

On the problem of the nuclear-transition γ -laser

V. S. Letokhov

Institute of Spectroscopy, USSR Academy of Sciences

(Submitted November 3, 1972)

Zh. Eksp. Teor. Fiz. **64**, 1555-1567 (May 1973)

The possibility of constructing a γ -laser based on recoilless transitions between the lowest nuclear levels is considered. A distinctive feature of the device is that its operation is based on relatively short-lived isomeric nuclear states with lifetimes of 0.1 to 10 sec. The minimum number of excited nuclei necessary for obtaining appreciable amplification is estimated. The possibility of producing the requisite number of excited nuclei by irradiating a target with a thermal-neutron stream or with resonance γ -radiation is considered. Then a low-inertia laser extraction from the target of a relatively small fraction of excited nuclei of a given composition, using the method of two-stage selective photoionization of atoms with excited nuclei by the radiation of two lasers, is considered. It is concluded that in principle a γ -laser of this type can be realized in the 20–60 keV energy range by the considered scheme.

1. INTRODUCTION

After the construction of the optical-band quantum generators, the possibility of extending the principle of the quantum amplification of radiation to the shorter wavelength region on the basis of stimulated radiative transitions between nuclear levels was discussed in a number of papers^[1-3]. It was noted in these papers that the most favorable transitions are the Mössbauer transitions for which the radiative-transition cross section at the peak is equal to

$$\sigma_0 = \frac{\lambda^2}{2\pi} \frac{f}{1 + \alpha} \quad (1)$$

(where λ is the wavelength of the radiation and α is the internal-conversion coefficient of the transition), i.e., σ_0 coincides up to the Debye-Waller factor f with the limiting value of the cross section of an oscillator of wavelength λ . In this case even in the very shortwave region—for example, at $\lambda = 0.1 \text{ \AA}$ (this corresponds to a transition energy $\hbar\omega_\gamma = 120 \text{ keV}$, which is a typical energy for rotational nuclear levels^[4])—for the quite attainable transition parameters $f \approx 0.1$ and $\alpha \approx 1$, the radiative-transition cross section $\sigma_0 \approx 10^{-20} \text{ cm}^2$, i.e., significantly exceeds the photoelectric-absorption cross section for the heavy atoms in this region: $\sigma_e = 10^{-22} - 10^{-21} \text{ cm}^2$ ^[4]. Furthermore, an appreciable gain $\kappa \approx 10 \text{ cm}^{-1}$ can be attained in this case when the inverted-population density $n = \kappa/\sigma_0 = 10^{21} \text{ cm}^{-3}$, which corresponds to a few percent of the nuclear density in a condensed medium. It also follows from this that the use of narrow, recoilless transitions is in principle necessary, since in the opposite case, because of the broadening of the transition, the density of excited nuclei necessary for obtaining such an amplification exceeds the nuclear density in a solid by several orders of magnitude. This fundamental aspect was recently pointed out by Khokhlov^[5] and Gol'danskiĭ and Kagan^[6].

The main problem lies in the creation of a nuclear-level population inversion. In^[1-3,5] the authors consider the possibility of using for this purpose the long-lived isomeric nuclear states with lifetimes of the order of a day or more, lifetimes sufficient for the separation of the excited (radioactive) and unexcited nuclei, using the existing methods of radiochemistry. In this case the lines to be amplified should have a width $\Delta\nu \ll 10^{-4} \text{ Hz}$, i.e., the relative width of the Mössbauer transition with an energy of 10–100 keV should attain the value $\Delta\nu/\nu = 10^{-23} - 10^{-22}$. The production of such narrow Mössbauer

resonances, even in absorption, is an extremely serious problem, since we must eliminate the weakest possible perturbations. It seems more realistic to use isomeric states of shorter lifetimes, but this, of course, requires that we give up the existing methods of separation of nuclear isomers. It is precisely this aspect of the γ -laser problem that is the subject of the present article.

In Sec. 2 we estimate the minimum number of excited nuclei necessary for obtaining an amplification of the order of 10^2 per pass of the radioactive sample. This value lies in the range of 10^{15} nuclei at $\lambda = 0.4 \text{ \AA}$. Then (Sec. 3) we propose a scheme for a γ -laser based on the isomeric nuclear states with lifetimes of 0.1–10 sec. The idea behind such a laser consists in the rapid production of the requisite number of excited nuclei in a target (for example, by irradiating the target with a pulsed thermal neutron flux), followed by a rapid laser separation of a relatively small fraction (10^{-5}) of the excited nuclei in a gas stream produced by rapidly vaporizing the target, and the concentration of the excited nuclei onto a microtarget. The distinctive features of the preparation of the required quantity of excited nuclei ($\sim 10^{15}$) are considered in Sec. 4, while the laser separation of a small fraction of the excited nuclei is considered in Sec. 5.

2. THE MINIMUM NUMBER OF EXCITED NUCLEI

Let us first estimate the minimum number of excited nuclei necessary for obtaining an appreciable amplification under practically attainable conditions. This estimate arises from the requirements imposed on the minimum gain, i.e., on the length of the active medium for a given density of excited nuclei, and on the minimum transverse dimension of the active medium because of diffraction.

As the minimum length L_m we can choose the length for which the gain per pass $k = \exp(\kappa L_m) = 10^2$, where $\kappa = \sigma(n_2 - n_1 g_2/g_1) = \sigma \Delta n$ is the gain per unit length, n_1 is the population of the ground (1) and the excited (2) nuclear levels, and σ is the radiative-transition cross section at the peak. It must be borne in mind when working with long-lived isomers ($\tau > 10^{-5} \text{ sec}$) that the width of the Mössbauer line is determined not by the natural width $\Gamma_0 = (1 + \alpha)/\tau$ (the contribution of the decay of the lower state to the broadening is neglected), but by

the inhomogeneous broadening on account of the numerous perturbations of ultranarrow lines¹. Bearing in mind that we want to use transitions with lifetimes $\tau \approx 0.1-10$ sec, we must take this effect into account. Then, the transition cross section σ is equal to

$$\sigma = \sigma_0 \Gamma_0 / \Gamma_{\min}, \quad (2)$$

where σ_0 is the limiting radiative-transition cross section at the peak, determined by the expression (1), and Γ_{\min} is the minimum inhomogeneous width of the Mössbauer line. For the subsequent estimates, let us assume $\Gamma_m \approx 10^5 \text{ sec}^{-1}$, which conforms with the already attainable level of technology of γ -resonance experiments. In consequence, we obtain for the minimum length L_m the estimate:

$$L_m = 10\pi \frac{1 + \alpha}{f\lambda^2 \Delta n} \frac{\Gamma_m}{\Gamma_0} = 10\pi \frac{\Gamma_m \tau}{f\lambda^2 \rho n_0}, \quad (3)$$

where Δn is the difference between the lever-population densities, which constitutes the ρ -th part of the total nuclear density $n_0 = n_1 + n_2$, i.e., $\Delta n = \rho n_0$. In the optimum case the lower level is empty ($n_1 = 0$), and $\rho = 1$.

For cylindrical geometry of the active medium (a radioactive filament), the minimum radius a_m is determined from the condition of smallness of the diffraction losses, or from the condition imposed on the Fresnel number $a_m^2 / L_m \lambda \gtrsim 3$, i.e.,

$$a_m = (3L_m \lambda)^{1/2}. \quad (4)$$

Here we take into consideration only the diffraction resulting from the finite transverse dimension of the medium, and do not consider the possible effects of the diffraction by the periodic structure, which, at certain orientations of the crystal axis, can give rise to additional losses.

From the relations (1), (3), and (4) we find the minimum volume $V_m = \pi a_m^2 L_m$ of the active medium and the minimum number N_m of excited nuclei necessary for obtaining an amplification per pass $k \approx 10^2$:

$$N_m = n_2 V_m = \frac{1 + \rho}{2} \left(\frac{\Gamma_m \tau}{f\rho} \right)^2 \frac{10^4}{n_0 \lambda^3}. \quad (5)$$

For a transition with energy ~ 30 keV ($\lambda = 0.4 \text{ \AA}$) in a pure sample of nuclear density $n_0 = 3 \times 10^{22} \text{ cm}^{-3}$, we obtain for $n_2 \gg n_1$

$$N_m = 5 \cdot 10^6 (\Gamma_m \tau / f)^2, \quad (6)$$

where the factor inside the brackets determines the deviation of the parameters of the Mössbauer transition from the ideal case: $\Gamma_m \tau = 1$ and $f = 1$. As has been noted above, it is realistic to count on $\Gamma_m \approx 10^5 \text{ sec}^{-1}$, and then, for example, for $\tau = 0.1$ sec the factor $(\Gamma_m \tau)^2 = 10^8$. Thus, for a transition with $f \approx 1$ the minimum number of excited nuclei is equal to $N_m = 5 \times 10^{14}$, the minimum sample length is $L_m = 0.7$ cm, the minimum diameter $2a_m = 10^{-4}$ cm, and the gain $k = 10^2$.

In the case of plane geometry of the active medium (a radioactive film) the expression (4) determines the thickness $2a_m$ of the medium and, instead of the relation (5), we have

$$N_m = n_2 (2a_m b) = \frac{1 + \rho}{2} \left(\frac{\Gamma_m \tau}{f\rho} \right)^2 \frac{6 \cdot 10^2 b}{n_0^{1/2} \lambda^{3/2}}, \quad (7)$$

where b is the width of the film ($b \gg 2a_m$). For the previous parameters of the active medium and for $b = 10^{-2}$ cm, we have $N_m = 3 \times 10^{16}$, the minimum thickness of the film $2a_m = 10^{-4}$ cm, the length $L_m = 0.7$ cm, while the gain is $\kappa = 2.5 \times 10^2 \text{ cm}^{-1}$.

Thus, it follows from the relations (5) and (7) that the number of excited nuclei necessary to obtain a gain of the order of 10^2 is relatively low ($\sim 10^{15}$), but it increases substantially when: 1) the parameters of the Mössbauer transition—the factor $(\Gamma_m \tau / f)^2$ —deviate from the ideal case, 2) the inverted-population density differs sharply from the total nuclear density ($\rho \ll 1$), 3) the nuclear density n_0 decreases, and 4) the geometry of the sample deviates sharply from the optimum thin-filament geometry. The transition-energy region is dictated by the condition of existence of the Mössbauer effect with the factor $f \approx 1$ ($\hbar\omega_\gamma < 100$ keV) and by the region where a significant number of isomeric states with a not very large internal-conversion coefficient ($\hbar\omega_\gamma > 10$ keV) exists. The 20–60 keV energy range is the optimum region.

From the relation (5) follow two alternative ways of constructing a γ -laser based on the realization of the maximum value of the factor $(\Gamma_m \tau / f\rho)^2$. The first method is to maximize the quantity $\Gamma_m \tau$ by using short-lived isomeric states with $\tau < 10^{-5}$ sec, in which case $\Gamma_m \tau \sim 1$. This variant has been considered in detail by Gol'danskiĭ and Yu. Kagan^[6]. Its disadvantages are: a decrease of the factor ρ on account of the impossibility of promoting all the neighboring (with respect to the mass number) nuclei of the nucleus M_k (k denotes the number of neutrons in the nucleus M) to the excited level of the nucleus M_{k+1}^* produced upon the capture of a neutron, a decrease of the factor f because of the heating of the sample when bombarded by the neutrons, and the necessity to use neutron streams of extreme intensity. Because of the shortness of the time τ , the separation of the excited nuclei and their concentration in a separate sample is problematic in such a variant.

The second method, which is considered below, is to use isomeric states with $\tau \gtrsim 0.1$ sec. This leads to a loss in the factor $\Gamma_m \tau$, but it allows us to attain, through the separation of the nuclei, high values of the fraction ρ of excited nuclei and the Debye-Waller factor f and to considerably reduce the required intensity of the neutron streams. For the realization of the second method of constructing the γ -laser, we need a method for preparing a concentrate of excited nuclei in a sufficiently short time, even though the absolute quantity of radioactive material may weigh only 10^{-8} g.

In using microquantities of a substance, we must make sure that the momentum imparted to a microsample of finite mass $M_s = A n_0 V_m$ (A is the atomic weight of the nuclei) does not lead to additional broadening of the Mössbauer line. The condition for this is

$$(\hbar\omega_\gamma)^2 / 2M_s c^2 < \hbar\Gamma_m. \quad (8)$$

If we use lines with $\Gamma_m \approx 10^5 \text{ sec}^{-1}$ and $M_s \gtrsim 10^{-8}$ g, then this condition is certainly fulfilled.

3. SCHEME FOR A γ -LASER WITH OPTICAL SEPARATION OF THE EXCITED NUCLEI

The necessity for preparing a concentrate of excited nuclei forces us to give up the schemes that directly use the excitation of the sample itself. In this case it is difficult to count on simultaneously obtaining nuclear-level population inversions and the excitation of a significant fraction (several percent) of one sort of nuclei relative to all the other sorts in the target. It seems expedient to use a sample of nuclei of the requisite composition in the excited level. For such a separation of the nuclei ac-

ording to mass and excitation energy, we cannot use the existing (nuclear-chemistry) methods of separation, owing to their considerable inertia (in rare cases the time for the separation of isomers is 10^3 sec, but usually it is considerably longer^[7,8]).

Let us consider the following γ -laser scheme which meets the above-named requirements (Fig. 1). When the target 1 is bombarded by a stream of particles 2, a relatively small number of excited nuclei is produced. The excited nuclei are extracted from the target by, for example, rapidly vaporizing the target with a pulse of laser radiation 3. The next problem is how to rapidly extract the nuclei of a given composition in the excited state. For this purpose, the method of selective photoionization of atoms is, in our opinion, suitable. The idea consists in the following.

In the optical spectra of atoms there usually are non-overlapping lines of various isotopes, since the isotopic shift usually exceeds the Doppler width of the absorption (emission) lines^[9,10]. This allows the excitation by radiation of atoms with nuclei of a given composition. Moreover, the excitation of the nucleus in an atom also leads to a small shift of the optical spectral lines, due to the weak electron-neutron interaction. The isomeric shift in optics was first detected by Melissinos and Davis^[11] in radioactive Hg^{197*} atoms for the isomers with the lifetimes of 65 and 24 hours. The magnitude of the shifts for the Hg I 2537-Å line turned out to be equal to 0.3–0.6 cm⁻¹, i.e., appreciably larger than the Doppler line width.

The existence of the isomeric shift in atomic spectra allows us to selectively excite by laser radiation only the atoms with an excited nucleus of a given composition. For this purpose the wavelength λ_1 of the laser 4 should coincide with the optical absorption line λ_0^* belonging to the atom with an excited nucleus M* of the requisite composition. The power of the radiation should be such as to promote a significant fraction of the atoms A(M*) to the excited electronic state. The subsequent separation of the excited atoms $\tilde{A}(M^*)$ is accomplished by photoionizing them with an auxiliary laser pulse. For this purpose the wavelength λ_2 of the radiation of the second laser 5 is chosen such that the radiation brings about the photoionization of only the atoms $\tilde{A}(M^*)$ excited by the laser radiation. After the simultaneous irradiation of the products of the vaporization of the target by laser radiation at two wavelengths λ_1 and λ_2 , ions J(M*) of

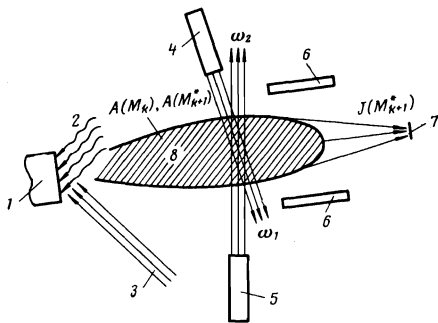


FIG. 1. A possible scheme for a γ -laser based on nuclear transitions: 1) target, 2) stream of bombarding particles (slow neutrons), 3) laser beam that vaporizes the irradiated target layer, 4) laser that excites the atoms $A(M_{k+1}^*)$ with excited nuclei M_{k+1}^* , 5) laser that photoionizes the excited atoms $A(M_{k+1}^*)$, 6) system for gathering the ions $J(M_{k+1}^*)$, 7) sample with the excited nuclei M_{k+1}^* , and 8) gaseous mixture of the atoms $A(M_k)$ and $A(M_{k+1}^*)$.

only a given isomer M* are produced which can be gathered by the electromagnetic focusing system 6 and deposited on a charged filament or film 7.

There are two most important problems connected with the above-described scheme for the γ -laser: 1) the preparation of a target containing $N_m \approx 10^{15}$ excited nuclei (for $\hbar\omega_\gamma = 30$ keV) across its thickness, which allows the vaporization of the target in a short time (of the order of 0.1 sec);

2) a laser separation of the excited nuclei that permits deposition of the ions with the excited nuclei on a microscopic sample.

Let us consider how we can solve the first problem and make estimates concerning the laser separation of the nuclei.

4. EXCITATION OF NUCLEI IN THE TARGET

For the excitation of the nuclear levels, several effective methods are available: Coulomb excitation, neutron capture, γ -irradiation, nuclear fission. In the scheme for the γ -laser being considered only those methods are suitable which allow us to produce in a time shorter than τ ($\tau = \Gamma_0^{-1}$ is the lifetime of the excited nuclei) N_m ($\sim 10^{15}$ for $\lambda = 0.4$ Å) excited nuclei in a thin target. For a target with N_m excited nuclei to be rapidly vaporizable in a time much shorter than τ , its thickness d should not exceed, say, 1 mm. For the scheme under consideration the most suitable nuclear levels are those with energies of 20–60 keV and lifetimes τ of from 0.1 to 10 sec. Scores of nuclei have excited levels with such parameters^[12].

If the depth l_0 of penetration of the bombarding particles is less than the characteristic thickness d , then the bombarding-particle flux density P at the surface of the target should satisfy the condition

$$\eta P S_0 \tau \geq N_m, \quad (9)$$

where S_0 is the area of the target, η is the quantum yield per particle of nuclei excited to the level in question. This means in practice that P should exceed the value 10^{14} particles/sec \times cm² for a penetration depth l_0 smaller than 0.1 cm. Of all the indicated methods of excitation only slow-neutron capture can really guarantee the excitation of nuclei in a thin layer. However, the required thermal-neutron flux density can apparently be attained only in pulse reactors.

If the depth l_0 of penetration of the bombarding particles is much larger than d , then the number of particles in the thin layer drops accordingly, and the required particle flux density increases:

$$P \geq \frac{N_m}{\eta S_0 \tau} \frac{l_0}{d}. \quad (10)$$

Such a situation is realized in the excitation of nuclei by charged particles and γ -quanta. Even in the region of the giant nuclear resonance the cross section for absorption of γ -quanta is much smaller than a barn, and their penetration depth into the sample $l_0 \gg 1$ cm. Therefore these methods are ineffective in the γ -laser scheme under consideration. An exception may, in principle, be the case of excitation by γ -radiation in the resonance absorption line, when the cross section can appreciably exceed a barn, although the narrowness of the absorption line at the transition directly to the isomeric state does not allow an effective direct excitation to the operating nuclear level to be carried out. The only pumping by

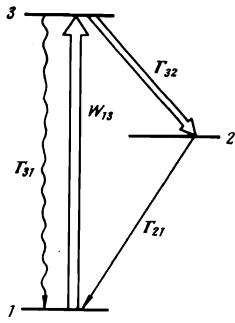


FIG. 2. Three-level scheme of excitation of the isomeric nuclear state 2 by γ -radiation at the transition $1 \rightarrow 3$.

γ -radiation worth discussing is the one employing the three-level scheme shown in Fig. 2. Let us consider it in greater detail.

The rate of radiative excitation to the nuclear level at the transition $1 \rightarrow 3$ with the transition probability Γ_{13} is given by the expression

$$W_{13} = \frac{\lambda_{13}^2 g_3}{2\pi g_1} \Gamma_{13} \frac{P_\nu}{2\pi}, \quad (11)$$

where P_ν is the spectral density of the pumping γ -radiation (in photons/cm² × sec² × Hz), $g_i = 2I_i + 1$ is the degeneracy of the i -th level, and I_i is the spin of the i -th level. Here it is assumed that the spectral width of the pump is much larger than the total width $\Delta\omega_{13}$ of the transition, including the broadening due to the decay of the excited state with probability $\Gamma_3 = (\Gamma_{13} + \Gamma_{32})(1 + \alpha_3)$ and other types of broadening (Γ_{32} is the probability of decay of the level 3 to the lower-lying isomeric state 2 and α_3 is the internal-conversion coefficient for the level 3).

The rate W_{13} is the sum of the rate of excitation in the narrow recoilless-absorption line and the rate of excitation in the entire absorption band $\Delta\omega_{13}$, the fractional contributions of these addends being respectively equal to f and $1 - f$, where f is the Debye-Waller factor. The rate of excitation does not depend on the presence of a narrow recoilless-absorption line. However, the absorption coefficients per unit length for the pumping radiation in the narrow line $\kappa_{13}^{(f)}$ and in the entire band $\kappa_{13}^{(1-f)}$ are significantly different:

$$\kappa_{13}^{(f)} = \frac{\lambda_{13}^2 f g_3 \Gamma_{13}}{2\pi g_1 \Gamma_3 n_1}, \quad \kappa_{13}^{(1-f)} = \frac{\Gamma_3}{\Delta\omega_{13}} \kappa_{13}^{(f)} \frac{1-f}{f} \quad (12)$$

(n_1 is the density of target nuclei in the ground state).

Let the excited state 3 decay to the lower-lying long-lived state 2, while the duration τ_p of the pumping pulse satisfies the condition

$$\Gamma_2^{-1} = \tau_2 \gg \tau_p \gg \tau_3 = \Gamma_3^{-1}. \quad (13)$$

Then a nuclear density n_2 is established in the metastable state 2:

$$\frac{n_2}{n_1} = \frac{\lambda_{13}^2 g_2}{2\pi g_1} \frac{\Gamma_{13}\Gamma_{32}}{(\Gamma_{13} + \Gamma_{32})(1 + \alpha_3)} \tau_p \frac{P_\nu}{2\pi}. \quad (14)$$

The case $\Gamma_{13} \lesssim \Gamma_{32}$ and $\alpha_3 \ll 1$ is the optimum case. In this case the rate of excitation of nuclei to the state 2 will be determined by the line width of the pumping transition $1 \rightarrow 3$, while the buildup time is determined by the pulse duration τ_p , i.e.,

$$\frac{n_2}{n_1} = \frac{\lambda_{13}^2 g_2}{2\pi g_1} \Gamma_{13} \tau_p \frac{P_\nu}{2\pi}. \quad (15)$$

In this lies the important advantage of the three-level scheme of excitation of the isomeric states by γ -radiation. It should be noted that the pumping of the isomeric states of a nucleus via a higher-lying ("activating"),

rapidly-decaying level with an energy ~ 1 MeV was realized long ago by Pontecorvo and Lazard^[13] and Korsunskii, Lange and Shpinel'^[14].

The depth of penetration of the broad-band γ -radiation is determined by the absorption coefficient $\kappa_{13}^{(1-f)}$, i.e.,

$$l_0 = \frac{1}{\kappa_{13}^{(1-f)}} = \frac{2\pi g_1 \Delta\omega_{13}}{\lambda_{13}^2 g_3 \Gamma_{13} n_1}. \quad (16)$$

For example, for $\lambda_{13} = 0.4 \text{ \AA}$, $g_1 \approx g_3$, $n_1 = 3 \times 10^{22} \text{ cm}^{-3}$, $\hbar\Delta\omega_{13} = 0.1 \text{ eV}$ (this corresponds, in order of magnitude, to the broadening of the absorption line of the nuclei in the crystal due to the thermal oscillations of the nuclei at $T = 300^\circ \text{ K}$), and $\Gamma_{13} = 10^{10} \text{ sec}^{-1}$ (the typical radiation width of a nuclear transition in the 100-keV energy region), the target will be excited to a depth $l_0 \approx 0.4 \text{ cm}$. We can decrease $\Delta\omega_{13}$ by decreasing the temperature of the target, but $l_0 \gg d = 0.1 \text{ cm}$ in every case.

The γ -radiation spectral density necessary for the production of N_m excited nuclei in a layer of thickness d is determined by the condition $n_2 d S_0 > N_m$ i.e., by

$$\tau_p P_\nu > \frac{N_m}{S_0 d n_1} \left(\frac{2\pi}{\lambda_{13}} \right)^2 \frac{g_1}{g_3} \frac{1}{\Gamma_{13}}. \quad (17)$$

For the example ($\lambda_{13} = 0.4 \text{ \AA}$, $N_m \approx 10^{15}$, $d = 0.1 \text{ cm}$, $n_1 = 3 \times 10^{22} \text{ cm}^{-3}$, $S_0 = 1 \text{ cm}^2$, and $\Gamma_{13} \approx 10^{10} \text{ sec}^{-1}$) under consideration in this paper, this condition reduces to $P_\nu \tau_p > 10^2 \text{ quanta/Hz} \times \text{cm}^2$ in a time shorter than $\tau = \Gamma_2^{-1}$. For nonmonochromatic-radiation sources this condition is in practice not fulfilled. If we use for pumping the nuclei the comparatively narrow lines of the x-ray emission of a high-temperature plasma heated by a high-power laser pulse^[15], then the condition can in principle be fulfilled. It should be noted that the x-ray yield from targets of large Z heated by a laser pulse already reaches 15–20%, as a percentage of the energy of the laser pulse^[16]. It is possible that the neutron emission of a high-temperature laser plasma heated by laser radiation^[17] will also attain intensities sufficient for the excitation of the target nuclei in the neutron-capture process.

5. LASER EXTRACTION OF EXCITED NUCLEI FROM A CONDENSED TARGET

Let us consider the extraction of nuclei of a given composition in an excited state from the target. We shall consider the most realistic method of excitation of the nuclei in a target—excitation upon the capture of slow neutrons. In this case from the primary nuclei M_k of the target is produced a relatively small number N_m of excited nuclei M_{k+1}^* of different isotopic composition in the target. The excited nuclei are produced in a layer $l_0 = (\sigma_{n\gamma} n_M)^{-1}$, where $\sigma_{n\gamma}$ is the cross section for the capture of a slow neutron and n_M is the density of the nuclei M_k in the target. In practice, $\sigma_{n\gamma}$ attains values greater than $10^4 \text{ b}^{[4]}$ and, in the case of a condensed target, the penetration depth $l_0 \lesssim 3 \times 10^{-3} \text{ cm}$ for $n_M = 3 \times 10^{22} \text{ cm}^{-3}$.

When an irradiated layer of thickness l_0 is vaporized, a gas stream containing $N_0 = n_M l_0 S_0 = S_0 / \sigma_{n\gamma}$ particles (S_0 is the irradiated area of the target) is produced which should contain not less than N_m ($N_m \sim 10^{15}$ for $\hbar\omega_\gamma \approx 30 \text{ keV}$) excited nuclei M_{k+1}^* of the heavier isotope that arises upon the capture of a neutron. We obtain in accordance with (9) an estimate for the minimum value of the integrated neutron flux at the surface of the target:

$$I > I_{\min} = (P\tau)_{\min} = N_m / \eta S_0. \quad (18)$$

This value is realized only when there are no restrictions when separating the nuclei and when it is comparatively low. For example, for $N_m \approx 10^{15}$, $\eta = 1$, and $S_0 = 10^2 \text{ cm}^2$, the quantity $I_{\min} \approx 10^{13}$ neutrons/cm², which is easily attainable in the times $\tau \gtrsim 0.1$ sec under consideration.

Let us appraise the possibility of extracting 10^{15} atoms with excited nuclei from a gas stream containing $N_0 \approx 10^{20} - 10^{22}$ atoms (for $S_0 = 1 \text{ cm}^2$ and 100 cm^2 respectively) in a time of the order of 10^{-1} sec.

The existence of the isotopic and isomeric shifts in the optical spectra of atoms allows the selective excitation of only the atoms with excited nuclei, the subsequent photoionization of the excited atoms, and the collection of the ions produced. We note that the two-stage selective photoionization was first accomplished in Rb atoms^[18]. Certain general aspects of it were considered in the paper by Ambartsumyan and the present author^[19]. In the present case the possibility of extracting a very small fraction of the atoms $A(M_{k+1}^*)$ with excited nuclei from a gas of the atoms $A(M_k)$ is important.

The process of excitation and photoionization of the excited atoms $A(M_{k+1}^*)$ is described by the equations

$$\begin{aligned} \dot{m}_0 &= -\sigma_e P_1 (m_0 - m_1) + \tau_{10} m_1, \\ \dot{m}_1 &= +\sigma_e P_1 (m_0 - m_1) - \tau_{10} m_1 - \sigma_{ph} P_2 m_1, \\ \dot{m}_i &= \sigma_{ph} P_2 m_1, \end{aligned} \quad (19)$$

where m_0 and m_1 are the densities of the atoms A in the ground ($A(M_{k+1}^*)$) and excited ($\tilde{A}(M_{k+1}^*)$) states; m_i is the density of the ions $J(M_{k+1}^*)$ with the nuclei M_{k+1}^* ; σ_e and σ_{ph} are the cross sections for the excitation and photoionization of the excited atoms by radiation fluxes of respective intensities P_1 and P_2 and frequencies ω_1 and ω_2 ; τ_{10} is the relaxation time of the excited atoms.

If the probabilities of excitation $W_e = \sigma_e P_1$ and photoionization $W_{ph} = \sigma_{ph} P_2$ are much larger than the relaxation probability $1/\tau_{10}$ for the excited atoms, or if the durations of the radiation pulses P_1 and P_2 are much shorter than τ_{10} , then the relaxation in (19) can be neglected, and, then, the number of ions under the action of the synchronous light pulses of intensities P_1 and P_2 , switched on at the moment of time $t = 0$, increases according to the law

$$\frac{m_i}{m_0} = 1 - \frac{a_1 + a_2}{2a_2} e^{-(a_1 - a_2)t} + \frac{a_1 - a_2}{2a_2} e^{-(a_1 + a_2)t}, \quad (20)$$

where m_0 is the initial density of atoms in the ground state,

$$a_1 = 1 + \frac{W_{ph}}{2W_e}, \quad a_2 = \left[1 + \frac{1}{4} \left(\frac{W_{ph}}{W_e} \right)^2 \right]^{1/2}.$$

The exponents in the expression (20) strongly differ from each other. The maximum of their ratio $(a_1 - a_2)/(a_1 + a_2)$ is attained at $W_{ph}/W_e = 2$ and is equal to ≈ 0.18 . The coefficient standing in front of the first exponential function weakly depends on W_{ph}/W_e . The maximum of the quantity $(a_1 + a_2)/2a_2$ is attained at $W_{ph}/W_e = 2$ and is equal to ≈ 1.2 . Thus, the quantity

$$T = \left\{ 1 + \frac{W_{ph}}{2W_e} \left[1 + \left(\frac{W_{ph}}{2W_e} \right)^2 \right]^{1/2} \right\}^{-1} \frac{1}{W_e} \quad (21)$$

is the characteristic time for the two-stage photoionization of the atoms $A(M_{k+1}^*)$, and the optimum duration of

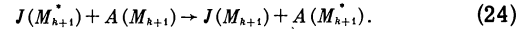
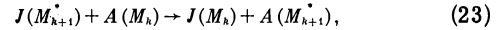
the laser pulses should be of the order of T , provided $T \lesssim \tau_{10}$.

The cross section for the resonance excitation of an atom exceeds by many orders of magnitude the photoionization cross section (typically, $\sigma_e = 10^{-12} - 10^{-10} \text{ cm}^2$, while $\sigma_{ph} = 10^{-19} - 10^{-18} \text{ cm}^2$). Therefore, to obtain $W_{ph} \approx W_e$, the power P_2 of the photoionizing radiation should exceed the power P_1 of the exciting radiation by a factor of $\sigma_e/\sigma_{ph} = 10^7 - 10^9$. The power P_1 is determined from the condition $W_e \tau_{10} > 1$, or

$$P_1 \geq \hbar \omega_1 / \sigma_e \tau_{10}. \quad (22)$$

For radiation in the region of the resonance lines of atoms (3000–4000 Å) and for a radiative mechanism of relaxation ($\tau_{10} \approx 10^{-8}$ sec), this corresponds to $P_1 \gtrsim 20 \text{ W/cm}^2$. Thus, for the totally attainable laser-pulse parameters, we can selectively photoionize only the atoms $A(M_{k+1}^*)$. Notice that the resonant transfer of excitation from the excited atoms $\tilde{A}(M_{k+1}^*)$ to the unexcited atoms $A(M_k)$ and $A(M_{k+1})$ does not jeopardize the preservation of the selectivity of the photoionization of the atoms $A(M_{k+1}^*)$, since the rate of photoionization of the excited atoms can always be chosen to be higher than the excitation transfer rate.

It is much more difficult to preserve the selectivity during the collection of the ions $J(M_{k+1}^*)$, owing to the resonant charge-transfer process, whose cross section for slow atoms usually exceeds considerably the gas-kinetic cross section^[20]. Two resonant charge-transfer processes are possible during the collection of the ions by an electric field:



The process (23) leads to the loss of the excited nucleus M_{k+1}^* decreases the effectiveness of the collection of the excited nuclei in the sample. The process (24) leads to a decrease in the inversion of the nuclear levels in the sample. Since the number of the nuclei M_{k+1} is smaller than the number of the primary nuclei M_k of the target by roughly a factor of 10^5 , the process (23) is the dominant process. In principle, the loss of the excited nuclei M_{k+1}^* can be made up for by repeating many times the two-stage photoionization process, but it is necessary to reduce in every case the gas-stream density in the ionization and ion-collection region to the value n_{\max} defined by the condition

$$n_{\max} = (\sigma_{ct} D)^{-1}, \quad (25)$$

where σ_{ct} is the cross section for the resonant charge transfer, D is the mean free path of an ion in the gas during the collection and is roughly equal to the transverse dimension of the gas stream, i.e., $D \approx \sqrt{S}$, where S is the cross section of the gas stream. For the cross section $\sigma_{ct} \approx 10^{-14} \text{ cm}^2$ ^[20] and $D = 10 \text{ cm}$, this corresponds to a maximum gas density $n_{\max} \approx 10^{13} \text{ cm}^{-3}$, i.e., to a pressure of only 0.3 mTorr.

The limitation on the gas density due to the resonant charge transfer leads to a restriction on the maximum number N_0^{\max} of atoms which can be separated by the method under consideration:

$$N_0^{\max} \approx n_{\max} S v \tau_{10}, \quad (26)$$

where v is the velocity of the gas stream and τ_{tr} is the time it takes to transport the atoms through the cross section S . The total time of transfer of an excited nucleus from the target to the sample, which is made up of the time for the vaporization, the transport time τ_{tr} , and the time for the collection and deposition of the ions on the sample, should be at least several times shorter than the decay time τ of the excited state. Here it is not assumed that the lower level decays more rapidly than the upper level, and, therefore, inversion exists only during a time of the order of τ . The slowest in the present case is the gas-transport process. Therefore, for estimates, the quantity τ_{tr} in the relation (26) can be replaced by $\sim \tau/5$. We then have

$$N_0^{\max} \approx v\tau/5\sigma_{ce}S^{1/2}. \quad (27)$$

Since N_0^{\max} should contain not less than N_m excited nuclei, the percentage of the isomer in the irradiated and vaporized target layer should not be less than N_m/N_0^{\max} . This imposes a subsidiary condition on the magnitude of the integrated neutron flux:

$$I = P\tau > \frac{N_m}{N_0^{\max}\eta\sigma_{ny}} = \frac{5\sigma_{ce}N_m}{\eta\sigma_{ny}S^{1/2}v}. \quad (28)$$

For decay times $\tau < \tau_1$, where τ_1 is determined by the expression

$$\tau_1 = 5 \frac{\sigma_{ce}}{\sigma_{ny}} \frac{S_0}{S^{1/2}v}, \quad (29)$$

the condition (28), which is specifically for the separation of the nuclei by the selective-photoionization method, is more rigid than the condition (18). Notice that in this region ($\Gamma_m^{-1} < \tau < \tau_1$), because of the dependence $N_m \tau^2$ (the expression (5)), the requisite neutron flux density does not depend on τ , and is equal to

$$P > 2.5 \cdot 10^4 \frac{\sigma_{ce}}{\sigma_{ny}\eta S^{1/2}v} \frac{1+\rho}{n_0\lambda^3} \left(\frac{\Gamma_m}{\rho f}\right)^2. \quad (30)$$

In conclusion, let us give the final estimates. For a target area $S_0 = 1 \text{ cm}^2$, a gas-stream cross sectional area $S = 10^2 \text{ cm}^2$; gas-stream velocity $v = 5 \times 10^4 \text{ cm/sec}$, slow-neutron capture cross section $\sigma_{ny} = 10^{-20} \text{ cm}^2$, and resonant charge-transfer cross section $\sigma_{ct} = 10^{-14} \text{ cm}^2$, the requisite neutron-flux intensity is determined by the relation (18) for $\tau > \tau_1 = 10 \text{ sec}$ and by the relation (28) or (30) for $\tau < 10 \text{ sec}$. In the latter case, for a γ -quantum energy $\hbar\omega_\gamma = 30 \text{ keV}$ ($\lambda = 0.4 \text{ \AA}$), the Mössbauer-line width $\Gamma_m = 10^5 \text{ sec}^{-1}$, $\rho \approx 1$, $f \approx 1$, and $n_0 = 3 \times 10^{22} \text{ cm}^{-3}$, the requisite neutron-flux density is equal to $P > 5 \times 10^{17} \text{ neutrons/cm}^2 \times \text{sec}$ in a time of the order of a fraction of τ .

Thus, the proposed γ -laser scheme with laser separation of the excited nuclei through selective photoionization is quite realizable for real physical parameters.

6. LASER EXTRACTION OF THE EXCITED NUCLEI FROM A GASEOUS TARGET

The limitation on the gas-stream density during the extraction of the excited nuclei by selective photoionization leads to the growth of the required neutron-flux intensity (the region $\tau < \tau_1$). Therefore, it seems advisable to look for other methods of separation in which there will be no such limitation. In particular, the following method of separation of the excited nuclei is possible when a molecular gaseous target is irradiated by a thermal-neutron stream. Upon the capture of a neutron and the subsequent emission of a cascade of γ -quanta by the compound nucleus M_{k+1}^* , because of the recoil

effect, the atom with the nucleus M_{k+1}^* breaks away from the molecule. This recoil-induced dissociation phenomenon is called the Szilard-Chalmers effect^[7,8]. As a result, the nuclei M_{k+1}^* turn out to be bound in atoms, while the remaining nuclei M_k remain bound in the molecules. The subsequent extraction of the nuclei M_{k+1}^* is carried out according to the scheme considered in Sec. 5, i.e., by a two-stage selective photoionization of the atoms $A(M_{k+1}^*)$ in the molecular gas stream issuing from the irradiated gaseous target. The basic difference lies in the fact that the charge-transfer process (23) is absent because of the absence of the atoms $A(M_k)$. The process of recombination of the atoms $A(M_{k+1}^*)$ with the radicals produced during the dissociation of the molecules is possible in the process of the separation, but the recombination cross section is considerably smaller than the resonant charge-transfer cross section.

The effective use of the neutron flux is attained for diverse dimensions d and densities n_M of the gaseous target (for $\sigma_{ny} = 10^{-20} \text{ cm}^2$ and $n_M = 3 \times 10^{19} \text{ cm}^{-3}$, the quantity $d \approx 3 \text{ cm}$). The quantum yield of the Szilard-Chalmers effect can attain several scores per cent^[7,8]. Therefore this scheme will apparently allow operation at neutron fluxes which satisfy the condition (18) and which are attainable under laboratory conditions.

7. CONCLUSION

The analysis carried out here shows that it is in principle possible to construct a γ -laser based on nuclear levels in the $\hbar\omega_\gamma \sim 20\text{--}60 \text{ keV}$ region, using the Mössbauer effect in isomeric transitions with lifetimes $\tau \approx 0.1\text{--}10 \text{ sec}$. We emphasize that in the estimates made above we used only the Mössbauer-transition parameters that have already been attained (in particular, the transition width $\Gamma_m \approx 10^5 \text{ sec}^{-1}$). Therefore, the possibility of constructing a γ -laser according to the scheme considered is, from the theoretical point of view, totally realistic. But for the practical development of such a laser, we must work out methods for solving the following important problems: 1) the production of a considerable number ($\sim 10^{15}$) of isomeric nuclei in a condensed or gaseous target in a time shorter than τ , 2) the rapid extraction of a small fraction of excited nuclei from among nuclei differing in isotopic composition and in excitation energy, and 3) the narrowing down of the Mössbauer-line widths to values smaller than $\Gamma_m = 10^5 \text{ sec}^{-1}$ by one-two orders of magnitude, which will allow a reduction of the neutron flux by two-four orders of magnitude. The solution of these problems, which stand in the way of the construction of the γ -laser, will lead to important developments in such directions as the production of high-intensity Mössbauer-radiation sources (with intensities of $\sim 10^3\text{--}10^4 \text{ Ci}$) and the rapid laser separation of isomeric and isotopic nuclei with a huge separation factor in one cycle. The current state of nuclear and laser technologies does, in our opinion, allow us to enter upon the practical solution of these complex problems.

The author recently became acquainted with the work^[6] in which the authors consider the possibility of producing directly the laser effect in a Mössbauer transition from a short-lived isomeric nuclear state ($\tau < 10^{-5} \text{ sec}$) during the pumping of a crystal by a pulsed thermal-neutron stream of extremely high intensity. Thus, the scheme proposed in the present paper is,

in a sense, a compromise scheme between the schemes considered in^[1-3,5] (independent preparation of the concentrate of isomeric nuclei of lifetime $\tau > 10^4 - 10^5$ sec) and in^[6] (the realization of the inversion directly in the sample itself during a pulsed neutron pumping at transitions with $\tau < 10^{-5}$ sec).

¹⁾This fact was pointed out to me by V. I. Gol'danskiĭ.

- ¹V. Vali and W. Vali, Proc. IEEE **51**, 182, 1248 (1963).
- ²G. C. Baldwin, J. P. Neissel, and L. Tonks, Proc. IEEE **51**, 1247 (1963).
- ³B. V. Chirikov, Zh. Eksp. Teor. Fiz. **44**, 2016 (1963) [Sov. Phys.-JETP **17**, 1355 (1963)].
- ⁴K. Siegbahn, Alpha-, Beta-, and Gamma-ray Spectroscopy, North-Holland Publ. Co., Amsterdam, 1965 (Russ. Transl., Vol. 1 and 2, Atomizdat, 1969).
- ⁵R. V. Khokhlov, ZhETF Pis. Red. **15**, 580 (1972) [JETP Lett. **15**, 414 (1972)].
- ⁶V. I. Gol'danskiĭ and Yu. Kagan, Preprint Instituta khimicheskoi fiziki (Preprint Institute of Chemical Physics); Zh. Eksp. Teor. Fiz. **64**, 90 (1973).
- ⁷M. Haissinsky, Nuclear Chemistry and its Applications, Addison-Wesley, 1962.
- ⁸G. Friedländer, J. Kennedy, and J. Miller, Nuclear and Radiochemistry, John Wiley, New York, 1964 (Russ. Transl. Mir, 1967).
- ⁹S. É. Frish, Opticheskie spektry atomov (Optical Spectra of Atoms), Fizmatgiz, 1963.
- ¹⁰I. I. Sobel'man, Vvedenie v teoriyu atomnykh spektrov (Introduction to the Theory of Atomic Spectra), Fizmatgiz, 1963 (Eng. Transl. Pergamon, New York, 1972).
- ¹¹A. C. Melissinos and S. P. Davis, Phys. Rev. **115**, 130 (1959).
- ¹²É. E. Berlovich, S. S. Vasilenko, and Yu. N. Novikov, Vremena zhizni vzbuzhdennykh sostoyaniĭ yader (The Lifetimes of Excited Nuclear States), Nauka, 1972.
- ¹³B. M. Pontecorvo and A. Lazard, C. R. Acad. Sci. **208**, 99 (1939).
- ¹⁴M. I. Korsunskiĭ, F. F. Lange, and V. S. Shpinel', Dokl. Akad. Nauk SSSR **26**, 145 (1940).
- ¹⁵V. S. Letokhov, in: Kvantovaya élektronika (Sov. J. Quantum Electronics), No. 5, 1973.
- ¹⁶P. J. Malozzi, H. M. Epstein, R. G. Jung, D. C. Applebaum, B. P. Fairand, and W. J. Gallagher, Report of Battelle Columbus Laboratories, USA, 1972.
- ¹⁷N. G. Basov, S. D. Zakharov, P. G. Kryukov, Yu. V. Senatskiĭ, and S. V. Chekalin, ZhETF Pis. Red. **8**, 26 (1968) [JETP Lett. **8**, 14 (1968)].
- ¹⁸R. V. Ambartsumyan, V. P. Kalinin, and V. S. Letokhov, ZhETF Pis. Red. **13**, 305 (1971) [JETP Lett. **13**, 217 (1971)].
- ¹⁹R. V. Ambartsumyan and V. S. Letokhov, IEEE J. Quant. Electr., QE-7, 305 (1971); Appl. Optics **11**, 354 (1972).
- ²⁰N. Mott and H. Massey, The Theory of Atomic Collisions, Oxford University Press, Oxford, 1965 (Russ. Transl., Mir, 1969).

Translated by A. K. Agyei

170