TITLE: UNRESOLVED PROBLEMS IN GRASER DEVELOPMENT

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UNRESOLVED PROBLEMS IN GRASER DEVELOPMENT

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Abstract

Although it has been recognized for two decades that the Mössbauer effect offers the possibility of stimulated emission from nuclear states, the contradictory requirements of an intense pump and a relatively undisturbed solid host continue to frustrate attempts to devise a workable gamma-ray laser system. This paper reviews the basic requirement for gain and some recent proposals for satisfying it, identifying the difficulties each proposal presents. Finally, it suggests the kinds of information, experience, and ideas that may enable further progress.

Nuclear transitions in cold, solid hosts, pumped into a lasing condition, would offer several conceptual advantages over atomic transitions for laser sources at and below one Angstrom wavelength.

In the energy region dominated by photoelectric absorption, the basic photon balance condition for gain

\[
\frac{N_2 - N_1}{N} \frac{\sigma_{\text{absorption}}}{\sigma_{\text{resonance}}} = \frac{\sigma_{\text{resonance}}}{\lambda^2 f_{\text{rad}}/T} \quad (1)
\]

predicts that the inversion density required for lasing should decrease with shortening wavelength. In crystals, photoelectric absorption can almost vanish for Bornmann modes, and the Mössbauer effect\(^3\) directs most of the emission into a recoilless line which, for lifetimes shorter than a few microseconds, has nearly the natural width; the resonance cross section then approaches its full theoretical maximum.\(^4\) Moreover, the power density needed for pumping long-lived nuclear isomers to inversion is far less than for short-lived atomic transitions of comparable energy. Finally, nuclear reactions, especially neutron capture, offer a distinct approach to pumping. Nevertheless, at such short wavelengths, pumping is still an unsolved problem.

Suppose, solving the photon balance relation Eq. (1) for excited-state concentration \(N_2\), we estimate the lasing threshold for a hypothetical gamma-ray transition to the ground state from a 40-keV isomeric state of 100-\(\mu\)s mean lifetime. The isomer is formed by neutron capture. The parent isotope is embedded in a beryllium host, which furnishes the low-absorption, highly rigid lattice needed for \(r\) strong Mössbauer emission line, in the proportion 1 atom of parent to \(10^6\) Be atoms. Inserting numbers,

\[
N_2 > 1.2(19) \quad \text{N}_h > 1.2(23) \quad N_2 = 6.38(18) \quad N_1 = 5.62(18)
\]

We find that even if there is no internal conversion to compete with gamma emission (viz., \(\alpha=0\)), the excited states must be formed in a concentration exceeding that of the lower states by 6 per cent of the total parent population. That is not easily accomplished by neutron irradiation.

The situation is much worse when one takes account of the time required for fast neutrons to moderate to energies at which they can be captured efficiently, of the finite incubation

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time of a wave with nearly natural bandwidth (during which inversion decays), and of depletion of the parent.

Figure 1 shows the result of a kinetics study that took account of these processes. An infinite solid hydrogen moderator containing $^{82}$Kr is assumed to be bathed suddenly in fast neutrons. When these have moderated to 40 eV, they undergo strong resonance capture to form the 147-ns isomer of $^{82}$Kr, which emits a 9.3-keV recoilless line. The lower set of curves (Fig. 1a) accounts for the population densities of the parent and the two nuclear states. We see that inversion lasts only for about one mean lifetime, and that the parent is by then fully depleted. The upper curves (Fig. 1b) show the time-dependence of gamma-ray amplitude at a succession of distances, measured in absorption lengths, from a spontaneous source. Because of the termination of pumping, amplification is abruptly quenched, and barely reaches a tenfold gain of amplitude at four absorption lengths—far less gain than the photon balance relation predicts. To reach this impressive performance, we assumed an utterly unrealistic neutron density $5 \times 10^{24}$ cm$^{-3}$. Had the computation taken account of moderator heating, this enormous number of neutrons could not have moderated to the 40-eV resonance.

Moderator heating prevents generation of neutrons in densities adequate for pumping a graser. If the neutrons are generated by fission with energies in the MeV range. This suggests that we consider a source of much lower neutron energy. For example, an electron storage ring could be equipped with an undulator designed to generate a highly collimated line spectrum of photons with energies sufficient to eject neutrons from beryllium. The narrow-filament geometry and traveling-wave excitation features are just those required for a graser, and the short burst duration would allow pumping nuclear states of nanosecond life times. However, the photon bandwidths are too great for radiative pumping, and the neutron yields are too low for neutron-capture pumping; unless some way is found to reduce the excitation requirements. Perhaps the periodic field structure of perfect crystals may allow narrower photon bandwidth for pumping by electron-excited radiation, but there is no experimental demonstration to date that the electrons would not destroy the conditions required for gain.

We therefore continue to face the same dilemma that has always confronted every graser concept: how to reconcile pumping requirements with the requirements for recoilless emission. Use of long lived isomers that can be prepared and concentrated radiocchemically, then crystallized, is unpromising because, for them, the narrow natural line is washed out by inhomogeneous broadening. Short lived transitions may not be unduly broadened, but they take high pump power, and it destroys the Bortmann and Mossbauer effects.

We must reduce the excitation requirements. There are several approaches.

Proposals made nearly a decade ago to employ techniques used in NMR spectroscopy to narrow the recoilless line are still untested. They are complicated and they await development of a method for measuring the widths of the very narrow lines that might be achieved. Moreover, there is a fundamental limitation: to achieve a narrowed line, whatever the method used, will require a time at least as long as the reciprocal of the final...
This conceptual grater system would be pumped by resonance-energy photons generated in beryllium by photons from relativistic electrons passing through a magnetic undulator. The required parameters are given to the left of the figure.

Fig. 2. Two-stage pumping to inversion. The slow pump could be a nuclear reaction with radiochemical separation. The fast pump, shown by heavy arrows, forms a short-lived state which lases to the ground state (wavy line).

Consider the nuclide $^{133}$Ba, with the energy levels shown in Fig. 4. The $28.84$-keV metastable state might be prepared by proton bombardment of $^{133}$Ba or by neutron capture in $^{132}$Ba. The short-lived $29.118$-keV level has opposite parity and diffracts by three angular momentum units. But how can we make the $280$-eV transition to it? Electron bombardment might succeed, but it is extremely unlikely that the step could be induced by multiphoton absorption with a laser. This is as close as any pair of excited states of disparate lifetime that we have been able to find. However, few energy levels are known to sufficient accuracy to permit a conclusive judgment of the idea, and very close pairs could easily have escaped detection.

It has been argued that a real second level is not required. There are proposals to use an intense laser to induce either a two-photon cascade or to stimulate an anti-Stokes transition, thereby to de excite a long-lived level prepared by a slow pump. This assumes that the nucleus can respond to two quite distinct frequencies: gamma-ray and optical. The possibility that an appreciable nonlinear susceptibility could combine these frequencies into a nonlinear charge polarization at the form:

$$P_{\text{NL}} = \chi^{(3)}(\omega_1, \omega_2, \omega_3) N L,$$

linewidth, during which excitation created initially by the pump will be decaying.

Two-stage pumping, in which neutron capture in a large volume generates resonance radiation that excites nuclei in a small volume, fails to give an intensity advantage over direct pumping, because of geometrical and kinetic losses.

In the remainder of the paper we consider two other possibilities for reducing the excitation threshold. One is to subdivide the pumping process into fast and slow steps. The other is to eliminate lower-state resonance absorption, by exploiting selection rules for the magnetic quantum number in a system of polarized nuclei. Each has two variants.

Consider a long-lived nuclear isomer, capable of isolation and concentration by laser-based chemistry after a nuclear production reaction has furnished the main part of the excitation energy. Let there be also a short-lived level of nearly equal energy, reached by inducing a transition from the long-lived state, perhaps with a laser (Fig. 3). This second, fast pump is then immediately followed by recoilless emission from the short-lived state, which induces similar transitions in other nuclei similarly excited. This idea has been around for over a decade; however, we have found no case of two adjacent nuclear levels with energies so nearly equal that they might be mixed by laser-induced transitions.

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seems remote because there is a great disparity in the nuclear radius, the gamma-ray wavelength, and the optical wavelength. The most intense optical fields in laser focal spots are weak in comparison with the Coulomb field at the nuclear surface. Nevertheless, another paper at this conference suggests that optical and nuclear frequencies can indeed be mixed, and it cites experimental evidence to support that suggestion.

Another approach is to eliminate terminal-state absorption by polarizing the nuclei. The selection rule for the magnetic quantum number in a gamma-ray transition that carries off $L = 1/2$ units of angular momentum is

$$\Delta M_1 \leq L$$

If, therefore, upper and lower states of the transition have magnetic quantum numbers that differ by more than the difference of the nuclear spin numbers, then a transition between those substates is forbidden. Such a condition might be realized in either of two ways: 1) by statically polarizing the nuclei and then exciting them in a reaction that preserves their orientation; or 2) by dynamically polarizing the nuclei, possibly by optical pumping, after the nuclear pump has excited them.

Static polarization in a strong magnetic field at low temperature requires a relaxation mechanism fast enough that orientation is completed before loss of inversion by excited-state decay, unless the excitation occurs after orientation is complete. To preserve the polarization, a radiative pump is necessary, but unless it is extremely narrow-band, the polarization could not survive heating by the pump. Again we confront the graver dilemma: Note, however, that if the upper- and lower-state magnetic moments have opposite sign, polarization might indeed be accomplished in a previously pumped population before inversion had decayed, if the relaxation mechanism is fast enough.

Dynamic polarization with an optical pump is conceptually more promising. To see how this might eliminate terminal-level absorption, consider the simple case (Fig. 5) of a hypothetical atom having a resonance line connecting two atomic states, each of electronic angular momentum $J = 1/2$ (viz., the F line of an alkali) and a nucleus with two states of spins, respectively $I = 1/2$ and $I' = 5/2$. Gamma-ray emission in this case involves an angular momentum change of two units and is therefore magnetic or electric quadrupole. Interaction of electronic and nuclear angular momenta splits each atomic state into two hyperfine sublevels. For the excited-nucleus atom, their total quantum numbers are $F = 0$ and $F = 1$. The Weiss field Zeeman sublevels are shown in the next part of the figure.
Using a sharply tuned laser, we pump at the wavelength of the (3,3) hyperfine component with right-hand circularly polarized light. Simultaneously, with a second laser, and left circular polarization, we pump at the (1,1) component. We see that saturating with the laser, followed by radiative relaxation, ultimately populates the \( M_L = +3 \) sublevel of the atoms having excited nuclei, while simultaneously populating only the \( M_L = -1 \) sublevel of the normal atoms. If, then, the magnetic field is increased sufficiently to decouple nuclear and electronic angular momenta, the lower-state nuclei, having magnetic quantum number \( M_L = -\frac{1}{2} \), cannot resonantly absorb radiation emitted by nuclei in the \( M_L = +\frac{5}{2} \) sublevel. We thereby have eliminated the need for inversion of the total population.

The Mössbauer effect is still required, and it in turn requires a solid host. Fortunately, atoms exist having sharp hyperfine structures in crystal hosts— namely rare earths and actinides, and some of them have Mössbauer isotopes. Unfortunately, their optical relaxation times are measured in milliseconds; the gamma-ray lifetimes, in nanoseconds.

Therefore, excitation would vanish long before pumping could orient the excited nuclei. There may be ways to accelerate the population process without destroying the sharpness of optical hyperfine structure; we are not prepared to discuss them at this time.

One might combine the approaches we have described, so that the excitation requirements might indeed by lowered by a large factor—if we can find candidates with a fortuitous combination of properties. The active medium must have suitable nuclear, solid state, and optical properties. At this stage, we cannot be certain that neutron capture is the only way to pump, but with other kinds of pump a separation stage may be necessary. If we are to pump in situ, then the side effects of the pump—damage, heating, line broadening, destruction of isomeric states—all must be considered. A laser system involves many aspects besides just a pump.

Although the graser problem still resists solution, we believe that ultimately some combination of approaches will solve it. Meanwhile, the problems we have outlined today suggest areas in which research is needed to develop data and experience, from which new ideas may arise that will enable the final solution.

References

2. Ibid., Appendix G.
3. Ibid., Appendix D.
4. Ibid., Appendix C.
5. Ibid., Appendix I.
11. Reference 1, Section V.5.
17. C. B. Collins et al., Paper 0.3, this Proceedings.
Appendix: Symbols

N  total concentration of atoms cm⁻³
N₂,N₁  concentration of atoms with (excited, unexcited) nuclei
Nₚ  concentration of atoms with parent nuclei
σ  cross section, cm²
Cs  a numerical constant depending on the electronic shell in which photoelectric absorption occurs
λ  wavelength of gamma radiation, cm
e  a number, less than unity, that accounts for the anomalous transmission of Borrmann-mode photons
Z  atomic number of host atoms
f  fraction of all photon emission events in the recoilless line
Γ  total frequency bandwidth of the recoilless line
Γ_rad  probability per unit time for emission of radiation
W  ratio of statistical weights (2l₂+1)/(2l₁+1)
α  internal conversion coefficient
β  branching ratio
T₂,T₁  mean lifetimes of nuclei in state (2,1)
B  increase of line breadth due to inhomogeneous broadening
P_NL  nonlinear part of total induced electric moment
ε₀  permittivity of vacuum
X_NL  nonlinear second-order electric susceptibility
E  amplitude of electromagnetic wave
ω  angular frequency of (optical, nuclear) radiation
l  nuclear spin quantum number
M_l  nuclear magnetic quantum number
J  electronic angular momentum quantum number
F  total quantum number of atom
M_f  magnetic quantum number of atom