

neutrons in the matrix itself [cross section  $(\sigma_g)_m$ ] presents no danger here if  $\sigma_{n\gamma} \gg (\sigma_g)_m$ . Estimates indicate that the relatively mild condition  $\sigma_{n\gamma} \gtrsim 100$  b must be satisfied for this purpose.

Thus, we find that it is possible to combine the objectively available parameters with the population-inversion requirement of the  $\gamma$  laser without excessive overheating. This means that "head-on" pumping is possible, at least in principle, because the necessary "window" of self-consistent physical parameters exists. However, the required integral neutron fluxes over a time  $t \ll \tau_0$  are enormous:  $J_t \sim 10^{19}-10^{20} \text{ cm}^{-2}$ , i.e., on a level presently attainable only in nuclear explosions (here, of course, we refer to the pumping of many needles at once).

Nevertheless, there is hope for the development of  $\gamma$  lasers operating at much smaller pump-neutron fluxes. In fact, the number of active nuclei in a single laser needle should be no greater than  $10^{13}-10^{14}$ . This suggests the idea of simultaneous excitation of the Mössbauer levels (the optimum variant is  $n\gamma$  capture) and transfer of the excited nuclei to some kind of receiver. It might then be possible to collect very short-lived nuclei and to separate the overheating region, where the levels are populated, from the region of the subsequent  $\gamma$  emission into the bargain.

Variants of the transplantation type require pickoff of Mössbauer nuclei from the surfaces of neutron-bombarded samples using the  $n\gamma$  capture recoil energy. In the bombardment of highly dispersed ( $\sim 100 \text{ m}^2/\text{g}$ ) systems, even when only 1% of the bombarded volume is filled (in the form dust),  $1 \text{ cm}^3$  contains  $10^{19}-10^{20}$  nuclei that are ready to escape from the surface upon  $n\gamma$  capture. About  $10^{15}$  excited nuclei can be obtained by bombarding  $1-10 \text{ cm}^3$  of such a dust even at  $J_t \approx 10^{15} \text{ cm}^{-2}$  when  $\sigma_{n\gamma} = 10^{-21}-10^{-20} \text{ cm}^2$ —a number sufficient to bring about stimulated  $\gamma$  emission. The emitting Mössbauer atoms might, for example, be collected quickly by converting them into oxide molecules (in an oxygen atmosphere) and expelling them in supersonic jets, or by ionization and focusing with an electric field. In the variants considered, therefore, the working body of the laser (needle, thin film, etc.) has already been formed from short-lived Mössbauer emitters, perhaps also with dilution by light nuclei. We should note that the production of integral fluxes on these scales during times  $t \ll \tau_0$  on the order of  $10^{-6}$  sec is even now quite feasible, even though such neutron sources do not yet exist. Special plasma sources that generate neutrons in a thermonuclear reaction, e.g., sources of the "plasma focus" type, may prove to be the optimum. The question as to selection of the isotope that is optimal with respect to the entire aggregate of requirements remains open. At the moment, we do not know all of the necessary characteristics, and the so-called isomer ratio in particular, for any of the appropriate transitions. This lends urgency to the performance of special nuclear-spectroscopy studies.

**Basic conclusions.** 1) The development of a gas laser for long-lived ( $\tau > 10^{-1}-10^{-2}$  sec) nuclear levels as proposed earlier is rendered unrealistic by inhomogeneous broadening of the resonant-transition line by defects—at least without the incorporation of ideas based on artificial line narrowing.

2) A pulsed variant of population of excited levels

with lifetimes much smaller than  $10^{-2}$  sec seems realistic.

3) Only transitions for which the Mössbauer effect occurs can be used.

4) Radiative neutron capture is the optimum pumping variant.

5) It is shown that stringent requirements regarding overheating of the working crystal can, in principle, be met by strong dilution of the working nuclei in a light matrix and by making the body of the laser in the form of slender needles. However, the required pulsed neutron flux is on the scale of  $10^{19}-10^{20} \text{ n/cm}^2$ , which can now be attained only from nuclear explosions.

6) The required pump-neutron integral fluxes can be lowered by many orders of magnitude by exciting and separating the excited nuclei simultaneously, e.g., by using the recoil of the nuclei and a branched target surface.

- <sup>1</sup>V. I. Gol'danskiĭ and Yu. M. Kagan, Zh. Eksp. Teor. Fiz. 64, 90 (1973) [Sov. Phys.-JETP 37, 48 (1973)].  
<sup>2</sup>Yu. M. Kagan, Zh. Eksp. Teor. Fiz. 47, 366 (1964) [Sov. Phys.-JETP 20, 243 (1965)].  
<sup>3</sup>G. E. Bizina, A. G. Beda, N. A. Burgov, and A. V. Davydov, Zh. Eksp. Teor. Fiz. 45, 1408 (1963) [Sov. Phys.-JETP 18, 973 (1964)].

**Yu. A. Il'inskiĭ and R. V. Khokhlov. The Possibility of Observing Stimulated  $\gamma$  Radiation.**

The question as to the possible use of long-lived isomers for the development of  $\gamma$  lasers was first posed back in the early 1960's (see<sup>1,2</sup> and others). This question was answered in the affirmative in the first papers. Later, however, apparently insurmountable difficulties made their appearance, with the result that no serious experimental work on the  $\gamma$  laser has been done in recent years. Now interest in the  $\gamma$  laser has been revived by a quick succession of developments in various branches of physics<sup>2,3</sup>. The present paper is devoted to ways to solve the problem of observing stimulated  $\gamma$  emission by the use of long-lived isomers.

If there is a column of matter of length  $l$ , in which some of the nuclei are in an excited state,  $\gamma$  quanta will be emitted from the end of the column at an intensity

$$I = I_0 G,$$

where

$$G = \frac{\exp\{(\beta - \delta)l - 1\}}{(\beta - \delta)l};$$

here  $I_0$  is the spontaneous-emission intensity of a slender column,  $\beta$  is the gain due to stimulated emission, and  $\delta$  is the absorption coefficient.

In the range of atomic weights around 100, which includes most isomer transitions for  $\gamma$  quanta with energies  $E$  from 1 to 200 keV, absorption is determined basically by the photoeffect and decreases with energy as  $E^{-7/2}$ . Under normal conditions in pure substances,  $\delta = 10^3 \text{ cm}^{-1}$  for  $E = 10 \text{ keV}$ , and  $\delta = 5 \text{ cm}^{-1}$  at  $E = 200 \text{ keV}$ . The amplification is determined by

$$\beta = \pi \lambda^2 f \frac{1}{\Gamma \tau} \frac{1}{1 + \alpha} N,$$

where  $\lambda$  is wavelength,  $G$  is the width of the emission spectrum,  $\tau$  is the lifetime of the excited nucleus,  $\alpha$  is the internal conversion coefficient,  $N$  is the population difference between the excited nuclei and the nuclei on the lower level, and  $f$  is the probability of Mössbauer emission without transfer of energy to the nucleus.

A combined analysis of absorption and gain indicates that the highest  $\gamma$ -quantum energies consistent with a high Mössbauer transition probability should be used under optimum conditions. Assuming  $\lambda = 10^{-9}$  cm,  $f = 1$ ,  $N = 10^{22}$  cm $^{-3}$ , and  $\alpha = 0$ , we find that the amplification coefficient is of the order of 1 cm $^{-1}$  at  $\Gamma\tau \lesssim 10^4$ . Recognizing that several days are required to prepare the radioactive sample and adjust it to the conditions of the experiment, we have  $\tau \approx 10^6$  sec. This means that stimulated emission can be observed at a line width  $\Gamma \approx 10^{-2}$  Hz. It must be noted that the smallest line width observed thus far is  $10^5$  Hz. Thus, the possibility of observing stimulated emission depends on the possibility of effective narrowing of the spontaneous  $\gamma$ -emission line to a width on the order of  $10^{-2}$  Hz.

There are several mechanisms that broaden the Mössbauer-transition line over its natural width, when  $\Gamma\tau = 1$ ; they are enumerated below together with an analysis of the possibilities for their reduction.

1. Temperature broadening, which has been studied theoretically in<sup>4,5,1</sup>. At low temperatures it is determined by

$$\Gamma = \frac{288\pi^7}{7} \left( \frac{E}{2mc^2} \right)^2 \left( \frac{T}{\Theta} \right)^6 \frac{kT}{h},$$

where  $\Theta$  is the Debye temperature and the other notation is standard. Analysis of this relation indicates that the necessary conditions for a width on the order of  $10^{-2}$  Hz can be met with relative ease.

2. Inhomogeneous broadening due to chemical shifts on lattice defects. Screw dislocations are the dominant defects that appear when nearly ideal crystals are grown. The lattice-atom displacement field around the axis of such a dislocation decreases as  $1/r$ . It follows from symmetry considerations that the chemical shift  $\Delta\nu$  is determined by the following relation far from the axis of a screw dislocation:

$$\Delta\nu = \Delta\nu_0 \left( \frac{a}{r} \right)^2,$$

where  $\Delta\nu_0$  is the chemical shift on the axis of the dislocation and  $a$  is the interatomic distance.

If we taken typical values around  $3 \times 10^7$  Hz for  $\Delta\nu_0$  and  $10^{-2}$  Hz for  $\Delta\nu$ , nuclei with the level shift cover a range  $r/a = 5 \times 10^4$ , which corresponds to a dislocation density of  $3 \times 10^5$  cm $^{-2}$ . Such dislocation densities are easily obtained.

Similar reasoning for edge dislocations leads to the relation  $\Delta\nu = \Delta\nu_0 a/r$ , whence it follows that a 1 cm $^3$  crystal must not contain edge dislocations. The number of point defects must not exceed  $10^{14}$  cm $^{-3}$ .

Although these last requirements are quite stringent, progress in the growing of defect-free crystals indicates that it may be possible to meet them.

3. Magnetic dipole-dipole interaction of nuclei. This interaction results in broadening of the order of  $10^4$ – $10^5$  Hz due to the fact that the magnetic moments of the nucleus in the states between which the transition occurs are different even when the nuclear moments are

fully oriented by external and internal fields. The same interaction determines the line width in NMR.

The line can be narrowed by applying to the sample a series of radio-frequency  $\pi/2$  pulses<sup>6,1</sup>.

In the presence of a strong static magnetic field, the expression for the dipole-dipole interaction takes the form

$$\mathcal{H}_d = \sum_{i < j} b_{ij} (I_{iz} I_{jz} - 3I_{iz} I_{jz}) + \sum_{i < j'} b_{i'j'} (I_{i'z} I_{j'z} - 3I_{i'z} I_{j'z}) - 2 \sum_{i < j'} b_{i'j} I_{i'z} I_{jz} \quad (1)$$

the indices  $i$  and  $j$  number the lattice points occupied by nuclei in the excited states, while  $i'$  and  $j'$  refer to the ground state (or the lower state if the lower state is not the ground state).  $I_{iz}$  and  $I_{jz}$  are the nuclear-spin operators, and the constants  $b_{ij}$  depend on the relative positions of the interacting nuclei, the direction of the external magnetic field, and the values of the magnetic moments in the ground and excited states. In a rigid lattice, the  $b_{ij}$  are not time-dependent.

Under the action of short  $\pi/2$  radio pulses, which set the nuclear moments in rotation around the respective axes, the operator  $I_{iz}$  (and, similarly,  $I_{i'z}$ ) is transformed to  $I_{ix}$  ( $I_{iy}$ ). It is possible to select a series of  $\pi/2$  pulses such that  $\mathcal{H}_d$  vanishes on averaging over the period of the pulse series. Here the line width decreases sharply, approximately in the ratio<sup>6</sup>  $6/(\Gamma_0\tau_0)^2$ , where  $\Gamma_0$  is the line width before averaging and  $\tau_0$  is the period of the pulse series. The pulse series must be somewhat different from those used in NMR so that both the resonant interaction of the form  $(I_{iz} I_{jz} - 3I_{iz} I_{jz})$  and the nonresonant interaction  $I_{i'z} I_{jz}$ , between nuclei in the different states will vanish. We might, for example, choose a series  $(90^\circ_x, 2\tau_1; 90^\circ_x, \tau_1; 90^\circ_y, 2\tau_1; 90^\circ_y, \tau_1)$  for one state and a series (synchronous with the first)  $(90^\circ_x, 2\tau_1; 90^\circ_x, \tau_1; 90^\circ_y, 2\tau_1; 90^\circ_y, \tau_1)$  for the second state. NMR experiments indicate that the line width can be reduced by several orders of magnitude in this way.

4. In crystals with structural defects, there is a substantial broadening of the line due to interaction of the nuclear quadrupole moment with the electric-field gradient. In the presence of a strong magnetic field, when the Zeeman interaction may be larger than the quadrupole interaction (for example, in cubic crystals), the Hamiltonian takes the form<sup>7,1</sup>

$$\mathcal{H}_Q = \frac{eQV_{zz}}{4I(2I-1)} [3I_z^2 - I^2], \quad (2)$$

where  $V_{zz}$  is the second derivative of the electric-field potential in the  $z$  direction, which corresponds with the direction of the external static magnetic field. The Hamiltonian (2) depends on  $I$  in the same way as the first two terms of Hamiltonian (1) and will be averaged along with the Hamiltonian of the dipole-dipole magnetic interactions. Thus, we have an opportunity for substantial narrowing of the Mössbauer emission line down to several hundredths of a hertz, thus opening the way to observation of stimulated  $\gamma$ -emission effects.

We note in conclusion that the traditional methods of Mössbauer spectroscopy are not applicable to investigation of lines narrower than  $10^5$  Hz. New methods must be found here. Correlation methods and methods based on the gravitational line-shift effect may be useful in this connection<sup>8,1</sup>.

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W. Vali and V. Vali, Proc. IEEE (Russ. transl.) 51, 223 (1963); G. C. Baldwin et al. *ibid.*, p. 849 (Russ. transl., as above, p. 1241).

<sup>2</sup>R. V. Khokhlov, ZhETF Pis. Red. 15, 580 (1972) [JETP Lett. 15, 414 (1972)].

<sup>3</sup>V. I. Gol'danskiĭ and Yu. M. Kagan, Zh. Eksp. Teor. Fiz. 64, 90 (1973) [Sov. Phys.-JETP 37, 48 (1973)].

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<sup>6</sup>U. Haerberlou and J. S. Waugh, Phys. Rev. 175, 453 (1968).

<sup>7</sup>A. Abragam, Principles of Nuclear Magnetism (Russ. transl.), IL, 1963, p. 225 [Oxford, 1961].

<sup>8</sup>C. A. Mead, Phys. Rev. 143, 990 (1966).

### V. S. Letokhov. Use of Lasers in Nuclear Spectroscopy

The paper considers the possible use of coherent laser light for: 1) production of narrow frequency-tunable emission and absorption resonances in nuclear  $\gamma$  emission, 2) highly accurate measurement of nuclear recoil energies on emission or absorption of  $\gamma$  quanta, 3) highly accurate measurement of the energy of metastable (isomeric) states of nuclei, 4) rapid separation of excited and unexcited nuclei, i.e., sorting of nuclei by states.

Two obvious physical facts underlie all of these effects:

- the state of the nucleus (spin, excitation energy, mass) is seen in its atomic and molecular spectra;
- motion of the atom or molecule simultaneously causes a Doppler frequency shift for both optical and nuclear transitions, together with one less obvious fact:
- buildup of molecular oscillations causes frequency modulation of the nuclear transition.

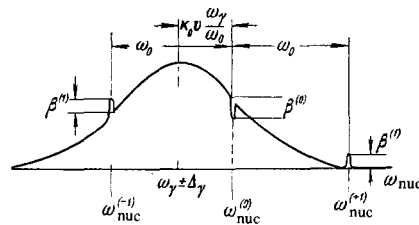
The production of narrow nuclear  $\gamma$ -emission resonances, unlike that of Mössbauer resonances<sup>[1]</sup>, is based on the action of a coherent light wave on an infrared vibrational transition of a molecule in a gas at comparatively low pressure ( $10^{-2}$ – $10^{-1}$  Torr). A coherent wave of frequency  $\omega$  builds up the vibrations of molecules having a certain projection of their velocity  $v$  onto the direction of the wave vector  $k_0$  ( $k_0 v = \omega - \omega_0 = \Omega$ ,  $\omega_0$  is the center frequency of the Doppler absorption line)<sup>[2]</sup>. The mean oscillator displacement  $\tilde{a}$  of the nucleus on excitation of molecular vibration is of the same order as or even larger than the wavelength  $\lambda_\gamma$  of  $\gamma$  quanta with energies from 10 keV to 1 MeV. The vibration of a nuclear radiator with amplitude  $\tilde{a} \gtrsim \lambda_\gamma$  should cause considerable frequency modulation of the nuclear emission. The result is distortion of the Doppler-broadened line of the nuclear transition as observed collinearly with the light wave<sup>[3,9]</sup> (see the figure). A resonance dip appears at the frequency

$$\omega_{\text{nuc}}^{(0)} = (\omega_\gamma \pm \Delta\gamma) + \frac{\omega_\gamma}{\omega_0} k_0 v$$

and resonance peaks at the frequencies

$$\omega_{\text{nuc}}^{(m)} = \omega_{\text{nuc}}^{(0)} \pm m\omega_0$$

( $\omega_\gamma \pm \Delta\gamma$  is the center frequency of the Doppler line with consideration of the recoil energy  $\Delta\gamma$ ). Vibrations



of molecules with a desired velocity projection  $k_0 v / k_0$  can be built up by changing the frequency deviation of the light wave with respect to  $\omega_0$ , thus effecting frequency retuning of the narrow resonances within the limits of the Doppler contour of the nuclear transition. Schemes for observing narrow  $\gamma$  resonances in emission and absorption have been examined.

A narrow  $\gamma$ -radiation resonance can be obtained in a nuclear Doppler line by the action of a light wave on an atomic electron transition (double  $\gamma$  and optical resonance<sup>[4]</sup>). Excitation of the atom's electron shell has no influence whatever on the nuclear transitions. The situation changes materially if "runoff" of atoms that have been selectively excited by the light wave is provided (atoms with  $k_0 v = \omega - \omega_0$ ) are excited. This can be done by photoionization, of excited atoms only, by additional laser radiation, with subsequent extraction of the ions by a small electric field. Because of the resulting shortage of nuclei with the resonance velocity, a narrow dip appears in the Doppler line of the nuclear transition and can be observed as an absorption minimum of the  $\gamma$  radiation from the Mössbauer source at  $(\omega_\gamma / \omega_0) k_0 v = -\Delta\gamma$ . This permits measurement of the recoil energy with a relative error of  $10^{-5}$ .

Excitation of an atomic nucleus in a molecule should result in a change in the vibrational frequencies of the molecule, since the excitation energy  $\Delta E$  is equivalent to an increase in nuclear mass by an amount<sup>[5]</sup>  $\Delta m = c^{-2} \Delta E$ . The resulting isomeric shift in the vibrational-rotational spectrum is smaller than or of the same order as the Doppler broadening. It can therefore be observed only by modern methods of laser spectroscopy within the Doppler width. This method can be used to resolve nuclear levels separated by  $\sim 1$  keV and to determine the energy of the nucleus accurate to  $\sim 10$  eV.

The isomeric shift due to excitation of the nucleus also appears in the atomic spectra as a result of the weak electron-neutron interaction. The magnitude of isomeric shifts in the atomic spectrum may greatly exceed the Doppler width, and this opens the way to selective laser excitation only of atoms with excited nuclei. Again, physical separation of the isomers can be accomplished by photoionization of the excited atoms with additional laser radiation and collection of the ions<sup>[6]</sup>. We note that selective two-stage photoionization of atoms of a given species by laser radiation was first demonstrated in<sup>[7]</sup>. This method can be used for fast separation of isomers with lifetimes down to  $10^{-4}$ – $10^{-6}$  sec, i.e., faster by a factor of  $10^7$ – $10^9$  than is possible by existing methods of nuclear chemistry. It can be used to create inversions on nuclear transitions with short excited-state lifetimes<sup>[8]</sup>.

<sup>1</sup>R. L. Mössbauer, Zs. Phys., 151, 124 (1958).

<sup>2</sup>V. S. Letokhov, ZhETF Pis. Red. 6, 597 (1967) [JETP Lett. 6, 101 (1967)].