be stimulated. That part of the pumping power that couples to unwanted states raises the plasma temperature, broadens the x-ray line, and increases the density of parasitically absorbing species and the intensity of background radiation.

It is difficult to envisage pumping mechanisms, even photoelectric absorption or internal conversion of gamma radiation, that are sufficiently specific for efficient pumping of K-shell vacancies. The lasing medium, being subjected to enormous pumping power, would therefore be either a plasma or a beam of ions, and pumping would necessarily be pulsed and preferably of the traveling-wave type (Shipman, 1967), to avoid waste of power.

It has been estimated that for operation at $\Lambda = 1$ A, the pumping power density must exceed 1(17) W cm^{-3} (Vinogradov and Sobel'man, 1972). Such power densities are presently attainable only in nuclear explosions (Belokogne and Chapline, 1978), but should eventually be reached also in laser-driven fusion reactions (Nuckolls, Wood, Thiessen, and Zimmerman, 1972). However, no mode-selective structure is likely to withstand such enormous power densities. Stimulated emission must occur before thermal relaxation, i.e., on a femtosecond time scale. The length of coherent wave trains therefore cannot approach that presently characteristic of Mössbauer radiation, and there will be an intense background of thermal radiation.

Thus the term "x-ray laser" connotes a range of photon energies much lower than those we consider in this article, unless some transitions amenable to nuclear-explosive pumping are found (Wood, 1980).[1] For lower x-ray energies, special mechanisms (e.g., charge exchange, level crossings, recombination, excimer formation) have been suggested (see the review by Waynant and Elton, 1976) which might favor certain excited species that, because of selection rules, are relatively long lived and so could accumulate beyond the point of inversion. Some claims of amplification have, in fact, been reported. We shall not attempt to discuss or evaluate them in this paper.

2. Unbound-electron lasers

When both states of the electronic transition are in a continuum, then the side effects of the pumping power need not be so serious as they are in the bound-electron type. In some versions of unbound-electron lasers under development, no material medium is required for the elementary oscillator.

The acceleration of an electron that leads to emission of radiation can be the result of application of either microscopic or macroscopic fields. The former category may include scattering by single atoms (bremsstrahlung) or photons (inverse Compton scattering), by assemblages of atoms (crystal lattices) or large numbers of photons. The latter category includes electric and/or deflecting magnetic fields (as in linacs or synchrotrons), as well as standing-wave optical fields.

A beam of monoenergetic electrons is obviously population inverted with respect to all lower energies, and the only photon loss mechanism in a pure electron beam is scattering by the beam electrons (Kroll and McMullin, 1978). Assuming that the pumping power problem is therefore soluble, we are still faced with the problem of selecting, from a continuum of transition energies, a narrow band of frequencies within which amplification is possible.

When one particular final state of momentum and polarization is already appreciably occupied by photons, the probability of scattering a photon into that final state is enhanced, i.e., the scattering is stimulated (Heitler, 1944). However, if the photons to be scattered into the coherent beam are from a continuous manifold, such as a Planckian distribution, then, because energy and momentum must be conserved in each scattering event, only a small fraction of the distribution is available for scattering into the coherent beam. On the other hand, with a monoenergetic unidirectional source of photons (i.e., a laser), nearly every electron and every photon could have the correct energy and momentum to be scattered into the common final state desired. However, experiments in which optical laser light is scattered off electron beams have produced only small yields of incoherently scattered photons (Fiocco and Thompson, 1963), and the conversion efficiency should go as the fourth power of the wavelength of the impinging radiation until intensities can be achieved at which stimulated scattering becomes probable.

Aperiodic deflection of electrons, whether by Compton scattering, bremsstrahlung, or single passage through a deflecting field, can give rise only to a very limited number of photons, distributed over a very broad spectral range. Even the periodic motion of orbital circulation of electrons in a synchrotron gives rise to a broad band of wavelengths (Elder, Langmuir, Gurewitsch, and Pollock, 1947; Lea, 1978). In order to enhance the electrons' spontaneous emission spectrum in a narrow band at very short wavelengths, it is necessary to provide a mechanism that forces the electrons in a beam to oscillate transversely to their direction of motion at a well-defined high frequency.

For this purpose, electron beam accelerating devices, both linear and circular, have been provided with so-called "undulator," comprising arrays of alternately directed transverse or helical magnetic fields, which superimpose a sinusoidal transverse oscillation on the electron paths (Winick, Brown, Halbach, and Harris, 1981). The dominant wavelength of spontaneous radiation from the resultant oscillating electric dipoles is then equal to the Lorentz-contracted

[1] Recent reports in the nontechnical literature (Aviation Week, Feb. 23, 1981, and Laser Focus, April, 1981) suggest that this may in fact have been accomplished. The Department of Energy has not officially confirmed these reports.

spatial period of the undulator. Unless, however, the undulator is impracticably long, the total number of oscillations of an electron in transit is relatively small, so that the spontaneous bandwidth is very great. In other words, the coherence length cannot exceed, and with relativistic electrons is far less than, the length of the undulator.

Moreover, the radiation generated by such a device is incoherent unless (1) the electrons are tightly bunched and so phased that work is done on the traveling electromagnetic wave as they oscillate, rather than vice versa, or (2) the electromagnetic wave and the beam are sufficiently intense that stimulated emission is appreciable. At the electron densities typical of circular electron accelerators and storage rings (see tabulation in Lea, 1978), however, the probability of stimulated emission per unit length of beam is extremely minute.

With undulator of spatial periods approaching the practicable limit of about 1 cm, x-ray photons have been generated with electron beams of a few GeV. However, the length of a "superradiant laser" and the beam current for appreciable gain would need to be impracticably high. In the absence of means to compensate for losses, a bunched monoenergetic electron beam entering one end of such a device would become dispersed in phase and energy, losing effectiveness as it proceeded.

Thus a resonator would seem to be an essential feature of a short-wavelength unbound-electron laser. The difficulty of providing a low-loss resonator that furnishes both a closed path for Bragg-reflected radiation and a segment of that path for an intense electron beam pump is such that, to our knowledge, no one has yet succeeded in developing one, despite numerous proposals (Sec. III.D.3).

An alternative to the undulator that might also avoid the resonator problem would use the periodic fields within optical laser cavities, or, in a still more extreme case, in perfect crystals (Vysotskii, Vorontsov, and Kuz'min, 1980). Short wavelengths could then be generated with lower electron beam energies, and the effective length of interaction would be far greater than for a macroscopic undulator. In the crystal case, at least, there are severe material limitations.

Winterberg (1979a) proposes another type of unbound-electron laser, for stimulating annihilation radiation from an assemblage of positrons and electrons. He predicts that, if two counter-rotating beams in a specially-designed magnetic storage ring were made to coalesce, the magnetic pinch effect would contract their common cross section, creating a medium inverted with respect to annihilation, with particle density exceeding that of ordinary matter by many orders of magnitude. Plasma instabilities and losses by Raman or Compton scattering are not discussed.

Bertolotti and Sibilia (1979) have calculated that a critical density of 4(20) cm^{-3} is required for observable stimulated emission of coherent annihilation radiation from positron-electron pairs in a resonant cavity.

C. Nuclear lasers

This approach would be based upon transitions between stationary states of nuclear isomers (Appendix B).

Atomic or molecular transition lasers that are pumped by radiation from nuclear sources (see, for example, Miley, 1977) are often miscalled "nuclear"—these should properly be termed "nuclear-*pumped* lasers."

The majority of nuclides, both stable and radioactive, have isomeric states, many with lifetimes longer than the atomic vacancy states discussed above, ranging up to many years (Segrè and Helmholz, 1949; Goldhaber and Hill, 1952). Transition energies range from a few electron volts, in the case of ^{235}U, to several tenths of MeV (Appendix B). The proposals that we consider in this article are based upon hopes that some nuclear transitions of intermediate lifetime (nanoseconds to hours), with transition energies in the 6- to 120-keV energy range, might be readily pumped, so as to enable development of sources of coherent radiation in the wavelength range between 0.1 and 2 A. If so, then nonlinear interactions (see, for example, Baldwin, 1969) might enable the generation of coherent radiation of lower photon energy as well, so as to bridge the gap to the far ultraviolet. In radiative transitions of nuclear isomers, multipole order higher than electric dipole is the rule, rather than the exception, and for any given multipole order, the transition lifetime is several orders of magnitude longer than that of a bound-electron transition of the same energy. Moreover, nuclear reactions that can lead to isomeric states are frequently exoergic. This reduces the demands on an external source of pumping energy, but at the expense of other, equally serious difficulties.

Free (or weakly bound) nuclei receive the full momentum of recoil of gamma-ray emission. The energy of recoil must be furnished out of that available for the transition and, therefore, at the expense of an emitted photon. Because of random thermal motion of source atoms, a constant increment of momentum is not a constant increment of energy, so that the gamma-ray spectrum is both shifted and broadened (Appendix E). This has a disastrous effect on the probability of induced transition (Appendix C), since it reduces the energy density per unit frequency interval U_ν of the inducing radiation. The Doppler-recoil breadth greatly exceeds the natural linebreadth, except for transitions so short lived that no advantage of pumping power would be offered by using nuclear, rather than x-ray transitions.

When nuclear isomers are bound in solids, the recoil momentum of gamma-ray emission is imparted to the remainder of the lattice (Appendix D) and, in a definite fraction of all emission events, the quantum state of the lattice vibrations is not affected by this impulse. For those emission events, the emitted radiation is confined within a spectral band much narrower than the Doppler-recoil breadth of unbound nuclei. The energy density per unit frequency interval U_ν, which, when multiplied by the Einstein B coefficient [Eq. (1)], gives the induced transition rate, is therefore anomalously high for a given total intensity of the inducing radiation. With isomers of sufficiently long lifetime, then, one might hope to achieve conditions that permit stimulated emission without excessively high pumping power. The product of linebreadth by

transition lifetime is the all-important consideration that determines the feasibility of the bound-nuclear approach, to which the following sections are chiefly addressed.

III. POSSIBILITIES

A. The cross section for stimulated emission

Einstein (1917) showed that the induced transition rate in a radiation field of spectral density U_ν is related to the spontaneous radiation rate coefficient A s^{-1} by Eq. (1), and therefore is

$$BU_\nu = (\Lambda^3/8\pi h)AU_\nu . \tag{3}$$

This can be recast into the form

$$B U_\nu = (\Lambda^2/2\pi)(A/\Delta\nu)(nc/4) \tag{4}$$

by replacing the energy density per unit frequency U_ν by the product of photon energy and photon density nE divided by the total bandwidth $\Delta\nu$. This form is recognizable by nuclear physicists as the product of a flux density of photons, $nc/4$, by an area, i.e., a cross section, that is inversely proportional to the bandwidth $\Delta\nu$.

In Appendix C we deduce the following generally applicable formula for the induced transition cross section,

$$\sigma_s = (\Lambda^2/2\pi)(\Gamma_r/\Gamma) , \tag{5}$$

where Γ_r is the radiative width, corresponding, except for a factor $\hbar$ when energy units are used (see Appendix A), to Einstein's A coefficient, and Γ is the total linewidth expressed as an angular frequency. It is customary to express widths of nuclear energy levels in energy units rather than as frequencies; the important quantity is the dimensionless ratio of the partial radiative to the total width, which we refer to as the "line-broadening factor".

The other, which we term the "area factor," is greater than the nonresonant photon removal cross section, although never by more than about five orders of magnitude in the short-wavelength region, except in the case of the Borrmann effect (Appendices F and G). Figure 2 compares these cross sections.

The line-broadening factor is therefore crucial. Its maximum possible value of unity occurs only when the transition is purely radiative and to a ground state. In that case

$$\Gamma = \Gamma_r = 1/T_2 . \tag{6}$$

If the transition is purely radiative, but the lower state is not stable,

$$\Gamma = T_2^{-1} + T_1^{-1} = (T_2 + T_1)/T_2T_1 . \tag{7}$$

When there are alternative modes of decay, the total, but not the radiative, width is increased; in particular, with internal conversion competing with radiative transitions from level 2 to level 1, and, in addition, with branching from level 2 to a different final state, then

$$\Gamma_r = \beta/T_2(1+\alpha) \tag{8}$$

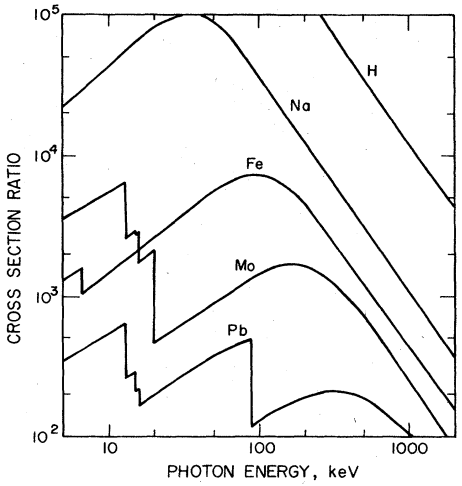

FIG. 2. Resonant versus nonresonant photon interactions. The ratio of the maximum possible resonant cross section of a purely radiative transition to the total nonresonant photon removal cross section is plotted as a function of photon energy for some representative elements.

and

$$\Gamma_r/\Gamma = [\beta/(1+\alpha)][T_1/(T_2+T_1)] . \tag{9}$$

Figure 3 illustrates this most general case.

If there are no other contributors to the linebreadth, then we say that the line is "homogeneously broadened" by branching, internal conversion, and lower-state decay, but that it is of "natural" width.

Several other effects that further reduce the cross section by decreasing the effective value of the linewidth factor are described in Appendix E.

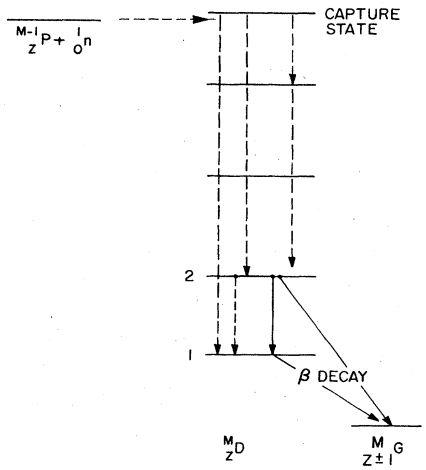

FIG. 3. Typical energy levels of an isomeric nuclide formed by neutron capture. The parent nuclide $^{M-1}_{Z}P$ captures a neutron to form the daughter nuclide $^{M}_{Z}D$ with emission of gamma radiation. In this case, the lowest state 1 is beta unstable, decaying to form the ground state of $^{M}_{Z+1}G$. In many cases, including all "Mössbauer isomers," the ground state of G lies above levels 1 and 2 of D, and the arrows indicating beta decay should be reversed.

B. Doppler-recoil broadening

Because of random thermal motion of molecules and recoil of the nucleus from photon emission, the emission spectrum is both shifted, on the average by (Appendix D)

$$R = 5.32(-4)E^2/M \quad \text{eV}, \tag{10}$$

and broadened by [Appendix E, Eq. (E10)]

$$\Gamma_D = 3.3(Rk\theta)^{1/2} \quad \text{eV}. \tag{11}$$

This, called the "Doppler-recoil," or, for brevity, the "Doppler breadth," reduces to the product of photon energy by the ratio of the rms thermal velocity in the direction of emission to the velocity of light. Thus the line's profile resembles the distribution of first-order Doppler shifts.

In the optical region, the recoil shift R is small compared with the Doppler-recoil width, and can be neglected. In the short-wavelength region, however, recoil shift can exceed the Doppler width. It displaces the emission line to lower, the absorption line to higher energy, so that the two do not necessarily overlap (Fig. 4).

For example, in the emission of a 30-keV photon by a nucleus of mass number 100, the recoil shift is 4.78(−3) eV. At room temperature, $k\theta = 2.6(-2)$ eV; at 4 K, it is 3.6(−4) eV. The full Doppler widths are, respectively, 3.71(−2) and 4.38(−3) eV. Thus at the lower of these temperatures the recoil shift exceeds the Doppler width of the line. The degree of overlap is a function of the temperature.

Collins, Olariu, Petrascu, and Popescu (1979a, 1979b) have suggested that all or part of the energy shift accompanying gamma recoil momentum might be compensated by simultaneous absorption or emission of photons from a strong laser-generated optical-frequency field, thereby enabling one to tune the radiation. Their estimate of the required laser intensity assumes the gamma-ray spectrum to be of natural width; use of the Doppler breadth would increase the requirement by many orders of magnitude.

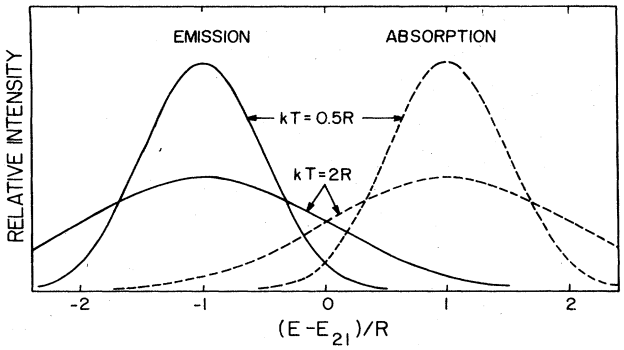

FIG. 4. Doppler-recoil-broadened distributions of gamma-ray energies. The solid curves show the emission spectrum, the dashed curves the absorption spectrum for nuclei in a gas with an isomeric transition energy E_{21}. Two sets of curves are shown, one for thermal energy equal to half the recoil energy, the other for thermal energy twice the recoil energy. Note the decreasing overlap of emission and absorption bands with lowered temperature.

C. Non-Mössbauer grasers

The fact that, at sufficiently low temperatures, resonance absorption and emission lines do not overlap, has been suggested as enabling development of stimulated emission devices that would not require population inversion (Marcuse, 1963).

It has also been suggested (Wood and Chapline, 1974) that if a plasma were "vigorously pumped" with fast neutrons from a laser-ignited thermonuclear reaction, excited nuclear states, with lifetimes of the order of picoseconds—longer than atomic vacancy states but considerably shorter than Mössbauer isomers—might accumulate inverted populations capable of lasing. It was noted that, in a fully ionized plasma, photoelectric absorption cannot occur and that, for such short-lived states, the Doppler breadth is comparable with the natural breadth. However, it has been shown (Gol'danskii and Namiot, 1975, 1976a) that even with unlimited pumping power this concept is not feasible. The plasma cannot be transparent to the radiation it is to amplify, owing to photon attenuation by plasma electrons (Thomson scattering, inverse bremsstrahlung).

For a graser, the essential factors are not the absolute magnitudes of linebreadths or of cross sections, but ratios: the ratio of radiative width Γ_γ to total width Γ, and the ratio of $\Lambda^2/2\pi$ to the total photon removal cross section σ_a. These ratios are favorable only in the case of recoilless gamma radiation, for which they are separated only by factors of the order 1(4) or less. The remainder of this article is therefore devoted entirely to the possibility that grasers might be based on the Mössbauer effect.

D. Recoilless transitions

1. The Mössbauer effect

For isomers in solids that show the Mössbauer effect (Appendix D), the gamma-ray emission spectrum exhibits a sharp component superimposed upon the Doppler-broadened background. Often this "recoilless" radiation is essentially of natural width, relatively intense, and only slightly, if at all, displaced from the energy of the nuclear transition. It is manifested by strong resonance absorption of radiation from excited states in one solid by ground states of the same nuclide in a second solid (Mössbauer, 1958), and its spectral distribution can be determined with the aid of the first-order Doppler effect, by imparting adjustable relative line-of-sight velocity to the two solids and observing the variation of transmitted intensity. As the resonance condition is reached, the transmission of the second solid for the recoilless component often decreases markedly, demonstrating both a high recoilless fraction and a high ratio of the resonant to nonresonant interaction cross section, and therefore of both of the factors of Eq. (5). Were the second solid pumped to a state of inversion, gain, rather than attenuation of the resonant radiation, would then be observed.

Table I lists a number of isotopes that emit recoilless radiation when in solid hosts.

The fraction of the total radiation emitted within the

TABLE I. Typical recoilless gamma transitions.[a] EC denotes orbital electron capture.

	Isomeric transition			Mössbauer parameters				Photon cross sections, b		Production reaction		
Nuclide	Transition energy	Half-life	Type	Internal conversion coefficient	Recoil energy meV	Debye[b] temp. K	No-phonon fraction $f(T=0)$	Removal[c]	Resonant[d]	Type	Parent	Half-life
^{57}Fe	14.4	97.8 ns	$M1$	8.2	2.0	490	0.93	5.5(3)	1.2(6)	EC	^{57}Co	267 d
	136.5	8.7 ns	$E2$	0.14	175		0.002	1.9(1)	2.3(2)	β	^{57}Mn	1.6 m
^{67}Zn	93.3	9.3 μs	$E2$	0.89	70	327	0.024	5.9(1)	3.6(3)	EC	^{67}Ga	78 h
										β	^{67}Cu	59 h
^{73}Ge	13.3	2.75 μs	$E2$	1100	1.3	370	0.94	1.5(4)	1.2(4)	EC	^{73}As	80 d[e]
	68.3	2.9 μs	$M1$	0.8	34		0.19	1.7(2)	5.6(4)	β	^{83}Ga	49 h
^{83}Kr	9.4	147 ns	$M1$	19.6	0.57	72	0.87	8.5(3)	1.2(6)	EC	^{83}Rb	86 d
^{107}Ag[f]	93.1	44.3 s	$E3$	20	43	225	0.035	3.2(2)	4.6(2)	EC	^{107}Cd	6.7 h
^{119}Sn	23.9	17.9 ns	$M1$	5.2	2.6	210	0.81	2.6(3)	5.6(5)	EC	^{119}Sb	38 h
^{151}Eu	21.5	9.7 ns	$M1$	28.6	1.66	195	0.86		1.6(5)	EC	^{151}Gd	120 d
										β	^{151}Sm	90 y
^{155}Gd	60.0	0.19 ns	$M1+E2$	8.7	12		0.33	3.0(3)	2.3(4)	EC	^{155}Tb	5.6 d
	86.5	6.3 ns	$E1$	0.43	26	195	0.10	1.2(3)	2.2(4)	β	^{155}Eu	5.0 y
	105.3	1.2 ns	$E1$	0.26	38		0.032	7.6(2)	5.6(3)			
^{161}Dy	25.7	28.2 ns	$E1$	2.9	2.2		0.83	6.5(3)	7.9(5)	β	^{161}Tb	6.9 d
	43.8	0.8 ns	$M2$	4.3	6.4	210	0.59	1.6(3)	1.4(5)	EC	^{161}Ho	2.4 h
	74.6	3.3 ns	$E1$	0.65	19		0.22	1.9(3)	5.7(4)			
^{181}Ta	6.2	6.8 μs	$E1$	82	0.11	240	0.99	9.7(4)	7.6(5)	EC	^{181}W	121 d
	136.3	40 ps	$M1+E2$	1.8	55		0.02	5.6(2)	8.6(2)	β	^{181}Hf	42 d
^{191}Ir	82.4	4.0 ns	$M1$	10.7	19	420	0.45	2.6(3)	1.4(4)	β	^{191}Os	15.4 d
	129.4	89 ps	$M1$	2.9	47		0.14	7.8(2)	5.3(3)	EC	^{191}Pt	2.8 d
^{237}Np	59.5	68.2 ns	$E1$	1.1	8.0	200	0.50	2.8(3)	1.6(5)	β	^{237}Pu	6.8 d
										EC	^{237}U	4.5 d
											^{241}Am	460 y

[a] Data from Stevens and Stevens (1976), except as noted.
[b] Assumed effective Debye temperature, usually that for the pure element. Most of these values are taken from the AIP Handbook, pp. 4–115–6.
[c] For the pure element as an undiluted host. Photon removal cross section from Storm and Israel (1970).
[d] Assuming a temperature of 4.2 K and the natural linewidth. No statistical weight factor is included.
[e] Pfeiffer (1977).
[f] Bizina, Beda, Burgov, and Davydov (1964).

recoilless component (and, therefore, capable of interacting resonantly in the second solid with an appreciable cross section) is closely related to the Debye-Waller factor of x-ray diffraction theory, which has the theoretical value

$$f = \exp(-4\pi^2\langle x\rangle^2/\Lambda^2) \tag{12}$$

and reduces to

$$f = \exp\left\{-\frac{3}{2}\frac{R}{k\Theta}\left[1+\frac{2}{3}\left(\frac{\pi\theta}{\Theta}\right)^2\right]\right\} \tag{13}$$

for a Debye solid of characteristic temperature Θ at temperature θ. The radiation width of Eq. (8) and, therefore, the resonant cross section must be multiplied by the recoilless fraction.

Close examination of the dependence of the transmission of a resonant solid for recoilless radiation often reveals the gamma-ray line to be shifted in position, split into several components, and broadened. Although the width of a broadened Mössbauer line always is still far less than the Doppler breadth at the ambient temperature, it can substantially exceed the natural breadth, and usually does in the case of long-lived (microseconds or more) transitions. The study of line-perturbing effects constitutes the remarkably precise field of Mössbauer spectroscopy, with which a considerable body of knowledge of the internal structure of solids has been acquired (Frauenfelder, 1962; Shenoy and Wagner, 1978). In the graser problem, however, these same effects have serious adverse consequences. We are particularly concerned with the line-broadening interactions. These are described in Appendix E.

2. Possibility of gain

Photons in the recoilless component can have a high probability of undergoing resonant interaction in solids while traversing distances in which the probability of their nonresonant removal is small. However, if there is to be net amplification, the gain by induced transitions must exceed removal by both nonresonant and resonant absorption. This is called the "photon

balance condition."

Stated quantitatively, the photon balance relation is

$$[N_2-(g_2/g_1)N_1]\sigma_s(E_{21})\geq N_0\sigma_a(E_{21}) \quad (14)$$

for a transition from excited state 2 to terminal state 1. Thus the ratio of the degeneracy-weighted population difference factor on the left, termed the inversion density, to the total population density of all atoms in the medium, N_0, must be not merely positive, but must exceed the ratio of nonresonant to resonant cross sections.

This is a necessary, but still not a sufficient, condition for a graser. Not only must stimulation add photons more rapidly than absorption removes them, but, if the stimulated photons are to be observable, there is a minimum requirement for the excess of gain over removal (Appendix H). Moreover, the time dependences of the inversion density and of the cross section (Appendices C and I) both are such as to increase the inversion density requirement, the former by decaying and the latter by growing asymptotically from zero to the value given by Eq. (5).

3. Possibilities for resonators

In principle, Bragg diffraction might offer a basis for developing a short-wavelength resonator. Where enormous pumping power requirements exist, as in the electron-transition and short-lived nuclear alternatives, we see little hope for such resonators. However, if nuclear isomers of lifetimes sufficiently long that they can be separately pumped and then assembled into perfect solids can be made to lase, then there could be considerable benefit from Bragg diffraction, even though the photon trajectories were not closed. Nonresonant interaction with atoms at regular lattice sites is greatly reduced for those radiation-field modes that satisfy the Bragg condition in a perfect solid (Appendix G). For them, the photon balance condition would be far less stringent than for gain in any other directions, so that these modes would be the first to lase.

The quality factor of a perfect crystal for Bragg modes has been estimated to approach 10^8 (Rivlin, 1963b).

The anomalous transmission of Bragg-directed modes is extremely sensitive to the presence of lattice defects (Borrmann, Hartwig, and Irmler, 1958), interstitial atoms (Patel and Batterman, 1963), and temperature. It is therefore imperative, if the effect is to be useful, that lattice damage in the pumping process, in crystal growth or doping, and in the subsequent radioactive decay be minimized or rapidly repaired; hope for the latter is offered by recent developments in laser annealing of thin solid layers (White, Narayan, and Young, 1979).

In the absence of a beam-defining resonator, the graser solid can be prepared as a thin filament of great length-to-diameter ratio; spontaneous emission directed along the major axis would then receive maximum amplification, so that a beam would be defined by the filament geometry (Baldwin, Neissel, and Tonks, 1963). Such a geometry has other advantages (Gol'danskii and Kagan, 1973a). The frequency bandwidth of the amplified radiation would be narrower than that of the spontaneous line, since the greatest gain is at the line center. If the spontaneous linewidth can also be nearly the natural width of the recoilless transition (i.e., negligible broadening), then it has been shown (Chirikov, 1963; Vorontsov and Vysotskii, 1974a; Baldwin and Suydam, 1977) that the group velocity of the developing wave is so low that a resonator would be unnecessary. The use of needlelike single crystals in a metallic matrix, with the nuclei aligned by internal field gradients, has been proposed as a means of enhancing directionality of emission (Jha and Blue, 1976).

Some other specific proposals for graser geometry will be described in Sec. V.

4. Possibility of inversion

Inverted populations are not uncommon in nuclear species. There are examples of nuclear reactions and of radioactive decay chains in which inversion results from favorable relationships among production rates and half-lives for spontaneous decay. In neutron capture, the initial event is formation of a highly excited daughter that decays by a chain of gamma-ray transitions, in which isomeric states (Appendix B) of relatively long lifetime can form. Inversions then can occur with respect to lower states.

The isolation of radioactive nuclides is a commonplace operation of nuclear chemistry, often carried out in the course of production reactions. Some isotopically selective reactions could be used for separating isomers as well. For example, the breaking of chemical bonds by reaction recoil, which has been employed for isotope separation (Szilard and Chalmers, 1934), might be used to prepare material enriched in an isomer (Sharpe and Schmitt, 1959), if the reaction has a high isomer ratio (Appendix B.4) and involves enough recoil impulse to break a chemical bond. When an active isotope has a chemically distinct radioactive parent, inversion can be achieved by a sequence of two or more chemical separations, separated by about one half-life (Vali and Vali, 1963a).

Isotopically selective photochemical techniques (Billings, Hitchcock, and Zelikoff, 1953; Gunning and Strausz, 1963; Liuti, Dondes, and Harteck, 1966) have been demonstrated. Recently, chemical reactions that are isotopically selective, based on magnetic isotope effects, have been reported (Podoplelov, Leshina, Sagdeev, Molin, and Gol'danskii, 1979), and it was noted that they might be isomerically selective as well. Spectroscopic features are known that are isomer specific (Melissinos and Davis, 1959), and it has been proposed to exploit them for the concentration of isomers, by an extension of laser photochemical techniques being developed for isotope separation (Letokhov, 1973a; Baldwin, Clark, Hakala, and Reeves, 1978).

When the upper- and lower-state magnetic moments are of opposite sign, it is possible that an *effective* population inversion might be created, even though the total population of ground states exceeds that of the excited states, by polarizing the nuclei in a strong

magnetic field, since selection rules on the magnetic quantum number would then limit the emission spectrum to hyperfine components that could not be resonantly absorbed (Carlson and Temperly, 1969; Vysotskii, 1979).

Atomic beam magnetic resonance experiments have demonstrated selective focussing of nuclear isomers in either state (Appendix K).

E. Possible applications

We shall not attempt to review the full range of speculation about specific applications of grasers on a case-by-case basis. Clearly, comparability of wavelength with atomic dimensions, and the penetration and ionizing ability of gamma radiation, must inevitably lead to a variety of novel uses, not only in fields already exploited by these properties of gamma radiation, such as inspection, diagnosis, therapy, crystallography, Mössbauer spectroscopy, etc., but in other fields as well, when directionality, monochromaticity, and coherence also characterize the radiation.

The utility of a graser would depend largely upon the forms in which it might be developed. The kinds of experiments and applications that would be possible for a graser pumped by a nuclear explosive are quite different from those that would be amenable to a semicontinuous-wave laboratory device that might be realized with long-lived transitions.

Moreover, it is a fundamental human quality to prophesy on the basis of analogy with existing technology, although, after the new technology has reached maturity, it is often found that the unanticipated applications are the dominant ones. This has certainly been the case with optical lasers, for example; in 1950, who might have expected extensions of coherent radiation technology to lead to use of optical radiation in surgery, to the production of defect-free crystals, or to laser fusion? The range of applications is often dictated as much by economics, the current state of other fields, and political vogue as it is by the laws of physics.

Still, recognizing the limitations of our ability to prognosticate, we are intrigued by possibilities suggested merely by analogy to optical lasers. For example, extension of interferometric techniques and of frequency-beating methods into the gamma-ray region would greatly extend the precision of measurement, not only of dimensions, energies, frequencies, etc., but also of the fundamental natural constants.

Nonlinear effects might be as useful to nuclear science and engineering as they have proved to optical technology, and even to communication. Here we are on very unsafe ground in extrapolating even from experience.

Still more uncertain—in fact, controversial—is the possibility of gamma-ray holography, particularly of molecules. For one thing there is the practical difficulty of recording such a hologram. More fundamental, however, is the objection that molecules would inevitably be destroyed before they could scatter enough radiation to form a hologram, since coherent scattering is far less probable than photoelectric absorption in this region (see, for example, Breedlove and Trammell, 1970 and Trammell, Chapline, and Wood, 1976).

We therefore suggest that further speculation on applications be deferred, confident that many uses will justify development of the sources. Readers interested in further speculations are referred to the review article by Waynant and Elton (1976).

F. The graser dilemma

Unfortunately, properties that should enable amplification and those that would permit obtaining nuclear inversion densities sufficient for lasing are not, at present, compatible.

The time required to activate, separate, concentrate, and crystallize an appreciable number of isomeric nuclei by conventional radiochemistry is at least a few seconds; commonly it requires several hours. If inversion is to persist for such a long time, the transition lifetime must be appreciably longer.

The Mössbauer effect, on the other hand, is presently observable (by conventional methods) only in transitions of lifetime T shorter than 10 μs (Appendix D) because nuclei are perturbed by locally varying internal fields in solids (Appendix E), so that the resonances of the longer-lived transitions are randomly displaced by more than their natural linewidths. A given subset of modes of the radiation field then can interact with only a small fraction of the isomer population. In short, the resonance cross section is so reduced by line broadening, in the case of long-lived transitions, that amplification is not possible; on the other hand, the intense pumping needed to invert isomers with lifetimes in the Mössbauer range may overheat, damage, or even destroy the solid that must support recoilless emission.

The specific proposals reviewed in this article are suggestions for overcoming or avoiding these difficulties.

IV. PROBABLE FEATURES OF GRASERS

A. Isomer lifetime range

We classify the possibilities, at first without regard for their practicability, before examining some specific proposals for graser systems. The most suitable single basis of classification is according to upper-level lifetime range (Baldwin and Khokhlov, 1975). It is convenient to distinguish three slightly overlapping ranges of upper-level lifetimes for the active isomer, as follows.

I. The Mössbauer range, which for a graser (but not—at least for the present—for Mössbauer spectroscopy) we define as 1(– 8) to 1(– 3) s.

II. The intermediate range [between 1(– 4) and 1(1) s].

III. Long (1 s or more).

That these boundaries overlap slightly recognizes the possibility that more than one distinct technique of pumping or of creating conditions favorable to lasing may apply in the regions of overlap.

B. Geometry

Any graser would necessarily require a crystal host, cooled to the lowest practicable temperature, since re-

coilless emission seems to be an essential feature.

The majority of proposals have assumed the acicular form typical of a single-pass amplified-spontaneous-emission (ASE) device, since this ensures access of pump radiation, rapid removal of heat, and escape of spontaneous radiation and by-products of pumping (Gol'danskii and Kagan, 1973a), as well as a well-defined output beam (Baldwin, Neissel, and Tonks, 1963).

Alternatives and elaborations have been suggested, including

(1) Closing the photon path by total external reflection at nearly glancing incidence at a large number of surfaces (Baldwin, Terhune, and Bredt, 1965).

(2) Achieving a closed photon path by a sequence of Bragg reflections (Deslattes, 1968).

(3) Reducing the nonresonant absorption by photon channeling in crystals (Baldwin, Neissel, and Tonks, 1963).

(4) Suppressing all but a small number of Bragg modes in a perfect crystal by an unspecified technique (Terhune and Baldwin, 1965).

(5) Surrounding the graser with an isotopically distinct but crystallographically identical blanket, such that total reflection occurs at the interface, thus creating a waveguide (Baldwin, Black, Goodale, Kasch, Neissel, and Terhune, 1962).

(6) Bending a whisker crystal into a circle to achieve a closed photon path (Vali and Vali, 1963b).

(7) Surrounding the graser filament with a coaxial blanket of fissile material and that by a reflector for neutrons, and generating a traveling wave of criticality by a sequence of laser-driven implosions (Baldwin, 1974a; Douglas, 1974; Baldwin and Khokhlov, 1975). See Fig. 5.

(8) Pumping in two stages, by enclosing the graser medium in a large convertor host, within which Mössbauer radiation is generated by neutron capture and used to pump the graser by resonant absorption (Goldanskii, Kagan, and Namiot, 1973).

(9) A variant of (8) in which the graser is a tapered cylindrical shell, to ensure focussing (Baldwin and Solem, 1980).

(10) Pumping the graser with neutrons from a moving moderator, so that the thermal neutron flux is upshifted by Doppler effect (Diven, 1970; Hemmendinger, 1970) to match the energy of strong resonance capture of neutrons in the parent isotope (Bowman, 1976).

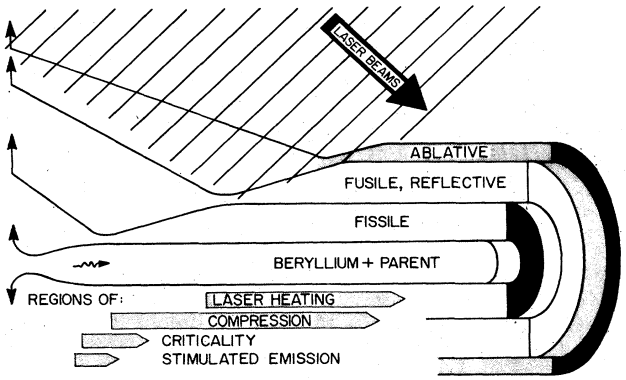

FIG. 5. Short-lifetime graser concept. This graser system would be pumped by neutrons generated in a traveling wave of laser-driven fission. The neutron-reflective material and the central parent-doped beryllium core would moderate neutrons generated in the fissile layer. The beryllium, last to be compressed, would also act as host for the Mössbauer emitter and as a flux trap for the neutrons. From Baldwin and Khokhlov (1975).

C. Minimum number of excited nuclei

Before considering various pumping reactions, it is necessary to estimate the minimum number of excited nuclei that, in an elongated filament geometry, would produce an appreciable effect of stimulated emission. [A similar estimate, but with a different condition for gain, is given by Letokhov (1973b); see Sec. V.B.4]. We invoke the condition (Allen and Peters, 1971b; Appendix H) that one net photon, in excess of those lost by nonresonant absorption and diffraction, be stimulated by a photon as it passes from one end of the laser body to the other. This condition establishes a minimum inversion density N^* for a given length L:

$$N^* \geq (N_0\sigma_a + L^{-1})/\sigma_s \,, \tag{15}$$

the total atom concentration N_0 being for undiluted graser isotope.[2] For a cylindrical body, the radius-to-length ratio is governed by the requirement of negligible diffraction loss

$$F \equiv a^2/L\Lambda \gg 1 \,; \tag{16}$$

the Fresnel number F should be at least three. In practice, the radius of the filament will be its limiting dimension. Thus the minimum volume of active graser material should be at least

$$V_{\min} = F\pi L^2\Lambda = \pi a^4/F\Lambda \tag{17}$$

and will contain $N_2 V_{\min}$ excited nuclei.

If the degree of inversion is written as

$$N^*/N_0 = (N_2 - wN_1)/N_0 = p \,, \tag{18}$$

then the excited-state concentration is

$$N_2 = N_0(w+p)/(w+1) \,, \tag{19}$$

where w is the statistical weight ratio of the two states.

Combining these relations with Eqs. (5) and (9), we find for the minimum number of excited nuclei

$$N_2 V_{\min} = \left(\frac{2\pi^2 a^2}{\Lambda^2}\right)\left(\frac{w+p}{pw+p}\right)\left(\frac{(1+\alpha)\Gamma T}{\beta f}\right)\left(1 + \frac{N_0\sigma_a a^2}{F\Lambda}\right). \tag{20}$$

The second factor accounts for less-than-complete inversion, the third for both homogeneous and inhomogeneous line broadening. The final factor gives the required excess of one photon over the number lost by nonresonant absorption. If the transition is between two excited states, then the effective lifetime in the third factor is

$$T = T_1 T_2/(T_1 + T_2) \,, \tag{21}$$

and the branching ratio β may be less than unity.

Assuming, in the most favorable conceivable case, that the radius is only 1(– 4) cm, the recoilless frac-

[2]Strictly, the coefficient of L^{-1} should be $\log_e 2 = 0.69\ldots$. The difference is trivial.

tion, statistical weight factor, and degree of inversion all are unity, and that internal conversion, line broadening, and nonresonant absorption are all negligible, then the absolute minimum number of excited nuclei would be only $1.3(7)E^2$, a very small number indeed. However, it increases rapidly when allowances are made for nonresonant absorption, internal conversion, line broadening, reduction of recoilless fraction, increased filament diameter, and appreciable gain above threshold (Letokhov, 1973b). Moreover, if the line is not appreciably broadened, then the decay of inversion during establishment of resonance must also be taken into account (Vorontsov and Vysotskii, 1974a). Still, even with generous allowances for these factors, the actual number of excited nuclei remains quite modest, of the order of 1(16) or less.

The concentration required is another matter, since the assumed volume is very small indeed. For 30-keV gamma radiation, $\Lambda = 0.41(-8)$ cm, the volume is only $3(-8)$ cm^3, and the absolute minimum concentration is therefore $4(17)$ cm^{-3}.

Because of the discontinuous dependences of photoelectric absorption (Storm and Israel, 1970) and internal conversion (Way, 1973) coefficients on photon energy and atomic number, associated with the electronic shell structure of atoms (cf. Fig. 2), a simple general statement cannot be made about the possibility of meeting the excitation requirements. For a specific case, at $E = 10$ keV in material with $Z = 50$, the nonresonant removal cross section σ_a is 2.74(4) b (Storm and Israel, 1970), and the resonance cross section is $\sigma_s = 2.45(7)(\Gamma_r/\Gamma)$ b. Therefore gain should be possible for unity statistical weight ratio when the degree of inversion lies in the range $1.12(-3)(\Gamma/\Gamma_r) < p \leq 1$. Whether or not this can be achieved depends on the total line-broadening factor and how it might be affected by the pumping process. Decay of inversion, when the line is not broadened (Vorontsov and Vysotskii, 1974a), must also be reckoned with. Thus the actual net inversion must exceed one percent of the total population by a substantial margin even in favorable cases.

The task of the pumping process is, therefore, not simply that of producing an adequate *number* of excited states but of producing them in high enough *concentration* in a solid line that *then* allows a Mössbauer (more precisely, recoilless) line. The total energy represented by even so small a total number of excited states is impressive. In the second of the above examples, the graser volume is only $8.5(-9)$ cm^3, but the 1.28(9) excited nuclei contribute 20 ergs of stored energy—a concentration of 256 J cm^{-3}.

D. Pumping methods

A graser might be pumped

(1) *in situ*, to form the active species by exposing the assembled body (parent + host) directly to a pumping radiation (Gol'danskii and Kagan, 1973a; Solem, 1979; Wood and Chapline, 1974);

(2) by separation, i.e., radiochemically (Baldwin, Neissel, and Tonks, 1963), photoelectromagnetically (Letokhov, 1973b) or mechanically (Gol'danskii and Kagan, 1973c) isolating the product of a pumping reaction, concentrating it, and then crystallizing or introducing it into a host before it has lost inversion;

(3) by a two-stage method (Gol'danskii, Kagan, and Namiot, 1973), in which a primary pumping radiation (e.g., neutrons) excites a secondary pumping radiation (e.g., resonant gamma radiation) that, in turn, generates the isomer when it reacts in an adjacent region;

(4) by radioactive decay of a parent, with a succession of chemical separations (Vali and Vali, 1963a);

(5) by hybrid methods that combine several other techniques (e.g., a sequence of neutron capture, chemical separation, ionization, acceleration, conversion in flight by appropriate reaction, and finally, deposition within a solid host).

E. Inversion mechanisms

An excitation reaction alone is of no avail without a mechanism to ensure that, when sufficiently intense, it will create an inversion above threshold. A variety of inversion mechanisms are conceivable, including:

(1) Preferential formation of the isomer in a nuclear reaction (Baldwin, Neissel, and Tonks, 1962). For example, the cross sections for thermal-neutron capture reactions $^{113}\mathrm{In}(n,\gamma)^{114}\mathrm{In}$ (72 s) and $^{113}\mathrm{In}(n,\gamma)^{114}\mathrm{In}^*$ (49 d), respectively, are 3.9 b and 4.4 b (Mughabghab and Garber, 1973). Formation of the 50-day isomer of ^{114}In is therefore 1.13 times more probable than of the lower state; the isomer ratio is 0.386. The respective half-lives, 72 s and 49 d, suggest yet another mechanism.

(2) Decay of the lower state at a rate faster than that of the upper (Baldwin, Neissel, and Tonks, 1962; Kamenev and Bonchev, 1975). In this case, homogeneous broadening of the gamma-ray line reduces the stimulation cross section. Thus the $^{114}\mathrm{In}^* \rightarrow {}^{114}\mathrm{In}$ transition cannot be stimulated (Vali and Vali, 1963b), despite the impressive degree of inversion that would exist after chemical isolation of the neutron capture product.

(3) Selective chemistry, e.g., a photochemical reaction that is isomerically selective (Letokhov, 1973a; Baldwin, Clark, Hakala, and Reeves, 1978), or a Szilard-Chalmers separation (Szilard and Chalmers, 1934) used in combination with an appropriate isomer-generating nuclear reaction (Baldwin, Neissel, and Tonks, 1962, 1966).

(4) An auxiliary induced transition from a long- to a short-lived metastable nuclear state (Eerckens, 1969; Baklanov, and Chebotyev, 1976; Rivlin, 1977a; Arad, Eliezer, and Paiss, 1979). (This suffers the disadvantage noted in case (2), above: The cross section of the auxiliary transition is diminished by homogeneous broadening.)

(5) A process inverse to internal conversion (Morita, 1973; Gol'danskii and Namiot, 1976b).

(6) Resonant absorption from a nondegenerate ground state (Gol'danskii, Kagan, and Namiot, 1973).

(7) Resonant absorption followed by branched decay from a higher to the lowest of several excited states (Baldwin and Solem, 1980).

(8) Optical hyperfine pumping into noncombining magnetic substates.

(9) Static polarization in a strong magnetic field, when the two states have $I \geq \frac{3}{2}$ and G factors of opposite sign (Carlson and Temperley, 1969; Vysotskii, 1979).

F. Pumping radiations

In principle, any radiation that can interact with a parent nuclide to form an isomeric state might serve for its pump. In practice, the pumping radiation must be (a) intense—to provide an adequate concentration of active isomer; (b) efficient—so that a negligible part of the pumping energy be expended in heating (Gol'danskii and Kagan, 1973a) or damaging (Schwenn, 1978) the host; (c) specific—so that the host not be contaminated with unwanted species that broaden resonance and increase nonresonant absorption.

The following pumping radiations have been suggested:

(1) Slow neutrons (Baldwin, Neissel, Terhune, and Tonks, 1963; Rivlin, 1963a; Gol'danskii and Kagan, 1973a; Letokhov, 1973b),

(2) Fast neutrons (Preiss, 1973),

(3) Bremsstrahlung (Marcuse, 1963; Bowman, 1976).

(4) Characteristic x radiation (Letokhov, 1973e; Vysotskii, 1979),

(5) Charged particles (Byrne, Peters, and Allen, 1974),

(6) Resonant ("Mössbauer") radiation (Gol'danskii, Kagan, and Namiot, 1973; Baldwin and Solem, 1980; Karyagin, 1980),

(7) Optical laser radiation (Eerckens, 1969; Baklanov and Chebotyev, 1975; Rivlin, 1977b; Arad, Eliezer, and Paiss, 1979),

(8) Synchrotron radiation (Dmitriev and Shuryak, 1974).

There are difficulties associated with the use of any of these radiations. Neutrons, for instance (see Appendix J), currently are available in high intensities only from fission chain reactions or explosive thermonuclear fusion reactions (laser-driven fusion may someday offer an alternative). Neither is energy efficient; in the case of fission, the neutrons have an average energy of 2 MeV and the reaction liberates approximately 133 MeV per net neutron produced. The average kinetic energy of fusion neutrons is much higher than that from fission, although the total energy cost per neutron is indeed much lower. Only a very small fraction of all the neutrons released by either process can be directed to impinge upon a small graser filament if it is not to be destroyed in the source reaction.

Inelastic neutron scattering can create isomeric states. It has been argued (Preiss, 1973) that it will favor production of isomers of high angular momentum; still, nonelastic fast neutron cross sections are seldom large in comparison with elastic scattering cross sections, which average 6–10 b. Considerable lattice damage (Schwenn, 1978) and heating by nuclear recoils (Gol'danskii and Kagan, 1973a) are inevitable accompaniments to fast neutron bombardment.

Slow neutrons, on the other hand, produce no direct material damage and have very high capture cross sections in certain nuclides, ameliorating the intensity requirement. Mössbauer isomers usually have higher capture cross sections than the parent isotopes, so that the effect of the pump on the daughter isotope must not be ignored. Gamma radiation emitted upon neutron capture, averaging 3–4 photons of about 2 MeV, generates recoils, ionization, and heating, but most of it can escape from a graser filament (Gol'danskii and Kagan, 1973a).

Unfortunately, slow neutrons can presently be obtained only by moderation of fast neutrons, and it has been shown that thermodynamic constraints on the moderation process (Appendix J; Baldwin and Solem, 1979a, 1979b) limit the density of neutrons that can be moderated to energies at which their capture becomes probable.

Bremsstrahlung (continuous x radiation) is inherently inefficient, since nuclei can respond only to those small portions of its spectrum that lie within Doppler widths of a small number of resonance lines (Appendix B.4). On the other hand, the entire spectrum interacts with electrons and atoms in the host (Appendix F), exciting, ionizing, and, therefore, heating it.

Characteristic x radiation (and nuclear gamma radiation) appears only as a finite number of lines of fixed energy. It has been noted (Kokorin and Los, 1973; Letokhov, 1973e; Vysotskii, 1979) that there are several cases for which the energy of an x-ray photon nearly agrees with that of a nuclear transition. However, a coincidence sufficiently close to permit efficient pumping seems extremely improbable, particularly because the x-ray lines are multiplets. If the energy mismatch were not too great, conceivably one might induce the nuclear transition by a combination of intense x and laser radiation, but whether an inversion could thereby be produced is not known.

Mössbauer radiation itself is an interesting exception (Secs. IV.D.3 and V.B.2.c).

The suggestion (Okamoto, 1977) that one might excite isomers by transfer of excitation from the electronic structure of an atom to its nucleus (Morita, 1973) invites the same objection as that applying to bremsstrahlung, even when the electronic excitation is created by photoelectric absorption, the only electronic excitation process other than internal conversion that is sufficiently specific in its action. There are, of course, even more serious objections to creating the electronic excitation by internal conversion of a more energetic gamma ray.

Charged particles, in addition to exciting nuclei by Coulomb interaction or transmutation, can also ionize and excite atoms, dissociate molecules, and create recoils in solid hosts. The average energy expended by an ion of energy U while creating one isomeric state with a cross section σ_x is given by

$$\Delta U = \frac{1}{N_p \sigma_x} \frac{dU}{dx} = \frac{M}{6.(23)\sigma_x} \frac{dU}{d(\rho x)} . \tag{22}$$

Assuming that the isomer formation cross section were 10 b, stopping power $dU/d(\rho x)$ near its minimum value for singly charged ions (Bethe and Ashkin, 1953) of 0.16 MeV cm^2 g^{-1}, and $M = 100$ g mol^{-1}, we estimate an energy cost of 2.7 MeV per isomer excitation event. It would be much greater if the ions were not at the (high) energy of minimum stopping power, and if the parent isotope were a diluent in a solid host.

The ion current density required to create a given density of excited states N_2 in a parent population of density N_p with a creation cross section σ_x is

$$J = (N_2/N_p)(e/T_2\sigma_x) \gg 1.6/T_2 \quad \text{A cm}^{-2} . \tag{23}$$

The population factor must not be less than $1(-4)$, owing to the photon balance condition (Sec. III.D.2), and the formation cross section cannot be much larger than 10 b. The experimental difficulties of creating the high current densities that therefore would be required to excite short-lived transitions are well known.

Several proposals have been made (Eerckens, 1969; Baklanov and Chebotyev, 1975; Rivlin, 1977b; Arad, Eliezer, and Paiss, 1979) to employ optical-frequency radiation from a laser to induce nuclear transitions from long-lived to short-lived isomeric states. However, although optical lasers can deliver energy in concentrations adequate for pumping a graser, the only known mechanisms by which energy at optical frequency has been converted to nuclear excitation proceed by first creating high local temperatures (Gol'danskii and Namiot, 1976b; Izawa and Yamanaka, 1979).

Synchrotron radiation (Elder, Langmuir, Gurewitsch, and Pollock, 1947) is becoming an important research tool for investigations in the far ultraviolet and soft x-ray regions (Lea, 1978) and, in fact, a measurable number of ^{57}Fe nuclei have been excited into the 14-keV isomeric state by resonant absorption of synchrotron radiation (Cohen, Miller, and West, 1979). However, a quantitative study of this method of graser pumping (Dmitriev and Shuryak, 1974b) concluded that no existing or presently planned synchrotron radiation source can generate adequate intensities for creating population inversion of Mössbauer isomers.

G. Time cycle and bandwidth

Although, by analogy with existing lasers, we can conceive of grasers having either continuous or pulsed outputs, the distinction is nebulous. Even with very short pumping pulses or fast Q-switching mechanisms, the duration of the amplified output pulse cannot be shorter than the reciprocal of its effective linewidth. The latter should not be much larger than the natural breadth; otherwise, amplification cannot occur because of the narrow latitude allowed by the photon balance relation.

Should it be possible to develop a method for narrowing the recoilless line of a long-lived transition nearly to its natural breadth (see Sec. V.5), the Uncertainty Principle would demand not only that this process require a time of the order of the mean lifetime of the transition (during which inversion would greatly diminish) but also that the rise time of the stimulated radiation be comparable with the mean lifetime of the isomer (Chirikov, 1963; Vorontsov and Vysotskii, 1974; Baldwin and Suydam, 1977).

Recent proposals to use nuclear alignment as a Q-switching mechanism (Wilson, 1977; Hora, Wilson, and George, 1977; Kopvillem and Choodnovsky, 1977) have not considered this aspect.

The side effects of pumping (Schwenn, 1978)—and even of the graser avalanche itself—may preclude sustained amplification by a steadily pumped graser system.

Thus a graser will almost certainly be a single-pulse system, but that "pulse" may be of appreciable duration.

H. Coherence of output

The term "coherence," applied to a beam of laser radiation, implies the presence of many photons within a very few modes of the radiation field (Scully and Jacobs, 1970). In the gamma-ray region, the density of electromagnetic modes is so enormous that this can never result from pure chance. Either stimulated emission or cooperative spontaneous emission is essential, and even then, without a highly selective resonator, the degree of coherence will be limited.

Nevertheless, recoilless gamma radiation is characterized by a significant degree of temporal coherence. The wave trains from individual transitions are of sufficient duration that they can be made to give rise to observable interference phenomena, under appropriate conditions [see Frauenfelder (1962), Sec. 5-8], and they can be modulated in frequency (Ruby and Bolef, 1960; Abragam, 1960), phase (Grodzins and Phillips, 1961), and amplitude (Isaak and Preikschak, 1972; Ruby, Preston, Skov, and Zabransky, 1973; Hauser, Neuwirth, and Thiesen, 1974).

The individual spontaneous emission events of nuclei within a Mössbauer source are uncorrelated, and their resultant wave trains are mutually incoherent. The same would be true of separate photon avalanches that might result from amplified spontaneous emission. Still, increased intensity, possibility of control, directional and temporal dependence, when added to the known properties of Mössbauer radiation, with even limited coherence, would have scientific value.

If it were to prove possible eventually to realize a true superradiant state (Terhune and Baldwin, 1965; Afanas'ev and Kagan, 1965a; Hung, 1980) in a pumped graser medium, i.e., one in which the individual nuclear transition currents are appropriately phased for cooperative emission, in the sense first described by Dicke (1954), then the limitations of coherence that we have just noted would be greatly relaxed, and the scientific value of the radiation generated would be correspondingly enhanced.

An analysis by Andre'ev (1977) of a single-pass mirrorless graser system that considered the possibility of Dicke superradiance concluded (Andre'ev, Il'inskii, and Khokhlov, 1977) that collective spontaneous emission rather than stimulated emission must play the main role in the generation of coherent Mössbauer radiation.

V. SPECIFIC PROPOSALS FOR GRASER SYSTEMS

A. Early concepts

Proposals for graser systems constitute two main groups. As has often happened in the history of science, the idea that a graser might be based upon the Mössbauer effect occurred independently and to many widely separated individuals. Several Russian and American groups are known to have made independent investigations, resulting in the nearly simultaneous appearance in 1963 of several publications. All discussed, but with slightly different emphases and varying assumptions difficulties in what had at first seemed an obvious solution to a straightforward problem. The long

delay between concepts (1960) and the first open publications (1963) is evidence of concern that the applications might include military uses.

Two of the early American studies (Babcock, Ruby, and Epstein, 1963; Podolsky, Mize, and Carpenter, 1963) did not publish in the open literature, but issued reports with limited distributions. The General Electric group published two notes (Baldwin, Neissel, and Tonks, 1963; Baldwin, Neissel, Terhune, and Tonks, 1963), together with detailed internal reports (Baldwin, Neissel, and Tonks, 1962; Baldwin, Black, Goodale, Kasch, Neissel, and Terhune, 1962), and an application for patent, which was issued in 1966. Others (Eerckens, 1969; Pickenbrock and Tibbals, 1971) applied for patents, issued only several years later.

Similarly, the first Soviet work on the subject was reported in patent applications (Rivlin, 1961). Open publication came later (Rivlin, 1963b) and the patent was issued only recently (Rivlin, 1979). Soviet publications commonly cite the patent applications rather than the accessible references.

All of the early studies envisaged chain reactions of induced transitions in radiochemically isolated isomeric nuclei of long lifetime, for which the pumping power is reduced to practically attainable levels. Stimulated radiation was to be generated at energies up to 100 keV by means of the Mössbauer effect.

Rivlin (1963a) pointed out also that Bragg reflections in perfect crystals might be the basis for developing resonant hosts with quality factors of the order of 1(8). Using photon balance (Sec. III.D.2), he derived a start-oscillation condition (Rivlin, 1963b) and combined it with rate equations for the excited-state population density, with a steady external source of radiation to serve as the pump, and showed that at least 1(5) kW of pumping power would be needed in a crystal of the order of 1 mm active dimensions. He noted that this power density could be obtained from nuclear reactions, including neutron capture, if the pumping were impulsive. Lattice damage might prove unacceptable, unless one could separate the pumping and lasing processes. If the separation were to be by a chemical procedure, then the isomer lifetime must be long, so that the gamma-ray line would be very narrow. This might demand methods for control of linewidth. Major problems were identified, but specific methods for dealing with them were not given.

Chirikov (1963) called attention to the time-dependent character of the stimulation cross section. In a laser that would operate on a line of nearly natural width, he noted, the radiation would develop very slowly, during a time of the order of the transition lifetime, and the group velocity of the wave would be so extremely slow that a resonator would be ineffectual (Fig. 6). However, he emphasized that this conclusion would apply only if an essentially unbroadened Mössbauer line could be obtained in a long-lived transition. He assumed that short-lived states cannot be pumped to inversion without inhibiting the Mössbauer effect.

That no one had succeeded in observing the Mössbauer effect in transitions of lifetime longer than about 10 μs was cited by the Westinghouse group (Babcock, Ruby, and Epstein, 1963) as demonstrating inevitable

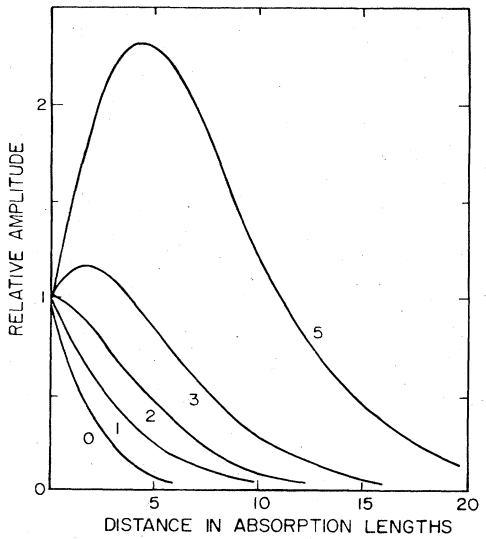

FIG. 6. Kinetics of induced Mössbauer radiation. The dependence of wave amplitude on distance from an initiating spontaneous source in a fully inverted medium is shown at a sequence of times in seconds (numbers affixed to the curves) after initiation, for a transition of 1(4) s mean lifetime. From Chirikov (1963).

broadening, of the order of 1(5) s^{-1}, that would make stimulated emission utterly unobservable in long-lived transitions. They assumed that, despite broadening, each successive member of a chain of stimulated emission events must involve a time delay of one mean lifetime, on the average, and that the stimulated radiation would be emitted isotropically. These premises led to extremely discouraging conclusions, culminating in the concluding statement that "further research on this subject would be fruitless."

Although time dependence in resonance absorption (Appendix C) had been observed experimentally (Lynch, Holland, and Hamermesh, 1960), all the other studies employed simple, constant cross section, photon-chain-reaction models, often in direct analogy with fission reactors, ignoring the time-dependent aspect.

The AVCO group reached no definite conclusions as to feasibility of any approach to gamma-ray lasers (Podolsky, Mize, and Carpenter, 1963).

Vali and Vali (1963a), reasoning that induced emission could be manifested by shortening of the apparent mean lifetime of an isomer, proposed to prepare a carrier-free sample of ^{103m}Rh ($E=40$ keV, $T=4550$ s, $\alpha=400$) by performing two successive chemical separations of Rh from ^{103}Ru ($T=5(6)$ s) and crystallizing the second sample as Rh metal, $\Theta=350$ K. Their discussion of the causes of line broadening in the Rh listed (1) chemical shift due to concentration fluctuations, (2) chemical shifts due to crystal defects, (3) gravitational red shift, (4) second-order Doppler shift. (These and other effects are treated in Appendix E.) They concluded that if one could produce a defect-free crystal with excited-state concentration uniform to within 200 ppm, at an average temperature of 4 K uniform to within 1.3(−9) K, then, by averaging out the spin-spin interaction through radio-frequency modulation of the Mössbauer

line, one might expect to observe a shortening of the mean lifetime by one percent and a detectable number of two-photon coincidence counts in the horizontal plane.

Their analysis of the lifetime reduction overlooked two important factors: (1) It implicitly assumed that the induced emission is isotropic and instantaneous. (2) Although they asserted that the gravitational shift would permit induced emission only in the horizontal plane, the minute solid angle within which emitted photons could still remain resonant was not considered in their calculation. The isotropic-emission assumption led to an underestimate of the effect of photon leakage from the crystal and, therefore, to an overestimate of the lifetime reduction.

Assuming instantaneous stimulation, they proposed that two-photon coincidences could confirm the lifetime reduction measurement. For this, they suggested a counting rate of 100 s^{-1} and a resolving time of $1(-7)$ s. The former would require only an extremely minute amount of material if the excited-state concentration were as high as they hoped to achieve, 0.25 of the total atom density. Their analysis suggested that, with careful technique, they might approach to within about two orders of magnitude of the natural width. However, since the stimulation process is not instantaneous when the line is not unduly broadened, instead of absolute time coincidence, there would be time delays of the order of minutes.

The General Electric (GE) group (Baldwin, Neissel, and Tonks, 1962, 1963; Baldwin, Black, Goodale, Kasch, Neissel, and Terhune 1962; Baldwin, Neissel, Terhune, and Tonks, 1963) also did not recognize the kinetic character of the stimulation cross section, but they did realize that stimulated emission occurs into the same mode as the stimulating radiation, so that the photon avalanche is therefore characterized by linear transport, rather than by diffusion (Baldwin, Neissel, and Tonks, 1963). Thus any analogy to fission chain reactors is misleading. Rather, the directionality would enable a beam to be formed by disposing the active medium in the form of a long filament; in particular, they suggested that whisker crystals be used for this purpose, grown rapidly from a vapor after chemical separation of neutron-bombarded material. Their analysis dealt with photon avalanches, each initiated by a randomly located spontaneous emission event, propagating at the velocity of light in the excited medium and multiplying by stimulated emission.

Lacking experimental evidence on line broadening, they proposed (Baldwin, Black, Goodale, Neissel, Kasch, and Terhune, 1962) to look for stimulated emission in the long-lived isomer ^{69}Zn ($E = 439$ keV, $T_2 = 13.0$ h, $T_1 = 58$ m), selected because of its ease of production, inversion maintained by decay, producibility by a Szilard-Chalmers separation during neutron bombardment, and ease of whisker crystal growth in zinc vapor. Mistakenly assuming that the induced and inducing photons would be time coincident, they predicted that one should observe, using a total-absorption gamma-ray detector, counts of photons apparently of twice the transition energy, emitted only in the direction of the long axis of the crystal, and only when aligned horizontally, so as to eliminate gravitational red shift. This proposed experiment was abandoned after (1) even optimistic estimates of the linebreadth and Debye-Waller factors demonstrated that resonance cannot be observed with this transition and (2) a report appeared of a direct measurement of linewidth in ^{107}Ag (Bizina, Beda, Burgov, and Davydov, 1963) which confirmed the existence of unacceptable broadening in long-lived transitions.

The GE group was the first to call attention to the significance of the Bormann effect (Bormann, 1941; Appendix G) and to the possibility that a superradiant state (Dicke, 1954) might form by spontaneous decay in a crystal prepared in an initial state of complete inversion (Terhune and Baldwin, 1965). Another suggestion (1963a, 1963b), that discrete but recoil-displaced Mössbauer lines might exist, for which population inversion would not be necessary, was shown to be untenable because of short phonon lifetimes (Vali and Vali, 1963b).

B. Recent proposals

1. Revival of interest

Between 1965 and 1972, no reports of new attacks on the graser problem appeared in the open literature, although several patents (Baldwin, Neissel, and Tonks, 1966; Vali and Vali, 1966; Eerckens, 1969; Pickenbrock and Tibbals, 1971) were granted. In 1972, Khokhlov, noting that recent developments in nuclear magnetic resonance (NMR) had included methods for reducing the inhomogeneous broadening of NMR lines, suggested that a graser based on long-lived transitions might be indeed feasible (Khokhlov, 1972). With Il'inskii (Khokhlov and Il'inskii, 1973; Il'inskii and Khokhlov, 1974a, 1974b), he proposed a method for averaging out the spin-dependent interactions that broaden Mössbauer lines (Appendix E) by applying a sequence of radio-frequency pulses to an aligned population of isomers.

In that same year, Gol'danskii and Kagan (1973a) presented calculations showing that some Mössbauer nuclei in appropriate hosts and geometries might be pumped above inversion by an intense burst of neutrons from an explosive source, without unacceptable temperature rise or lattice damage. Simultaneously, Letokov (1973a, 1973b) suggested that optical lasers can vaporize, selectively excite, and ionize atoms of isomeric nuclides that have been generated by nuclear reactions, and that the ionized species can be electrically separated and implanted on a substrate appropriate to support recoilless emission. This might enable the use of intermediate lifetimes, for which line broadening might not be unacceptable.

We first examine the proposals that do not suggest a separation procedure.

2. Neutron-pumped Mössbauer transitions

a. Basic proposal and variants

The proposal of Gol'danskii and Kagan (1973a, 1973b; 1973c) featured direct use of neutrons moderated from a nuclear explosion to generate, through radiative cap-

ture in a parent embedded in a Be host, an isomer in the Mössbauer range. A similar proposal, in an unpublished internal memorandum at Los Alamos (Solem, 1979), differed essentially only in the use of a Li host.

Variants of this proposal have included (1) a two-stage mechanism (Gol'danskii, Kagan, and Namiot, 1973), in which the graser would be excited by Mössbauer radiation generated by neutron capture in a "convertor" surrounding the graser; (2) a system that would combine the neutron source and the graser (Fig. 5) (Baldwin, 1974a; Douglas, 1974), based upon suggestions that laser compression of fissile material might greatly reduce the critical mass of a chain-reacting assembly (Winterberg, 1973b; Askar'yan, Namiot, and Rabinovich, 1974) and that the density of moderated neutrons can be increased by compressing the moderator (Askar'yan and Namiot, 1974); (3) a proposal to enhance the resonance capture rate of neutrons by pumping with a Doppler-upshifted thermal flux, generated by moving the moderator rapidly toward the graser (Bowman, 1975).

b. Requirements for direct neutron pumping

We consider first the directly pumped graser proposal of Gol'danskii and Kagan (1973a, GK).

Starting with the relation derivable from photon balance (Sec. III.D.2) for the minimum density of neutron captures N_c for which gain should be possible,

$$N_c \geq 2.6(14) E^2 (\Gamma/\Gamma_\gamma)[(1+\alpha)/f\xi]\, l^{-1}, \tag{24}$$

where

$$\xi = 2\eta - 1 \tag{25}$$

is the net contribution to inversion by a single neutron capture event, and l here is the photon mean free path in cm, GK derived the following requirements for a directly excited graser.

(1) The lifetime of the isomer must be in the range $1(-8) \leq T \leq 1(-4)$ s, in order to avoid excessive inhomogeneous line broadening, on the one hand, and excessive pump power on the other.

(2) The gamma-ray energy E must be in the range $10 \leq E \leq 100$ keV, in order to keep both the internal conversion coefficient α and the Mössbauer fraction f within reasonable limits (i.e., small α, high f).

(3) The total density of excited nuclei must be at least 1(18) to (19) cm^{-3}.

(4) The temperature of the graser body must not have risen by more than the Debye temperature by the time the critical density of excited nuclei has been reached.

(5) The excited nuclei must remain embedded within a rigid lattice of low nonresonant absorption (i.e., long photon mean free path l).

(6) The geometry of the graser body must favor (a) access of pumping radiation; (b) escape of unwanted radiations; (c) gain in the desired direction.

The last requirement dictates an elongated filament, as proposed earlier (Sec. III.B.3), or an assemblage of parallel filaments.

For meeting the fifth requirement, GK suggested that beryllium be doped with the parent isotope until the total nonresonant photon absorption was just double that of Be. In the Solem proposal, lithium was to serve as the host material.

To meet the third requirement, it was proposed to capture neutrons from nuclear explosions. That neutrons so generated are too fast for efficient capture imposed the further requirement that they be moderated to about 100 eV before impinging on the graser body—this energy was determined by an assumed inverse dependence of neutron capture cross section on neutron velocity (Appendix J) and by an estimate of heating by nuclear recoil [Requirements (3) and (4)]. GK showed that, if the graser filament diameter were at most only a few micrometers, then the additional heating by capture gamma radiation would be negligible. Neutron capture by the isomeric nuclei was not considered.

Although the requirements on the graser transition and temperature were simply those for the Mössbauer effect (Appendix D), it was recognized that, in an amplifying situation, isomers of somewhat longer lifetime and, therefore, having smaller resonance cross sections than those useful in Mössbauer spectroscopy, could be used. An illustrative estimate (Gol'danskii and Kagan, 1973a), made for the well-known ^{181}Ta Mössbauer transition (see Table I), showed that not enough information was available for identifying any particular isotope or transition as a graser candidate; in particular, the isomer ratio (Appendix B) is rarely known. Moreover, transitions between excited states, which cannot show the Mössbauer effect, might prove suitable for graser application (unless the lower level's lifetime were too short), and even moderately radioactive nuclei might serve as parents of useful isomers. GK therefore urged that intensive research be undertaken to ascertain unknown parameters (viz., isomer ratios, neutron cross sections of radioactive species), to improve values of known parameters (viz., conversion coefficients, branching ratios) and to discover new transitions, particularly of low energy and intermediate lifetime.

c. Two-stage neutron-gamma pumping

This proposal was followed by a brief paper (Gol'danskii, Kagan, and Namiot, 1973) suggesting that the extremely high neutron fluence requirement could be reduced by orders of magnitude by taking advantage of the fact that the probability of Mössbauer resonance absorption (Appendix D) can be much higher not only than that of nonresonant gamma-ray absorption (Appendix F), but of neutron capture events (Appendix J) as well. In the proposed "two-stage" method, neutron capture occurs in a parent-doped convertor, where it generates Mössbauer radiation, which in turn is absorbed by ground-state nuclei in the graser. However, since resonance absorption between a single pair of levels can at most saturate, not invert, their populations, it is necessary, if the two-stage mechanism is to lead to inversion, that additional states be involved; either there must be more than one excited state, or there should be a relatively unoccupied sublevel of the ground state (Sec. V.B.7.d). The authors also acknowledged that the numerical advantage offered by the high ratio of resonant to nonresonant cross section would be reduced by geometric and kinetic factors. The former,

they estimated at ≤0.5; the latter as ~0.2. Still, it was expected that a net orders-of-magnitude reduction should be obtained for the neutron fluence, in comparison with direct pumping. There should also be a geometrical advantage, in that pumping radiation enters from the side of the filament, and a shielding advantage, in that the neutrons are attenuated in the convertor before reaching the graser, reducing deexcitation of the graser states by neutron bombardment.

d. Difficulties with neutron pumping

The following considerations apply not only to the two proposals just described but also to their more elaborate variants.

The estimates of GK were criticized by Vorontsov and Vysotskii (1974a) and by Hannon and Trammell (1975) on grounds that they ignored the decay of excitation that occurs during the development of stimulated emission. In any narrow-line resonance process, as first noted by Chirikov (1963) and later suggested in the optical laser context by Hopf, Meystre, Scully, and Sealy (1975), the response evolves on a time scale established by the linebreadth (Appendix I). When inversion is created essentially instantaneously, in a brief burst of pump radiation, then it will disappear (in the case of equal degeneracy factors and full initial inversion) in one half-life of the isomer (unless the lower state also decays). Thus, when the line is nearly of natural width, inversion may be dissipated before appreciable gain can be developed. However, others (Baldwin, Pettit, and Schwenn, 1975; Baldwin and Suydam, 1977) have pointed out that statistical fluctuations of the time for a burst of fast neutrons to moderate down to energies at which capture is probable exceed the lifetimes of all but the very longest-lived transitions admitted by GK, so that the pumping would, in fact, be sustained, not instantaneous.

In order to ascertain the neutron source requirement for a directly excited graser when these considerations are taken into account, a computer study was undertaken by Baldwin and McNeil (1977). The Mössbauer transition in ^{83}Kr ($E = 9.3$ keV, $T = 140$ ns, $\alpha = 20$) was investigated, following a suggestion by Bowman (1975). Various admixtures of ^{82}Kr to Be host, and pure solid Kr at 0 K were assumed to be bombarded by neutrons moderating from a delta-function burst of fast neutrons injected uniformly throughout an infinite hydrogenous moderator. Using a kinetic model developed by Baldwin and Suydam (1977) that incorporated the time dependences of both the neutron spectrum and the stimulated emission process, the amplitude of a plane electromagnetic wave, initiated by a spontaneous recoilless event, was calculated for a sequence of times after source burst and distances from source point. Various values of doping, linebreadth, and temperature were assumed as inputs. This computation was deliberately simplified and therefore somewhat unrealistic, since it did not take account of heating of either the graser or the moderator, transient changes of linebreadth, nor depopulation of the graser states by neutron bombardment. Figure 7 is typical of the results of the kinetics study.

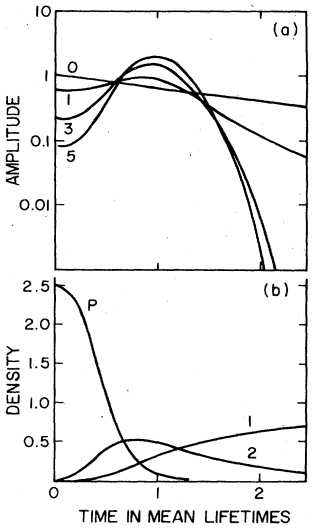

FIG. 7. Kinetics of a neutron-pumped solid-krypton graser. This shows the computed time dependences in a graser system pumped by capture of neutrons in the 40-eV resonance of ^{82}Kr to form the 9.3-keV Mössbauer isomer. Shown are (a) amplitude of the vector potential of a plane wave emitted at $t = 0$, observed at a sequence of distances from the spontaneous source measured in nonresonant absorption lengths; (b) population density of the parent nuclide P and of the upper 2 and lower 1 of the isomeric states of ^{83}Kr. Note the initial delay of amplification owing to neutron moderation time and inertia of resonance, the rapid depletion of the parent material, and the rapid termination of inversion as ground states accumulate. The source burst was 5(22) n cm^{-3}, moderator water, isomer ratio unity, linebreadth ten times natural. Heating of the moderator and neutron capture by the isomer were neglected. From Baldwin and McNeill (1977).

It was found that

(1) Significant departures from purely exponential attenuation and decay can be obtained only if the initial density of source neutrons exceeds 1(22) cm^{-3}, i.e., if the proportion of neutrons to moderator atoms is of the order of 1:60.

(2) The role of the Be host ($\Theta = 1440$ K) is purely that of providing an effective Debye temperature (Appendix D)

$$\Theta_{\text{Be-Kr}} = (M_{\text{Be}}/M_{\text{Kr}})^{1/2}\Theta_{\text{Be}} = 474 \text{ K} \quad (26)$$

higher than that of pure solid Kr (72 K); its parasitic absorption is undesirable.

(3) Depletion of the parent isotope and accumulation of ground states significantly increase the neutron fluence requirement.

(4) Most of the pumping was via the 40-eV resonance of ^{82}Kr (Mann, Watson, Chrien, Zimmerman, and Schwartz, 1959); thermal capture was negligible.

It has been asserted (Baldwin and Solem, 1979a) that it is thermodynamically impossible to meet such high neutron fluence requirements with neutrons moderating from any existing pulsed sources, however intense, and that the maximum possible neutron capture probability is therefore insufficient to generate inversion in any known nuclide (Baldwin and Solem, 1979b). This is

because all known pulsed sources of neutrons (except the relatively weak photonuclear sources) generate those neutrons with average energies of 2 MeV or more. This energy, transferred during moderation to the atoms of the moderator, raises its mean thermal energy. Moderation ceases when the neutron and moderator atom energies have equalized. There is, therefore, an upper bound to the density of neutrons that can be moderated to a given energy. When account is taken of the decrease of probability for neutron capture with increasing neutron energy, the above conclusion follows. (For further details, refer to Appendix J).

It is pertinent, then, to consider ways to increase the densities of neutrons after they have moderated. One is by compressing the moderator (Askar'yan and Namiot, 1974). For those nuclides whose neutron capture cross sections vary inversely with neutron velocity, the rate of capture varies directly with the neutron density, i.e., adiabatic heating of a compressed neutron-moderator system increases the collision rate, but by a smaller factor than that by which it decreases the capture cross section. For other dependences, it would be necessary for the compression ratio greatly to exceed even the factor needed for inertial fusion (Nuckolls, Wood, Thiessen, and Zimmerman, 1972), before the capture rate could thereby be augmented sufficiently to overcome the orders-of-magnitude discrepancy between the inversion requirement and the thermodynamic limit to moderated neutron density.

Another possibility is diffraction focussing, by means of a zone plate, of neutrons anomalously transmitted (Appendix G) by an appropriate crystal (Indenbom, 1979). The neutrons must be in a very narrow band of wavelength and direction if they are to be focused so strongly, so that their intensity at the focal point or line would be limited. Perhaps such a nearly monoenergetic beam of neutrons could be obtained by using a neutron storage ring (Paul and Trinks, 1978) combined with electron-injection, beam-cooling techniques. Whether a storage ring could be designed to accumulate the neutron intensities necessary for graser pumping within the neutron's lifetime is yet to be established.

Finally, intense sources of synchrotron radiation may eventually be developed that can generate high yields of photoneutrons from Be, with energies in the resonance region (Eremeev, 1978; Hofmann, 1980).

It remains to estimate the actual advantage afforded by the two-stage proposal. Baldwin and Solem (1980) have analyzed that form of the two-stage method in which two or more resonant Mössbauer lines are involved, dismissing as unrealistic the other variant, involving nondegenerate ground states. Evaluating the kinetic and geometrical factors involved in pumping a three-level transition, they calculated values for the interstage coupling coefficients of ^{57}Fe, ^{73}Ge, ^{161}Dy, and ^{181}Ta, relating neutron flux in the convertor to pumping rates in the graser. The scheme is illustrated in Fig. 8. For this it was necessary to know the isomer and branching ratios; "optimistic" values were assumed. Using the calculated coupling factors they solved rate equations numerically to determine degree of inversion in the graser as a function of neutron capture rate in the convertor. The neutron densities required for inversion were shown to be far above the thermodynamic limits found in their earlier studies, cited above, for any of the four isotopes listed. Figure 9, showing degree of inversion p as a function of the neutron capture rate coefficient for ^{72}Ge, is typical of

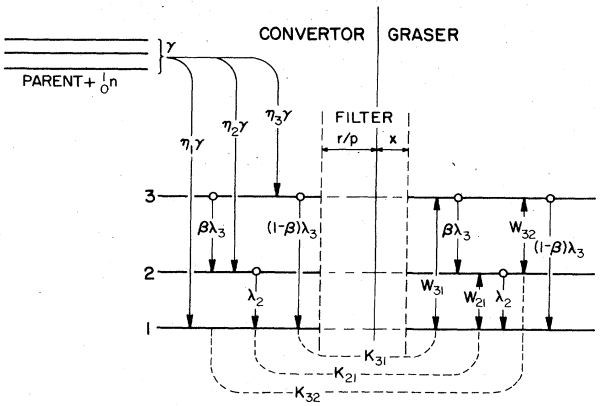

FIG. 8. Outline of a two-stage three-level pumping calculation. The figure displays a computational scheme for evaluating the kinetics of a three-level Mössbauer system pumped by a two-stage mechanism. Neutron capture at rate γ by the parent isotope in the convertor region forms three isomeric states in proportions η_1, η_2, η_3, respectively. Their relaxation generates three recoilless radiations; after penetrating thickness r/p at an obliquity $\cos^{-1} p$ in the convertor and a thickness x in the graser, these radiations interact resonantly with ground-state nuclei in the graser. The respective pumping rates W_{ij} in the graser are proportional to γ through coupling coefficients K_{ij}, expressing the effects of geometry and resonant scattering. From Baldwin and Solem (1980).

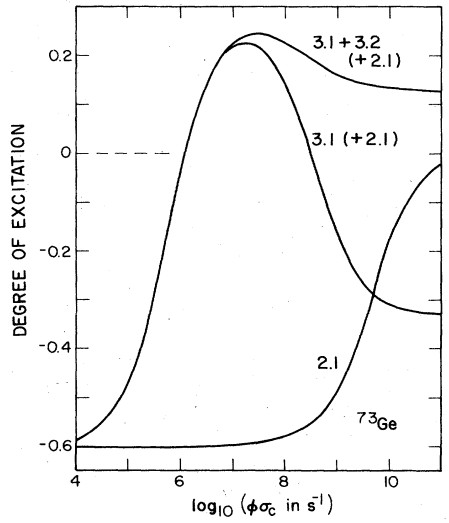

FIG. 9. Excitation via two-stage three-level pumping. The computational scheme of Fig. 8 is applied to the 2.6-μs, 13.5-keV and the 1.9-ns, 68.8-keV levels of ^{73}Ge. The curves indicate degree of inversion of the 13.5-keV level for pumping with only one (2-1), two (3-1 and 2-1), or all three recoilless lines, on the assumption that such selection can be made by an appropriate filter (Fig. 8). The abscissa is the neutron capture rate coefficient γ of Fig. 8. Pumping via the third level permits inversion to be achieved, but only if the pumping rate coefficient exceeds 1(6) s^{-1}. From Baldwin and Solem (1980).

their results. A hypothetical isotope was then "invented" having "unrealistically optimistic properties." They still found that, despite a coupling coefficient far higher than that of any of the known isotopes, the critical thermal neutron density for inversion via a two-stage, three-level mechanism is at least three orders of magnitude above the thermodynamic limit.

These authors were not optimistic that this negative conclusion could be reversed, either through development of intense sources of resonance energy neutrons, or by discovery of new transitions with more favorable parameters. Thus they concluded that if a graser is to be developed, it must incorporate means for rapidly separating and concentrating the excited material by a large factor between the pumping and lasing operations.

3. A fast-separation, short-lifetime graser proposal

In addition to his proposal to pump a short-lifetime graser *in situ* with neutrons from a moving moderator, Bowman (1976) has suggested a method for physically separating the pumping and lasing processes of a short-lived Mössbauer transition.

Essentially, the proposed device would resemble a reverse-biased cylindrical proportional counter. An axially centered wire of beryllium would collect ions of the Mössbauer isotope formed in a surrounding cylindrical region by a pumping radiation. Bowman proposed the 9.3-keV transition in ^{83}Kr, noting that it can be pumped either by $^{82}\mathrm{Kr}(n,\gamma)^{83}\mathrm{Kr}$ (i.e., with neutrons) or by $^{84}\mathrm{Kr}(\gamma,n)^{83}\mathrm{Kr}$ (i.e., with high-energy bremsstrahlung). From considerations of the angular momenta of the various states involved, he predicted a high isomer ratio. The parent krypton isotope(s) might be liquid, gaseous, or frozen as a condensate upon a radially disposed array of foils coplanar with the central wire. Upon transmutation, each ion would be drawn immediately toward the central wire. Its momentum would implant it in the Be, forming a graser similar to that envisaged by Gol'danskii and Kagan (1973a, 1973b) and others, but with the advantage of a high concentration ratio in the convergent collecting field. With a brief pumping pulse, it was suggested, the ion arrivals at the wire might even be so timed that the graser would function as a traveling-wave amplifier.

Quantitative estimates of losses by charge exchange, sputtering, and decay during collection were not given, nor were the pumping requirements calculated.

4. Fast-separation, intermediate-lifetime grasers

a. Optical effects of nuclear isomerism

Conventional radiochemical procedures for separating and concentrating an active isomer usually require appreciable time (minutes to hours), so that they are applicable only to long-lived transitions. For those, the recoilless line would be far broader than its radiation width. Lasing would be impossible, unless methods discussed in a later section, that have been proposed for line narrowing, can be developed. Here we discuss the possibility that the separation and concentration steps can be accomplished so rapidly that some isomeric transitions in the lifetime range that we have termed "intermediate" could be stimulated despite appreciable broadening.

It has been suggested (Letokhov, 1973a) that laser techniques presently under development for the separation of isotopes are applicable to this problem. Letokhov noted that measurable shifts in the positions of optical resonance absorption lines, well in excess of their Doppler breadths, have been observed for atoms whose nuclei are in isomeric states (Melissinos and Davis, 1959); this effect originates both in the altered nuclear mass and in the change of nuclear volume that is also responsible for the monopole shift of Mössbauer lines, through the electron-nuclear electrostatic interaction (Appendix E).

Attention has been called also (Reeves and Strong, 1973; Zel'dovich and Sobel'man, 1975) to still greater—in fact, qualitative—differences in the optical spectra of isomeric species of the same element that were observed in the same experiments. Nuclear isomerism is accompanied by appreciable differences in angular momentum of the two states. Interaction of the nuclear and electronic orbital magnetic moments is therefore manifested by unequal numbers and displacements of hyperfine components in the optical spectra of atoms having nuclei in different states. For example, in the particular case cited, ^{197g}Hg and ^{197m}Hg, the hyperfine structures of the 2537 A resonance lines are entirely distinct, since the nuclear spins are, respectively, $\frac{1}{2}$ and $\frac{13}{2}$ (Melissinos and Davis, 1959).

Thus those atoms containing a selected one of the isomers can be separately excited by resonance absorption of optical radiation from a tunable laser or from an appropriately filtered lamp. The excited atoms have chemical and physical properties unlike those of the ground state—in particular, lower ionization potentials, unequal magnetic moments, and higher chemical reactivity.

b. Isomer separation by selective photoionization

Letokhov suggested (1973b) a system with which selective optical excitation could be exploited for the rapid removal and concentration of isomeric nuclides of intermediate lifetime (Fig. 10). After the nuclear and atomic excitation steps, he proposed to photoionize, electrically extract, and deposit the upper-state isomeric atoms in a line geometry on an appropriate substrate. In order to maintain isomeric purity during the ion collection process, one must form and accelerate the ions in a gas or vapor of low density and collect them so rapidly that charge exchange with untransmuted atoms would be negligible. This was shown to demand that a high proportion of atoms in the sample be transmuted and, therefore, to determine the pumping parameters.

Letokhov's analysis addressed the following questions: (1) the minimum number of isomeric nuclei required for perceptible amplification; (2) alternative reactions for exciting the nuclei; (3) the laser photon flux for quantitatively exciting optical levels of the isomeric atoms; (4) the photoionization requirements; (5) efficient collection of the ions.

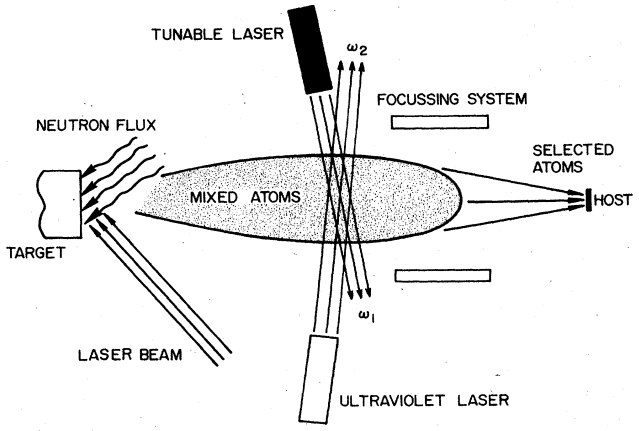

FIG. 10. Concept for an intermediate-lifetime graser. The concept features an isomer-separation operation, as follows: (1) neutrons transmute parent nuclei into the graser isomer; (2) a laser beam vaporizes the activated mixture; (3) a tunable laser selectively excites those atoms containing upper-level isomers; (4) a third laser photoionizes the excited atoms; (5) an electric field extracts and epitaxially deposits the ions onto the host crystal to form the graser rod. From Letokhov (1973).

In estimating the number of excited nuclei, Letokhov determined the dimensions of the graser by considerations like those of Sec. IV.C, but he specified that the single-pass gain be 100 (rather than 2) and neglected nonresonant photon removal. He found that the pump must excite at least 1(15) isomeric nuclei within a rapidly vaporizable volume of the solid target. From analyses of both radiative and neutronic pumps, he chose thermal neutron capture as the preferred reaction.

Vaporization of the activated portion of the target was assumed to be a straightforward step, using intense laser irradiation, so that no estimates were made of the power or time requirements for this step. It was remarked that, if the target for neutron activation were a molecular gas or vapor, then a recoil separation (Szilard and Chalmers, 1934) would be possible.

The flux of resonant optical photons required to ensure complete excitation of the isomeric atoms was estimated from the excitation cross section σ_{exc}, in the range $1(-12)$ to $1(-10)$ cm^2, the atomic relaxation time T_a, typically $1(-8)$ s, and the optical resonance photon energy E_a by means of the formula

$$\Phi_a \geq E_a/\sigma_{exc}T_a . \tag{27}$$

It was found to correspond to a few W cm^{-2}, provided the rate coefficient for ionization were made approximately equal to that for excitation. Because the photoionization cross section is far lower, the ionization step calls for much more intense radiation, the ratio of cross sections being of the order of 1(4). Given these conditions, excitation and ionization processes can be completed extremely rapidly, in only a small number of atomic relaxation times.

The most critical step was stated to be that of collecting the ions without appreciable loss of the excited nuclear species through charge exchange with untransmuted atoms. This calls for limiting the vapor density to a maximum value of

$$N_g \leq (\sigma_{cx}D)^{-1}, \tag{28}$$

where D is the diameter of the vapor cloud. The charge exchange cross section σ_{cx} is expected to be of the order of $1(-14)$ cm^2.

Collection of the required minimum number of isomeric ions must be completed before appreciable decay; Letokhov specified $0.2T$ for this step. Thus, if the drift velocity of the ions in the collecting field is v, the distance for collection cannot exceed $0.2vT$, and since the vapor density is limited, the maximum number of ions that can be collected in this time is

$$n_{max} = 0.2\, vTD/\sigma_{cx} . \tag{29}$$

That part of this number which can be deposited within the graser volume must exceed the minimum required by graser threshold relations. This, rather than the concentration of isomers in the neutron-bombarded target, is the limiting factor in establishing the neutron fluence requirement, except for the longest-lived transitions (lifetime longer than 10 s). Numerical values given for illustration by Letokhov are presented in Table II.

Letokhov assumed extremely optimistic parameters for the transition and for the parent isotope. For example, the recoil energy of a nucleus with $A = 120$ in

TABLE II. Design parameters of a photochemically pumped graser (Letokhov, 1973b).

Transition:		
Energy	E	30 keV
Lifetime	T	10 s
Recoilless fraction	f	1
Conversion coefficient	α	0
Linewidth	Γ	1(5) s^{-1}
Degree of inversion	p	1
Number of isomerides	N_{min}	1(15)
Parent neutron capture cross section	σ_c	1(4) b
Atomic properties:		
Photoexcitation cross section	σ_{exc}	1(−12) cm^2
Photoionization cross section	σ_{ion}	1(−20) cm^2
Relaxation time	T_a	1(−8) s
Charge transfer cross section	σ_{cx}	1(−14) cm^2
Nonresonant absorption cross section	σ_a	0
Physical dimensions:		
Target area	S_0	1 cm^2
Activated depth		3(−3) cm
Vapor cloud diameter	D	10 cm
Ion drift velocity	v	5(4) cm s^{-1}
Ion collection time	$0.2T$	2 s
Graser diameter	$2a$	5(−5) cm
Graser length	L	7(−1) cm
Input-output parameters:		
Neutron fluence	Φt	5(17) cm^{-2} for ~1 s
Excitation laser power	P_1	4(3) W for ~1(−8) s
Ionization laser power	P_2	5(10) W for ~1(−8) s
Graser single-pass gain		100

emission of a 30-keV gamma ray is 4 meV. The Debye-Waller factor, assuming a typical rigid lattice of $\Theta = 300$ K, cannot then exceed 0.9, even at 0 K; at 300 K, it would be only about 0.16. For $Z = 50$, internal conversion would be barely possible in the K shell, but the total conversion coefficient would nevertheless be of the order of 5 for electric dipole transitions (Way, 1973). However, a lifetime of 10 s implies higher multipolarity, either $M3$ or $E3$ (Wapstra, Nijgh, and van Lieshout, 1959). For $M3$, the internal conversion coefficient in the K shell alone is expected to be of the order of 2000; it would be higher for $E3$ transitions (Way, 1973). Moreover, the nonresonant absorption cross section for 30-keV radiation in $Z = 50$ is over 8000 b (Storm and Israel, 1970), while the value of the "area factor" of Eq. (5), $\Lambda^2/2\pi = 270$ b. This is further reduced by the linebreadth factor, homogeneously by a factor of the order of thousands, and inhomogeneously by a still greater factor. Clearly, the low stimulation cross section for a transition with these parameters excludes it from consideration. The neutron capture cross section assumed for the parent is unusually high; still, were it much lower, the required neutron fluence could not be generated (Appendix J).

These considerations emphasize that, although optimistic parameters can be useful to draw attention to a new principle, it is unlikely that the parameters for any transition will ever all be favorable. We must use realistic parameters of actual isotopes in estimating the feasibility of any proposed approach to an actual graser system. We do not mean to imply that the photoionization approach is unfeasible; in fact, it may be the most promising of the proposed alternatives. But it can produce a graser only on paper until an isotope-by-isotope evaluation discovers an appropriate candidate.

Even in the event that a transition can be identified in an element which, fortunately, combines favorable sets of both nuclear and optical properties, a number of other questions must still be answered. For example, what strength of electric field(s), and what configuration, are required to collect the ions and focus them onto so small a target in the required time, and what is the effect of their heat of impact on that target? Would an accelerate-decelerate system be necessary to avoid target damage? Will a discharge be triggered that can further heat and perhaps even destroy the deposited layer? What are the effects of space charge and ion recombination on the collection process? How can the optical lasers be protected from an intense neutron pump? Or could it be used to pump them (Miley, 1977) as well?

c. Isomer enrichment by optical pumping

Although no definite proposals for a graser system have yet appeared that would employ the nuclear magnetic moment as a basis for rapid separation of isomers, it is pertinent here to remark that isomers with lifetimes as short as 10 ms have actually been enriched by selectively exciting and then magnetically sorting atoms containing them with light from optical lasers. The currents involved were extremely small; the significant fact is that selective optical excitation of isomers has indeed been demonstrated. These experiments are briefly described in Appendix K, and a possible basis for a graser system based upon their general principle is developed in Sec. V.B.7.d.

5. Use of long-lived isomers

a. Need for line narrowing

Khokhlov's suggestion (1972) that improvements in crystal growth technology, combined with newly developed techniques of nuclear magnetic resonance (NMR) might enable inhomogeneous broadening to be eliminated, revived interest in the possibility that long-lived isotopes could yet be used in grasers. Soon a number of suggestions for Mössbauer line narrowing appeared (Khokhlov and Il'inskii, 1973; Il'inskii and Khokhlov, 1974a, 1974b; Gol'danskii, Karyagin, and Namiot, 1974a, 1974b; Namiot, 1973; Kagan, 1975; Karyagin, 1976, 1977).

The recoilless linebreadth (Tables III–V below) must be reduced by a very large factor indeed if truly long-lived transitions are to be useful. For example, if the lifetime is 1(4) s and the inhomogeneous linebreadth is 1(5) s^{-1}, the factor $\Gamma T > 1(9)$. The width Γ must be reduced to the order of no more than 1–0.1 s^{-1} before a graser application is conceivable. No known Mössbauer line is that narrow.

If narrowing can be accomplished, then one can expect the development of grasers to follow. There would be many advantages of such an approach, if it were to prove not too complicated in implementation. With time for chemical operations, substantial concentration of the isomeric activity would be possible. There would be wide latitude in the type and specification of the pumping source. For example, the method (Vali and Vali, 1963a) of extraction from a long-lived radioactive parent would become possible; the isomer might be produced directly by reactor or accelerator bombardment; or Letokhov's photoionization method (1973b) could be used. Inefficient collection of the ions would then be tolerable. Moreover, the graser's output might then be quasicontinuous, with only a low background of spontaneous radiation.

On the other hand, it would be necessary to work with highly radioactive material, and it is not yet known whether highly radioactive crystals can be grown, or, if so, how long they would retain their structure, particularly if it were desired to exploit Bragg-angle effects for reducing the inversion requirement (Appendix G).

The first problem to be solved is that of reducing the breadth of the recoilless line. Here difficulty arises in that several distinct phenomena contribute to the inhomogeneous linebreadth.

b. Sources of the broadening

The origins and magnitudes of the line-broadening interactions are discussed in more detail in Appendix E. Some numerical values are given in Tables III and IV. Here we note that they fall into the following distinct

TABLE III. Linebreadths of some Mössbauer radiations.[a]

Nuclide	Energy (keV)	Half-life (s)	Natural Γ_0[b] eV	Γ_0 s^{-1}	Measured Γ_1[c] $m\,s^{-1}$	Γ_1 s^{-1}	$\Gamma_1 - \Gamma_0$ s^{-1}	Reference
^{153}Eu	97.4	0.21(−9)	2.17(−6)	3.30(9)	1.5(−2)	7.4(9)	4.1(9)	d
^{155}Gd	86.5	6.3(9)	7.20(−8)	1.1(8)	1.25(−3)	5.5(8)	4.4(8)	d
^{57}Fe	14.4	9.78(−8)	4.67(−9)	7.1(6)	4.3(−4)	3.1(7)	2.4(7)	e
^{73}Ge	13.3	2.95(−6)	1.55(−10)	2.35(5)	1.4(−5)	9.4(5)	7.0(5)	f
^{181}Ta	6.24	6.80(−6)	6.71(−11)	1.02(5)	5.3(−5)	1.7(6)	1.6(6)	g
^{67}Zn	93.3	9.15(−6)	4.96(−11)	7.53(4)	0.81(−6)	3.8(5)	3.1(5)	h
					1.9(−6)	9.0(5)	8.2(5)	i
^{107}Ag	93.3	4.43(1)	1.03(−17)	1.56(−2)	See text	1.2(4)	1.2(4)	j
^{109}Ag	88.0	3.98(1)	9.3(−18)	2.04(−3)	See text	6.1(−2)	5.9(−2)	k

[a] Energy and half-life data from Stevens and Stevens (1976).

[b] Calculated from the listed half-life.

[c] Column 6 lists the narrowest line reported in the referenced work as a Doppler scan result. Often this is sensitive to sample preparation and geometry. The seventh column converts the Doppler result of Column 6 into an equivalent frequency.

[d] Henning, Baehre, and Kienle (1970).

[e] Fe in a Cu host; Schiffer, Parks and Heberle (1964).

[f] Pure germanium. Pfeiffer and Raghavan (1974).

[g] Dornow, Binder, Hiedemann, and Kalvius (1979). This transition shows unusually large isomer shifts and quadrupole interactions. The narrowest line was obtained with a tungsten host and tantalum metal absorber. For further data on this transition see Table IV.

[h] Zinc oxide. De Waard and Perlow (1970).

[i] Zinc metal. Potzel, Forster and Kalvius (1978).

[j] This measurement did not employ Doppler scanning. The linewidth was inferred from a cross section, calculated from measurements of activity induced by resonance activation (Bizina, Beda, Burgov, and Davydov, 1964).

[k] Wildner and Gonser (1979). The linewidth was inferred from the change of self-absorption with temperature in the range 4.2–78 K, using a single crystal of Ag into which ^{109}Cd had been diffused.

groups: monopole, or spin-independent interactions; spin-related interactions, including magnetic dipole-dipole; interactions of quadrupole and higher moments of the nuclear charge with appropriate derivatives of crystal fields; temperature, or second-order Doppler broadening; and gravitational broadening.

It can be assumed that the temperature and gravitational contributions can be controlled, and we note that manipulation of these effects has even been proposed as means for controlling the onset of lasing (Vali and Vali, 1963b). Thus our major interest is in the monopole, also known as the "chemical" or as the "isomer" shift, and in the multipole, or spin-dependent interactions.

Monopole broadening results from variations in the local density of the electron cloud within which the nuclear volume is immersed. Since the nuclear volume is not in general equal for two isomeric states, a change in the electrostatic potential energy of the nucleus accompanies the isomeric transition, slightly shifting the energy of the emitted gamma ray. Within a lattice containing defects or impurities, the electron density varies from site to site, thereby broadening the recoilless line. In a perfect crystal, monopole broadening would be small, but still not entirely absent, because the two states of the isomer, having unequal magnetic moments, would affect the electron density unequally.

The multipole interactions all depend upon the relative orientation of the nuclear spins, which are ordinarily disordered by thermal agitation. For example, the most serious interaction in many cases, magnetic dipole-dipole broadening, results from distribution of interaction energies of pairs of randomly oriented nuclear magnetic moments. There is a similar interaction between the electrons and nuclei: In certain cases, the local spin density of electrons at the nuclei differs from zero and gives rise to hyperfine splitting. Another electron-nucleus magnetic dipole interaction results from paramagnetic impurities. The range of interaction of an unpaired electron may be great enough to affect even nuclei beyond nearest-neighbor sites. Electric multipoles interact with derivatives of the internal crystal fields, including high-order derivatives of complex origins (Pfeiffer, 1977). Note particularly that, even in perfect crystals, inhomogeneous broadening by the multipole mechanism is unavoidable, since the two isomeric states have different magnetic dipole and electric quadrupole moments.

Some specific estimates of broadening in long-lived transitions are given in Table V.

c. Proposals to reduce multipole broadening

Various techniques have been proposed for artificially reducing the nuclear multipole broadening. All involve causing the nuclear moments to precess with respect to the crystal lattice. The possibility of using nuclear alignment techniques was first pointed out by Vali and Vali (1963a), but the first detailed proposal came only ten years later (Khokhlov and Il'inskii, 1973), after NMR line narrowing had been demonstrated (Waugh,

TABLE IV. The ^{181}Ta 6.2-keV Mössbauer line in various hosts. All the sources listed employed ^{181}W as the parent activity. For all but the last two, it was prepared by the ^{180}Ta(n,γ) reaction. In reference f, the activation was by deuteron bombardment of ^{181}Ta, and the activity was separated carrier free. For further details the references should be consulted.

Source	Host Absorber	Linewidth μm s^{-1}	Isomer shift μm s^{-1}	Reference
Ta	Ta	1000	+900	a
W	Ta	260	+900	
W	TaC	no resonance observed		
W	Ta	91	+860	b
Ta	Ta	400	+40	
W	Ta	71	+839	c
Pt	Ta	380	−2650	d
W	W	69	+900	
V	Ta, 99.996%	5000	−33200	e
Ni	Ta, 99.996%	500	−39500	
Nb	Ta, 99.996%	130	−22600	
Ru	Ta, 99.996%	1300	−27500	
Rh	Ta, 99.996%	3400	−28800	
Pd	Ta, 99.996%	1300	−27800	
Hf	Ta, 99.996%	1600	−600	
Ta	Ta, 99.996%	184	−75	
W	Ta, 99.996%	69	−860	
Re	Ta, 99.996%	600	−14000	
Os	Ta, 99.996%	1800	−2350	
Ir	Ta, 99.996%	1600	−1840	
Pt	Ta, 99.996%	300	+2670	
W	Ta, 99.97%	76	+860	f
W	Ta, 99.99%	53	+836	

[a] Steyert, Taylor, and Storms (1965).
[b] Sauer, Matthias, and Mössbauer (1968).
[c] Kaindl and Salamon (1970).
[d] Wortmann (1971).
[e] Kaindl, Salamon, and Wortmann (1973).
[f] Dornow, Binder, Heidemann, Kalvius, and Wortmann (1979).

Huber, and Haeberlen, 1968). That proposal suggested a cycle of pulses designed to reorient the nuclear moments periodically in a strong magnetic field and thereby cause the dipole-dipole interaction to average to zero. Other proposals (Namiot, 1973; Andreev, Il'inskii, and Khokhlov, 1974) have suggested the use of continuous radio-frequency perturbing fields. Rather than discuss each in detail, we here consider their common principle.

For simplicity, we consider first a nucleus having a large magnetic moment in its excited state, but zero or negligible moment in its ground state. Because the numbers of excited and ground states in the neighborhood of each lattice site vary statistically and their spin directions are thermally disordered, the local magnetic fields, and therefore their energy of interaction with each nucleus, vary slightly about an average. Now cause the excited nuclei to precess, with a common period short compared with the lifetime of the isomeric state. In a semiclassical sense, we can consider this to modulate the energy of the gamma ray, with an excursion equal to the dipole-dipole interaction energy and a period equal to the precession period. Thus each emitting nucleus becomes a frequency-modulated source of radiation; the modulation frequency is constant, but the amplitudes of modulation (i.e., the intensity of the sidebands but not their widths or spacings) vary from site to site. The nucleus now emits a comb of discrete lines, characteristic of frequency modulation; the carrier in each case is the same gamma-ray energy [except for variations of monopole shift and the temperature-kinetic shift (Karyagin, 1977)]. It is readily seen that all higher multipole (i.e., spin-related) interactions will be similarly modulated, whether the interaction is electric or magnetic and whether the source of the local field is other nuclei, lattice imperfections, or trapped impurities. The case of nonzero multipole moments for both isomeric states is more complicated; both must be made to precess. Although the frequency modulation patterns become more involved, the essential feature of separation of the carrier from sidebands is preserved.

Il'inskii and Namiot (1974) have shown that the net gain of the graser transition, for the resultant comb of narrowed lines, is the same as that for an unsplit but narrowed transition.

Actually, it turns out that very few of the excited states are actually lost from the central line so as to emit, instead, in the sidebands. Classically, the vector potential of the frequency-modulated wave of central frequency ω_0 and modulation frequency ω is

$$\mathfrak{A} = \mathfrak{A}_0 \sum_n J_n(U_m/\hbar\omega_0) \exp[i(\omega_0 + n\omega)t], \tag{30}$$

i.e., the envelope of the sidebands is given by the Bessel function $J_n(U_m/\hbar\omega_0)$. Thus the ratio of the first sideband to the carrier is

$$\left(\frac{J_1}{J_0}(U_m/\hbar\omega_0)\right)^2 = U_m/2\hbar\omega_0 \tag{31}$$

for small values of the argument. Since the magnetic interaction energy U_m is only a very minute fraction of the gamma-ray energy [typically $1(-13)$ or less], the fraction of nuclei emitting in the sidebands is trivial.

The original Il'inskii-Khokhlov proposal was based upon pulsed-NMR techniques pioneered by Waugh, Huber, and Haeberlen (1968), in which the nuclear moments are systematically reoriented at appropriate intervals by application of a certain periodic sequence of brief pulses of radio-frequency magnetic field applied normally to a strong orienting field. The directions and magnitudes of the pulses are such as to effect 90° reorientation of pairs of spin components (x and y, y and z, z and x). The objective is to cause the time-averaged value of the Hamiltonian of the dipole-dipole interaction

$$H_{dd} = \tfrac{1}{2}\sum_{i\neq j} b_{ij}(\mathbf{I}_i \cdot \mathbf{I}_j - 3I_{iz}I_{jz}) \tag{32}$$

to vanish (Haeberlen and Waugh, 1968).

Pulsed-NMR operations have been described that reduce certain NMR linewidths to 200 s^{-1} (Vaughan, Elleman, Stacey, Rhim, and Lee, 1972), from initial widths approximately 100 times greater. Further reduction would be difficult because of stringent demands on the uniformity of the main magnetic field and on the amplitudes and timing of the spin-rotating field pulses (Rhim, 1976), and on heating by the rf pulses. To avoid the

TABLE V. Estimates of broadening for long-lived recoilless transitions. Numbers, unaccompanied by other reference, indicate the condition for which the gamma-ray line is broadened by one natural width. Bracketed integers refer to explanatory notes, below. A dash (-) indicates that the mechanism was not considered. A zero (0) indicates that the mechanism was believed to cause no significant broadening, in comparison with other mechanisms. A question mark (?) indicates that existing information was deemed inadequate to permit a reasonable estimate. An equal sign (=) preceding a number indicates that the natural width would be exceeded by the factor given in the table.

Transition:								
Nuclide	^{107}Ag			^{103}Rh			Unspecified	
Half-life, s	44			3.36(3)		1	1(4)	1(6)
Energy, keV	93			40			Unspecified	
Reference	a	b	c	d	e		f	g
Broadening mechanisms:								
Gravitation [h]	--	1(−4) cm	--	5(−6) cm	3(−6) cm	1(−3) cm	1(−5) cm	--
Fundamental length	--	0	--	--	=1(4)	--	--	--
Doppler								
Vibration [i]	1(−7)	3(−12)	--	--	0	--	--	--
Thermal gradient	2 K m^{-1}	1.7(−4) K	--	1.3(−9) K	0	1(−5) K	1(−9) K	--
Temperature [o]	--	--	=1(4)	--	--		s	0
Monopole								
Chemical [j]	0	? [m]	--	0	--		t	--
Concent. gradient	--	1.5(−4)	--	--	--	--	--	--
Strain	--	0	--	--	--	--	t	--
Point defects [k]	--	1.5(−4)	--	1.5(−4)	2(−10)	1(−6)	1(−10)	3(−19)
Screw disloc.	--	--	--		--	10 cm^{-2}	none	3(5) cm^{-2}
Edge disloc.	--	--	--		--	1 cm^{-2}	none	none
Surface [l]	--	--	--	--	--	200 layers	4000 layers	--
Inner-shell ionization	--	--	--	--	--	--	--	--
Electron-nuclear	=1(3)	0	--	p	T_e=8 mK [r]		u	u
Nuclear-nuclear	=1(5)	? [n]	--	p	T_n=1 mK		u	u
Quadrupole	--	--	--	q	--		u	u

[a] Bizina, Beda, Burgov, and Davydov (1963).
[b] Carlson and Temperley (1969).
[c] Kagan (1964).
[d] Vali and Vali (1963a).
[e] Mead (1966).
[f] Gol'danskii and Kagan (1973a).
[g] Il'inskii and Khokhlov (1974a).
[h] Gives the vertical distance for which resonance is shifted by one linewidth.
[i] Gives the acoustical vibration velocity of two separate hosts, in m s^{-1}, for which resonance is shifted by one linewidth.
[j] Refers to inequality of average S-electron density associated with large-scale differences of host composition (e.g., source-absorber difference).
[k] Relative concentration of electron density variations from interstitial or substitutional impurities and vacancies. For Refs. b–f, inclusive, to shift by one linewidth; in Ref. g, to broaden by 0.1^{-1}.
[l] Refers to altered lattice spacing in the vicinity of a crystal boundary.
[m] Implies concern for the effect of accumulating decay products.
[n] Mentions, but gives no quantitative estimates of, the hyperfine splitting associated with lattice imperfection.
[o] Evaluates a formula, derived in Ref. c, for temperature broadening.
[p] Asserts that magnetic hyperfine interactions can be averaged out, but gives no details.
[q] Asserts that quadrupole broadening does not occur in dislocation-free crystals.
[r] Assumes that broadening is caused only by that fraction of all electrons having thermally disordered spins. The indicated temperature is that for which these would suffice to broaden by one natural linewidth.
[s] Notes that homogeneous broadening due to the thermal-fluctuation mechanism treated in Ref. c amounts to 1–100 s^{-1} at room temperature, independently of the lifetime.
[t] Estimates that a fractional change $\Delta V_0/V_0$ of unit-cell volume, or an equal strain due to shearing stresses, should produce a broadening by $2(7)\Delta V_0/V_0$ s^{-1}.
[u] Asserts that all spin-dependent broadening mechanisms can be averaged to zero by methods developed for NMR spectroscopy. A specific procedure is suggested in Ref. g.

heating difficulty, Karyagin (1977) suggests "Fermi-amplification," i.e., employing the Fermi contact interaction (Fermi, 1930) between electrons and nuclei in some specially selected compounds to amplify the rf effect.

In contrast to NMR, narrowing of a Mössbauer line requires reducing the unequal perturbations of two nuclear levels, while their populations are changing because of radioactive decay. The Hamiltonian to be averaged contains three, rather than one, set of double sums, enumerating the interactions of upper-with-upper, upper-with-lower, and lower-with-lower states, in the case of a crystal of pure isomer. There are more terms if the crystal contains additional components (e.g., an isotopically or chemically distinct host).

Khokhlov and Il'inskii (1973) suggested certain pulse

cycles that would reorient the upper- and lower-state spins separately, thereby causing the three-sum form of the Hamiltonian to be reduced by a factor $\sim \Delta t/T$, where Δt is the time to complete one pulse cycle. They noted that, since the electric quadrupole interaction involves a similar product of spin components in its Hamiltonian, both causes of broadening would be simultaneously alleviated.

Other proposals, involving continuous radio-frequency field perturbations (Namiot, 1973; Andreev, Il'inskii and Khokhlov, 1974), soon followed.

d. Reduction of inhomogeneous monopole breadth

Even if multipole broadening could be completely suppressed, other mechanisms of line broadening would still suffice to inhibit graser action (Table V; Appendix E). In particular, the broadening due to local variations of the monopole electron-nuclear interaction ("chemical" or "isomer shift") must be simultaneously reduced.

Crystal defects constitute a major source of inhomogeneous monopole broadening (Vali and Vali, 1963; Mead, 1966; Khokhlov and Il'inskii, 1973; Gol'danskii and Kagan, 1973c), particularly in the case of damage to the lattice by the pumping process (Schwenn, 1978) or by implantation after photoionization (Letokhov, 1973b).

Hope that the stringent specifications on the number of permissible defects (Khokhlov and Il'inskii, 1973; Gol'danskii and Kagan, 1973c) may be met has been revived by recent developments of techniques for annealing lattice damage to ion-implanted semiconductors, using optical lasers (White, Narayan, and Young, 1979). In this method, intense optical radiation is employed to melt highly localized regions near the surface of a solid that has been implanted with a dopant; after the laser pulse, as heat is conducted away by the underlying substrate, the melted region recrystallizes to assume the crystal form of the unmelted substrate, with the dopant atoms occupying regular lattice sites. The latter fact is significant in view of the sensitivity of the Borrmann effect (Appendix G) to lattice defects.

Still, the fast annealing technique would not entirely eliminate chemical broadening, since a partially inverted isomer population is itself a source of electron density variation.

It has been shown that, in principle, the monopole broadening can be canceled (Gol'danskii, Karyagin, and Namiot, 1974a, 1974b; Kagan, 1974a; Andreev, Il'inskii, and Khokhlov, 1974) by taking advantage of the fact that, for certain atomic species, both the monopole shift and the hyperfine splitting of nuclear states vary approximately in proportion to the density of S electrons at the nucleus, when the hyperfine splitting is caused by an internal magnetic field from paramagnetic atoms or by the Fermi contact interaction (Fermi, 1930).

Although the expectation values of total electron number density

$$|\psi_{up}(0)|^2 + |\psi_{down}(0)|^2$$

and of net spin density

$$|\psi_{up}(0)|^2 - |\psi_{down}(0)|^2$$

at the nucleus are not proportional, *small* variations of these quantities arising from variations of unit cell volume do vary proportionately from site to site. Thus the variable isomer shift is accompanied by a proportional variation of hyperfine splitting.

Gol'danskii, Karyagin, and Namiot (1974a, 1974b) note that, if the changes in electron density and in net spin density were related by just the right factor of proportionality, then the two effects would cancel for one of the hyperfine components of the isomeric transition spectrum.

Nature is unlikely to provide such a perfect match. The proposal suggested that this condition might nevertheless be forced by controlling the effective energy of the hyperfine interaction, through external manipulation of the internal magnetic field. To this end, it was proposed to apply a conically precessing magnetic field which, under appropriate conditions, would align the electron magnetic moments, causing them to execute a similar precession, thereby adjusting the effective average of the local electron-spin-generated magnetic field that creates the hyperfine splitting.

The precessing resultant of a static field with a rotating radio-frequency magnetic field describes a cone. Let its angular velocity be ω and inclination be α. The effective electron contact field follows this externally applied field if (1) thermal fluctuations do not destroy the stability of rotation and (2) the electron-spin precession in the field $\mathfrak{B}_{ext}$ is repeatedly averaged during the period of complete rotation $2\pi/\omega$. The combined requirement is

$$k\theta \ll \hbar\omega \ll \mathfrak{B}_{ext}\mu_e. \tag{33}$$

Under these conditions, the magnetic inductions from the external field $\mathfrak{B}_{ext}$ and the contact field $\mathfrak{B}_f$ will remain collinear and precess together. Their resultant, $\mathfrak{B}_{eff}$, therefore precesses at the relatively low angular velocity ω of the applied radio-frequency field. The nuclear magnetic moments, in turn, precess about $\mathfrak{B}_{eff}$, at the much higher Larmor frequency $\Omega \gg \omega$ corresponding to $\mathfrak{B}_{eff}$.

It can be shown (Gol'danskii, Karyagin, and Namiot, 1974a, 1974b; Pettit, 1976) that under these circumstances the part of the nuclear-state energy owing to hyperfine splitting is given by

$$E_m = \hbar(I\omega \pm m\Omega + \delta), \quad |m| \leq I, \tag{34}$$

the sum of a precession shift, which depends only on the variable parameter ω, and a multiple of the hyperfine splitting $\hbar\Omega$, which depends on four parameters G, ω, θ, and $\mathfrak{B}_{eff}$, and the monopole shift $\hbar\delta$.

By choice of the variable parameters, it should therefore be possible for the experimenter to compensate the monopole broadening by adjusting the hyperfine splitting, for one of the components of the gamma-ray line.

Figure 11(a) shows the contributions from monopole shift, rotation shift, and hyperfine splitting to an energy-level diagram for upper-state spin $I_2 = \frac{3}{2}$ and lower-state spin $I_1 = \frac{1}{2}$. In Fig. 11(b), we show the spectra of transitions between the levels of Fig. 11(a). The

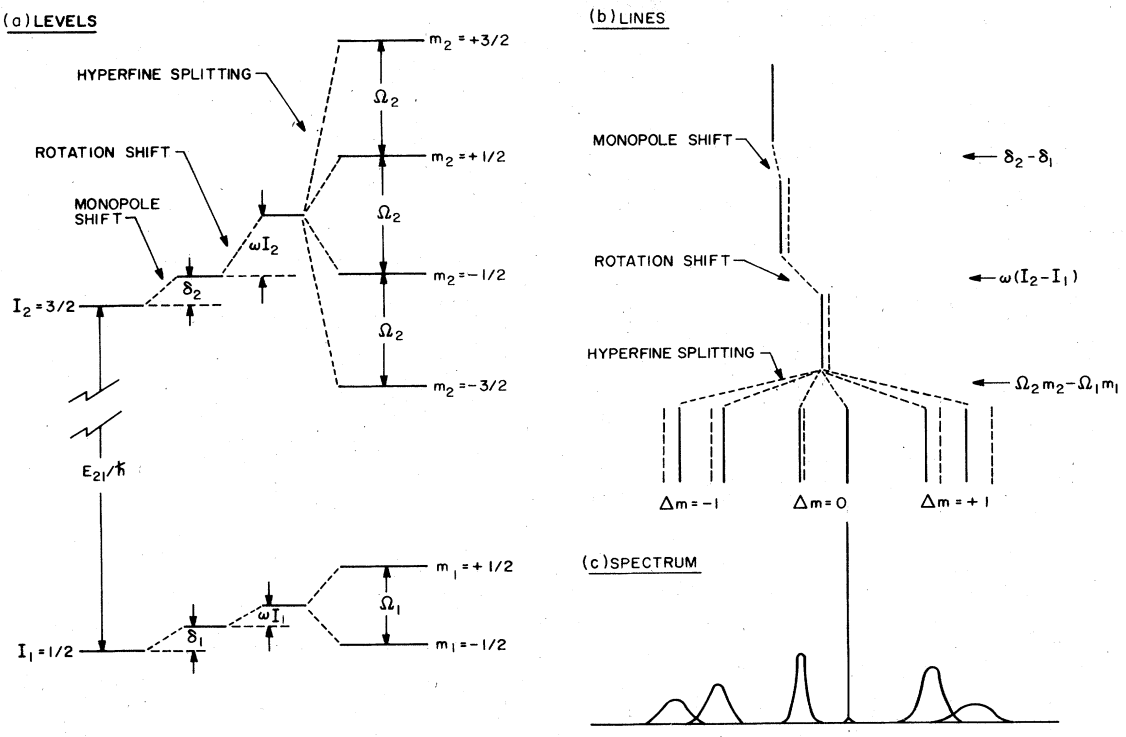

FIG. 11. Cancellation of monopole broadening. (a) Energy levels of a hypothetical isomeric nuclide ($I_2 = \frac{3}{2}$, $I_1 = \frac{1}{2}$) that is subjected to the line-narrowing operation proposed by Gol'danskii, Karyagin, and Namiot (1974), showing successively, on a frequency scale, the monopole shifts δ_i due to electron-nuclear electrostatic interaction, the rotation shifts $I_i\omega$, and the hyperfine splittings Ω_i in the effective magnetic field. (b) Relative positions of components of the recoilless radiation arising from allowed transitions between the levels of (a), for two slightly different values of the local S-electron density, represented, respectively, by solid or dashed lines. Note the unequal magnitudes of the two monopole shifts and of the spacings of the two sets of hyperfine components. Because of a judicious choice of parameters for the applied magnetic fields, acting in combination with the electron-spin-generated field, one of the $\Delta m = 0$ components is unshifted. (c) Mössbauer spectrum that would be observed under the above conditions in the presence of a random distribution of values of local S-electron density.

variables ω, θ, and $\mathcal{B}_{\rm eff}$ have been so adjusted that the change in monopole shift is exactly compensated by the change in hyperfine splitting for the $m_2 = +\frac{1}{2} \rightarrow m_1 = +\frac{1}{2}$ transition. Note that both levels are shifted; their respective shifts must be adjusted to achieve compensation by making the total difference in energy between one pair of their substates insensitive to local variations of electron density.

That part of the monopole shift of a chosen component of the gamma-ray line, caused by small local deviation from the average S-electron density, is $\delta_2 - \delta_1$; the local value of the contact field induction deviates from average by a proportionate amount,

$$\Delta\mathcal{B}_f = K(\delta_2 - \delta_1) . \tag{35}$$

The constant of proportionality K depends on the disposable parameters. Compensation of the two variations is accomplished when these parameters satisfy the following relation (Gol'danskii, Karyagin, and Namiot, 1974a, 1974b):

$$K = (m_2/I_2\Omega)(G_n\mathcal{B}_{\rm eff} + \omega\cos\alpha)_2 - (m_1/I_1\Omega)(G_n\mathcal{B}_{\rm eff} + \omega\cos\alpha)_1, \tag{36}$$

$$\Omega = [(G_n\mathcal{B}_{\rm eff})^2 + 2G_n\mathcal{B}_{\rm eff}\,\omega\cos\alpha + \omega^2]^{1/2}. \tag{37}$$

e. Combined line-narrowing methods

It has been asserted (Gol'danskii, Karyagin, and Namiot, 1974b) that the compensation method, having more than one disposable parameter which affects the hyperfine interactions at the control of the experimenter, should in principle be capable of simultaneously reducing all of the line-broadening interactions that arise from local variations of internal magnetic and electric fields.

Kagan has summarized a number of possibilities for compensating inhomogeneous shifts (1974a, 1975). They include adjustment of an external field in the case of nontransition metals, of an external field and temperature in the case of paramagnetic impurities, shifts from exact NMR frequency in more complicated cases, and resonant optical repopulation of electronic states in a transparent crystal. No experiments to test any of the above proposals have been reported.

f. Comments on line narrowing

Whether it may prove possible to reduce the inhomogeneous broadening by a large factor through application of principles that have been outlined awaits experi-

mental determination. It has been found possible, in favorable cases, to reduce the dipole-dipole broadening of nuclear magnetic resonance lines by factors approaching 1(4) (Rhim, 1976); there, however, only a single, stable ground state is involved. For the graser, experimental problems of magnetic field homogeneity, radio-frequency penetration and heating, sample geometry, etc. that limited the NMR work are magnified by radioactive decay of a highly inverted graser population.

Moreover, there is a natural limit to the degree of line narrowing that one could ever hope to achieve, because of radioactive decay. An initially inverted population loses inversion within one half-life (somewhat more or less, depending upon the statistical weight ratio of the two isomeric states); the narrowing process must have increased the stimulation cross section sufficiently for lasing to get under way well before this time. However, the time-frequency complementarity principle demands that, whatever the particular mechanism of narrowing employed to accomplish it, it will require at least as long as the reciprocal of the final linewidth it achieves. Thus it would be futile to attempt to approach the natural width, even if that were possible in practice. The proposals that suggest application of a cycle of $\pi/2$ pulses are further limited by the need for many cycles to elapse before averaging can be effective.

It is essential that some experimental effort be initiated in this field. First, however, one must overcome difficulties other than those associated with the design and conduct of the line-narrowing operation itself; in particular, the measurement of very narrow recoilless lines presents a major experimental challenge. The widely used Mössbauer technique, based on the first-order Doppler effect for scanning gamma-ray resonances, becomes utterly impracticable for lifetimes longer than about 10 ms, because the relative source-absorber velocities that must be generated and measured are so minute, of the order of mm h^{-1}. The absorbers must be thin and are therefore easily perturbed by extraneous sources of vibration.

Recently it has been proposed (Baldwin, 1979a) to make measurements in the time domain with stationary sources and filters, and infer the spectral distribution of the gamma radiation from its time dependence, rather than by scanning in frequency. There are both practical and fundamental questions raised by this proposal, which has yet to receive a definitive experimental test. One difficulty, pointed out when the time-domain approach was suggested, that of accidental coincidence background, can be resolved by use of statistical correlation methods (Baldwin and Gol'danskii, 1980). More serious is the question whether measurements solely of the time dependence of intensity can be transformed to determine a unique frequency dependence, without some simultaneous determination related to phase.

Until those or some other new technique of high-precision gamma resonance can be demonstrated, the proposals for line narrowing remain purely speculative, and their eventual role in graser development cannot be evaluated realistically.

It is important to realize, however, that narrowing of recoilless lines would be a boon to Mössbauer spectroscopy, whether or not a graser application materialized, if the corresponding improvement of spectroscopic technique could also be developed.

6. Conversion from long to short lifetime

There are proposals (Eerckens, 1969; Baklanov and Chebotayev, 1975; Kamenov and Bonchev, 1975; Rivlin, 1977a, 1977b; Arad, Eliezer, and Paiss, 1979) to transform a long-lived, radiochemically producible isomer into a shorter-lived species capable of lasing. Most of these would employ optical laser radiation to effect the transformation.

Suppose that a nuclide exists having two isomeric states separated by only a few electron volts, but with greatly different lifetimes (Fig. 12). Assuming the shorter-lived to be the higher level,

$$E_2 - E_3 = \Delta E \ll E_2, \tag{38}$$

$$\lambda_3^{-1} \gg \lambda_2^{-1}. \tag{39}$$

Let the longer-lived isomer be prepared, radio- or photochemically, in the form of a transparent crystalline filament, and subjected suddenly to an intense radiative pump at the wavelength corresponding to the energy separation of the two levels, so as to induce transitions between them. If the induced rate coefficient of the pump can be made sufficiently intense that

$$W_{23} \gg \lambda_2, \tag{40}$$

then this transition will saturate before the ground-state population will have accumulated appreciably, and nearly half of the initial population of long-lived states will have been transformed into an initially full population inversion of the shorter-lived transition. If the latter had a recoilless line, not too greatly broadened, and its other nuclear parameters were favorable, it could lase. The pump would replenish inversion from the number three-state reservoir until enough ground states had accumulated to terminate lasing.

The apparent advantage of this situation is that, instead of supplying the full excitation energy of level 2 at once, we need supply only the far smaller energy difference ΔE within a time short in comparison with λ_2^{-1}. The much larger remainder of the excitation energy would already have been furnished by a prolonged irradiation. We now estimate the pumping requirement.

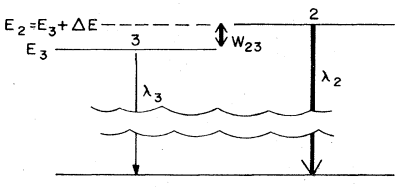

FIG. 12. Conversion from long to short lifetime. Energy level diagram for a fictitious nuclide having two isomeric states separated by only a few electron volts, but with greatly different decay coefficients $\lambda_2 \gg \lambda_3$. If the long-lived isomer, level 3, were prepared at high concentration and embedded in an appropriate host, graser operation might be initiated on the 2-1 transition by intense radiation saturating the 3-2 transition. Either level might be the higher in energy.

The induced rate coefficient is

$$W_{23} = \Phi_{23}\sigma_{23}, \tag{41}$$

where the photon flux density Φ_{23} of the fast optical pump is related to the power density in the optical beam by

$$\Phi_{23} = P/(e\Delta E). \tag{42}$$

The cross section for the pumping transition, by Eq. (5), is

$$\sigma_{23} = (\Lambda_{23}^2/2\pi)(\Gamma_r/\Gamma)_{23}. \tag{43}$$

Since we wish to saturate the 3-2 transition, we require that

$$W_{23} = [2.45(-15)P/(e\Delta E)^3](\Gamma_r/\Gamma) \gg \lambda_2, \tag{44}$$

so that the optical beam power flux must exceed

$$P \gg 6.5(-5)[(\Delta E)^3](\Gamma/\Gamma_r)\lambda_2 \quad \mathrm{W\,cm^{-2}}. \tag{45}$$

As usual, it is the linewidth factor that is crucial. If the 2-1 transition is to be capable of lasing, then, as we have seen in earlier sections,

$$\lambda_2 \geq 1(4)\cdot\ \mathrm{s^{-1}} \tag{46}$$

is desirable. The radiative width will be less than λ_2, since very-low-energy nuclear transitions are known to be highly internally converted [viz., the 73-eV isomeric level of ^{235}U decays entirely by internal conversion (Neve de Mevergniens, 1968)]. Finally, for the total width, we must use the Doppler breadth.

Thus, even if such an unusual near-coincidence of two levels of greatly disparate lifetime were found to exist, it is not at all certain that this would enable a graser to be developed. It is important that the possibility be recognized, however.

The first three proposals referred to differ in detail, but the basic idea for dividing the pumping process into slow and fast operations is a common feature of all of them. Three (Baklanov and Chebotayev, 1975; Rivlin, 1977b; Arad, Eliezer, and Paiss, 1979) liken the system to a Raman laser. However, the excited levels must both be real, since nonlinear response of a nucleus, analogous to that of Raman-active molecules (Baldwin, 1969; Bloembergen, 1965), cannot be expected at optical field strengths that can be generated by even the most intense optical lasers.

Eerckens's early proposal recognized that internal conversion would be the principal route for transition between levels 2 and 3. Nevertheless, he suggested (Eerckens, 1969) that a properly chosen radio- or optical-frequency pump could resonate this process, exciting the electron, which would then transfer its excitation to the nucleus. This assertion was not supported by any published analysis. More recently, Morita (1973) presented a quantitative theory for the excitation of nuclear levels by transitions in the electron shells, and Okamoto (1977) suggested that this be considered as a graser pumping mechanism. He did not, however, explain how electron transitions of nearly the same energy as the nuclear transitions to be stimulated were to be excited efficiently. The low $3 \rightarrow 2$ transition energy postulated in the lifetime-conversion scheme above would not present that difficulty.

It has long been known that internal conversion is sensitive to the electronic environment of the nucleus (De-Benedetti, Baros, and Hoy, 1966). Gol'danskii and Namiot (1976b) have predicted that inverse internal conversion can excite an isomeric nuclear state, in particular, the 73-eV isomer of ^{235}U. Observation of the effect has been reported (Izawa and Yamanaka, 1979).

The proposal of Kamenov and Bonchev (1975) assumes without proof that a two-step cascade from a long-lived isomer to the ground state via a short-lived intermediate state can be induced by radiation resonant with either step, and with a cross section essentially equal to the larger of the two cross sections for inducing the separate transitions.

7. Miscellaneous proposals

a. Mössbauer effect in a magnetized plasma

Winterberg (1979b) has suggested that an impulsively pumped graser could work at high temperatures and transition energies as high as several MeV if an extremely strong magnetic field were used. In his scheme, a high-current linear discharge would generate the field, which might be intensified by compression, and isomeric nuclei embedded in the plasma would emit without recoil in the direction parallel to the discharge.

The analogy to the Mössbauer effect postulated for this plasma configuration is based upon an assumed close correspondence between Alfvén waves in a plasma (Alfvén, 1950) and lattice vibrations of a solid. In the plasma, a recoil impulse directed normal to the field lines is opposed by the resultant of tension along the lines, and can thereby generate Alfvén waves.

Winterberg derives an expression for the fraction of recoilless emission events by calculating the density of modes of Alfvén waves in the plasma, noting that there is a maximum frequency for those waves, and formally identifying the frequency with the Debye cutoff frequency for lattice vibrations in a solid. Thus the plasma is assumed to have a well-defined Debye temperature, which is then used in the conventional formula, Eq. (13), for the recoilless fraction of a solid representable by a Debye continuum. The result suggests that there might be appreciable recoilless, Mössbauer-type radiation even for energies as high as 1 MeV and plasma temperatures of the order of 10 keV.

The neutron-pump limitation discussed in Sec. V.B.d and Appendix J applies to this proposal. There are more serious objections, however. The analogy of a highly anisotropic plasma to an isotropic Debye solid is untenable. A correct expression for the recoilless fraction cannot be based upon Eq. (13), which is a special form applicable only to a Debye solid. Strictly, in order to determine the recoilless fraction, all forms of ion motion should be quantized, a Hamiltonian function written for the isomeric ion, and the transition probability for gamma-ray emission unaccompanied by exchanges of quanta with any of the modes of ion motion determined. It is possible to make an estimate, however, by using the fundamental Debye-Waller relation, Eq. (12), according to which the mean square amplitude of ion vibration is related to the recoilless fraction by

$$\langle x^2\rangle = 4\pi^2\Lambda^2 \ln(1/f). \quad (47)$$

In the very dense plasma under consideration, ion motion is constrained by collective effects. It can be shown that the mean square ion amplitude is limited to a given value $\langle x^2\rangle$ when the ion density N_+ exceeds a value given by

$$N_+ \geq \frac{4\pi\varepsilon_0}{(Zed)^2}\left(\frac{3k\theta}{2\langle x^2\rangle} + \frac{h^2}{8Mm_1\langle x^2\rangle^2}\right), \quad (48)$$

where the coefficient d accounts for weakening of Coulomb internuclear repulsion by interaction with electrons: usually $0.05 \leq d < 1$ (Karyagin, 1980b).

The corresponding pressure p averaged over the cross section of the plasma column is related to the magnetic induction $\mathfrak{B}$ at the surface of the column by

$$p = (Ze)^2 N_+^{4/3}/4\pi\varepsilon_0 = \mathfrak{B}^2/2\mu_0 . \quad (49)$$

Table VI evaluates these relations for several sets of plasma parameters and compares them with values given by Winterberg. As expected, the values based upon Eq. (12) are several orders of magnitude higher than those obtained by Winterberg, who used Eq. (13) and considered only the Alfvén waves. Still, even were it possible to suppress all the other instabilities, the density and current requirements would be quite unrealistic at any plasma temperature likely to be obtained.

b. Phonon injection

The possibility is often discussed that one might increase the probability for recoilless emission by modifying the spectrum of lattice vibrations in the host, so as to increase the Debye-Waller factor. An early paper (Baldwin, Neissel, and Tonks, 1963) suggested that discrete, recoil-displaced lines in solids, which would not require population inversion for lasing, might be created by injection of phonons of a single frequency (Jacobsen, 1961), and that if their frequency were close to that equivalent to the free nuclear recoil energy, the change in f would be considerable, in consequence of the Lipkin sum rule [Appendix D, Eq. (D4)].

Phonon injection has been investigated, both theoretically (Abragam, 1960; Vorontsov and Vysotskii, 1974b; Sadykov, 1977) and experimentally (Ruby and Bolef, 1960). It frequency-modulates the gamma radiation, transferring a fraction of the energy from the recoilless line into a number of sidebands. These comprise pairs of equal amplitude, equally spaced from the central line.

That the cross section for stimulation would be affected favorably has been denied on grounds that the sidebands would be unacceptably broad because phonon lifetimes in solids (Vali and Vali, 1963b) are very short.

c. Nonlinear parametric interaction of optical and gamma radiation

Recognizing that a major stumbling block to creating a source of coherent gamma radiation is the excitation of the nuclei, Vysotskii and Vorontsov (1977) have proposed an entirely distinct approach that avoids this problem, based upon the parametric interaction of three electromagnetic waves in an optically transparent nonlinear medium. Such interactions are well known in the microwave and optical regions (Armstrong, Bloembergen, Ducuing, and Pershan, 1962).

In their ingenious proposal, an intense Mössbauer source pumps a nonlinear medium that has been excited optically beyond inversion of an optical-frequency transition. A weak "signal" wave is introduced at the difference of Mössbauer and optical-line frequencies. Although it was not stated, one presumes that the signal could be derived from the Doppler-broadened portion of the gamma radiation. Simultaneous interaction of atoms of the nonlinear medium with the pump and signal waves generates a component of the charge polarization of the medium that varies at their difference frequency, and this acts as a volume-distributed source of an "idler" wave at the optical frequency. The latter, propagating in the inverted medium, is amplified by stimulated emission. In turn, the growing idler wave interacts in combination with the Mössbauer pump to create nonlinear polarization sources at the signal wave frequency in the medium. In this way, it is said, energy, introduced at the frequencies of a Mössbauer and an optical transition, is converted into a coherent wave of increasing intensity at the signal frequency. The authors show, by a detailed analysis of the general problem of classical three-wave nonlinear interaction, that the threshold for the postulated process depends

TABLE VI. Parameters for recoilless emission from a magnetized plasma.

Parameter	footnote a	footnote b			
Gamma-ray energy E keV	1000[a]	1000			100
Recoilless fraction f	1.0	0.9			0.9
Mean square amplitude $\langle x^2\rangle$ m^2		6.6(−26)			6.6(−24)
Atomic number Z		80		1	30
Mass number M	200	200		1	60
Plasma temperature k keV	10	10	1	1	1
Ion number density N_+ m^{-3}	5(30)	3(34)	3(33)	2(37)	2(32)
Compressing pressure P J m^{-3}		1(22)	5(20)	1(22)	2(18)
Magnetic induction V s m^{-2}	1(5)	2(8)	4(7)	2(8)	2(6)
Column current in A for 0.1 mm radius		8(10)	2(10)	8(10)	1(9)

[a] From Winterberg (1979a).
[b] Calculated with Eqs. (47) and (48), with $d = 1$.

on the absorption coefficient for optical but not for gamma radiation.

The authors point out that certain rare-earth species (viz., Sm, Eu, Dy) happen to have not only readily invertable optical transitions, but also isomers that show the Mössbauer effect. Estimating the nonlinear susceptibility for such species, and considering the strong anomalous dispersion near the Mössbauer line, they conclude that, with a strong (1 Ci) Mössbauer source and a strong laser pumping at the optical frequency, a 100-fold gain might be realized in the signal intensity, with an active medium length of only 6–8 cm.

How the optical inversion is to be maintained, without being immediately dissipated through amplified spontaneous emission or lasing, was not indicated.

Although this proposal may not be feasible as presented, it suggests a possibly fruitful alternative approach to generation of coherent gamma radiation. Nonlinear polarization of nuclei and core electrons is unlikely, but loosely bound valence and conduction electrons do contribute second-order nonlinear susceptibility, owing to the $\mathbf{v}X\mathbf{B}$ term in the Lorentz force. This has been demonstrated both theoretically and experimentally (Freund and Levine, 1969; Eisenberger and McCall, 1971). A parametric amplifier also requires provision for sustained matching in phase of the interacting waves; for x rays, phase matching is accomplished by Bragg scattering, through the dependence of refractive indices on direction in the vicinity of the Bragg angle (Freund, 1968; Maier and Sukhorukov, 1979).

d. Use of polarized nuclei

In addition to proposals to employ magnetic alignment for line narrowing (Sec. V.B.5.c) or for isomer separation (V.B.4.c), there are two proposals that suggest its use for reducing excitation requirements. Essentially, the idea is to prevent lower-state nuclei from resonantly absorbing recoilless radiation that is emitted by the upper states (Carlson and Temperley, 1969).

An ensemble of nuclei having their angular momenta oriented along a preferred direction may be achieved either statically, through thermal relaxation in an intense magnetic field at low temperature, or by dynamic methods, as in the optically pumped atomic beam magnetic resonance (ABMR) measurements mentioned in Sec. V.B.4.c. Their gamma-ray emission is anisotropic, having an angular distribution peculiar to the multipole order of the transition and to the degree of polarization (Murnick and Feld, 1979).

One proposal (Wilson, 1977) suggests that nuclei be oriented during or immediately following their excitation, in such a sense that the probability of emission in the direction of the graser axis remains small until a critical density of excitation is reached. The axis of polarization is then rapidly rotated into coincidence with that of the graser rod. How the nuclei are to be pumped and simultaneously oriented was not considered.

A more detailed proposal (Vysotskii, 1979) would exploit selection rules for radiative transitions between magnetic substates of the nuclear levels, so as to reduce the excitation requirements by eliminating lower-state resonant absorption.

Vysotskii's proposal was illustrated by quantitative reference to a particular nuclide, ^{161}Dy, which has Mössbauer transitions from the following levels to the ground state (1):

level 4: $E = 103.07$ keV, $I = \frac{7}{2}^+$, $T = 0.5(-12)$ s,
level 3: $E = 74.57$ keV, $I = \frac{3}{2}^-$, $T = 3.3(-9)$ s,
level 2: $E = 25.65$ keV, $I = \frac{5}{2}^-$, $T = 2.9(-8)$ s,
level 1: $E = 0$, $I = \frac{5}{2}^+$, stable.

In the strong internal magnetic field (700 T) known to arise from contact interaction in this element, and at a temperature of 0.01 K or less, the ground state is statically polarized, essentially all nuclei being in its $m_{I1} = -\frac{5}{2}$ substate. Vysotskii proposes to pump them with characteristic x radiation ($K-\beta_2$) of Ra, 103.05 keV, which happens to agree very closely in energy with the 103.07-keV transition of ^{161}Dy. Invoking the selection rule for the magnetic quantum number, $\Delta m_I = \pm 1, 0$, he shows that only the $m_{I4} = -\frac{3}{2}$ substate of the 103-keV level will be excited appreciably, and that it will decay to populate the $m_{I3} = -\frac{1}{2}$ substate of the 74.57-keV level.

Assuming that the $m_{I2} = -\frac{1}{2}$ substate of the 25.65-keV level is not appreciably occupied, because the pumping radiation is not enhanced at that particular energy and because the static polarization of the ground state has left few nuclei in substates that can combine with it, Vysotskii concludes that lasing should be possible on the $3 \rightarrow 2$ transition, between substates $m_{I3} = -\frac{1}{2}$ and $m_{I2} = +\frac{1}{2}$, when the population of the 74.57-keV level is made to exceed the fraction σ_a/σ_s, approximately $6(-4)$, of the total atom population. To meet this requirement, he estimates that the spectral brilliance of the x-ray pump at the Ra K-β_2 line should be at least 1(11) photons cm^{-2} s^{-1} Hz^{-1}.

This analysis neglects the buildup of population in higher sublevels of the ground state by radiative relaxation while the 103-keV state is being pumped, the buildup of population in the relatively long-lived 25.65-keV level, and heating by nonresonant absorption of the pumping radiation. Assuming the Doppler breadth for 1000 K, approximately 3(13) s^{-1}, a single-line pump of 103-keV photons would have an intensity of the order of 50 Gw cm^{-2}; the actual intensity would be far higher, since one cannot generate the K-β_2 line without simultaneously exciting all the other line transitions that fill K-shell vacancies, together with continuous bremsstrahlung. These radiations all contribute to raising the temperature, so as to destroy both the Mössbauer conditions and the nuclear polarization.

Shortcomings of one specific suggestion should not discourage further development of this basic idea of exploiting the Δm_I selection rule. The difficulty with Vysotskii's specific proposal is that polarization is to *precede* the pump, which therefore must be radiative. The pump will inevitably destroy the polarization.

It is therefore pertinent to consider polarizing the nuclei *after* their excitation, as in the optically pumped ABMR experiments (Appendix K). We do not know whether orientation created in an atomic or ionic beam by an optical pump can survive the process of implantation into a solid to a depth sufficient for a sharp Mössbauer line. A more intriguing possibility

is to employ optical pumping to orient isomeric nuclei after they have been deposited within the Mössbauer host. In particular, the frequencies and senses of circular polarization of the optical radiation might be so chosen that upper- and lower-state nuclei were oriented simultaneously, but in magnetic substates of maximum $|m_I|$ of opposite signs (e.g., in ^{161}Dy, the level-2 nuclei might be in their extreme $+\frac{5}{2}$ substate, the ground-state nuclei in the $-\frac{5}{2}$ substate, so that their angular momenta would differ by five units). The lower states would then be incapable of resonantly absorbing gamma radiation emitted by the upper states. Not only would the inversion requirements thereby be drastically reduced, but, if this could be done quasicontinuously, inversion would be effectively maintained despite lower-state accumulation.

For this to be possible, the optical spectra of the active atoms in both nuclear states must have clearly resolvable hyperfine structures in the solid environment. This is known to be true of certain rare-earth ions in crystals (Crosswhite and Moos, 1967). Unfortunately, the lifetimes of excited optical states of rare-earth ions are longer than those of Mössbauer isomers, so that inversion would be lost before polarization became complete. Optical pumping must take place in a weak field (Zeeman effect of hyperfine structure) so that, since electronic and nuclear magnetic moments remain coupled, a torque applied by laser radiation to the electronic structures of the atoms can reorient the nuclei as well. The selection rules for Δm_1 apply in strong fields, however, so it would be necessary to increase the magnetic field rapidly before appreciable decay of the excited population or relaxation of the orientation had occurred. Whether this could be done cyclically, so as periodically to reduce the lower-state resonant absorption, has yet to be determined.

e. A low-temperature in situ graser

Karyagin (1980) has examined the feasibility of a low-temperature *in situ* graser that combines some novel features with previously proposed measures for lowering excitation requirements. Karyagin focuses on the vicious circle of inhibiting effects that cannot be defeated by increased pump intensity because they are also exacerbated by increased pump intensity; for example, heating by the pumping radiation, which reduces the Mössbauer fraction and prevents the use of polarized nuclei; or spontaneous decay, prethreshold lasing, and superfluorescent noise, all of which further heat the material. He proposes a combination of schemes that would alleviate these inhibiting effects with the intention of either breaking or even reversing this vicious circle.

First, he notes that the Borrmann effect (Appendix G.1) would allow the parent-host concentration ratio to be reduced to less than 10^{-5}. He refers to the utilization of the Borrmann effect in this context as "anomalous dilution." The effect mitigates pumping requirements not only because it reduces nonresonant absorption but also because it reduces spin-spin interactions between active nuclei, so that linebreadths on the order of 0.1 to 0.01 Hz may be achieved. Furthermore, low spin temperatures, on the order of 0.01 K, can be obtained, producing conditions favorable to polarization of nuclei.

Karyagin further proposes to separate, both in space and time, the phases of (1) preparation of excited isomeric states, (2) inversion or removal of active nuclei from the lower state of the amplifying transition, and (3) amplification by stimulated emission. Inversion is to be accomplished by either radio-frequency pulse separation or optical pulse separation. Nuclear polarization is essential to the radio-frequency scheme but not to the optical scheme. In the optical scheme, all the nuclei that reach the ground state are removed from resonance through the operation of selection rules, as discussed in Sec. V.B.7.d. The optical scheme maintains total inversion throughout the process of wave generation. Karyagin discusses various types of radio-frequency and optical schemes, delineating the advantages and disadvantages of each.

Karyagin proposes temporal and spatial separation of the three zones by a scheme for traveling-wave or "flowing-zone" pumping. He shows that the envelope of the gamma-ray wave train acquires the same asymptotic shape, regardless of its initial form. (This asymptotic traveling-wave pulse shape is generally called a "π pulse.") Then, by means of rapidly opening shutters, called "fast resonant switches," Karyagin proposes to isolate sections of the graser, and thereby eliminate premature lasing and reduce superfluorescent noise. These fast resonant switches would open to admit the π pulse as it passed down the active grasing medium. Finally, Karyagin shows that the active medium may be formed into a closed circle without increasing the losses, and thereby a resonator may be achieved (Vali and Vali, 1963b).

Karyagin concludes that the combination of all these measures would have the following advantages: (1) reduction of pumping threshold by three orders of magnitude; (2) reduction of the heat dissipation in the active medium by five orders of magnitude, in both direct and two-stage variants; (3) reduction of inhomogeneous linebreadth; (4) separate control of pumping and generation; (5) increased amplification; (6) controlled output, with various programed regimes of gamma-ray generation; and (7) increase in the number of candidate nuclides, which he has listed in the paper.

VI. PROGNOSIS

The quest for the gamma-ray laser resembles that for controlled nuclear fusion in many respects. Neither goal appears to be denied by any fundamental principle, both offer enormous rewards for success, both present challenging problems in many disciplines, and both have been maddeningly elusive. Both have been pursued for many years, each with an effort proportional to its generally perceived payoff. In each, just when a particular approach has been shown to be fruitless, another concept has emerged, and the quest has been resumed.

On the other hand, the scientific potentiality of the graser, which we believe is comparable to that of controlled fusion, was not as widely recognized until re-

cent laser developments began to focus attention on the x-ray region. The nuclei that will play important roles in fusion are known; in the graser, where nuclear parameters play much more involved roles, there are as yet no clearly identified candidates. Fusion's problems are largely those of plasma physics, the graser's of solid-state physics. The quest for fusion has been a steadily sustained effort; that for the graser has seen two periods of intense activity, each succeeded by a hiatus.

The first phase of graser research (Sec. V.A), emphasizing use of crystallized radiochemically prepared isomers, ended when the graser dilemma (Sec. III.F) and the inertia of resonance (Appendix C.2) were encountered. After a six-year hiatus, it became apparent that these difficulties might be circumvented by impulsive pumping (Sec. V.B.2) of short-lived isomers, by rapid separation (Secs. V.B.3 and V.B.4) of isomers of intermediate lifetimes, or by artificial control of the linewidth (Sec. V.B.5) in long-lived isomers. In the ensuing second phase, it has become apparent that (1) it is not possible to achieve gain on Doppler-broadened gamma-ray lines (Sec. III.C), i.e., the Mössbauer effect is an essential feature; (2) it is not possible to pump a graser directly *in situ*, since the only radiation that would not inevitably destroy the Mössbauer host (Sec. IV.D) cannot be generated in adequate intensity (Appendix J) from known sources; (3) line narrowing (Sec. V.B.5.e) will be extremely difficult and complicated, and there are reasons (Sec. V.B.5.f) to suspect that it may have only limited advantage.

These negative conclusions of the second phase are offset, however, by hopes raised by recent rapid progress in a number of technologies, viz., laser-based isotope separation, atomic beam hyperfine spectroscopy, nuclear polarization, production of defect-free solids, and the generation of coherent radiation in the soft-x-ray region.

Heretofore the electronic and nuclear approaches have had no common feature. They may change in the near future, since there is hope that radiation of sufficiently narrow bandwidth and high intensity may become available for pumping a statically polarized population, as a result of new approaches to the x-ray laser problem (Wood, 1980).

Recent work in atomic beam hyperfine spectroscopy has demonstrated the feasibility of isomer-selective optical pumping and separation of isomers based on magnetic moments—in weak currents.

Optical pumping offers the hope that a population of separately pumped nuclei can be not only rapidly concentrated, implanted, and annealed, but also polarized so as to eliminate lower-state absorption (Sec. V.B.7d).

We are convinced that an intermediate concentration step must separate the pumping and lasing stages, so that the best hope for the future lies in rapid separation and careful deposition in a host. Although it now appears that impulsive pumping *in situ* cannot by itself create an adequate density of excited states, and that the actual advantage of two-stage pumping (Sec. V.B.2c) is considerably less than one might have expected, some combination of approaches that individually cannot suffice to create perceptible gain may yet enable the goal to be reached. In particular, polarization approaches may relax the enormous pumping requirement. To prove them feasible, we must determine (1) whether it is possible to separate enough of the isomer (Sec. IV.C) and then implant it to sufficient depth without unacceptable damage or heating of the host; (2) whether a polarized population can indeed be created during either the pumping or the separation stage and, if so, whether it can then be implanted without destroying its polarization; (3) whether an isomer can first be implanted and then polarized by optical- or radio-frequency pumping so as to eliminate lower-state resonant absorption. To these basic questions must be added the practical question of the actual existence and identity of isomers that combine all essential nuclear, optical, and chemical properties.

Should these questions be answered in the negative, the line-narrowed long-lived isomer approach still remains. Again, a number of technical questions cannot yet be answered, viz, (1) Can a Mössbauer line be narrowed sufficiently? (2) What will be the effects of complementarity and radioactive decay on the degree of narrowing that can be achieved before loss of inversion (Sec. V.B.5.f)? (3) How will the width of the narrowed line be measured (Sec. V.B.5.f)? (4) To what actual transitions can these techniques be applied?

Despite the negative conclusions we have found for the more obvious proposals, there are more potential approaches to the graser problem today than ever before. Their technological bases are less secure, however, and the precise forms of systems that might implement them are not at all obvious.

Further progress toward the graser goal is therefore dependent upon research and technological development in many areas. In particular, we urge further study of the following subjects.

(1) Nuclear isomerism—level schemes, decay parameters, production reactions, and cross sections for destruction (as well as for production) of the isomer by the pump.

(2) Optical properties of atoms and of crystals containing isomeric nuclei.

(3) Hyperfine spectroscopy of atoms containing isomeric nuclei, to measure magnetic and quadrupole moments.

(4) High-yield isotope and isomer separation.

(5) Mössbauer investigations of the effect of optical pumping to orient nuclei in solid hosts.

(6) Techniques to extend Mössbauer spectroscopy to long-lived transitions, including development of defect-free hosts (probably by laser annealing) and development of time-correlation methods for measuring the linewidth.

(7) Techniques for rapid growth of isomer-bearing solids, including ion epitaxy and implantation, and observation of the effects of radioactive decay during growth.

(8) Thorough theoretical analyses of the kinetics of the graser system and of the dynamics of producing, separating and depositing, implanting, or crystallizing an inverted species, using realistic models and parameters.

(9) Devising unambiguous tests of lasing that are ap-

plicable to pulsed outputs of gamma radiation.

New proposals should consider the complete system, not simply a particular aspect. The authors hope that this review will assist in giving the perspective that this final suggestion requires.

The gamma-ray laser presents as difficult a challenge as any that Man has ever undertaken. It spans a vast range of disciplines and state-of-the-art technologies. We cannot truly say whether we are closer to the goal today than in the past, since it is not even possible to assert with certainty that the goal will ever be reached. We can say that no scientific principle has yet been shown to prohibit grasers, that the scientific and technological rewards of success would be immeasurable, and that undertaking difficult technological challenges has usually been fruitful. Many investigations suggested by the graser problem are useful for further extending the arts of Mössbauer spectroscopy, solid-state physics, nuclear physics, and quantum electronics, whether or not they finally lead to useful sources of coherent gamma radiation.

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APPENDIX A: CONVENTIONS, CONSTANTS, AND SYMBOLS

1. Conventions

Throughout this article, except where it is explicitly noted, we have used absolute SI units of time, length, and mass, but for convenience have expressed energies in keV or in eV and, usually, given cross sections in barns [$1(-24)$ cm^2]. We characterize electromagnetic radiation, ordinarily, by photon energy, except where explicitly noted, rather than by wavelength or frequency. Linebreadths are given in angular frequency units. The conversion factor to their equivalent energy, Planck's constant divided by 2π, is given in the next subsection.

Statistical weight factors for degenerate levels are applied to the populations of the nuclear states, as in laser practice, rather than to cross sections, the usual nuclear convention.

Powers of ten are given in the "floating point" notation; thus, $a.bc\ldots(d)$ denotes $a.bc\ldots\times10^{d}$.

2. Symbols and values of special units and constants

With the above conventions, the following approximate values of constants and conversion factors will be useful:

b	Unit of nuclear cross section	$1.00(-28)$ m^2
c	Vacuum velocity of electromagnetic radiation	2.998(8) m s^{-1}
e	Ratio of energy in absolute SI units to energy in keV	$1.602(-16)$ J keV^{-1}
g	Gravitational acceleration at sea level	9.806 m s^{-2}
h	Planck constant, ratio of energy to frequency	$4.136(-18)$ keV s
$\hbar$	Ratio of energy to angular frequency	$6.582(-19)$ keV s
k	Boltzmann constant, ratio of energy to temperature	$8.617(-8)$ keV K^{-1}
$E\Lambda$	Product of photon energy by wavelength	$1.240(-9)$ keV m
m_1	Mass of an atom of unit atomic weight	$1.660(-27)$ kg
	Energy equivalent to m_1	9.315(5) keV
ε_0	Permittivity of vacuum	$8.854(-12)$ C m^{-1} V^{-1}
μ_n	Nuclear magneton	$5.051(-27)$ A m^2
μ_0	Permeability of vacuum	$4\pi(-7)$ V s A^{-1} m^{-1}
G	Universal gravitational constant [in Eq. (E5)]	$6.670(-11)$ J m kg^{-2}
N_a	Avogadro number	6.023(23) mol^{-1}

3. Symbols and units for variables

A	Probability for a spontaneous radiative transition (identical with Γ_r); when used as a unit, denotes "ampere"	s^{-1}
$A(E)$	Probability for a spontaneous transition to emit radiation within a unit energy interval centered at photon energy E	s^{-1} eV^{-1}
B	Probability that a radiative transition be induced by resonance radiation of unit spectral energy density	J^{-1} s^{-2} m^3
C	Molar heat capacity of a host	J mol^{-1} K^{-1}
D	Diameter of a gas cloud	m
E	Energy of photon, neutron, or other particle	keV or eV
E_a	Energy of photons from an optical-frequency atomic transition	eV
E_{ij}	Difference of total energy of two nuclear eigenstates	keV
$E_{K,L\ldots}$	Energy to remove a $K, L\ldots$-shell electron	keV
E_s	Average energy of neutrons generated in a fission or fusion reaction	eV
F	Number of Fresnel zones in a diffracting aperture	
$F(v)$	Normalized probability distribution function for atomic velocities	m^{-1} s
G	Magnetic moment of a nuclear	

	state, divided by the nuclear spin, in units of the nuclear magneton μ_n	
$G(E)$	Normalized spectral distribution function of radiation per unit photon energy range	keV^{-1}
H	Hamiltonian function of a quantum-mechanical system	J
I_i	Total angular momentum, in units of $h/2\pi$, of the ith nuclear state	
I_{iz}	Projection of I_i on the direction of an aligning magnetic field	
I_n	Modified Bessel function of the first kind and nth order	
J	Current density of a beam of singly-charged particles	$A\,m^{-2}$
J_n	Bessel function of the first kind and nth order	
K	A proportionality constant	
L	A linear dimension	m
M	Mass number of a nuclide; in Appendix B, also designates a metastable state	amu
$M(E)$	Density of discrete normal modes of electromagnetic radiation per unit energy range	$m^{-3}keV^{-1}$
N	Number density of atoms or nuclei	m^{-3}
N_g	Molecular number density of a gas or vapor	m^{-3}
N_0	Number density of all atoms in a given medium	m^{-3}
N^*	Net, degeneracy-weighted excess number density of excited resonant nuclei ("inversion density")	m^{-3}
N_i	Number density of nuclei in the ith quantum state	cm^{-3}
N_+	Number density of ions in a plasma	m^{-3}
P	Power density of pumping radiation: in Sec. V.B.7.a, pressure in a pinched plasma	$W\,m^{-3}$ $J\,m^{-3}$
Q	Electric quadrupole moment of a nuclear state	b
R	Kinetic energy imparted to a nucleus initially at rest by the momentum of gamma-ray emission	eV
$\mathbf{R}_{ij}$	A vector extending from the ith to the jth of an assemblage of nuclei	m
$\mathbf{R}_0$	A unit vector parallel to vector $\underline{R}_{ij}$	
T	Effective lifetime corresponding to the sum of the total widths of two combining unstable levels	s
T_a	Average lifetime of an atomic level	s
T_i	Average lifetime for decay of the ith nuclear state	s
$T_{1/2}$	Half-life of a nuclear state	s
U	Kinetic energy of particles in a beam	eV
U_m	Interaction energy of a magnetic dipole moment with the local magnetic induction	J
U_ν	Spectral energy density (energy per unit volume in a unit range of frequency) of radiation, centered at frequency ν	$J\,s\,m^{-3}$
V	Active volume of the graser	m^3
W_{ij}	Probability rate coefficient for induced transitions between states i and j	s^{-1}
Z	Atomic number of an atom or nuclide	
a	Radial dimension of a graser host	m
a_n	Probability amplitude of the nth eigenstate of a quantum-mechanical system	$m^{-3/2}$
$\mathbf{b}$	Vector characterizing orientation and reciprocal spacing of a set of parallel crystal lattice planes	m^{-1}
b_{ij}	Numerical dimensional constants, relating energy to spin and magnetic field	J
d	A dimensionless coefficient	
f	Fraction of all radiative transitions between a pair of isomeric states that are not accompanied by phonon exchange with the host	
g	Acceleration due to local gravity	$m\,s^{-2}$
g_i	Statistical weight $(2I_i+1)$ of the ith nuclear state	
$\mathbf{k}$	A vector in the direction of phase advance of a wave, of magnitude $2\pi/\Lambda$	m^{-1}
m	Magnetic quantum number of a hyperfine state	
n	Density of particles (photons or neutrons)	m^{-3}
n	(As a subscript or hyphenated prefix) order of a sideband, Bessel function, etc.; quantum number of an eigenstate	
$\mathbf{n}$	Unit vector directed normal to a plane	
p	Ratio of inversion density N^* to total atom density N_0	
r	Radial dimension of a cylindrical or spherical body	m
t	Time coordinate	s
u_n	Wave function of a quantum-mechanical system in its nth eigenstate	$m^{-3/2}$
v	Velocity; e.g., drift velocity of ions, Mössbauer scan velocity	$m\,s^{-1}$
w	Ratio of statistical weights (degrees of degeneracy of two combining nuclear states): $w=(2I_2+1)/(2I_1+1)=g_2/g_1$	
x	A spatial coordinate; displacement of an atom from equilibrium position; distance measured along a beam	m
$\langle x^2\rangle$	Mean square displacement from equilibrium due to thermal agi-	m^2

	tation	
$\mathfrak{A}$	Vector potential amplitude of an electromagnetic wave	$V\,s\,m^{-1}$
$\mathfrak{B}$	Magnetic induction	$V\,s\,m^{-2}$
$\mathfrak{D}$	Electric displacement	$C\,m^{-2}$
$\mathfrak{E}$	Electric field intensity	$V\,m^{-1}$
$\mathfrak{H}$	Magnetic field intensity	$A\,m^{-2}$
$\mathfrak{L}$	Angular momentum of a photon in units of $h/2\pi$	
l	Mean free path between collisions	m
$\mathfrak{M}_{ij}$	Matrix element for transition between eigenstates i and j	J
$\mathfrak{P}$	Electric dipole moment per unit volume	$C\,m^{-2}$
$\mathfrak{P}_{ij}$	Matrix element of the electric dipole moment of a quantum system for transition between eigenstates i and j	A m
$\mathfrak{Y}$	Nonexponentially-varying part of the vector potential	$V\,s\,m^{-1}$
Γ	Total difference of angular frequency between half-intensity points of single-line radiation	s^{-1}
Γ_D	Angular frequency (radians) interval between half-intensity points of a Doppler-recoil broadened gamma-ray line profile	s^{-1}
Γ_g	Contribution of gravitational red shift to the total linebreadth	s^{-1}
Γ_{ij}	Sum of the widths of two nuclear levels, also called the "homogeneous width"	s^{-1}
Γ_r	Partial width of an energy level for radiative transition—identical with A	s^{-1} (unless stated as eV)
Δ	An increment; e.g.,	s^{-1}
	$\Delta\nu$ a frequency bandwidth	J or eV
	ΔU an energy expenditure	
Θ	Characteristic temperature of a Debye-model solid	K
Λ	Wavelength of electromagnetic radiation	m
Φ	Neutron flux (track length per unit volume and time)	$m^{-2}\,s^{-1}$
Φ_a	Flux of optical-frequency photons resonant with an atomic transition	$m^{-2}\,s^{-1}$
Φ_{ij}	Flux of γ-ray photons resonant with a transition between states i and j	$m^{-2}\,s^{-1}$
Ω	Angular frequency of precession or modulation	s^{-1}
α	Ratio of the probabilities of internal conversion and gamma emission	
β	Fraction of all decays from a given level that terminate on a second given level	
γ	Increase in the semibreadth of a gamma-ray line associated with inhomogeneous interactions	s^{-1}
δ	Magnitude of monopole shift of the ith nuclear level associated with a localized departure of S-electron density from average	s^{-1} or eV
ε	Ratio of the imaginary part of the atomic x-ray scattering amplitude at the Bragg angle to that for forward scattering	
ζ	A numerical measure of line asymmetry	
θ	Absolute temperature	K
η	Isomer ratio; fraction of all events in a particular pumping reaction that form an isomeric state	
κ	An exponential coefficient expressing combined temporal and spatial growth rates in an active medium	$m^{-1}\,s^{-1}$
λ_i	Probability per unit time for decay of the ith nuclear state	s^{-1}
λ_p	Probability per unit time for a pumping reaction in a parent nucleus	s^{-1}
μ	Magnetic moment ($G\mu_n I$) of a nucleus	$A\,m^2$
μ_e	Magnetic moment of an electron	$A\,m^2$
ν	Frequency of electromagnetic radiation	s^{-1} (Hz)
ξ	Net contribution to the inversion density of a single neutron capture; in Appendix J, the increment of lethargy associated with moderating collisions	
ρ	Fraction of incident pumping power that is absorbed (Sec. II.B.); density of host (Sec. III.F.)	$g\,cm^{-3}$
$\rho_{e,n}$	Electric charge density of an electron or nuclear charge distribution (Appendix E)	$C\,m^{-3}$
σ_a	Average cross section of a medium for absorbing or scattering a photon	m^2 or b
σ_{cs}	Total cross section per atom for Compton scattering	m^2 or b
σ_{cx}	Cross section for ion-neutral charge exchange	cm^2
σ_{exc}	Cross section for excitation of an atomic level by resonance absorption	b
σ_{ph}	Total cross section per atom for photoelectric absorption	m^2 or b
σ_s	Cross section for stimulated emission	m^2 (unless stated as b)
τ	Absorption coefficient ($N_0\sigma_a$)	m^{-1}
ϕ_{zz}	Axial component of electric field gradient	$V\,m^{-2}$
χ	Dielectric susceptibility	
ψ	Electron wave function of an atom	$m^{-3/2}$
ω	Angular frequency of radiation or of an applied magnetic field	s^{-1}

APPENDIX B: NUCLEAR ISOMERS

1. Nomenclature

The term "nuclide" designates a particular nuclear species, characterized by atomic, or proton, number Z and mass number A. "Isobars" are nuclides of different Z but with a common value of A; "isotopes," of different A but with equal values of Z. Each nuclide has a characteristic spectrum of excited states in addition to its lowest, the "ground state" (Fig. 3). The ground state may or may not be stable against beta decay; if not, it may emit an electron-neutrino pair or capture an atomic electron. The term "isomer" signifies an alternative configuration of a system. As applied to nuclei, it is often employed to designate those excited states of a nuclide that have noticeably long lifetimes, i.e., metastable states (see, for example, Segrè and Helmholtz, 1949; Goldhaber and Hill, 1952). We often use the qualifying words "long-lived" or "short-lived." In this paper, they relate to the Mössbauer effect (Appendix D), which is observed, by conventional methods, only in transitions to the ground state from certain isomers of lifetimes less than about $1(-5)$ s.

2. States of excitation

The various states of a particular nuclide are characterized by excitation energy E_i, probability of decay λ_i, angular momentum $I\hbar$, magnetic moment μ, and electric quadrupole moment Q. Their volumes are not, in general, equal. In their relaxation to lower states of the same nuclide, they may undergo branched decay, by gamma-ray emission or by internal conversion. Internal conversion (ejection of bound electrons) is followed by x-ray and Auger electron emission.

Excitation to higher states of a nuclide may result from gamma-ray absorption, Coulomb excitation, inelastic scattering of neutrons or charged particles, and electron-shell transitions. Nuclear isomers are also formed in nuclear transmutation reactions, in particular, by neutron capture or loss in an adjacent isotope. The binding energy of the captured neutron, of the order of 5–10 MeV, is released in a cascade of capture gamma radiation, with a photon multiplicity averaging 3–4 (Appendix J). In neutron-loss reactions, this energy must be supplied by the bombarding particle or photon.

Compilations (Lederer, Hollander, and Perlman, 1967; Lederer and Shirley, 1978; Nuclear Data Group, 1973) of nuclear data list the half-life $T_{1/2}$, excitation energy E, angular momentum I, ratio of magnetic moment to angular momentum (the "g factor"), and quadrupole moment of some isomers; and the energy E, branching ratio β (fraction of all nuclei making a particular transition), internal conversion coefficient α (ratio of bound electrons ejected to gamma-ray photons emitted) of some transitions. Usually it is more convenient to employ the mean lifetime,

$$T = T_{1/2}/\log_e 2 , \tag{B1}$$

or its reciprocal, the decay coefficient per unit time λ rather than the half-life in calculations.

3. Transitions

For an isomer that deexcites only by gamma-ray emission, the mean lifetime is the reciprocal of the Einstein A coefficient; the radiative width of the transition is the energy equivalent to this quantity. Internal conversion and branched decay to other states or to an isobar further reduce the lifetime of the isomer, by a factor $(1+\alpha)^{-1}$. These possibilities are indicated in Fig. 3.

Each of the excited states of a nuclide has an intrinsic energy width, related to its mean lifetime by the uncertainty principle. The gamma radiation emitted in an isomeric transition therefore is not precisely monochromatic (that would require an infinitely long wave train of constant amplitude), but has a spectral bandwidth given by the sum of the widths of the two combining levels

$$\Gamma_{21} = T_2^{-1} + T_1^{-1} \quad \mathrm{s}^{-1} . \tag{B2}$$

Although the detailed structure of nuclei is not yet known well enough to permit accurate calculation of radiation widths or lifetimes, order-of-magnitude estimates can be based on general principles that account for the observed range of lifetimes (Blatt and Weisskopf, 1952).

The nuclear radius, approximately $1.5(-15)\ A^{1/3}$ m, is small in comparison with the wavelengths Λ of gamma radiation, permitting the latter to be represented by a multipole expansion in powers of Λ^{-1}, each term corresponding to a particular value of the change in angular momentum and parity of the nucleus. The expansion converges rapidly, so that only the lowest-order terms are dominant. Allowed transitions conform to the selection rule

$$|I_2 - \mathcal{L}| \leq I_1 \leq (I_2 + \mathcal{L}) , \tag{B3}$$

where $\mathcal{L}$ is the difference in the angular momenta of initial and final states, carried by the emitted (or absorbed) photon. In the energy range under consideration, electric dipole transitions ($\mathcal{L} = 1$) are uncommon, in contrast to the atomic case. Isomeric pairs are known having very large spin differences and, correspondingly, very long lifetimes. Some typical properties of isomers useful in Mössbauer spectroscopy are given in Table I.

The competing process of internal conversion has been well studied theoretically, and tabulations exist of values of α for various multipole orders and parity changes of the nuclear transition (Way, 1973) in atoms of various atomic numbers. Internal conversion, being sensitive to the electronic structure of the atom as well as to the electric charge configuration of the radiating nucleus, is sensitive to chemical combination (Roggwiller and Kündig, 1975; Neve de Mevergniens, 1968).

4. Isomer formation

Nuclear isomers cannot be formed in appreciable yield by the process inverse to their radiative decay, when their isomerism is a consequence of the fact that a dipole transition to a lower state, and therefore its inverse, are forbidden. Moreover, gamma radiation emitted in isomeric decay of similar nuclei is not of

precisely the correct energy for resonance absorption by the ground state, owing to recoil shift. The Mössbauer effect is a conspicuous exception to these statements (Appendix D).

Isomer formation by absorption of x radiation from fast electrons has nevertheless been observed (Collins, Waldman, Stubblefield, and Goldhaber, 1939). The process is shown in Fig. 13. The radiation excites the nucleus to a level E, higher in energy than the isomer M, which is then formed by a prompt transition $E \rightarrow M$ (or $E'' \rightarrow E' \rightarrow M$). The electron energy must therefore exceed a threshold higher than the excitation energy of the isomer. The yields are very small because (1) only a very small fraction of a continuous spectrum of x radiation from fast electrons (typically of the order of 1 MeV) lies within the Doppler width of the absorption line (typically of the order of 0.1 eV); (2) the radiative width is usually less than the Doppler width; (3) the return transition from the initial level E to the ground state competes strongly with the transition $E \rightarrow M$. The theory of the process has been discussed by Guth (1941), with special reference to the excitation of ^{115}In by x radiation.

The process inverse to internal conversion—capture of an electron from the ionization continuum—can also occur under certain very special conditions, and it has been suggested (Gol'danskii and Namiot, 1976b) that the nucleus can be excited to an isomeric state by this mechanism; the conditions required are very difficult to realize, but the effect has been reported (Izawa and Yamanaka, 1979) for the 26-min 75-eV isomer of ^{235}U (Asaro and Perlman, 1957; Huizenga, Rao, and Englekermier, 1957).

It has also been suggested (Morita, 1973; Okomoto, 1977) that a nucleus can be excited into an isomeric state by transfer of energy of excitation in the electronic shells to the nucleus.

Of particular interest to the graser question is the formation of isomeric states by neutron interactions, either excitation from the ground state by inelastic scattering of fast neutrons or formation in the course of deexcitation of the compound nucleus formed by neutron capture (Fig. 3). Because the latter is accomplished with slow neutrons, it is the mechanism most commonly proposed for gaser pumping. (See Appendix J).

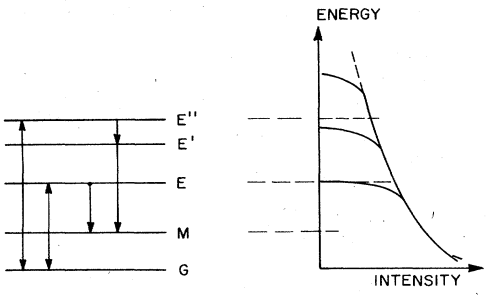

FIG. 13. Isomer formation by electron bremsstrahlung. On the left, some low-lying levels of a nuclide, including a metastable level M; on the right, spectra of bremsstrahlung from electrons of several different energies in thin targets. Absorption of x-ray photons of energies E, E'' is followed by cascades that promptly form the isomeric state M, provided the electron energy is high enough. Yields are low, both because only a small fraction of the intensity lies within the widths of absorption lines and because the continuous spectrum of bremsstrahlung consists mainly of low-energy photons.

From Fig. 3, it is apparent that the cascade of capture gamma radiation may, in general, produce both members of an isomeric pair of states. Therefore, if an inversion is to be produced, either the probability of forming the higher state must exceed that for forming the lower, or the latter must be of much shorter lifetime. The first alternative is preferable, because the cross section for stimulation of the transition is reduced when the lower state is unstable (Appendix C). The fraction of all captures that form the isomer, termed the "isomer ratio," has been investigated theoretically by Huizenga and Vandenbosch (1960). They assume that the initial compound state formed by neutron capture deexcites by a chain of electric dipole transitions; thus the probability of forming a particular intermediate state depends upon the difference between the angular momentum of that state and that of the state initially formed by neutron capture; the isomer ratio will be high if the spin of the upper isomeric state is the closer of the two to that of the capture state. Thus very high or very low isomer ratios are more likely to occur in the case of long-lived isomers, for which the spins of the two states differ by large amounts.

Table I lists a number of nuclear isomers that have been either considered as candidates for graser application or mentioned elsewhere in this article.

APPENDIX C: CROSS SECTION FOR INDUCED ISOMERIC TRANSITIONS

1. Asymptotic value

Consider a pair of combining nuclear levels, of which the lower is not necessarily the ground state.

Recoilless spontaneous radiative transitions that end on the lower level constitute a fraction $\beta f/(1+\alpha)$ of all upper-level decays. The two levels, if unperturbed, have energy widths $\hbar\Gamma_2$ and $\hbar\Gamma_1$, respectively, which, according to the uncertainty principle, are given by

$$\hbar\Gamma_i = \hbar/\Gamma_i \text{ keV}. \tag{C1}$$

The spontaneously emitted radiation therefore has an intrinsic or "natural" spread in angular frequency

$$\Gamma_{21} = \Gamma_1 + \Gamma_2 = T_1^{-1} + T_2^{-1} \quad \text{s}^{-1} \text{ keV}, \tag{C2}$$

which exceeds the partial width for recoilless radiative transitions

$$\Gamma_r = \beta f/T_2(1+\alpha) \quad \text{s}^{-1}. \tag{C3}$$

The probability per unit time that a photon will be emitted spontaneously with energy in the range (E, dE) is

$$A(E)dE = \Gamma_r G(E)dE \quad \text{s}^{-1}, \tag{C4}$$

where

$$G(E) = \frac{\hbar\Gamma}{(E-E_{21})^2 + (\hbar\Gamma)^2/4} \quad \text{keV}^{-1} \tag{C5}$$

is the normalized line-shape function (Heitler, 1944, Sec. 12), often called the Lorentzian function.[3]

This radiation is shared, with equal probability, by all of the

$$M(E)dE = \left(\frac{8\pi\nu^2}{c^3}\right)\left(\frac{d\nu}{dE}\right)dE \quad \text{m}^{-3} \tag{C6}$$

normal modes of the electromagnetic field (Jeans, 1921; Fowler, 1955) in the differential energy range dE. Thus the spontaneous emission rate into a single one of these modes is

$$A_1(E) = \frac{A(E)dE}{M(E)dE} = \frac{\Lambda^2}{4}\hbar\Gamma_r\, cG(E) \quad \text{m}^3\text{s}^{-1}. \tag{C7}$$

An important case is that of thermal equilibrium, for which the average occupation number of each mode is (Heitler, 1944, p. 108)

$$\langle n\rangle = [\exp(E/k\theta) - 1]^{-1} \quad \text{photons.} \tag{C8}$$

These quanta induce transitions in either sense at rates such that the population densities of the two interacting states at equilibrium are in the ratio

$$N_2/N_1 = (g_2/g_1)\exp(-E/k\theta) \tag{C9}$$

prescribed by Maxwell-Boltzmann statistics. For these relations to be consistent (Einstein, 1917), the resonant absorption probability from a single mode must be given by

$$W_a = (g_2/g_1)\langle n\rangle\, A_1(E) \quad \text{m}^3\text{s}^{-1}, \tag{C10}$$

and the stimulated emission probability into a single mode by

$$W_s = \langle n\rangle\, W_{21}(E) \quad \text{m}^3\text{s}^{-1}. \tag{C11}$$

The total rate coefficient for transitions from the upper to the lower state that contribute photons to a single mode is therefore

$$N_2W_{21} = (n+1)\, N_2A_1(E) \quad \text{s}^{-1}, \tag{C12}$$

while that for the reverse transition is only $N_1A_1ng_2/g_1$. These expressions contain no reference to the temperature.

Hence the induced rate in either sense between a single pair of nondegenerate states, due to a current nc of photons (which need not be within the same mode, so long as their energies are sensibly equal) is

$$W_{21} = nA_1(E) = nc(\Lambda^2/4)\hbar\Gamma_rG(E) \quad \text{m}^3\,\text{s}^{-1}, \tag{C13}$$

and, therefore, the apparent cross section for emission induced by photons having a sharply defined energy is

$$\begin{aligned}\sigma(E) &= nA_1(E)/nc \\ &= (\Lambda^2/4)\hbar\Gamma_rG(E) \\ &= \frac{(\Lambda^2/8\pi)\hbar^2\Gamma\Gamma_r}{(E-E_{21})^2 + (\hbar\Gamma)^2/4}\end{aligned} \tag{C14}$$

and, if the inducing radiation is at exact resonance with the transition,

$$\begin{aligned}\sigma(E=E_{21}) &= (\Lambda^2/2\pi)(\Gamma_r/\Gamma) \\ &= [2.446(-7)/E_{21}^2][\beta f\, T_1/(1+\alpha)(T_1+T_2)]\,.\end{aligned} \tag{C15}$$

Often, the inducing radiation is not of narrow bandwidth. If its intensity is practically constant over the extent of the spontaneous line, then the effective cross section for stimulation by a band of radiation of energy width ΔE centered at the resonance is

$$\sigma_s = (\Lambda^2/4)(\hbar\Gamma_r/\Delta E)\,. \tag{C16}$$

In the commonly encountered case of an inducing line having Lorentzian shape but of much greater breadth Γ than the nuclear line itself, one can still use the above formula, by taking for the equivalent square bandwidth the value

$$\Delta E/\hbar = 2\Gamma/\pi \tag{C17}$$

so that

$$\sigma = \frac{\Lambda^2}{8\pi}\frac{\Gamma_r}{\Gamma}\,. \tag{C18}$$

An inducing line equal in width but displaced in energy by ΔE, presents a reduced cross section that is essentially a Lorentzian function of ΔE, but an apparent width twice that of the natural line. This is seen, for example, in Mössbauer spectroscopy (Appendix D) using natural-line sources and absorbers.

2. Time dependence of the cross section

The induced transition cross section deduced in Appendix C.1 was obtained by considering an equilibrium situation, in which dynamic aspects of the transition probability were not considered. We therefore refer to it as the "asymptotic cross section." It contains the linebreadth Γ as a factor, a quantity which requires finite time for its determination. The act of transition is not instantaneous and, while it is occurring, the field intensity and excited-state populations change (Chirikov, 1963).

Classically, a resonant system driven by a periodic perturbation builds up forced oscillations slowly, to saturate eventually at an amplitude and time determined by the damping constant, i.e., by the sharpness of resonance.

For the nuclear analog, we must resort to time-dependent quantum-mechanical perturbation theory (Heitler, 1944, Sec. 11). The nuclear system, with unperturbed Hamiltonian H_0, stationary-state wave functions $u_n(\mathbf{r})$, and eigenenergies E_n, is assumed to be perturbed by a monochromatic electromagnetic plane wave of vector potential $\mathfrak{A}(t)\exp(i\omega t)$ and frequency ω. The wave function of the perturbed system satisfies the Schrödinger equation

$$i\hbar\dot{\psi} = [H_0 + \mathfrak{A}\cdot\dot{\mathfrak{P}}\exp(i\omega t)]\,\psi\,, \tag{C19}$$

where $\mathfrak{P}$ is the internal polarization current of the nucleus.

Expanding ψ in the set of unperturbed wave functions and invoking their orthogonality, we obtain differential

[3] This same functional dependence characterizes other resonant reactions, e.g., resonant neutron scattering or capture. It is then called the Breit-Wigner function.

equations for the amplitudes a_n of the expansion:

$$\dot{a}_k = (i/\hbar) \sum_n a_n(t) \mathfrak{M}_{kn} \exp[i(\omega_{kn} - \omega)] , \tag{C20}$$

where

$$\omega_{kn} = (E_k - E_n)\hbar \tag{C21}$$

is the apparent frequency of transition between states k and n, and $\mathfrak{M}_{kn}$ is the matrix element of the perturbation Hamiltonian. Since the size of the nuclear system is small compared with the wavelength of gamma radiation, the matrix element is factorable:

$$\mathfrak{M}_{21} = \mathfrak{A} \cdot \mathfrak{P}_{21} \tag{C22}$$

for the two-level system under consideration. The amplitude $|\mathfrak{A}|$ of the wave field is related to its Poynting energy flux by

$$ncE = |\mathfrak{A} X (\nabla X \mathfrak{A})/\mu_0|$$
$$= (\omega|\mathfrak{A}|)(2\pi|\mathfrak{A}|/\Lambda\mu_0) , \tag{C23}$$

where n is the photon density.

The cross section is defined as the ratio of induced transition probability to the incident photon flux nc:

$$\sigma_s = \frac{1}{nc|a_2|^2} \left(\frac{d}{dt}|a_1|^2\right). \tag{C24}$$

In the usual "small-signal" approximation, one solves the two-level amplitude equations by assuming $\mathfrak{A}(t)$ to be independent of the time (i.e., that the externally applied field is unaffected by the nuclear transitions). The initial conditions are

$$a_2(t) = \exp(-\lambda t/2) , \tag{C25}$$

$$a_1(0) = 0 . \tag{C26}$$

The result is then integrated over the Fourier spectrum of $\mathfrak{A}$ and evaluated at times long compared with $2\pi/(\omega_{21} - \omega)$, leading to the well-known Fermi "golden rule" for the transition amplitude. The cross section, so found, is identical with that found in the previous section.

When, as in a laser device, the field is built up by a chain of induced transitions from an initial spontaneous event, both the wave amplitude and the cross section are time dependent. If we wish to describe the initial kinetic behavior of a graser system prepared in a state of full initial inversion, we begin by taking

$$a_1(0) = 0 , \tag{C27}$$

$$a_2(0) = 1 , \tag{C28}$$

and obtain, for exact resonance,

$$\sigma(t) = \frac{2\pi\mu_0\Lambda|\mathfrak{P}_{21}|^2}{|\mathfrak{A}(t)|} \int_0^t \mathfrak{A}(t')dt' . \tag{C29}$$

This result (Chirikov, 1963; Fig. 6) is valid only for times short compared with the reciprocal linebreadth. When $\mathfrak{A}(t)$ is large and constant, it corresponds to the undamped classical case, in which the amplitude of resonantly driven oscillation increases linearly with time.

An exact formulation of the complete time dependence of the cross section therefore requires knowledge of the history of the radiation field, which depends partly but not entirely on the time dependences of the cross section and of the excited-state populations, since each may be changed by external interactions (e.g., photon removal by nonresonant absorption, excitation of the upper state by the pump). The finite bandwidths of the perturbing radiation and of the nuclear resonance (corresponding to the classical damping constant) also must be properly accounted for.

When, as in the graser problem, operation must be with a line of nearly natural breadth (since otherwise the balance relation cannot be satisfied), the cross section cannot develop rapidly. During its development, both the radiation intensity and the nuclear excitation can change.

It is therefore preferable to avoid the cross section formulation in time-dependent problems. In laser theory, the photon occupation numbers of the various modes and the populations of the combining states are related by coupled differential equations. These are considered in Appendix I.

APPENDIX D: THE MÖSSBAUER EFFECT

1. Recoil from gamma-ray interaction

When an unbound nuclear isomer deexcites by emitting a gamma ray, the nucleus must recoil to conserve momentum. To conserve energy as well, the energy of the gamma ray must be less than that of the total transition, by the kinetic energy of the recoil of the nucleus. For a nucleus of mass number M initially at rest (see Appendix E), the latter is given by

$$R = 5.32(-4)\, E^2/M \quad \text{eV} . \tag{D1}$$

For nuclei not initially at rest, the recoil energy differs from R. In particular, it can be readily shown that, given a Maxwellian distribution of initial energies characterized by absolute temperature θ, the recoil energies are distributed normally about R with standard deviation $\sqrt{Rk\theta}$. The line is then said to be "Doppler broadened."

2. Recoilless interaction

If, however, the nucleus is bound in a solid, the recoil impulse is given to a system having a discrete vibrational spectrum, characterized by the various atomic masses, the structure of the crystal, and the strengths of the bonds between the atoms. The emission (or absorption) impulse may or may not result in a change of the occupation numbers of the vibrational states.

Such no-phonon events are termed "recoilless," although the recoil momentum is actually taken up by an appreciable portion of the entire solid, whose excitation involves motion of a very large number of atoms, the spatial distributions of whose wave functions remain fixed with respect to one another. Since imparting recoil momentum to this block involves a much smaller transfer of energy than in the free-nuclear case, the gamma-ray photon appears with almost exactly the energy of the isomeric transition and, in many cases, with essentially the natural linewidth.

Although there is no Doppler broadening of the re-

coilless component in first order, there is a second-order Doppler effect, manifested as a temperature-dependent shift of the resonance (Pound and Rebka, 1960a). This is often ascribed to the change of nuclear mass in the emission event (Josephson, 1960); the mass change is accompanied by a corresponding change in the mean square velocity of thermal agitation, and therefore, of the second-order Doppler term. In a solid with $3N$ degrees of freedom, the shift is

$$\frac{1}{E}\left(\frac{dE}{d\theta}\right)=\frac{3Nk}{2NMm_1c^2}=\frac{3k}{2Mm_1c^2} \tag{D2}$$

in the temperature region of constant specific heat; at low temperatures, according to the Debye theory, this is reduced by the factor $(\theta/\Theta)^3$.

3. The recoilless fraction

The probability f that the occupation numbers of lattice states will not change (i.e., that no phonon will be emitted or absorbed) is related to the ratio of the wavelength of the gamma ray to the root-mean-square amplitude $\langle x^2\rangle^{1/2}$ of vibration:

$$f=\exp(-4\pi^2\langle x^2\rangle/\Lambda^2)\,, \tag{D3}$$

a function of the absolute temperature of the system.

The fraction f of all emission or absorption events in which no phonon is exchanged with the solid, called the Lamb-Mössbauer factor (Lamb, 1939; Mössbauer, 1958), is closely related to the Debye-Waller factor of x-ray diffraction theory (James, 1950). Its precise value [Eq. (D3)] depends upon the lattice temperature and the spectrum of lattice vibrations (Visscher, 1960). It has been shown that the expectation value of the energy transferred to the solid is just the free-nuclear recoil energy (Lipkin, 1960), i.e.,

$$f\cdot 0+(1-f)\langle\Delta E\rangle=R\,, \tag{D4}$$

and, therefore, f will approach unity if the lattice is sufficiently rigid that phonons very energetic in comparison with R can be exchanged. For a lattice that can be described by the Debye model, with a characteristic temperature Θ, the recoilless fraction at temperature θ is given by

$$f=\exp\left\{-\frac{3R}{2k\Theta}\left[1+\left(\frac{2\theta}{\Theta}\right)^2\int_0^{\Theta/\theta}\frac{x\,dx}{e^x-1}\right]\right\}. \tag{D5}$$

For a pronounced Mössbauer effect, it is therefore necessary that the gamma-ray energy be low enough that the energy of free recoil R be not large in comparison with the Debye energy of the lattice (Fig. 14).

In many solids, the Debye model is not a good approximation, except for a crude estimation of f, even though it is quite satisfactory for explanation of the temperature dependence of the specific heats of solids. This is because the majority of phonons exchanged in heat transfer are of much lower energy than in the case of gamma-ray emission (Lipkin, 1960). The Debye model formulas are especially untrustworthy for diatomic solids, characterized by optical branches (Kagan and Maslov, Table 3-2 1961). Thus the recoil-free fraction must be found experimentally. Often, an effective Debye temperature may be reported which differs from that found from other methods (Frauenfelder, 1962).

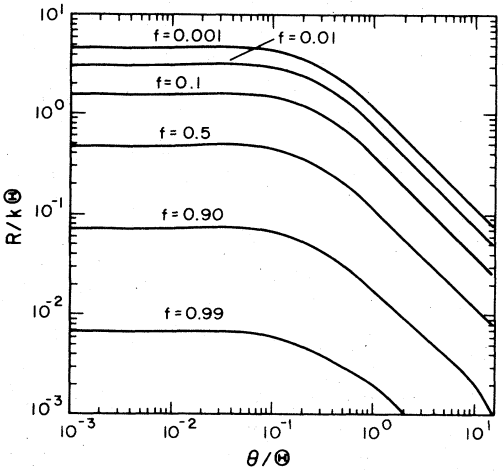

FIG. 14. Recoilless fraction. Contours of constant recoilless fraction f for a Debye-model solid are shown as functions of the free-recoil energy $R/k\Theta$ and the lattice temperature θ, both in units of Debye temperature Θ.

4. Mössbauer spectroscopy

Experimentally, the Mössbauer effect is manifested by observation of strong resonance absorption of gamma radiation from isomer sources in a solid by ground states of the isomer in a second, solid body. The source isomer is formed by radioactive decay. In order to demonstrate resonance, the absorber is given an adjustable velocity, of the order of a few mm s^{-1} relative to the source, and the intensity of the transmitted gamma radiation is determined with a suitable detector as a function of the relative velocity. By this means, the emission spectrum of the source is progressively displaced in energy with respect to the absorption spectrum of the absorber, through the first-order Doppler shift. When both spectra are single lines of natural width, the resulting transmission function is a superposition of the spectra of emission and absorption by nonresonant processes—a Lorentzian function of width twice that of the transition. In general, internal fields in either or both of the solids can displace, split, or broaden the observed resonance (Appendix E). Thus the Mössbauer effect has provided a powerful probe for the investigation of the internal structures of solids. Some typical Mössbauer isomers are listed in Table I.

For further information, the compilations of Stevens and Stevens (1976) should be consulted.

5. Relevance to the graser

The resonance cross sections that are measured in Mössbauer spectroscopy often are found to be far greater than the nonresonant absorption cross section, particularly when the recoilless line is nearly of natural width (Table I). Thus, if the absorber foil could be replaced by a foil containing an inverted population of the isomer, amplification instead of absorption would occur.

APPENDIX E: LINE BROADENING

1. Types of broadening

The total linebreadth Γ in the formulas of Appendix C is applicable to every transition in the ensemble of identical nuclei. In general it exceeds the radiative breadth of the upper state, and therefore the stimulation cross section is less than $\lambda^2 f/2\pi$. For the purposes of this paper we distinguish three types of broadening: (1) Homogeneous broadening, (2) Doppler broadening, and (3) Inhomogeneous broadening.

Homogeneous broadening occurs (1) if the lower state is unstable, so that its energy, according to the uncertainty principle, is not precisely defined; (2) if there are alternative ways for the upper state to deexcite, such as internal conversion, branched radiative decay to another lower level, or beta decay; or (3) if the lifetime of the transition is so long compared to its energy that its natural linewidth is determined by a fundamental length—should such a quantity exist (Mead, 1966).

Doppler broadening results from thermal vibrations even in a perfectly homogeneous lattice.

Inhomogeneous broadening occurs when the various nuclei are subject to perturbing fields that act unequally. A distribution of transition energies, frequently Gaussian, is mathematically equivalent to a situation in which the inducing radiation, rather than the transition energy, is spread out into a spectrum as described in Appendix C. Since a graser can tolerate only a modest amount of line broadening, we can still regard an inhomogeneously broadened line as having approximately the Lorentzian form and use Eq. (5) for the effective cross section.

2. Homogeneous broadening

a. Unstable lower state

The homogeneous ("natural") linewidth is simply the sum of the widths of the two combining energy levels (Heitler, 1944, Sec. 12) related, through the Heisenberg principle, to their mean lifetimes:

$$\Gamma_{21} = \Gamma_2 + \Gamma_1 = T_2^{-1} + T^{-1} \quad \mathrm{s}^{-1}. \tag{E1}$$

Note that we use angular, rather than cyclic, frequency. The energy equivalent is found by multiplication by Planck's constant $\hbar$ (Appendix A).

Tabulations of nuclear data usually list half-lives rather than average lifetimes:

$$\Gamma_{21}(\mathrm{s}^{-1}) = (\log_e 2)/T_{1/2}(s). \tag{E2}$$

b. Alternative deexcitation

We are interested primarily in the ratio of the radiative width of the upper state to the total width of the line, rather than in their absolute values. The radiative width Γ_r is the product of the fraction of all decays of the upper level that lead to the lower (called the branching ratio β), the fraction of all such transitions that proceed by gamma radiation rather than by internal conversion, and the total probability of decay per unit time of the upper state:

$$\Gamma_r = \beta/(1+\alpha)T_2. \tag{E3}$$

Accordingly, the line-broadening factor for a homogeneously broadened transition is

$$\frac{\Gamma_r}{\Gamma} = \frac{\beta}{T_2(1+\alpha)}\left(\frac{1}{T_2}+\frac{1}{T_1}\right)^{-1} = \left(\frac{\beta}{1+\alpha}\right)\left(\frac{T_1}{T_2+T_1}\right). \tag{E4}$$

c. Fundamental length

Mead (1964) has suggested that the infinite divergences of quantum field theory could be eliminated by postulating an intrinsic indeterminacy in the measurement of position ("quantum of length") having the value

$$(G\hbar/c^3)^{1/2} = 1.6(-35) \quad \mathrm{m}. \tag{E5}$$

He has given a number of arguments, based upon *Gedankenexperimente* to support this hypothesis.

In another paper (Mead, 1966) he argued that the existence of this fundamental length would be manifested by corresponding uncertainties in the energy of quantum states and, in particular, of the frequency of gamma radiation. The relative broadening was predicted to be of the order of the ratio of the fundamental length to the radius of the radiating system; for gamma radiation, this would be of the order of 1(−20), provided one grants Mead's assumption that the radiating system is a single nucleus. A more realistic viewpoint views the entire crystal lattice as the source of Mössbauer radiation; the broadening would be far less under that assumption.

Fundamental-length broadening, if it does exist to the extent Mead assumes, should exceed the natural breadth of radiation from transitions for which the product of photon energy in keV multiplied by lifetime in s is of the order of 100 or more. No presently observable Mössbauer transition satisfies this criterion, so his hypothesis is as yet untested. Mead has shown that the effect he predicts might be observable for the 56-m, 40-keV radiation from ^{103}Rh, in the absence of other broadening.

Mead's analysis has been criticized for using the first power, rather than the square of the ratio of fundamental length to system dimension in estimating the broadening (Sharp, 1980). With this correction, fundamental length ceases to contribute significantly to line broadening.

3. Doppler broadening

a. First-order Doppler breadth

The fraction of all nuclei in a gas at temperature θ, having line-of-sight components of velocity in the range (v, dv), is

$$F(v)dv = \left(\frac{Mm_1}{2\pi k\theta}\right)^{1/2} \exp\left(-\frac{Mm_1 v^2}{2k\theta}\right) dv. \tag{E6}$$

When a nucleus with this velocity component emits or absorbs a gamma ray of energy E keV, receiving an increment of momentum $\pm E/c$, its energy is changed by an amount

$$\Delta E = Ev/c \pm E^2/2Mm_1c^2 \tag{E7}$$

$$= Ev/c \pm R \tag{E8}$$

at the expense of the photon energy. R is the energy of recoil of a nucleus initially at rest; the positive sign applies in the case of absorption. Since v has the distribution (E6), the energy shifts are distributed normally about $\pm R$. The mean square dispersion is

$$\langle(\Delta E \mp R)^2\rangle = \int_0^\infty (Ev/c)^2 F(v)dv = Rk\theta\,, \tag{E9}$$

and the full width between half-intensity points of either distribution is (in energy units)

$$\hbar\Gamma_D = 4\sqrt{\log_e 2}\,\sqrt{Rk\theta}\,, \tag{E10}$$

called the "Doppler breadth."

In the optical region, where $\hbar\Gamma_D \gg R$, the Doppler-broadened emission and absorption lines of a transition overlap at any practicable temperature, but this is not necessarily true in the gamma region. Whenever $\hbar\Gamma_D < 2R$, they do not appreciably overlap, so that population inversion would not be necessary for stimulated emission (Marcuse, 1963). Figure 4 shows both cases.

In a solid, the effect of a recoil impulse depends upon the spectrum of lattice vibrations. Although the Doppler-broadened line profile is not of the same form as for a gas, the spread of energies is of the same order of magnitude (Visscher, 1960). The most significant difference of form is the appearance of the recoilless (Mössbauer) line.

b. Second-order Doppler effect

The time average of the thermal velocity of the emitting nucleus is practically zero when the nucleus is bound to a solid. However, the square of the thermal velocity does not average to zero. Thus there is a second-order Doppler shift of the resonance toward lower frequencies as the temperature of the host is increased (Pound and Rebka, 1960a). This was discussed in Appendix D.

4. Mechanisms of inhomogeneous broadening

a. Temperature-gradient broadening

The second-order effect shifts the gamma-ray energy by an amount proportional to the absolute temperature; hence, in the presence of a temperature gradient, the gamma resonance energy depends on the nuclear location and the line is therefore broadened. If the range of temperature variation over the host is $\Delta\theta$, then the breadth of a gamma ray of E keV is increased by

$$\Gamma_{\Delta\theta} = 3.4(4)(E\Delta\theta/M)(C/N_a ke) \quad \text{s}^{-1}. \tag{E11}$$

Temperature is a statistically defined quantity; the exact amount of the second-order Doppler shift is thereby subject to fluctuation. According to Kagan (1965), this contributes broadening; in an ideal crystal,

$$\Gamma_\theta = 1.3(E/M)^2(\theta/\Theta)\theta \quad \text{s}^{-1} \tag{E12}$$

at normal temperatures and

$$\Gamma_\theta = 4.7(3)(E/M)^2(\theta/\Theta)^6\theta \quad \text{s}^{-1} \tag{E13}$$

at extremely low temperatures. Although this is not significant in the Mössbauer range, and therefore has not been reported experimentally, it can greatly exceed the natural linewidth for long-lived transitions.

b. Electrostatic hyperfine interactions

The Mössbauer line derives from those events that do not exchange phonons with the lattice of the host. In a perfect solid with no internal or applied fields, it would have the homogeneous breadth, and for many short-lived transitions this is observed, in fact (Table III). Long-lived transitions, on the other hand, always show some additional width. For example, the narrowest line reported for ^{181}Ta (Table IV) is eight times wider than the natural width (Dornow, Binder, Heidemann, and Kalvius, 1979), a fact ascribed to several causes, all indicative of local variations in the nuclear environment provided by the host. These variations are of (1) electron density, caused by physical defects or chemical impurities, (2) magnetic induction, arising from thermally disordered electronic and nuclear magnetic moments, and (3) electric field gradients.

Site-to-site variations in the electrostatic interaction of electrons with the nuclear charge density include those called "monopole," "isomer," or "chemical" shifts, which are independent of the nuclear angular momentum, together with those that are dependent upon its orientation with respect to the local electric field or its derivatives.

The magnitude of the total electrostatic electron-nuclear energy is given formally by (Dunlap and Kalvius, 1978)

$$U_i = -e^2 \int\int \frac{\rho_e(\mathbf{r}_e)\rho_n(\mathbf{r}_n)}{\mathbf{r}_e - \mathbf{r}_n}\, d\tau_e\, d\tau_n\,, \tag{E14}$$

where integration is extended over the entire volume of the solid. Expansion of the denominator in spherical harmonics produces a series of terms, describing, respectively, the monopole, quadrupole, and higher (even)-order electrostatic interactions (Ramsay, 1955).

Since the nuclear volume and shape in general may differ for two isomeric states, the energy available for the emitted recoilless photon differs from the total energy of the transition by an amount that depends on the local value of the electronic charge density or, equivalently, of the electric potential it creates within the nuclear volume.

Assuming the nucleus to be approximately spherical, but to change radius r in the isomeric transition, the Coulomb interaction energy is changed by

$$\Delta E_c = (1/10\varepsilon_0)Ze^2\rho_e(0)(\Delta r_{21})^2\,, \tag{E15}$$

where

$$(\Delta r_{21})^2 = \langle r_2^2\rangle - \langle r_1^2\rangle \tag{E16}$$

is the difference in the mean squares of the two nuclear radii. The electron density ρ_e, evaluated at the nucleus, is proportional to the square of the normalized S-electron wave function. Variations of this quantity arise from lattice defects (which alter the volume of the unit cell) and from impurities (which also can donate or accept electrons). Vacancies in inner electron shells

should also affect the interaction. The resultant monopole, or chemical, shift between two nuclei is approximately

$$\Delta^2 E_c = (1/10\varepsilon_0) Z e^2 \Delta\rho_e(0)(\Delta r_{21})^2 . \quad \text{(E17)}$$

A more precise formula has been given by Bodmer (1961). A distribution of values of $\Delta\rho_e(0)$ leads to inhomogeneous broadening.

The second nonvanishing term of the expansion of (E9) is a scalar product of the quadrupole moment Qe of the nuclear charge distribution with the gradient of the electric field at the nuclear site, found by operating twice on the electrostatic potential. The resulting nine-component tensor is reduced by crystal lattice symmetry: in the special case of axial symmetry the interaction energy is

$$\Delta E_q = eQ(3\cos^2\theta - 1)\phi_{zz}(0) , \quad \text{(E18)}$$

where ϕ_{zz} is the axial component of electric field gradient and θ the orientation of the nuclear quadrupole moment (which is that of the spin vector I) with respect to the axis. This interaction is space-quantized, the Hamiltonian function taking the form

$$H = eQ\phi_{zz}(3I_z^2 - I^2)/8I^2 , \quad \text{(E19)}$$

with eigenvalues

$$E_{Q,m} = eQ\phi_{zz}[3m^2 - I(I+1)]/4I(2I+1) , \quad \text{(E20)}$$

where

$$-I \le m \le I . \quad \text{(E21)}$$

An isomeric pair of nuclear levels, differing in angular momentum and in quadrupole moment, are therefore split unequally in energy by interaction with the local electric field gradient. Where there are site-to-site variations of electric field gradient, each of the $2I+1$ components of a degenerate level varies slightly about the average value of the displaced energy. If the quadrupole splitting is less than the natural linewidth, the gamma-ray line appears single, but broadened, whether or not there are site-to-site field variations.

Higher terms in the expansion of (E9) are so small that they are usually ignored in Mössbauer radiation; even where they may have been observed (Pfeiffer, 1977), they are far less than the natural width. There are no odd-order electric moments. The highest observable even-order electric multipole is that of order 2^{2I} (Ramsay, 1955).

c. Magnetic hyperfine interactions

The interactions of internal magnetic fields in solids, arising from unpaired electron spins and from nuclear magnetic moments, as well as from external sources, can be described formally by an integral analogous to (E9), in which charge densities are replaced by divergences of vector potentials (Ramsay, 1955). The lowest-order term in its expansion, describing interaction of the nuclear magnetic dipole moment with the local magnetic induction, is the most significant part of this interaction. It splits each nuclear state of spin I into $2I+1$ components and divides the corresponding gamma-ray line into hyperfine components, each having the width established by all other broadening effects. When the splitting does not exceed the natural width, the line appears further broadened. Each hyperfine component is also broadened by site-to-site variations of the splitting, including those that arise from a random distribution of the orientations or magnitudes of the nuclear magnetic moments.

The magnetic induction $\mathfrak{B}_{ij}$ at lattice site j created by the magnetic moment μ_i of a nucleus located at the ith site is

$$\mathfrak{B}_{ij} = [3(\mu_i \cdot \mathbf{R}_0)\mathbf{R}_0 - \mu_i](\mu_0/4\pi)/R_{ij}^3 , \quad \text{(E22)}$$

where $\mathbf{R}_{ij}$ is the vector connecting site i to site j and $\mathbf{R}_0$ is a parallel vector of unit length. The factor $\mu_0/4\pi$ relates magnetic field to induction. Typical values of the local induction, in the absence of an externally applied field, range from zero to 1(-3) V s m^{-2} (10 G), depending on nuclear angular momenta and G factors and on the internuclear spacing and arrangement. A nuclear moment of, say, $2I$ nuclear magnetons and spin of $I=5$, which in SI units amounts to

$$\begin{aligned}\mu &= G\mu_N[I(I+1)]^{1/2} \\ &= 2\times 5.05(-27)\sqrt{5\times 6} \\ &= 5.5(-26) \quad \text{A m}^2 ,\end{aligned} \quad \text{(E23)}$$

therefore has an interaction energy with a neighboring dipole

$$U = G\mu_N I\mathfrak{B}_{ij} \quad \text{(E24)}$$

that can range in value anywhere from zero up to 1.0(−28) J, or 6.3(−10) eV, equivalent to a frequency of 0.15 MHz.

Usually the thermal agitation energy greatly exceeds this value, and so, if there is no externally applied field, the nuclear moments are completely disordered. Thus there will be broadening of gamma-ray lines, arising from site-to-site variations of the magnetic dipole-dipole interaction, approaching a substantial fraction of 1 MHz, even if all the nuclei are in the same isomeric state.

As in the electrostatic case, the interaction of nuclear magnetic dipoles with the magnetic induction is space quantized. In the presence of a strong aligning field, the interaction Hamiltonian function is a sum of terms containing factors of the form

$$\mathbf{I}_i \cdot \mathbf{I}_j - 3I_{iz} \cdot I_{jz} ,$$

in which the individual spins and their axial projections include those of isomeric nuclei in either state, those of the host, and those of impurities. Clearly, randomness in locations of the excited and ground-state nuclei in the host crystal inevitably leads to broadening, even when the temperature is extremely low. Even an initially pure isomer crystal becomes increasingly "randomized" as radioactive decay proceeds.

d. Gravitational broadening

Nuclei situated at points of different gravitational potential but otherwise in identical environments are not precisely in resonance. The Einstein Principle of Equivalence calls for a "gravitational red shift" of frequency equivalent to the difference of potential energy. This has been verified for Mössbauer radiation (Pound

and Rebka, 1960b; Pound and Snider, 1965).

In standard gravity, the nuclear resonances in an extended body of vertical dimension δz are dispersed uniformly over a band of frequencies of width

$$\Gamma_g = (gE/\hbar c^2)\delta z = 166\,Ez \quad \mathrm{s}^{-1}. \tag{E25}$$

e. Line distortion by interference

Resonant absorption of gamma radiation, forming the upper isomeric state, is followed by internal conversion, in a fraction $\beta(1+\alpha)^{-1}$ of all events, to form an ion with its nucleus in the ground state and an ejected electron, indistinguishable from the result of photoelectric absorption of the same multipole order in the same electronic shell. The total probability of that result, proportional to the square of the sum of the quantum-mechanical probability amplitudes for the two indistinguishable processes, therefore contains their cross product. Thus an interference phenomenon takes place, resulting in an asymmetric distortion of the natural line (Trammell and Hannon, 1969; Kagan, Afanas'ev, and Voitovetskii, 1969). The line-shape factor is the ordinary Lorentzian function [Eq. (C5)] multiplied by a term of the form

$$1 - 2\xi(E - E_{21})/\Gamma ,$$

where Γ is the total inhomogeneous linewidth and ξ a factor of order unity.

Photoelectric absorption (Appendix F), predominantly an electric dipole interaction, affects the line shape significantly only if the nuclear transition has $E1$ multipolarity, viz, the ^{181}Ta transition (Sauer, Matthias, and Mössbauer, 1968). A slight effect has, however, also been observed in the 13-keV electric quadrupole transition of ^{73}Ge (Pfeiffer, 1977).

Under ordinary laboratory conditions, the interference results in only a slight asymmetry of the gamma-ray line, which appears to shift its centroid (not its maximum), but does not broaden the line. However, in a highly excited solid, necessarily bathed in intense gamma radiation, such as would be the case in a pumped graser, an appreciable fraction of the atomic cores would be photoionized, so that the gamma-ray line would be slightly broadened. No estimates of the magnitude of broadening to be expected from this effect have been given. We suspect that it would be inappreciable in comparison with the monopole broadening accompanying intense photoionization of the host.

5. Magnitude of inhomogeneous line broadening in long-lived transitions

a. Experimental evidence

It is instructive to examine the data presented in Table III, comparing the natural and measured linewidths of some selected Mössbauer transitions of various lifetimes.

Mössbauer measurements involve a convolution of source and absorber resonances. If their lines are unsplit and equally broadened, the measured line has twice the width of either resonance; if they are unequally broadened, their sum. The experimental results are reported as the difference of the two relative velocities in m s^{-1} at which the resonant part of the absorption is half of its maximum value. The column labeled s^{-1} assumes equal linewidths of source and absorber.

Short-lived transitions have nearly the natural breadth; the longest-lived transitions are generally broadened. A noteworthy exception is ^{73}Ge, for which ideal single crystals of extremely high purity are readily prepared. Broadening usually amounts to several tens of kHz in even carefully prepared samples, and it is an increasing proportion of the total breadth as the lifetime is increased.

The importance of the chemical and metallurgical nature of the host is illustrated by Table IV, which compares various measurements of the 6.23-keV line of ^{181}Ta. This isomer has high nuclear spin ($\frac{9}{2}$) and a large electric quadrupole moment (3.9 b), and the nuclear volume changes substantially in the transition, so the line is especially sensitive to monopole and hyperfine broadening. Thus even the narrowest line reported so far is over four times the natural width.

A survey of experimental and theoretical information concerning the line-broadening effects of radiation damage, with especial reference to the graser application, has been made by Schwenn (1978).

The final entry of Table III is controversial. The conventional Doppler scanning technique of Mössbauer spectroscopy would be hopelessly impracticable for so narrow a line. Instead, the linewidth was inferred (Bizina, Beda, Burgov, and Davydov, 1963) from measurement of the delayed gamma-ray activity induced by recoilless resonant absorption, when an activated silver source foil was exposed to a similar, but initially inactive, silver absorber foil (Mössbauer spectrometers using conversion electron detection are analogous except for the time scale). The source foil first was activated by proton bombardment; the ^{107}Ag$(p,n)^{107}$Cd reaction created a volume distribution of the 6.5-h electron capture activity to serve as a slowly decaying source of the 44-s ^{107}Ag isomer. In order to eliminate accumulated hydrogen and repair lattice damage from the bombardment, the foil was annealed at 560 °C for two hours, then cooled to liquid helium temperature, and finally exposed to the absorber. The short-lived activity of ^{107}Ag induced in the absorber was determined by summing counts, in corresponding time channels, in repeated exposures. Control experiments verified that activity was not transferred by physical contact and that it was induced only if source and absorber were both at low temperature. Thus it was assumed that the detected activity resulted solely from resonant absorption of the 93.3-keV Mössbauer radiation. From the yield, a cross section of $7.4 \pm 2.0(-31)$ cm^2 was inferred. Assuming that $f = 0.029$, according to the Debye model, Bizina *et al.* concluded that the experimental cross section is only $1.33(-6)$ of the maximum possible theoretical cross section for this reaction. They attributed the reduction to inhomogeneous broadening, by the magnetic dipole-dipole interaction, of 2.7 kHz—a reasonable result in the light of the other data of Table III.

Nevertheless, the authors' assumptions, analysis,

and conclusions have been criticized on several grounds (Carlson and Temperley, 1969). For example, Bizina *et al.* completely ignored the gravitational red shift in their analysis, although its effect may have been as large as a factor of 1(4). The source activity was determined at room temperature, when the gamma-ray attenuation is purely nonresonant. In the activation transfer exposure, both resonant and nonresonant attenuation occur; nevertheless, Bizina *et al.* employed a common value for the photon mean free path for each case. It was assumed that temperature and isomer shifts were negligible, despite the different histories of the two foils and the likelihood that residual damage and chemical impurities from the bombardment would not be completely eliminated in a two-hour annealing at only 560 °C.

Recently, Wildner and Gonser (1979) have claimed to observe recoilless resonance in the similar isomer ^{109}Ag ($T = 57.5$ s, $E = 88$ keV, $\alpha = 26$). They employed a single silver crystal into which 470-day ^{109}Cd parent was introduced to a limited depth by thermal diffusion, and observed the counting rate of 88-keV gamma radiation as a function of temperature. Over the temperature range 4.2–78 K, the observed dependence was attributed mainly to change in the nuclear resonant self absorption, corresponding to temperature dependence of the recoilless fraction. To estimate a linewidth from their measurements, they assumed a Debye temperature of 226 K and a distribution in depth of the source activity; they do not state whether corrections for gravitational shift were made. From the observed change (0.1%) in counting rate, they estimated an effective linewidth 30 times the natural width. The reasons for such remarkably small broadening, and its probable origin, were not discussed.

Such questions emphasize the need for a new technique for linewidth measurement of higher resolution than Doppler scanning. Temperature differential and gravitational scanning (Mead, 1966) are obvious possibilities, but appear to offer great experimental difficulties. It has recently been proposed to employ time-domain Fourier transform methods to determine the line shape (Baldwin, 1979a), in conjunction with correlation counting techniques (Baldwin and Gol'danskii, 1980). At present, lacking more direct information, one can resort only to theoretical estimates of the linewidth for long-lived transitions.

b. Theoretical estimates

A number of papers have given estimates of the broadening to be expected for recoilless lines from long-lived isomers. These are summarized in Table V and the accompanying notes.

One is immediately struck by the diversity of mechanisms that have been identified and by the fact that none of the references has considered all the conceivable line-broadening interactions. In particular, monopole broadening associated with a high degree of inner-shell ionization from nonresonant absorption in a highly excited solid has been completely overlooked.

The large number of independent sources of monopole broadening should be especially noted. Nonuniformity of doping of a host with a graser isotope which is chemically distinct from the host can lead to substantial broadening. Even crystals which are absolutely defect free can still exhibit broadening if internal strain exists, and there is evidence that measurable numbers of microscopically strained regions exist within even perfect crystals of germanium (Hunter, 1959). Even the proximity of a crystal boundary can broaden the line.

APPENDIX F: NONRESONANT PHOTON ABSORPTION

1. Processes of photon removal

For photon balance calculations, any process that results in even a slight change of photon energy must be considered as equivalent to its complete removal.

In the 5–100 keV energy region, the important interactions of radiation with matter are photoelectric absorption and Compton scattering: the former with bound electrons in atoms, the latter with free (or relatively loosely bound) electrons.

The theory of these processes may be found in texts of quantum electrodynamics (e.g., Heitler, 1944). Tabulations of photon cross sections have been given by Storm and Israel (1970), Hubbell and Berger (1965), and Koch and Wyckoff (1959), among others. Extensive bibliographies may be found in each of these tabulations.

The tabulations list several kinds of cross section. With the exception of recoilless Rayleigh ("coherent") scattering from solids, all processes of scattering displace the photon energy by more than the resonant bandwidth, and therefore contribute to removal. Compton scattering is incoherent and replaces a photon by a scattered photon of lower energy and altered direction of propagation, the remaining energy being imparted to a recoiling electron. Photoelectric absorption completely removes the photon, replacing it with an ion and ejected electron. The "energy absorption cross section" refers only to the transfer of energy from radiation to the medium, considering the secondary Compton-scattered photon as not having been removed.

Therefore, in using tabulated cross sections, one must use the total "narrow-beam" cross section for photon balance calculations.

2. Empirical formulas

Since amplification is appreciable only if the photon balance criterion is satisfied by a wide margin, empirical relations of limited accuracy are useful.

A rough estimate of the total Compton scattering cross section for energies in the Mössbauer range can be obtained from the formula

$$\sigma_{cs} = 0.658Z \exp[-2.95(-3)E] \quad \text{b} \qquad \text{(F1)}$$

if tabulated cross sections are unavailable.

Each electron shell for which the binding energy does not exceed the photon energy participates in photoelectric absorption, but the strongest interaction is with the most tightly bound electron shells. The cross section therefore exhibits discontinuities at photon energies equal to subshell binding energies. The K-shell edge is

located approximately at

$$E_K = 9.67(Z/30)^{2.16} \quad \text{keV} \tag{F2}$$

and the higher of the L-shell edges at

$$E_L = 1.18(Z/30)^{2.6} \quad \text{keV}. \tag{F3}$$

The photoelectric absorption cross section can be approximated by

$$\sigma_{ph} = KZ^{4.5}E^{-3.0} \quad \text{b}, \tag{F4}$$

where the constant K has the approximate values

$$\begin{aligned} K &= 8.5, \quad E_K < E \\ &= 0.70, \quad E_L < E < E_K \\ &= 0.115, \quad E < E_L. \end{aligned} \tag{F5}$$

The linear absorption (removal) coefficient τ of the medium is obtained by multiplying the sum of the photoelectric and total scattering cross sections by the atom density and summing for each element.

3. Relationship to nuclear-resonance cross section

In the energy range of interest, matter becomes increasingly transparent to radiation as the wavelength is shortened, except for the discontinuities at absorption edges. The reason to hope for development of lasers in this spectral region is that the photoelectric cross section is far less and diminishes more rapidly with increasing photon energy than the stimulation cross section at energies for which the Compton-scattering cross section is relatively unimportant. As one approaches relativistic energies, on the other hand, Compton scattering eventually becomes the dominant removal process, since it falls off less rapidly with energy than either the photoelectric or the stimulation cross section; an energy is eventually reached above which no material medium can be sufficiently transparent to amplify radiation. This is exhibited by Fig. 2 (Baldwin, 1973b), which displays the ratio $\Lambda^2/2\pi(\sigma_{cs}+\sigma_{ph})$.

4. Photon removal in fully ionized matter

The relationships given here apply to ordinary or weakly ionized matter under terrestrial conditions. In a fully ionized plasma, however, photoelectric absorption no longer is possible. Inverse bremsstrahlung, in which the photon interacts inelastically with an electron in the Coulomb field of an ion, assumes an increasingly dominant role as the degree of ionization and the density are increased. Compton scattering remains important, since the full complement of Z electrons per atom is free to scatter photons. Aside from noting that the photon balance condition is more difficult to satisfy in dense plasmas than in solid media (Gol'danskii and Namiot, 1976a), we shall not investigate this subject, which, although fundamental to the realization of x-ray lasers, is outside the scope of the present article. See also Sec. III.C where the question of "vigorously pumped" nuclear transitions is briefly discussed.

APPENDIX G: PHOTON CHANNELING IN CRYSTALS

1. Anomalous transmission

The processes of photon attenuation described in Appendix F include absorption (by transfer of energy to atoms and electrons) and extinction (by scattering out of a beam), and the cross sections of Eqs. (F1) and (F4) apply for arbitrary directions of propagation in ordinary amorphous, polycrystalline, or fluid matter.

A remarkable departure from the predicted attenuation was observed during World War II (Borrmann, 1941), in the case of single crystals with monochromatic x rays propagating in directions related to the wavelength by the Bragg law of diffraction. The effect elicited little notice until it was subsequently rediscovered (Campbell, 1951).

For the effect to be observable, the x radiation must be highly monochromatic and well collimated, the crystal must be single and free from defects, and the Laue geometry employed (Fig. 15). When these conditions are satisfied, the effective attenuation coefficient for Bragg-directed s-polarized radiation can be two orders of magnitude less than for other directions. In addition to strong Bragg reflection on the entrance side, the incident beam excites two equally intense beams on the emergent side, leaving the crystal at points where the plane of incidence and the diffracting crystal planes that received the entrance beam meet the emergent surface (Fig. 15). Thus the radiation evidently is channeled between the lattice planes, and the phenomenon exists because of Bragg diffraction.

2. Standing-wave fields in crystals

a. Interference of a direct and a reflected wave

When an electromagnetic wave strikes a plane conductive surface, the reflected wave's propagation vector $\mathbf{k}'$ is directed symmetrically with respect to the incident wave vector $\mathbf{k}$ and a unit vector $\mathbf{n}$ normal to the

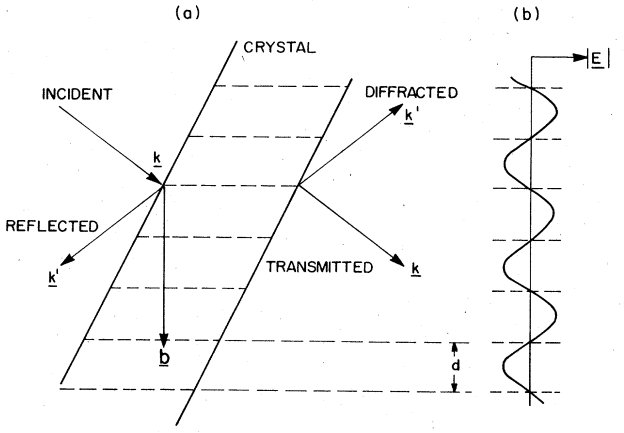

FIG. 15. Borrmann-Campbell effect. (a) Geometry for Laue diffraction, showing the Bragg-reflected, anomalously transmitted, and diffracted beams. (b) The standing-wave pattern of electric field amplitude in the crystal for the anomalously transmitted mode.

conductive surface, i.e.,

$$\mathbf{k}-\mathbf{k}'=\text{const}\,X\,\mathbf{n}. \tag{G1}$$

In the region of overlap of the two waves, if the incident wave is monochromatic and coherent, an interference pattern forms. When the incident wave is polarized normal to its plane of incidence ("*s* polarization"), the amplitude of the electric vector in the resultant total field varies sinusoidally with distance from the reflective plane, with period $|\mathbf{k}-\mathbf{k}'|^{-1}$. The magnetic vector is also spatially modulated, but does not vanish, and it has both longitudinal and transverse components. The Poynting vector is everywhere parallel to the line of intersection of the plane of incidence and the surface, varies sinusoidally, and vanishes at planes uniformly spaced parallel to the reflecting surface.

b. Interference of Bragg-diffracted radiation in crystals

When a crystal scatters an incident plane wave, the scattered wavelets combine, according to Huyghen's principle, into a weak reflected plane wave. The unscattered remainder of the incident radiation penetrates further into the crystal, with gradually diminishing intensity.

Waves scattered from the various lattice planes tend to cancel by interference, unless the incident wave vector fulfills the Bragg condition:

$$\mathbf{k}-\mathbf{k}'=m\mathbf{b},\quad m=1,2,\ldots, \tag{G2}$$

where **b** is a vector normal to the lattice planes equal in magnitude to the reciprocal of their spacing. The standing-wave patterns arising from reflections by each of the atomic planes then all coincide. The reflected wave field can then approach the amplitude of the incident wave, and interfere with it, just as in ordinary reflection. The *s*-polarized component of the incident wave excites two orthogonal modes, one having electric antinodes, the other nodes, at the atomic planes. In the second mode, energy flow within the crystal is channeled between the lattice planes. The magnetic vector and the electric field gradient do not vanish, however (nor does any other field property), in the case of *s* polarization. Interaction of the radiation with atoms in lattice sites is therefore suppressed to an extent that depends upon their sizes, their amplitudes of thermal excursion, the multipole order of their interaction, the degree of polarization of the radiation, and its angular and spectral dependences.

Photoelectric interaction of the *s* component, largely electric dipole, is strongly suppressed, and the absorption coefficient of the anomalously transmitted mode is reduced by a factor

$$\sigma_a(\mathbf{k}_{\text{Bragg}})/\sigma_a=1-\varepsilon, \tag{G3}$$

where ε is the ratio of the imaginary part of the amplitude for atomic scattering at the Bragg angle to that for forward scattering; for an ideal isotropically scattering point atom, ε is unity. In cold near-perfect crystals ε can exceed 0.99, giving over 100-fold reduction in the absorption coefficient.

In practice, finite atom size, zero-point motion of the atoms, and finite angular and frequency breadths of the radiation further reduce the transmission. Resonant nuclei contribute to the scattering. For detailed treatment, the dynamical theory of x-ray diffraction (James, 1950) is required, and is outside the scope of this Appendix. The reader is referred to the review by Batterman and Cole (1964), which accounts for the phenomena observed in the ordinary case, where the source of radiation is external to the crystal and the nuclei are not resonant to the radiation.

3. Effects of crystal imperfections, strain, and temperature

Crystal imperfections that place atoms in regions of nonvanishing electric field both increase the absorption and disturb the standing-wave field. Anomalous transmission is therefore sensitive to dislocations, interstitial impurities, and internal stress, and can be used for detecting such imperfections. For example, microstrains of the order of 1(−5) have been demonstrated in nearly perfect crystals of Ge (Hunter, 1959); Borrmann, Hartwig, and Irmler (1958) have made shadowgraphs of crystal defects; the presence of 1 ppm of oxygen impurity in silicon crystals has been detected (Patel and Batterman, 1963), and its removal by annealing was demonstrated by monitoring the x-ray transmission. Of particular interest to graser development are the observations that radiation damage in single crystals of Cu by 1(17) fast neutrons cm^{-2} did not appreciably change ε from values of 0.96 (Nicklow, Sherrill, and Young, 1965), and that careful bending of a good crystal does not destroy anomalous transmission, since the wave field can adjust its direction of propagation to maintain the Bragg relation (Cole and Brock, 1959); it was suggested that this might enable development of a focussing device.

The effect of temperature is closely approximated by multiplying ε by the Debye-Waller exponential factor (Appendix D).

4. Internal sources

Electronic transitions of inner-shell-excited atoms and nuclear transitions of isomers within atoms of a crystal can emit narrow-line radiation into modes obeying the Bragg relation, Eq. (G2). Propagation within the crystal of internally generated radiations depends upon the multipole orders of their sources, the direction of polarization, and the source location relative to the lattice planes. Characteristic x radiation, originating in electric dipole transitions, excites only the strongly absorbed mode, with antinodes at the lattice planes, so its propagation in Bragg directions is suppressed. Sources of higher multipole order can also excite the anomalously transmitted mode, and this radiation can emerge from the crystal (Hannon, Carron, and Trammell, 1974b).

To a distant observer, the directions satisfying the Bragg relation define cones with axes normal to the lattice planes; thus internally generated radiation that is able to emerge forms bright lines in the form of conic sections on photographic plates, superimposed upon the diffuse background of incoherently scattered radiation. Similarly, dark lines indicate anomalous absorption. These are called Kossel lines, and are useful for the study of crystal structures (Hannon, Carron,

and Trammell, 1974b).

In crystals composed of isomeric nuclei, the resonant nuclear scattering may be strong enough to interfere appreciably with the electronic scattering, and so to affect the value of ε. Anomalous dispersion (Bernstein and Campbell, 1963) and internal fields further complicate the phenomena. For example, magnetic fields split the resonance, each Zeeman component having its own set of Bragg angles and polarization. Furthermore, some crystals have a magnetic structure that does not coincide with the atomic structure. Finally, Faraday rotation can transform one polarization mode into the other.

With commonly used Mössbauer sources, the Mössbauer ground state accumulates as the parent isotope decays; the Bragg modes are affected by the increasing nuclear contribution to scattering of the recoilless component, but not of the recoil-broadened component.

5. Suppression of inelastic processes

The interference of wave modes in crystals gives rise to anomalous effects in which the spatial arrangement of atoms or nuclei determines their response to radiation, not only for electromagnetic waves but also for neutrons.

Afanas'ev and Kagan (1965b) have noted that when two wave modes interfere in a crystal, their respective probability amplitudes for forming a compound nucleus by resonant absorption at a lattice site may sometimes be opposite in sign, though equal in amplitude. The nuclear reaction (e.g., internal conversion from gamma-ray absorption, capture radiation from neutron absorption) is then suppressed (Knowles, 1956; Smirnov, Sklyarevskii, and Artem'ev, 1970; Voitovetskii, Korsunskii, and Pashin, 1968).

6. Bearing on the graser problem

The reduction of photoelectric absorption in the Borrmann-Campbell effect reduces the critical inversion density needed for net gain by stimulated emission (Baldwin, Neissel, Terhune, and Tonks, 1963)—provided interaction with the excited nuclei is not reduced to the same extent. In addition to mitigating the pumping problem, the effect would also improve the directionality of the stimulated emission (Kagan, 1974b). Moreover, it offers the possibility for developing a resonator (Vali and Vali, 1963b; Fisher, 1974; Yariv, 1974).

Kagan (1974b, 1975), Andreev and Il'inskii (1975a, 1975b, 1976), and Hannon and Trammell (1975) have considered the problem of amplifying recoilless radiation in Bragg modes by stimulated emission. They concluded that nuclear transitions of multipole order higher than electric dipole can indeed interact with the anomalously transmitted radiation, but that there must be no suppression effect (see the preceding section) nor mixing of the two states of polarization, in nuclear transitions of low energy and high multipolarity. Andreev and Il'inskii (1975b) showed that diffraction spreading of the transversely bounded beams in a needle-shaped crystal should not inhibit anomalous transmission if the crystal's diameter exceeds 100 μm.

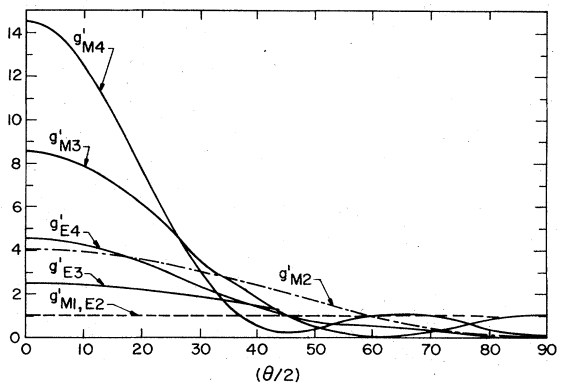

FIG. 16. Coupling of Bragg-wave modes to nuclear multipoles. Coefficients of coupling, calculated for transitions of various multipole orders, in nuclei located at regular lattice sites, as a function of the Bragg angle $\theta/2$. From Hannon and Trammell (1975).

Hannon and Trammell (1975) made quantitative estimates of the coupling of Bragg two-wave modes to the nuclear multipole moments, and showed that the coupling is poor for low multipole orders. Figure 16, taken from their paper, shows the dependence of the coupling factor on the Bragg angle, assuming that the nuclei are located at lattice sites. They also give values of the net factors by which inversion density requirements are reduced for Bragg modes of several Mössbauer transitions, taking account of zero-point motion as well as of the reduced coupling to the wave field. They obtain factors ranging from only about 5 for ^{119}Sn to 50 for ^{169}Tm. In the case of ^{73}Ge, $E = 13.5$ keV, for which with normal absorption it is impossible to satisfy the balance condition, they obtain a reduction factor of 15.8, which would enable an otherwise unsuitable transition to realize stimulated emission gain—provided, of course, that the anomalous transmission modes could survive damage associated with the pumping process.

Kagan (1975) has noted that multiwave interference (for propagation in directions which simultaneously satisfy the Bragg condition for more than one set of lattice planes) would enable higher values of the coupling factors to be realized.

APPENDIX H: THE CRITERION FOR STIMULATED EMISSION GAIN

A necessary condition for a medium to amplify radiation by stimulated emission is that photons be added more rapidly than they are absorbed. In a single passage through a thickness dx the balance of stimulated emission, resonant absorption, and nonresonant removal processes is

$$N_2\sigma_s nc\,dx - N_1(g_2/g_1)\sigma_s nc\,dx > N_0\sigma_a nc\,dx\,, \tag{H1}$$

leading to the "photon balance condition"

$$N^*\sigma_s > N_0\sigma_a\,, \tag{H2}$$

where

$$N^* \equiv N_2 - (g_2/g_1)N_1 \tag{H3}$$

is the inversion density.

Allen and Peters (1971a) have shown that this necessary condition is not sufficient, if gain is to be perceptible; the appropriate condition for observable amplification is not merely that absorption losses be balanced by stimulated emission, but that, in passage from one end of the active medium to the opposite end, each photon should, on the average, give rise to at least one additional photon. This leads to the amplified-spontaneous-emission (ASE) condition

$$N^*\sigma_s - N_0\sigma_a \geq 1/L \tag{H4}$$

for a medium of length L in the direction of propagation. Peters and Allen (1971) have shown experimentally that the ASE condition accurately predicts the observed threshold of mirrorless gas lasers for amplifying Doppler-broadened lines.

When a graser must operate on a line of near-natural breadth, the ASE condition also is not sufficiently restrictive, because it neglects the kinetic character of the stimulation cross section (Appendix C). The threshold inversion density then may be over an order of magnitude higher than the ASE condition predicts (Vorontsov and Vysotskii, 1974a; Hannon and Trammell, 1975; Hopf, Meystre, Scully, and Seely, 1975), unless excitation is continuously regenerated by the pump (Hopf, Meystre, Scully, and Seely, 1975; Baldwin and Suydam, 1977). This problem is considered in Appendix I.

APPENDIX I: NARROW-LINE LASER KINETICS

1. Semiclassical treatment of the radiation field

The following analysis is adapted from Baldwin and Suydam (1977).

In terms of the vector potential $\mathfrak{a}$, the Maxwell field equations, with the following constitutive relations

$$\mathfrak{B} = \mu_0 \mathfrak{H}, \tag{I1}$$

$$\mathfrak{D} = \varepsilon_0 \mathcal{E} + \mathcal{P}, \tag{I2}$$

$$\mathcal{P} = \varepsilon_0 \chi \mathcal{E}, \tag{I3}$$

$$\mathbf{J} = \dot{\mathcal{E}}/\rho, \tag{I4}$$

for a nonmagnetic active dielectric medium, yield a propagation equation

$$\nabla \times (\nabla \times \mathfrak{a}) + (1/c^2)\ddot{\mathfrak{a}} = -(\mu_0/\rho)\dot{\mathfrak{a}} + \mu_0 \ddot{\mathcal{P}}, \tag{I5}$$

where

$$c = (\mu_0 \varepsilon_0)^{-1/2} \tag{I6}$$

is the velocity of light waves *in vacuo*.

This has plane-wave solutions modified by the right-hand terms, the first accounting for losses, and the second for forcing by the charge polarization of the medium. When the change of amplitude over one wavelength is small, an approximate solution is the plane-polarized plane transverse wave with amplitude varying in the direction of propagation (taken to be the x axis):

$$\mathfrak{a} = \{\mathfrak{a}(x,t)\exp[i(\omega t - kx) + \text{c.c.}]\}\underline{x}_1 . \tag{I7}$$

Substituting Eq. (I7) into (I5) and neglecting the second spatial derivative of $\mathfrak{a}$ reduces the propagation equation to the form

$$\frac{\partial \mathfrak{a}}{\partial x} + \frac{1}{c}\frac{\partial \mathfrak{a}}{\partial t} = -\frac{N\sigma_a}{2}\mathfrak{a} + \frac{\dot{\mathcal{P}}}{2\varepsilon_0 c_\omega} . \tag{I8}$$

Because of strong anomalous dispersion, the wave is slowly propagating (Fig. 6; Chirikov, 1963), so that the retardation term is also negligible. Here $N\sigma_a$ is the absorption coefficient, giving the loss rate per unit distance of propagation due to interaction with electrons. The second term on the right is the driving force of time-varying polarization currents in the medium.

The polarization current $\dot{\mathcal{P}}$ $[A/m^{-2}]$ induced by the field of the developing wave acting on the nuclear charge obeys the dispersion relation

$$\frac{\partial \dot{\mathcal{P}}}{\partial t} + [i(\omega - \omega_{12}) + \Gamma/2]\dot{\mathcal{P}} = \frac{fN^* |\dot{\mathcal{P}}_{21}|^2}{\hbar}\mathfrak{a} , \tag{I9}$$

in which the linebreadth Γ appears as a damping coefficient. A fraction f of the driving-wave field acts to induce transitions between states 1 and 2 at a rate proportional to the square of the total matrix element of the polarization current. The net effect, positive or negative, is proportional to the inversion density N^*, which acts as either a source or sink of energy.

When Eqs. (I9) and (I8) are combined by eliminating the polarization, a partial differential equation results for the spatial and temporal behavior of the vector potential. For the important case of exact resonance,

$$\left(\frac{\partial}{\partial t} + \frac{\Gamma}{2}\right)\left(\frac{\partial}{\partial x} + \frac{\tau}{2}\right)\mathfrak{a} = \frac{N^*\Gamma\sigma}{4}\mathfrak{a} \tag{I10}$$

in which σ is the asymptotic cross section evaluated for the natural line and τ is the absorption coefficient. Note that the two differential operators do not commute.

A similar equation for the field amplitude was derived by Chirikov (1963), based on the time-dependent cross section (Appendix C) for an unbroadened line:

$$\frac{\partial}{\partial t}\left(\frac{\partial}{\partial x} + \frac{\tau}{2}\right)\mathfrak{a} = K\mathfrak{a} . \tag{I11}$$

He assumed that N^* was constant, so his solution (Fig. 6) describes only the early course of amplification.

2. Rate equations for state populations

The time dependence of the inversion density N^*, which depends not only on the net induced transition rate, but also on the spontaneous decay and pumping rates, must be known before Eq. (I10) can be solved.

One of many possible upper-state rate equations is

$$\frac{\partial N_2}{\partial t} + \lambda_2 N_2(t) = \eta_2 R(t) - nc\sigma N^* . \tag{I12}$$

Correspondingly, the lower state obeys

$$\frac{\partial N_1}{\partial t} - \beta\lambda_2 N_2(t) + \lambda_1 N_1(t) = \eta_1 R(t) + nc\sigma N^* . \tag{I13}$$

These describe a system of two states with decay constants, respectively, λ_2 and λ_1, created by transmutation at respective rates $\eta_2 R$ and $\eta_1 R$, and exchanged by induced transitions. It is possible that the pump could destroy as well as create the graser states; if

so, additional terms are needed.

Equations (I10) through (I13) constitute a coupled nonlinear set that can be solved, numerically, if the pumping function $R(t)$ and isomer ratios η_i are known and if the photon density n is expressed in terms of the field amplitude $\mathfrak{a}$.

3. Case of freely decaying, initially full inversion

Let us assume for simplicity that the excited state is not generated continuously by the pump, but instead, that an inverted population has been suddenly created (as by chemical separation) and rapidly assembled, afterwards to decay to a stable terminal state.

We adopt the "weak signal" approximation, i.e., neglect induced changes of population. The inversion density is therefore

$$N^* = N_0 [e^{-\lambda t} - w(1 - e^{-\lambda t})]. \tag{I14}$$

At all points along the path of propagation, it remains positive only for a time interval

$$t_{\text{inv}} = \frac{1}{\lambda} \log_e \left(\frac{g_2}{g_1 + g_2} \right). \tag{I15}$$

The kinetic equation (I10) is to be solved with the initial conditions

$$\mathfrak{a}(x, 0) = \mathfrak{a}_1 e^{-\tau x/2}, \tag{I16}$$

$$\mathfrak{a}(0, t) = \mathfrak{a}_1 e^{-\lambda t/2}, \tag{I17}$$

corresponding to exponential attenuation of a damped wave from a decaying source at $x = 0$.

Substitution of

$$\mathfrak{a} = \mathcal{Y} e^{-\gamma t/2} e^{-\tau x/2} \tag{I18}$$

reduces the kinetic equation to

$$\frac{\partial^2 \mathcal{Y}}{\partial t \, \partial x} = \kappa \mathcal{Y} \tag{I19}$$

and the initial conditions to

$$\mathcal{Y}(x, 0) = \mathfrak{a}_1, \tag{I20}$$

$$\mathcal{Y}(0, t) = \mathfrak{a}_1 e^{\gamma t}, \tag{I21}$$

where

$$\kappa = N^* \Gamma \sigma_s / 4 \tag{I22}$$

characterizes the resonant interaction and

$$\gamma = (\Gamma - \lambda)/2 \tag{I23}$$

the extent of line broadening.

The solution for the unbroadened-line case ($\gamma = 0$) with neglect of the time dependence of N^* is readily found (Vorontsov and Vysotskii, 1974a):

$$\mathfrak{a} = \mathfrak{a}_1 \exp[-(\lambda t + \tau x)/2] I_0(2\sqrt{\kappa x t}). \tag{I24}$$

When the inversion density is positive, the monotonically decreasing exponential factor is multiplied by a divergent Bessel function, which may or may not suffice to overcome attenuation and decay, depending on the value of $\kappa x t$. If decay has proceeded sufficiently that κ has become negative before the medium is made resonant, the modified Bessel function is replaced by the ordinary Bessel function of the first kind, $J_0(2\sqrt{k x t})$, and $\mathfrak{a}$ takes the form of a damped oscillation superimposed on the attenuating decay curve, the medium acting as a resonant filter. This latter case has been observed in experiments with Mössbauer isomers (Lynch, Holland, and Hamermesh, 1960).

For a wave to grow in the direction of propagation, i.e.,

$$\frac{\partial \mathfrak{a}}{\partial x} > 0, \tag{I25}$$

it is required that

$$\frac{I_1 (2\sqrt{\kappa x t})}{I_0 (2\sqrt{\kappa x t})} > \frac{\tau \lambda}{4\kappa}, \tag{I26}$$

a condition that can hold over only a limited range of values of x and t, since the ratio of the Bessel functions is always less than unity.

The behavior of a broadened-line system is less restricted, and the ASE condition of Allen and Peters (Appendix H) applies, since the resonant cross section then varies rapidly in comparison with the inversion density, quickly reaching its asymptotic value.

For a slightly broadened line, the solutions become complicated. If decay is neglected, so that κ is constant, one obtains a solution (Vorontsov and Vysotskii, 1974a; Baldwin and Suydam, 1977)

$$\mathfrak{a} = \mathfrak{a}_1 \left[e^{\kappa x/\gamma} - e^{-\gamma t} \sum_{n=0}^{\infty} \left(\frac{\kappa x}{t \gamma^2} \right)^{n/2} \times I_n(2\sqrt{\kappa x t}) \exp[-(\Gamma t + \tau x)/2] \right], \tag{I27}$$

from which it can be seen that, when absorption is small, the behavior approaches exponential growth with x when γ is increased, the medium becoming a broadband amplifier.

In optical-frequency lasers, lines are Doppler broadened by many orders of magnitude; the exponential term suffices to describe the behavior of the wave field except for extremely short pulses. In the Mössbauer case, however, the behavior is dominated by the inertia of resonant response, the pump, and decay of inversion. The Bessel functions then are dominant.

APPENDIX J: NEUTRONS AS PUMPING RADIATION

1. Advantages

Neutrons have been frequently proposed for graser pumping because, in contrast with other nuclear radiations, they are capable of exciting and transmuting nuclei even when their kinetic energies are only thermal, so that they produce no direct damage and little heating in solids. Other pertinent properties are briefly described below. For more detail, see any text on nuclear reactor physics or engineering, e.g., Lamarsh (1966), Chap. II.

2. Nuclear reactions

Free neutrons undergo beta decay with a 10.6-minute half-life if not first absorbed into a nucleus. Therefore the reactions necessary to generate neutrons are just as important in the graser context as the reactions

they undergo for pumping.

Besides the reaction type, it is necessary to know the net reaction energy ("Q value") and the dependence of the cross section for the reaction upon the energy of the bombarding particle. Charged particles must surmount a Coulomb barrier. Neutrons have the unique properties of being uncharged and absorbed exoergically. Photons, also uncharged, must have energy sufficient to remove a bound particle or, in the case of pure electromagnetic excitation, energy exactly matching, within the bandwidth, a radiative transition of the target nucleus.

In the usual convention for identifying a particular nuclear reaction, bombardment of beryllium with alpha particles to produce neutrons is written as $^{9}\mathrm{Be}(\alpha, n)^{12}\mathrm{C}$, and its bombardment with gamma rays, also to produce neutrons, as $^{9}\mathrm{Be}(\gamma, n)2\,^{4}\mathrm{He}$. These are useful laboratory sources, the initial alpha or gamma radiation being obtained from radioisotopes or accelerator sources; the yields, however, are far less than would be needed for graser excitation. It has been suggested that synchrotron sources may eventually be capable of generating x radiation above the 1.665-MeV threshold of the beryllium photonuclear reaction in intensity sufficient for copious generation of neutrons with energies of only a few keV (Eremeev, 1978; Hofmann, 1980).

Although many reactions are known that generate neutrons, viz., (p, n), $(d, 2n)$, (d, n), spallation reactions, and fission, only the last two are at present sufficiently intense to be considered for graser pumping. Yields of neutrons from reactions with accelerated particles are limited by the accelerator parameters and by energy loss in the intended target. The thermonuclear (d, n) reactions $^{2}\mathrm{D}(d, n)^{3}\mathrm{He}$ and $^{3}\mathrm{T}(d, n)^{4}\mathrm{He}$ may someday be copious sources of fast neutrons.

Every known neutron-producing reaction generates neutrons having energies averaging far more than those (characteristic of thermal equilibrium) at which they react most efficiently. In the case of fission, the average neutron energy is 2 MeV, with a distribution corresponding to evaporation from fission products excited to a comparable temperature.

Controlled chain reactions are capable of producing thermal neutron fluxes up to 1(16) $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$. Fast burst reactors can produce approximately 2(17) fast neutrons per burst. Nuclear explosions produce far more, but even they have obvious limits to the neutron yield; for example, the complete consumption by fission of 1 kg of $^{235}\mathrm{U}$ can release at most $6(23)\times(2.5-1)\times(1/0.235) = 3.8(24)$ fast neutrons.

3. Pumping reactions

Although fast neutrons are capable of producing many nuclear reactions [e.g., (n, p), (n, α), (n, n') inelastic scattering, and (n, f) fission], whenever the energy requirements of the reaction can be met, the cross sections are only of the order of the geometrical area of the nucleus or less. On the other hand, slow neutrons, particularly neutrons in thermal equilibrium, have large de Broglie wavelengths and can be captured [i.e., undergo the (n, γ) reaction] with cross sections many times the geometrical area of the nucleus. Resonances exist for neutron capture at intermediate energies. Far from resonance, the capture cross sections vary inversely as the neutron velocity. This same law of variation also commonly characterizes thermal neutron capture, leading to a capture rate proportional to neutron density. The factors of proportionality vary widely from one nuclide to another.

4. Neutron moderation

Since neutrons are generated with high energy, they must be moderated, before they can be used for pumping by capture, by thermal contact with a heat reservoir at low initial temperature. Since the infinite thermal reservoir of abstract thermodynamic theory is not available, neutron moderators are necessarily finite bodies with limited heat capacity.

Moderators employ nuclei of any species having a low cross section for capture but a substantial cross section for scattering, having dimensions large enough to ensure that only a small fraction of the neutrons escape across boundaries before they have moderated. Neutrons from compact sources within such a moderator diffuse outward, so that, even with negligible leakage, their density rapidly diminishes as they moderate; meanwhile, the energy they transfer raises the moderator's temperature and, therefore, increases the final energy to which the neutrons can be moderated.

It has been shown (Baldwin and Solem, 1979a) that there is a theoretical upper bound to the density of moderated neutrons, given by the energy balance that would apply in an infinite solid of initial temperature θ into which neutrons, initially of energy E_s and density n, are injected uniformly:

$$n(E) = N_m(E - 3k\theta)/(E_s - E), \tag{J1}$$

where N_m is the density of the moderator atoms and E the final neutron energy. In actual, finite moderators, the practically attainable neutron density is lower by several orders of magnitude because of leakage.

Moderation requires a finite time, which, on the average, is comparable with or greater than the lifetimes of most observable Mössbauer transitions. This is also true of the statistical fluctuations of the moderation time.

It is readily shown (Lamarsh, 1966) that the time for neutrons of initial energy E_s to be moderated to final energy E is given by

$$t_{\mathrm{mod}} = 1.446(-4)(l/\xi)(E^{-1/2} + E_s^{-1/2}), \tag{J2}$$

where the factor ξ is the average decrease in the logarithm of the neutron energy in a single moderating collision, and l is the mean free path between collisions. For hydrogen, ξ has the value unity, so that to thermalize neutrons from fission sources in water takes approximately 1(−5) s. In heavier moderators, the process is much slower; e.g., ξ for Pb is 0.003.

5. Spectrum of moderated neutrons

The spectrum of fully moderated neutrons is approximately Maxwellian, characterized by a temperature slightly higher than that of the moderator. Its amplitude slowly decreases with time because of absorption

and leakage.

The spectra of neutrons while in the course of moderation can be described only approximately, and then only for certain special cases. In general, numerical computations are necessary.

In an infinite hydrogenous moderator, uniform injection at $t=0$ of a burst of n_s very fast neutrons per unit volume gives rise to the spectrum (Williams, 1966)

$$n(v,t)=n_s(1/v)(vt/l)^2\exp(-vt/l) \quad \text{(J3)}$$

in which, after the first collision, neutrons of all velocities are present simultaneously. This formula neglects the heating of the moderator expressed in Eq. (J1). In the opposite extreme, a heavy-atom moderator, the neutrons tend to remain closely bunched in energy, while their average energy slowly diminishes with time (Bergmann, Isacoff, Murin, Shapiro, Shtranikh, and Cazarnovsky, 1956).

6. Neutron capture rate

For describing the kinetics of a neutron-pumped graser system (Appendix I.2) the time dependence of the pumping rate must be known. This is a convolution of the time and velocity dependences of the neutron density with the velocity dependence of the cross section for capture by the parent nuclide:

$$\lambda_p(t)=\int_0^\infty n(v,t)v\sigma_c(v)dv. \quad \text{(J4)}$$

Here σ_c is the capture cross section, v the neutron velocity, n the density per unit velocity range at velocity v, and $\lambda_p(t)$ the total neutron capture rate coefficient for a parent nucleus at time t.

Baldwin and Solem (1979b) have asserted that, when the neutron source is a single burst of fission or fusion neutrons, the velocity dependences of the neutron capture cross section and the limiting density of moderated neutrons [Eq. (J1)] are such that λ_p cannot approach values comparable with the decay coefficients of isomers in the Mössbauer range. This conclusion has been questioned by Karyagin (1979) and by Bowman (1980). The latter has noted errors in their numerical results.

APPENDIX K: ISOMER ENRICHMENT BY ATOMIC BEAM METHODS

Recently a combination of optical laser spectroscopy with atomic beam magnetic resonance (ABMR) techniques has been developed for precise measurement of nuclear properties (spin, magnetic moment, volume change, quadrupole moment), in which the numbers and wavelengths of optical resonance lines are determined (Murnick and Feld, 1979; Jacquinot and Klapisch, 1979). In certain versions of this general technique nuclear isomers as well as short-lived (10 ms) radioactive isotopes of elements in columns I and II of the Mendeleef chart have actually been enriched. Although the currents employed are extremely small [$\leq 1(10)\ s^{-1}$ or less], it is pertinent to this review to describe how the enrichment is accomplished.

The essential components of the ABMR method (Fig. 17) are (1) an accelerated ion beam, bombarding a target (2) within an oven (3) having a collimator (4) forming an atomic beam (5) of the escaping products of nuclear reactions in the target. From the oven, the atoms enter an interaction region to which a weak magnetic field (6) is applied and through which a beam (7) of circularly polarized light from a tunable laser (8) passes, exciting optical transitions in those atoms that are in resonance. The atomic beam then enters a strong, inhomogeneous magnetic field (9), having the property of focussing those atoms having their magnetic moments aligned in the direction of propagation while defocussing the remainder. Finally, the atoms reach a detector (10), which registers an increased current whenever a particular species of atom is being focused.

In the Letokhov proposal (Sec. V.B.4.b), the laser is also tuned to resonate with a selected hyperfine transition, but its beam is not polarized; those atoms having excited isomeric nuclei are ionized before they can relax, then focused and collected as ions. In the ABMR research, the selected atoms first relax into states that are magnetically oriented, then are focused as atoms, although they may then be ionized and mass-selected for identification immediately prior to detection.

Alignment of the nuclear angular momentum is effected by optical radiation acting on the electronic structure, through the coupling of nuclear and electronic angular momenta. Magnetic interaction of the electrons with the nucleus in a weak magnetic field replaces the quantum numbers J and I by F and m_F. For example, in the alkali Na, each of the levels giving rise to the D_1 resonance line ($^2S_{1/2}$, $^2P_{1/2}$) has two hy-

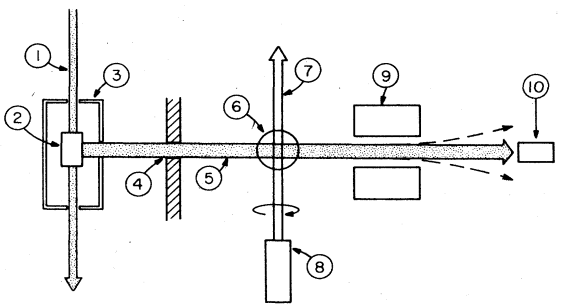

FIG. 17. Atomic beam magnetic resonance spectrometer with optical pump. The method of optical hyperfine spectroscopy described in Appendix K and illustrated here might be adapted for separation of nuclei in isomeric states from those in their ground states. A beam of accelerated ions (1) transmutes or excites nuclei in a target (2), located in an oven (3). Reaction products are collimated by an aperture (4) to form an atomic beam (5). In a weak magnetic field (6), traversed by polarized light (7) from a tunable laser (8), atoms containing isomeric nuclei are selectively pumped into substates of maximum total quantum number M_F. In the spectrometer, the oriented atoms are then analyzed by a six-pole magnet (9) which decouples the nuclear and electronic angular momenta, focussing those having positive M_J onto the detector (10), while defocussing those with negative M_J. For isomer separation, the analyzer might be modified so as to deliver atoms with selected M_I to a collector or to implant them into a host; an ionizer and ion-optical focussing stage might be required as well.

perfine components of total angular momentum, respectively,

$$F = I + \tfrac{1}{2} \quad \text{or} \quad F = I - \tfrac{1}{2},$$

each having twofold degeneracy that is removed by the applied magnetic field (Zeeman effect of hyperfine structure). Allowed optical transitions then conform to selection rules

$$\Delta F = \pm 1, 0$$

$$F = 0 \neq F = 0$$

$$\Delta m_F = 0, \quad s \text{ polarization}$$

$$\Delta m_F = \pm 1, \quad p \text{ polarization},$$

the signs being determined by the sense of circular polarization. If, say, right circularly polarized light is applied in the field direction with the appropriate frequency, bandwidth, and intensity, all allowed $\Delta m_F = +1$ transitions will be saturated; since spontaneous optical emission also occurs from each excited level to all permitted lower levels, eventually all the resonant atoms are pumped into the $m_F = I + \frac{1}{2}$ substate of the $^2S_{1/2}$ ground state. Similarly, left circularly polarized light pumps into the $m_F = I - \frac{1}{2}$ substate.

In the strong magnetic field of the analyzing magnet, however, the coupling of I and J is broken (Back-Goudsmit effect), and the appropriate quantum numbers are now m_I, m_J. Since the magnetic moment associated with J is far stronger than the nuclear magnetic moment, the analyzing magnet selectively focuses those atoms having the proper sign of m_J, irrespective of m_I. However, the optical pump and the adiabatic transition into the strong field will have assured that both m_I and m_J have their extreme permitted values.

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