Possibility of nonthreshold γ amplification in a system of polarized nuclei

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(Submitted 28 December 1978)
Zh. Eksp. Teor. Fiz. 77, 492–497 (August 1979)

The influence of polarization of a nuclear system on the satisfaction of the threshold condition for γ amplification on Mössbauer nuclear transitions is considered. It is shown that at sufficient degree of polarization of the ground and excited states it is possible to realize a nonthreshold amplification, i.e., at pump parameters smaller by several orders of magnitude than the value that follows from the ordinary threshold condition. Such an effect, but with somewhat modified parameters, can take place also if only the unexcited nuclei are polarized. The possibility of developing a γ laser on polarized nuclei of the isomer Dy^{161} when the system is excited by radiation from a real x-ray installation is considered on the basis of the observed effect. It follows from estimates that by using the characteristic radiation of such an installation it is possible at present to develop a γ laser with polarized nuclei on short-lived ($T_e \approx 10^{-6}$–10^{-8} sec) Mössbauer γ nuclei.

PACS numbers: 42.55.Bi

It is customarily assumed that to realize γ amplification it is absolutely necessary to satisfy a threshold condition that includes the requirement (which is necessary but not sufficient) that the concentration of the excited nuclei exceed that of the unexcited ones. In a system of long-lived isomers this condition can be satisfied, but the need for narrowing down the line by 5–8 orders of magnitude makes the model unrealistic at the present time. For short-lived systems ($T_e \leq 10^{-6}$ sec), the last requirement is unnecessary, but the principal task, inversion (with a threshold concentration exceeding 50%) within a short time $\tau < T_e$, is exceedingly complicated.[1,2] We consider below a realistic model that makes use of the advantages of short-lived nuclei (non-broadened line), but calls for its realization a pump with parameters weaker by many orders of magnitude than the value that follows from the ordinary threshold condition. This allows us to speak of non-threshold amplification, the meaning of which in this particular case is that the condition for the amplification (threshold) of the γ transition are not satisfied as a whole, since amplification on one of the frequencies of the nuclear multiplet of the polarized nuclei is possible in the case when the total concentration of the excited nuclei is smaller by several orders of magnitude than the concentration of the unexcited nuclei, and the medium as a whole is not inverted. This polarization mechanism was never before considered in the γ-laser problem (Ref. 3 deals with the long-known problem of the anisotropy of emission of polarized nuclei). The gist of the effect is made clear by a simple example.

Consider the possible γ transitions in polarized nuclei of two types with pairwise identical spins of the ground state ($J_g = 1/2$) and of the excited state ($J_e = 3/2$), but with different directions of the projections of the magnetic moments μ: case a, when $\mu_g > 0, \mu_e > 0$, as in Au^{197} (only the sublevels $m_g = -1/2, m_e = -3/2$ are populated), and case b, when $\mu_g < 0, \mu_e > 0$, as in Fe^{57} ($m_g = 1/2, m_e = -3/2$ are populated). In case a we can have transitions upward (absorption): $m_g = -1/2 \to m_e = -3/2, -1/2, 1/2$ and downward (amplification): $-3/2 \to -1/2$. It is seen that the frequency ν_{amp} of the amplifying transition coincides with ν_{abs}. The amplification conditions in this case have the same threshold as before, i.e., they cannot be satisfied. The situation is different for the nuclei of type b. They can absorb a quantum on transitions $1/2 \to -1/2, 1/2, 3/2$, and emit (i.e., amplify) on $-3/2 \to -1/2$, so that $\nu_{amp} \neq \nu_{abs}$. The population of the excited sublevel $m_e = -3/2$ can be many times smaller than that of the unexcited $m_g = 1/2$. Nonetheless, induced γ emission (amplification) is possible on the $-3/2 \to -1/2$ transition, but resonant absorption is impossible–the lower state $m_g = -1/2$ is populated, i.e., such a system is indeed capable on nonthreshold (with respect to total inversion) γ amplification. The same situation is realized on a transition of any multipolarity L between sublevels m_e and m_g such that $|m_e - m_g| \leq L$ but $|m_e - m_{g0}| > L$, where m_{g0} is the magnetic quantum number of the lower sublevel. We now examine the extent to which such systems are realistic.

It is clear, above all, that nonthreshold γ amplification is realized even if only nuclei in the ground state are polarized. In fact, all the sublevels m_e are equally populated, but nonthreshold amplification takes place only on transitions from m_e levels for which $|m_e - m_{g0}| > L$, and the coefficient of this amplification turns out to be smaller by a factor $2J_2 + 1$. This possibility is of fundamental importance for the development of systems for γ amplification on short-lived transitions, for which the time of relaxation to the equilibrium state at the given temperature T is $t_r \sim T_e$, with usually $t_r \sim 1/T$. Then at $T \sim 0.1$–0.01 K the time is $t_r \sim 1$–10^3 sec and polarization of the unexcited nuclei by a direct method is impossible. Some decrease of t_r, by introducing paramagnetic impurities,[4] is insufficient, and the presence of these impurities decreases strongly the γ-amplification cross section. On the other hand, the ground state (stable or long-lived with $T_g \gg t_r$) can be "prepared" beforehand and manages to relax to the equilibrium state of the polarization. Taking this into account, we find that nonthreshold γ amplification is pos-

 0038-5646/79/080250-03$02.40

sible on short-lived transitions (and on nuclei of either type, a or b), and the γ-amplification coefficient can remain the same in the case of selective population of the sublevels with definite m_e via the higher-lying levels.

How realistic are such systems? It is known that very strong magnetic fields of exchange origin exist at nuclei of many elements.[5] These fields reach values from $H\approx 6\times 10^5$ Oe for Fe to $H\approx 7\times 10^6$ Oe for Tm and Dy. Then, for example, the magnetic splitting between sublevels of the ground state of the isomer Dy^{161}, with spin $J_g=5/2$ and $\mu_g=-0.37\mu_N$, takes on a value $\mu_g H \approx 3.2\times 10^{-6}$ eV. When such a system is cooled to $T\approx 10^{-2}$ K, the populations of the sublevels $m_{g0}=-5/2$ and $m_g=1/2$ (it will be shown below that nonthreshold amplification is possible when the latter takes part) have a ratio $1:8\times 10^{-6}$. In the absence of nonresonant absorption, even at equal population of the sublevels m_e, the threshold concentration of the excited nuclei is 5×10^{-5} of that of the unexcited ones. In the case of dynamic polarization[4] it is possible to use weaker fields H and higher temperatures T.

Allowance for nonresonant absorption makes the problem more complicated. The amplification condition $\sigma_{amp}(n_3-n_2)\gtrsim \sigma_{n}n$ is the presence of the polarization p on the transition between levels 1 and 2 (i.e., between m_{g0} and m_g), without allowance for the spontaneous decay at level 3, takes the form

$$n_3 \geqslant n[\delta+(1-p)/(1+p)],\ \delta=\sigma_{nr}/\sigma_{amp},$$

where n is the total concentration, σ_{amp} and σ_{nr} are the cross sections of the resonant amplification and of the nonresonant absorption, $n_{2,3}$ are the concentrations (populations) of the lower and upper levels of the resonant transition m_g and m_e, respectively. Recognizing that δ is determined by the properties of the system, optimization of the problem calls for $(1-p)/(1+p)<\delta$, which is always attainable. The concrete value of δ varies in a wide range. Thus, for the γ transition in Al^{28} with $E\approx 32$ keV, $T_e=3\times 10^{-9}$ sec, $\sigma_{nr}\approx 5\times 10^{-24}$ cm^2 and $\sigma_{amp}\approx 6\times 10^{-19}$ cm^2 (at $T\ll\Theta$, where $\Theta=390$ K is the Debye temperature), we have $\delta\approx 8\times 10^{-6}$, while for Dy^{161} with $E=25.6$ keV, $T_e=2.8\times 10^{-8}$ sec, $\sigma_{nr}\approx 2\times 10^{-21}$ cm^2, and $\sigma_{amp}\approx 3.7\times 10^{-18}$ cm^2 the parameter is $\delta\approx 6\times 10^{-4}$. With increasing T_e, the parameter δ increases as a rule. It follows from the estimates that when polarized nuclei are used, it suffices to invert 10^{-5}–10^{-3} of all the nuclei in order to satisfy the γ-amplification condition in the case of short-lived isomers, and 10^{-1}–10^{-3} in the case of long-lived isomers (for the latter, in addition, it is necessary to eliminate the nonradiative broadening[1,6,7]). Such a relaxation of the requirements on the necessary concentration of the excited nuclei makes it possible to use excitation sources with intensity lower by the same factor (if non-broadened γ transitions with $IT_e\approx 1$ and $T_e\sim 10^{-6}$–10^{-8} sec are used it is possible to use existing pulsed x-ray installations).

To estimate the necessary pump parameters we consider the equivalent four-level scheme. In the case of a relatively weak (i.e., the only realistic) pump, at which no substantial change of the population of the ground level 1 takes place during the pump time τ and the condition $t_r\gg T_e$ discussed above is satisfied, the change of the populations is described by the system of equations

$$\frac{dn_2}{dt}=n_3\Gamma_{32},\quad \frac{dn_3}{dt}=n_4w_{43}-n_3\Gamma_3,\quad \frac{dn_4}{dt}=n_1w_{14}-n_4w_{41}-n_4\Gamma_4,$$

under the initial conditions

$$n_2(0)=n_1(1-p)/(1+p),\ n_3(0)=n_4(0)=0.$$

Here, w_{nm} is the radiative-transition probability;

$$\Gamma_n=\sum_m \Gamma_{nm},$$

where Γ_{nm} is the partial probability of the $n\to m$ transition. From the solutions of the system we obtain expressions for the inverted population and for the condition of amplification on the transition $3\to 2$ (with allowance for the condition $\Gamma_4\ll\Gamma_3$):

$$\Delta=n_3-n_2=n\left[\frac{w_{14}\Gamma_{43}}{\Gamma_4\Gamma_3}\left(\frac{\Gamma_3+\Gamma_{32}}{\Gamma_3}(1-e^{-\Gamma_3 t})-\Gamma_3 t\right)-\frac{1-p}{1+p}\right],\quad \Delta\geqslant n\delta.$$

The maximum inversion is reached at

$$t=t_0=\Gamma_3^{-1}\ln[(\Gamma_3+\Gamma_{32})/\Gamma_{32}].$$

The radiative-excitation probability w_{14} of the activation level 4, by a source with a spectral width $\Delta v\gg\Gamma_4+2(RK\Theta/h)^{1/2}$ (R is the recoil energy) is given by

$$w_{14}=\int_0^{\nu_{max}}\sigma(\nu)P(\nu)d\nu\approx\lambda_{14}^2\Gamma_4 P(\nu_{14})/4,\quad \nu_{14}<\nu_{max},$$

where $P(v_{14})$ is the spectral density of the pump-photon flux at the frequency v_{14}.

We now estimate the parameters of the system, using as an example the Mossbauer isomer Dy^{161}. By analyzing the structure of the levels and of the allowed transitions for a polarized Dy^{161} nucleus[5] we easily verify that nonthreshold γ amplification can be realized only on the transition $3\to 2$ with participation of the following levels:

$$1)\ J_g={}^5/_2,\quad m_{g0}=-{}^5/_2,\qquad 2)\ J_g={}^5/_2,\quad m_g={}^1/_2,$$
$$3)\ J_e={}^5/_2,\quad m_e=-{}^1/_2,\qquad 4)\ J_a={}^7/_2,\quad m_a=-{}^3/_2$$

(the remaining levels and transitions do not influence the dynamics of the system in the weak-pump approximation, when $n_1\gg n_{2,3,4}$).

The partial values Γ_{nm} are determined using Clebsch-Gordan coefficients:

$$\Gamma_{43}=0.5\Gamma_4(C^{J_3m_3}_{J_4m_4L_{43}(m_3-m_4)})^2,\quad \Gamma_{32}=\Gamma_3(C^{J_2m_2}_{J_3m_3L_{32}(m_2-m_3)})^2.$$

The values for Dy^{161} are $\Gamma_{43}\approx 0.25\Gamma_4$, $\Gamma_{32}\approx 0.51\Gamma_3$, $t_0\approx 1.1T_3$. From the amplification condition, with account taken of the maximum inversion at $t=t_0\lesssim\tau$, we get

$$P_{min}(\nu_{14})\approx\frac{4\delta}{\lambda_{14}^2}\frac{\Gamma_3}{\Gamma_{43}}\left(1-\frac{\Gamma_{32}}{\Gamma_3}\ln\frac{\Gamma_3+\Gamma_{32}}{\Gamma_{32}}\right)^{-1}.$$

If $\tau<t_0$, then the necessary value is $P>P_{min}$. Substituting the concrete parameters $E_{14}=103.2$ keV and $T_4=0.5\times 10^{-12}$ sec (Ref. 8) and the highest values $\sigma^{polar}_{amp}\approx\sigma^{unpol}_{amp}\approx 3.7\times 10^{-18}$ cm^2 for the given nucleus (Δm

$=1, L=1)$ for quantum motion along **H**, we find that since $\tau > t_0$ it follows that $P_{min} \sim 10^{11}$ photons/sec · cm² · Hz.

Let us estimate the possibilities of contemporary pulsed x-ray installations. The spectral flux of bremsstrahlung photons is described by the expression

$$I_T(\nu) = 2W(E_{max} - E)\eta/\tau E_{max}^2 E,$$

where $E = h\nu$, $E_{max} = eV$ is the maximum energy of the electron (the end point of the bremsstrahlung spectrum), $W = NE_{max}$ is the energy of all the beam electrons, $N = i\tau/e$, i is the beam current, $\eta = \eta(E_{max})$ is the efficiency of x-ray generation in the anode[9,10] and depends on the accelerating voltage V and on the atomic number Z of the anode material; $\eta \approx 1.5\times10^{-9} ZV$ if $\eta \ll 1$ and $\eta \approx 0.5$ if $ZV = 10^9$ V. Taking into account the relativistic compression of the radiation pattern in the solid angle $\Delta\Omega$ in the direction of the electron beam

$$\Delta\Omega \sim (m_0 c^2/E_{max})^2,$$

we obtain for the spectral density of the bremsstrahlung photon flux at a distance R from a focal spot of area S

$$P_{br}(\nu) = 6\cdot 10^4 i(E_{max} - E)/E_{max}E(R^2\Delta\Omega + S) \text{ photons/sec} \cdot \text{cm}^2 \cdot \text{Hz}.$$

Here i is the current in amperes, E and E_{max} is the energy in electron volts, and R and $\sqrt{S}$ are in centimeters. The best of the modern x-ray installations produce current pulses $i \approx 3\times10^6$A at $V \approx 10^7$ V and $\tau \approx 10^{-7}$ sec.[11] At these parameters, and at the typical values $Z \sim 70-90$, $S \sim 0.1$ cm², $R \sim 1-5$ cm, and $E_{14} \sim 100$ keV we get $P_{br}(\nu_{14}) \sim 10^7$ photons/sec · cm² · Hz. It is seen that the value $P_{br}(\nu_{14})$ attained in the case of bremsstrahlung is insufficient to ensure the amplification condition. At $E_{max} > 200$ keV, however, besides the generation of bremsstrahlung, there is always excitation of the characteristic spectrum. We note that at $E_{max} \gg 200$ keV the total integrated flux of bremsstrallung photons greatly exceeds the flux of the characteristic radiation photons, so that the value $P_{br}(\nu_{14})$ cited above remains approximately the same in magnitude also when the characteristic radiation is excited.

Usually the spectral flux of the characteristic-radiation photons $I_x(\nu)$ is larger than $I_{br}(\nu)$ by four orders of magnitude. Assuming a spatial dependence of the radiation of the characteristic photons in the form customarily used for dipole transitions (this is the worst case), we can find the value of $P_x(\nu_{14})$ in the characteristic spectrum. At the same parameters and at $R \sim 0.3$ cm we obtain $P_x(\nu_{14}) \sim 10^{11}$ photons/sec · cm² · Hz. In contrast to diffraction experiments, where a parallel photon beam is necessary, a γ laser can be pumped by an isotropic flux. Then, using reflectors or a focusing system (via Bragg diffraction or glancing reflection), it is possible to increase by several times the value of $P_x(\nu_{14})$ and realistically obtain $P_x(\nu_{14}) \sim (1-3)\times 10^{11}$ photons/sec · cm² · Hz. Some decrease in the indicated value can be caused by absorption in the anode material, but at realistic values of anode thickness 1–2 mm of pulsed x-ray installations with relativistic electrons and at an absorption cross section $\sigma_{nr} \lesssim 10^{-22}$ cm² at $E \sim 100$ keV in a medium with $Z \sim 70-90$, the attenuation of P_x is small.

Returning to the isomer in question, we find that the transition energy $E_{14} = 103.2$ keV in Dy^{161} agrees (when account is taken of the K-line width) with the energy of the $K_{\beta 2}$ line in radium ($E_k = 103.05$ keV). Consequently, if the anode of the x-ray tube is made of radium (or if the traditional refractory anodes of platinum and tungsten are enriched with radium), then it is possible to obtain the specified radiation parameters and produce the required degree of inversion in the amplifying medium for the γ laser. If the pump is focused and the characteristics of the x-ray installations are improved by one order ofmagnitude, it is possible to use many other nuclei. Such an increase, as indicated in Ref. 11, does not entail any difficulties in principle. An increase of $P_x(\nu_{14})$ by 20% over P_{min} ensures in Dy^{161} a linear amplification coefficient of approximately 10 cm⁻¹. We note that, without using polarization in the considered system, to satisfy the γ-amplification condition it is necessary to increase $P(\nu_{14})$ by 2×10^3 times.

The main difficulties in the development of a polarization γ laser lie in the heating of the active medium (which increases δ and decreases p). The processes of polarization of the medium and the subsequent pumping and amplification can be separated in time and in space, owing to the large t_r. In addition, a short-time heating ($t \lesssim t_0, \tau$) does not lead to a substantial decrease of the polarization during the γ-radiation pulse generation time.

We note in conclusion that schemes with characteristic pumping were considered previously,[12,13] but their use in two- and three-level versions is not effective (small cross section for photoexcitation and activation), and their use in ordinary (non-polarization) four-level scheme with the three radiative transitions ($4\to3\to2\to1$) instead of two leads in the considered polarized system ($4\to3\to2$) to a large energy E_{14} which is as a rule beyond the limits of the characteristic spectrum, so that such a mechanism cannot be used.

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Translated by J. G. Adashko