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GRASERS: PROPOSALS, PROBLEMS, AND PROSPECTS*

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Abstract The possibility and potentiality of amplification of recoilless radiation emitted by nuclear isomers has long been recognized; nevertheless, development of gamma-ray lasers continues to await resolution of the pumping vs. linebreadth dilemma. We identify problems that accompany proposals for reducing the excitation requirements and suggest areas of investigation that may contribute to resolving the dilemma; several involve use of optical lasers.

INTRODUCTION

Einstein's demonstration that stimulated emission[1] is necessary to account for thermodynamic equilibrium in any kind of quantum system in the presence of electromagnetic radiation was published in 1917, but it was many years before, not only the importance, but also the wide scope of that derivation was recognized. In particular, the fact that his treatment applies to nuclear transitions seems not to be generally appreciated.

Einstein considered the rate equation for a two-level system

$$\dot{N}_2 = -AN_2 + BUN_1 w - BUN_2 \qquad (1)$$

in the presence of thermal radiation, for which the spectral density is

$$U = (8\pi h\nu^3/c^3)/(\exp\{h\nu/k\theta\}-1) \text{ J m}^{-3}\text{ Hz}^{-1} \qquad (2)$$

according to Planck's law. At equilibrium, we have

$$\dot{N}_2 = 0 \ ,$$

$$N_2/N_1 = wBU/(A + BU) = w \exp\{-h\nu/k\theta\} \ , \qquad (3)$$

the Boltzmann factor. Hence

$$B = A\lambda^3/8\pi h \qquad m^3 J^{-1} s^{-2}. \qquad (4)$$

The induced transition rate

$$\begin{aligned} BU &= (A\lambda^3/8\pi h)(nh\nu/\Delta\nu) \\ &= (\lambda^2 A/2\pi\Delta\nu)(nc/4) \end{aligned} \qquad (5)$$

turns out to be a cross section multiplied by a photon flux.

Except for the degeneracy factor w, any system whose ground state can resonantly absorb radiation can, when in its excited state, be stimulated to emit that same radiation. The Mössbauer effect demonstrates that stimulated emission is possible from nuclear states--in principle. The problem is to make it observable, and, eventually, useful.

After Einstein's fundamental investigation, 16 years elapsed before stimulated emission was first observed in the laboratory. At last, in 1933 Ladenburg[2] showed that a negative absorption term is necessary to account for the dispersion of optical resonance radiation in the positive column of a neon glow discharge. It is fascinating to speculate upon what might have happened had Ladenburg merely added a pair of mirrors to his apparatus and, thereby, created the first laser, long before nuclear fission was discovered. Perhaps nuclear physics might have developed much more slowly, and, today, protestors would be demanding that we "ban the laser." We would not be meeting in Oak Ridge to discuss applications of lasers in nuclear physics, nor the hoped-for applications of nuclear physics to lasers--which is the subject of this paper.

As it turned out, however, stimulated emission waited over two more decades for application in devices, although it was manifested quite clearly in microwave phenomena. Thus, the maser came first, then the optical laser, before the ultraviolet was reached. Today the soft x-ray region is under active attack. The nuclear member of the laser family still lies in the future, exciting today only occasional interest, except for a few die-hard enthusiasts who persist in believing that it, too, has tremendous potential and that, given an adequate level of effort, grasers will eventually be operating in the 5-100-keV range. In my allotted time, I can only briefly review the formidable array of problems that first must be overcome, and refer those who wish more-detailed information to several recent review articles.[3-5]

BASIC REQUIREMENTS

In Table I we list the essential features of optical lasers, which fall into two main types according to the nature of the host. Also, we show that lasers for very much shorter wavelengths either must use inner-shell electronic transitions of atoms or ions having picosecond or shorter excited-state lifetimes, or they must use nuclear transitions, which offer a wide range of lifetimes, because they include multipole orders higher than electric dipole.

Short lifetime and high quantum energy both demand high pumping power; therefore, an x-ray laser is inevitably a hot plasma. The nuclear analogue, on the other hand, can be pumped by nuclear reactions, but must remain a cool solid. It must, necessarily, operate with a spontaneous linewidth approaching the "natural width" determined by the lifetimes

SPECTRAL RANGE	OPTICAL		SHORTWAVE (> 1 keV)	
TYPE	SOLID	GAS	X-RAY	γ-RAY
ACTIVE MEDIUM	Cr^{+++}, Nd^{++}...	He/Ne, Co_2, Xe_2...	ION	NUCLEAR ISOMER
HOST	SAPPHIRE, GLASS	TUBE, BUFFER GAS	HOT PLASMA	COOL SOLID
PUMP	FLASHLAMP	ELECTRON BOMBARDMENT CHEMICAL REACTION	?	NUCLEAR REACTION (n,γ), (γ,γ')...
TRANSITION TYPE	ELECTRONIC, USUALLY E1			NUCLEAR, E > I OR M
	INNER VALENCE	VALENCE	INNER-SHELL	
LIFETIME	~ ms	~ μs	fs ⇔ ps	ps ⇔ My
LINEWIDTH	DOPPLER			~ NATURAL

TABLE I Comparison of essential elements and characteristics of lasers for optical radiation with those for shorter wavelengths.

of the two combining nuclear states, rather than with the broadened lines that characterize all the other types. That, essentially, is the reason that grasers have proved so difficult to develop. It also means that their kinetic behavior will be unlike that of atomic-transition lasers.

For lasing, stimulated emission must add photons into the radiation field more rapidly than absorption can remove them. The inequality

$$(N_2 - wN_1)\sigma_s > N_i\sigma_i \qquad (6)$$

known as the photon balance condition, is a necessary (but not a sufficient) condition. It demands a population inversion and a transparent medium.

From Mössbauer spectroscopy, we know that the cross section for interaction of monochromatic photons with a resonant system at exact resonance is

$$\sigma_s = (\lambda^2/2\pi)(\Gamma_r/\Gamma) \qquad (7)$$

where λ is the wavelength, Γ_r the probability per unit time for a radiative transition, and Γ is the total bandwidth of the spontaneously emitted radiation [see Eq. (5)]. We omit the statistical weight factor w from the cross section, so that the latter has the same value for both upward and downward transitions.

In this spectral region, photons are lost mainly by photoelectric absorption, with cross section

$$\sigma_i = C_i Z_i^{9/2}\lambda^3(1-\varepsilon) \qquad (8)$$

dependent on the atomic number Z and wavelength λ, with a coefficient C_i whose value depends on the particular electron shell involved in absorption.

The factor ε accounts for the Borrmann effect. In perfect crystals, photons can channel between the lattice planes if the Bragg condition is satisfied by interfering direct and scattered waves.[6] The resultant wave, whose electric vector vanishes at the lattice planes, is transmitted without undergoing photoelectric absorption, an electric dipole interaction, but its interaction with higher-order multipoles (nuclei) is not suppressed. This can reduce the absorption losses by nearly two orders of magnitude in favorable cases, while still permitting stimulation of the pumped nuclei. However, it is extremely sensitive to lattice imperfections, and it resembles the Mössbauer effect in its sensitivity to temperature.

Even without the Borrmann effect, the ratio of resonant to nonresonant cross sections lies in the range 10^2 to 10^5, and it is higher at short than long wavelengths. Matter is sufficiently transparent for a pumped medium to amplify recoilless gamma radiation; that is demonstrated by the Mössbauer effect. The Borrmann effect gives still more latitude for satisfying the balance requirement.

Next, we consider the pumping and linewidth problems. In Figure 1 we have expanded the photon balance inequality, listing separately all the contributions to the linewidth: intrinsic widths Γ_1 and Γ_2 of the two combining states; Γ_c, the chemical or monopole broadening; Γ_m, the magnetic dipole broadening; Γ_q, the quadrupole-electric field gradient broadening; and Γ_r, the recoil or Doppler broadening. Mössbauer and Borrmann effects are included via the factors f and ϵ. The possibility that the transition is between excited states calls for the branching ratio β, and internal conversion is accounted for by $1 + \epsilon$. We could also include gravitational red-shift broadening, but it is unimportant except for very long-lived transitions.

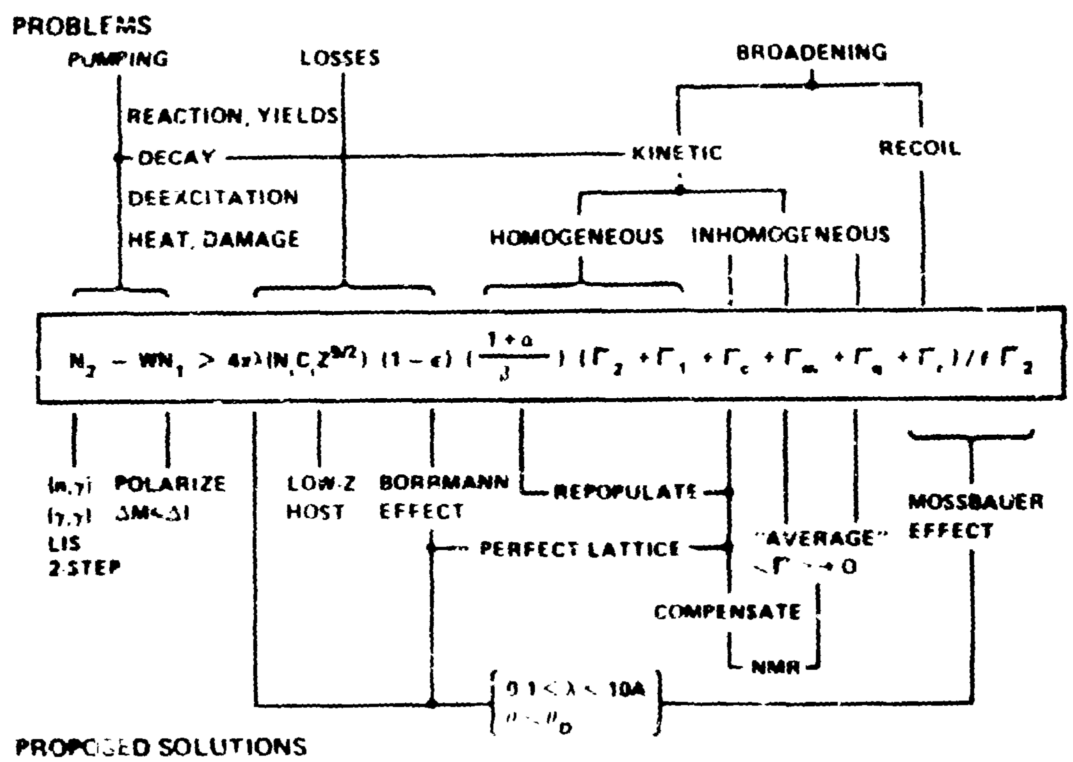

FIGURE 1 The photon balance condition in expanded form (box), problems that it presents (upper part), and proposed solutions to those problems (lower part). Lines point out relationships.

PROBLEMS

The problem categories of pumping, losses, and broadening are given in Figure 1 above the box detailing the photon balance inequality: the lines relate them to other problems and to the appropriate items in the photon balance condition.

Note the term "kinetic." It recognizes the Uncertainty Principle. Because linewidth is an inverse time, it requires a finite time for its determination.[8] Mössbauer experiments, in fact, show that the gamma-ray line is broader shortly after the excited state forms.[9] The resonant cross section is therefore a time-dependent quantity. If then, the ultimate linewidth is to approach that of the natural line, excitation by the pump decays, and the inversion disappears before resonance is established. Thus, the kinetic factor increases the pump requirements beyond those given by photon balance.[10]

Figure 2 shows, however, that unbroadened lines are emitted only from isomers of lifetimes less than 1 us. For them, the pumping power required to reach inversion is so enormous that it is natural to consider separating the pumping and lasing processes. This was the earliest approach suggested,[11] in fact; radiochemical methods are known which, in combination with nuclear reactions that might produce the excited state, can create inverted populations, but the chemical manipulations must be completed and the solid host must be assembled before decay has destroyed the inversion. For long-lived isomers, however, the spontaneous line is greatly broadened, so the resonant cross section is reduced. It has been proposed that selective laser excitation and ionization of atoms containing only the upper-state nu-

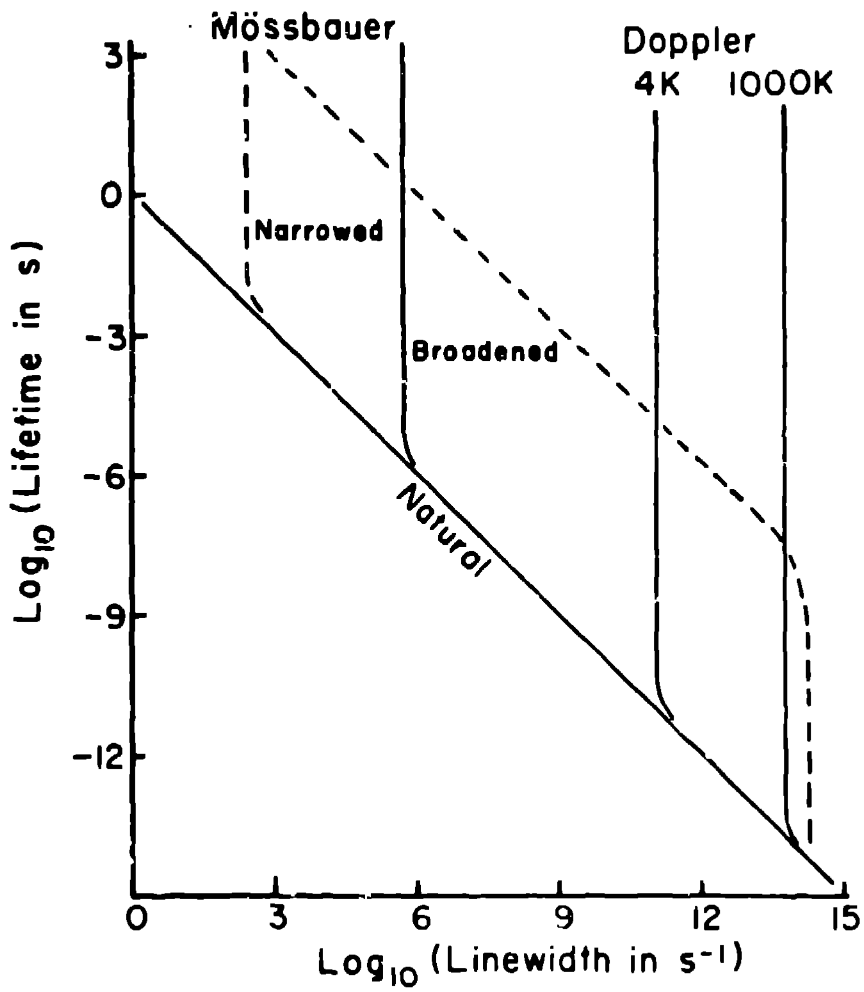

FIGURE 2 Linewidths vs. lifetimes for isomeric nuclear transitions. The natural linewidth is represented by the straight line inclined to the left. Doppler and Mössbauer widths are also shown. The dashed line indicates the maximum extent of line broadening allowed by photon balance.

clei would enable much shorter lifetimes to be used in a separation laser.[12] In a contributed paper, Ms. Dyer and I discuss the problems that accompany that approach with ev a long-lived isomer.[13]

If we are to use transitions of near-natural width, there can be no time for separation and assembly. We must pump them directly _in situ_. Proposed pumps include:

1) Neutron capture,[14] in which the capture gamma radiation is allowed to escape and, fortuitously, the reaction preferentially forms the isomeric nuclide;

2) Absorption of resonance radiation generated in an adjacent body by neutron capture -- so-called "Two-stage" pumping;[15]

3) Two-step pumping, in which two adjacent levels of widely differing lifetimes are required.[16]

Both of the latter two approaches require a third level. The first two _in situ_ pumping methods encounter a fundamental difficulty--that neutron sources generate fast neutrons. But we cannot moderate fast neutrons to the low energies at which capture is efficient, in the densities needed to create inversion.[17] Gol'danskii and Kagan[14] have estimated that at least 10^{18} neutrons per cm^2, of energy below 100 ev are required. Others, considering the kinetic factor, increase this estimate by nearly two orders of magnitude.[10,18] Our own studies[19] identified an additional factor: the time-spread that accompanies neutron moderation,[20] which exceeds the lifetimes of all known Mössbauer isomers. Therefore, only an intense source of neutrons that have energies much lower than fission neutrons would suffice.

Bowman has suggested that neutrons from a nuclear explosion could be thermalized and then Doppler-upshifted to match a capture resonance.[21] Others have suggested using photoneutrons; for example, electrons in a storage ring could be passed through a magnetic undulator to generate photons of energy just above the 1.665-MeV threshold for the γ,n reaction in beryllium or deuterium.[22] This would provide ideal line-geometry and traveling wave excitation, and it lends itself to both direct and two-stage pumping. It seems unlikely that enough neutrons can be generated in this way, however.

Thus, a persistent dilemma confronts us: recoilless emission and pumping seem basically incompatible, unless ways can be found to reduce the excitation requirements. Various suggested approaches are categorized, in Figure 1, beneath the photon balance inequality.

PROPOSALS

First, there is the two-stage principle, mentioned above.[15] The graser body is immersed in a large volume of convertor, in which neutrons are captured to excite recoilless resonance radiation. The resonance cross section of ground-state nuclei in the graser region is much higher than either the neutron-capture or the nonresonant absorption cross section. Therefore the density of excitation there is much higher than the density of neutron captures in the convertor. A third level is necessary to reach population inversion. However, geometrical losses at the interface, kinetic losses in the convertor, and, of course, the thermodynamic limitation on neutron density[17,23] prevent this otherwise ingenious idea from resolving our dilemma.

Various proposals exist for averaging out and for compensating the inhomogeneous interactions that broaden the line. Averaging[24] borrows a technique used for narrowing nuclear magnetic resonances. It brings the difficulty that both nuclear states are broadened, and, in general, they have unequal Larmor frequencies. The chemical, or monopole broadening, which accompanies the nuclear volume change, might be compensated with controlled hyperfine splitting in an internal magnetic field,[25] or reduced, if we can grow defect-free crystals or reduce the density of s-electrons in the atoms by laser techniques. These proposals, aside from serious questions about their implementation and effectiveness, suffer from two major problems: 1) present methods for measuring the linewidth, using first-order Doppler scanning, lack the sensitivity needed to demonstrate that a line has indeed been narrowed;[26] and 2) the kinetic principle, already noticed, sets a lower limit on the factor by which the line can be narrowed before decay leads to loss of inversion.[1]

One might hope even to reduce the homogeneous width. For instance, the internal conversion coefficient might be reduced by changing the density of s-electrons, which we just noted would also reduce the monopole broadening. At Los Alamos, in fact, we[27] plan to attempt to observe an effect of laser irradiation on the very low energy isomer (73-eV) of U-235; but we see little hope of having any significant effect on higher-energy isomers.

Attention is now focused mainly on the other side of the inequality. In particular, there are suggestions to exploit selection rules for the magnetic quantum number in a system of polarized nuclei. For example, Vysotskii calls attention to the case of Dy-161, which can be completely polarized at 0.02 K in a strong internal magnetic field.[28] Using a fortuitous approximate coincidence of energies, he suggests that the Ra-Kβ2 line can pump this isotope into an effectively inverted substate, from which the emitted radiations cannot be resonantly absorbed by nuclei left in lower states (Figure 3). Again, an ingenious idea is accompanied by a serious problem--how to pump without destroying the polarization.

But--suppose we pump before polarizing the nuclei, so as to ensure that the selection rules can be exploited? In this conference, we have seen how an isomeric nucleus can be optically pumped. Suppose that we could pump all the upper-state nuclei into states with an extreme positive value of M_I, while simultaneously pumping all lower states into extreme-negative M_I. Selection rules on the nuclear magnetic quantum number would eliminate resonant absorption

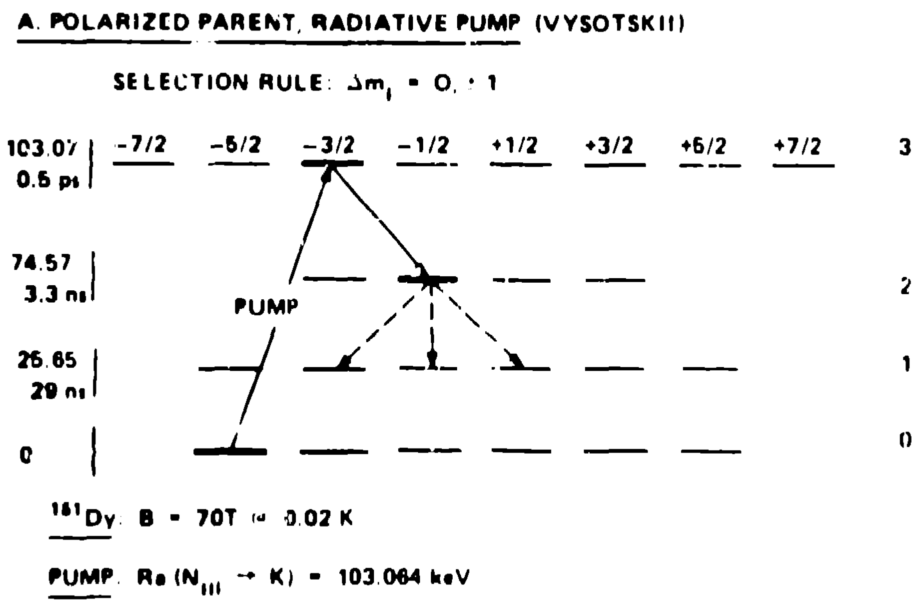

FIGURE 3 Energy levels of the nuclide Dy-161 in a strong magnetic field. At low temperature, only the -5/2 sublevel of the ground state is occupied; the sublevels that would be excited by a 103-keV radiative pump are indicated.

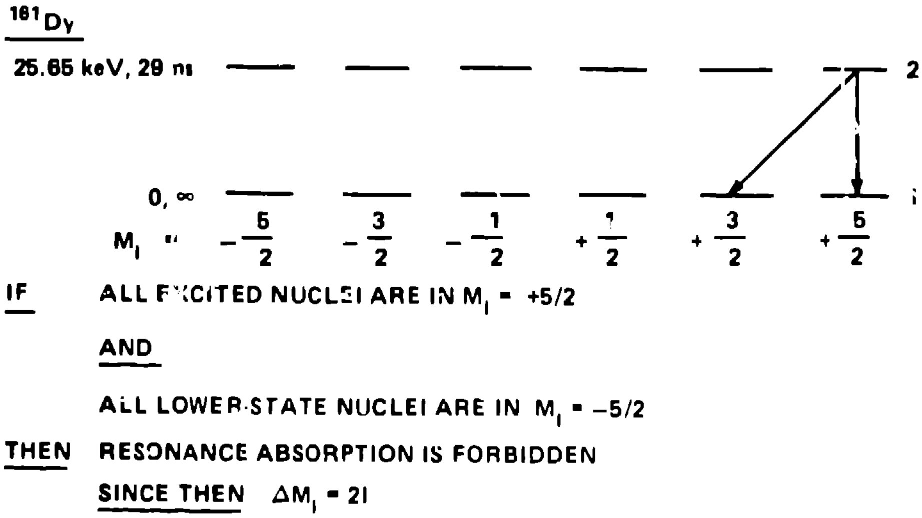

FIGURE 4 Magnetic substates of ground and 25,65-keV levels of Dy-161. Allowed transitions from the m = +5/2 substate are shown. If the lower-state nuclei were all in their −5/2 substate, they could not absorb that radiation.

(Figure 4). Then, population inversion would be unnecessary, and the excitation needed for lasing would be only that sufficient to overcome nonresonant absorption, several orders of magnitude less.

Again, difficulty. Unbroadened recoilless gamma radiation demands nuclear states of lifetimes no greater than 100 ns, and the nuclei must be in a solid host. Species that might be pumped by optical means, having sharp hyperfine structure in their resonance lines when in solid hosts,[29] include several rare earths with known Mössbauer lines.[30] Unfortunately, the nuclear lifetimes are of the order of nanoseconds; the optically excited state lifetimes are milliseconds. Although we are not optimistic, we are still hopeful that very brief laser pulses, or some other pumping method, can resolve this dilemma.

Two-step pumping remains for our consideration (Figure 5). Suppose that a long-lived nuclear state is separated from a short-lived state by only a very small en-

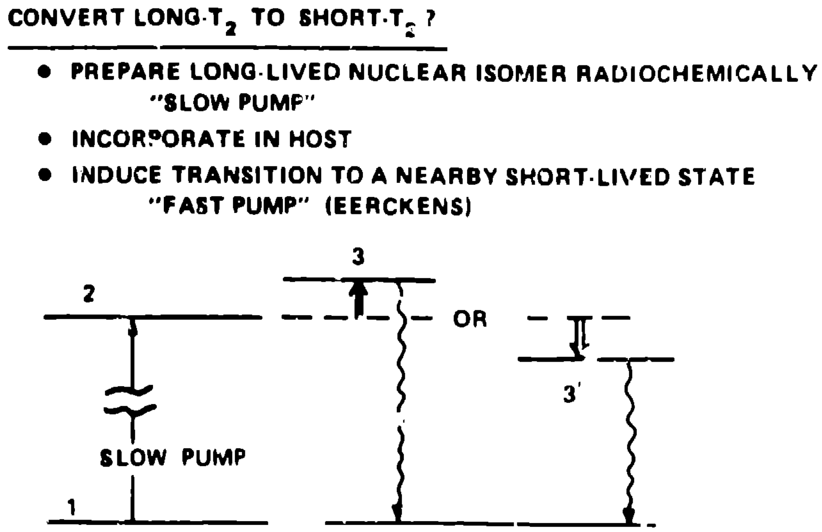

FIGURE 5 Basic requirement of two-step pumping: a long-lived isomer that can be prepared radiochemically and that has an adjacent short-lived level, to which a transition can be induced with only moderate input power.

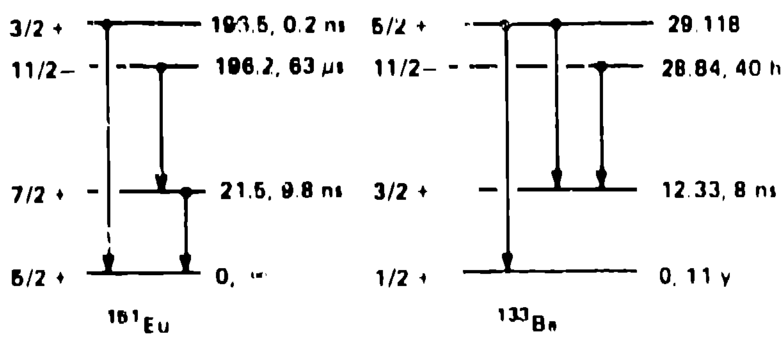

FIGURE 6 Energy levels of the nuclides Ba-133 and Eu-151. Note the pairs of levels spaced 300-eV apart, but having widely differing half-lives.

ergy gap. We first supply most of the excitation energy by a slow radiochemical procedure, then assemble the long-lived nuclei in an appropriate solid host. Lastly, by some means, we induce a fast transition that supplies only the energy needed to transform these nuclei into the short-lived species--a much more modest pump.

In searching for a suitable candidate, however, once again we meet with frustration. The closest pair of levels we find having widely differing halflives[31] is in Ba-133; they are 280 eV apart (Figure 6). How are we to induce the transition? Can it be done with synchrotron or undulator radiation? Are there closer pairs? Can multiphoton absorption induce the transition? We do not know.

There is a suggestion that a pair of real levels is not needed, if we can induce a two-photon transition, one of the photons being from an optical laser. This requires that a nucleus respond in a nonlinear fashion to the laser field, enabling the optical and gamma-ray frequencies to be mixed. When one considers the great disparity between nuclear radius and optical wavelengths, and between the most intense laser field strengths and the Coulomb fields of protons in a nucleus, there seems little reason to hope for this approach --in the case of an isolated nucleus. But the Mössbauer effect reminds us that a bound nucleus is not isolated. It may indeed be possible to mix optical and nuclear gamma radiation.[32] But, once again, we do not know, and it would be unwise to be dogmatic.

CONCLUDING COMMENTS

Grasers present a major challenge that only further work by many people in several distinct disciplines can finally

TABLE II Questions central to the development of gamma-ray lasers.

<u>Active Nuclide</u>:
Z, A
Transition parameters (energy, lifetime, etc.)
Possible chemical forms
Atomic spectrum, including HFS parameters
<u>Host medium</u>:
Z, A
Transparency
Debye temperature
Crystallographic form and perfection
Compatibility
<u>Pump</u>:
Type of radiation
Intensity available
Reaction type
Excitation mechanism
Cross sections for formation, destruction
Side effects
<u>Gain</u>:
Cross sections
Linewidths
Dependences (threshold, temperature, damage)
Kinetics
<u>Control</u>:
Hold-off
Trigger
Directionality

TABLE III Subjects needing investigation

Nuclear:

- Accurate level and transition parameters
- Isomer ratios
- Cross sections for formation and destruction
- Nuclear moments of isomeric nuclides
- Can nuclear and optical radiation be mixed?
- Can 300-eV transitions be induced? How?

Mössbauer:

- Linewidth measurement techniques
- Line-narrowing experiments
- Optical pumping (double resonance)
- Growth and behavior of radioactive crystals

Spectroscopic:

- HFS of atoms with isomeric nuclei in solid hosts

Isomer separation:

- Recoil ("Szilard-Chalmers") separation
- Laser photochemical
- Laser photoionization
- Other?

Neutron sources:

- Thermal, Doppler-upshift to match resonance
- Resonance, electron-undulator + Be

overcome, and I hope that many of you will begin to think seriously about this subject. I am confident that the challenge can be met by combining some of the approaches already considered with new ideas. To this end, Tables II and III list some of the areas in which research is necessary if further progress and new ideas are to be forthcoming.

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