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Effect of thermal conditions on the generation threshold of Mössbauer γ radiation in a system of excited nuclei

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We consider the influence of thermal expansion and of the relaxation processes in the establishment of thermodynamic equilibrium on the attainment of the threshold condition for γ amplification in a system of excited short-lived Mössbauer nuclei. It is shown that at an initial crystal temperature $T \gtrsim 10^{-2}$ K the threshold condition is not attainable because of the inhomogeneous Doppler line broadening due to the γ-laser working-medium linear thermal expansion by the heat released in the pumping and by the absorption of the energy of conversion electrons and of the resonant γ quanta. At infralow temperatures $T < 10^{-3}$–10^{-5} K the phonon-electron and phonon-phonon relaxation times can exceed the lifetime of the excited states of the nuclei. As a result, the activation and emission of the nuclei will proceed more rapidly than the heating and linear thermal expansion of the sample, so that the attainment of the threshold condition for γ amplification in a system of polarized Mössbauer nuclei is feasible in principle.

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An exhaustive analysis of various γ-laser models makes it possible to specify concretely the lasing conditions, as well as the parameters of the working isotope and of the pump. This stimulates in final analysis experimental research in this field. In this article we estimate the temperature regime of the operating medium of a γ laser using short-lived Mössbauer isotopes ($\tau \sim 10^{-6} - 10^{-8}$ sec) in the pumping and emission processes, and demonstrate that the rate of change of the temperature with time, $T_t = \partial T/\partial t$ influences substantially the possibility of realizing the laser process.

The dependence of the resonant gain on the temperature was considered earlier only from the point of view of the "static" action of the heating, i.e., of the inhomogeneous and homogeneous temperature broadenings of the spectral lines[1] and of the decrease of the probability f of the Mössbauer effect with increasing temperature.[2] However, the sample temperature varies with time because of the heating of the laser working medium by the pumping as well as by absorption of the energy of the conversion-electron and of the resonant γ quanta upon decay of the excited nuclei. The linear thermal expansion causes a relative motion of the nuclei with velocity

$$v=\gamma l T_t,$$

where $\gamma(T)$ is the thermal-expansion coefficient and l is the distance between nuclei. The corresponding Doppler shift of the frequency leads to an inhomogeneous broadening of the emission line and to a decrease of the gain. Since noticeable lasing is possible only if the active medium is extensive enough, the gain length must be larger than or of the order of the length l_{ph} of the photoabsorption of the resonant γ quanta. If the threshold value of the line width is denoted by Γ^*, then at $l = l_{\text{ph}}$ the critical rate of change of the temperature is determined by the condition

$$T_t^*=\Gamma^* c/\nu\gamma(T) l_{\text{ph}}, \tag{1}$$

where c is the speed of light and ν is the frequency of the resonant transition.

In the γ-laser models of Refs. 1–3, appreciable cooling of the working medium is proposed, but since the relaxation processes in a solid slow down with decreasing temperature, and the characteristic lasing time is of the order of the lifetime of the short-lived transitions, we must, before proceeding to estimate the influence of thermal deformations on the possibility of lasing, answer the following question: can heating and thermal expansion of the crystal take place within a time of the order of τ?

In the case of sufficiently pure metals, the principal mechanism whereby the electron-gas energy is transferred to the lattice is the electron-phonon interaction. Heating of the sample by absorption of the energy of conversion electrons and of x-ray and γ quanta is effected entirely via this mechanism. The time of establishment of thermodynamic equilibrium in the system made up of the electron gas and the lattice is characterized by the phonon-electron relaxation time τ_{pe}.[4,5] Since the relaxation of the phonon distribution function to an equilibrium form is determined also by the phonon-phonon relaxation time τ_{pp}, it is obvious that these two parameters are the ones that characterize the time of establishment of an equilibrium crystal temperature.

In the quasiharmonic approximation, the process of thermal expansion of solids can be described in the following manner: The violation of the thermal equilibrium in the phonon system leads to a change of the lattice free-energy function $F(\omega_q)$, and this causes also a change in the volume V of the body, in order that the corresponding variation of the phonon-mode energy $\hbar\omega_q$ lead to satisfaction of the thermodynamic-equilibrium condition $(\partial F/\partial V)_T = 0$. The end result of this chain of transformations is the establishment of new equilibrium values of F and of the phonon distribution function. Thus, finding the characteristic time of thermal expansion reduces to a determination of the time of relaxation of the phonon distribution function, i.e., to a determination of the times τ_{pp} and τ_{pe}.

To calculate τ_{pe} we use the theory of thermal conductivity of a solid,[4,5] which yields in the quasiharmonic approximation

$$\varkappa_{pe}={}^1/_3 c_p v_s^2 \tau_{pe}, \tag{2}$$

where $\varkappa_{pe}$ is the lattice thermal conductivity due to the phonon-electron interaction, v_s is the average phonon speed and is assumed equal to the speed of sound, and c_p is the lattice specific heat per unit volume.

Direct utilization of relation (2) is made complicated by the fact that in the case of metals the thermal conductivity is determined principally by the electronic component. In addition, at low temperatures, ($T<0.1\Theta_D$, where Θ_D is the Debye temperature), it is due to the scattering of the electrons by the crystal defects, and the quantity $\varkappa_{pe}$ is not determined in experiment. Estimates of $\varkappa_{pe}$ at low temperatures were based on the semi-empirical formula[4]

$$\varkappa_{pe}\approx\frac{3.67}{n_a^2}\left(\frac{T}{\Theta_D}\right)^2\varkappa_{e\infty}, \tag{3}$$

where n_a is the number of free electrons per atom, and $\varkappa_{e\infty}$ is the electron thermal conductivity at high temperatures. Since $c_p \propto T^3$ at low temperatures, the relaxation time τ_{pe} varies like $1/T$.

Equation (2) is generally speaking valid also for a description of phonon-phonon interactions, but there are no published reliable experimental data for a number of parameters needed to calculate $\varkappa_{pp}$. At the same time there exist unique experiments on Poiseuille flow of phonons, in which a value of $v_s\tau_{pp}$ in bismuth was directly measured.[6] If it is assumed that the relaxation time τ_{pp} obtained in this manner is close in order of magnitude to τ_{pp} in metals, then we can estimate τ_{pp} at ~ 10^{-8} sec to be $T = 4$K. With decreasing temperature, τ_{pp} increases rapidly like $1/T$.[4,6]

We consider now the model of a neutron-pumped γ laser. Following Ref. 2, we choose a working isotope with atomic number $A = 150$, $\tau = 10^{-7}$ sec, $h\nu = 15$ keV, and $f = 0.5$, introduced at a concentration ~ 10^{-3} into a beryllium matrix whose specific heat at $T = 4$K is $c_p = 1.3\times 10^{-5}$ J/g·K, $\Theta_D = 1160$ K, $\varkappa_{e\infty} = 0.8$ W/cm·K, and $v_s = 10^6$ cm/sec.[7,8] Calculations by Eqs. (2) and (3) lead to the inequality $\tau_{pe}\approx 10^{-12}$ sec $\ll \tau_{pp} \ll \tau$. This in turn means that the beryllium sample manages to become heated within a time on the order of τ to a (thermodynamically) definite temperature and its volume changes to a corresponding optimal value. The macroscopic heat-conduction equation is then valid.

We consider the thermal processes in a thin long beryllium rod of diameter $d = 10^{-4}$ cm and length $l_{\rm ph} = 2.5$ cm,[9] placed in a helium thermostat. For neutron pumping of the upper resonant levels, the main heating source is absorption of high-energy captured γ quanta. This source releases heat at a density $Q = 1.7\times 10^8$ W/cm^3 (Ref. 2). The approximate solution of the heat-conduction equation with allowance for the temperature dependence of the specific heat $c = aT^3$ is then

$$T(t)=[\Theta^4+4Qt/a\rho]^{1/4}, \tag{4}$$

where $\rho = 1.84$ g/cm^3 is the density of the beryllium, and $\Theta = 4$K is the initial temperature. Using the handbook data,[8] we can estimate the coefficient a at 2×10^{-7} J/g·K^4. It follows from (4) that at the instant $t = 0.1\tau$ when the pumping is stopped the relative temperature is $T_0 = 67$K.

We assume that the pumping is followed by a weak-amplification regime, wherein the decay of the excited nuclei hardly differs from ordinary spontaneous decay. The density function of the thermal sources takes the form

$$Q(t)=(Q_0^{(\gamma)}+Q_0^{(e)})\exp(-\Gamma_0 t), \tag{5}$$

where $\Gamma_0 = 1/\tau$. The initial heat-release density, due to absorption of the resonant γ quanta is, at a conversion coefficient α and an excited working-isotope density n_0,

$$Q_0^{(\gamma)}=n_0\Gamma_0 h\nu\frac{J}{1+\alpha}\frac{\pi d}{4l_{\rm ph}}\cdot \tag{6}$$

The energy contribution of the conversion electrons of energy E_i, absorbed from the i-th shell at an absorption coefficient μ_i is

$$Q_0^{(e)}=\frac{n_0\Gamma_0}{1+\alpha}\sum_i \alpha_i E_i\frac{\pi d\mu_i}{4}. \tag{7}$$

It is easy to show, using the handbook data,[9] that at E_i ~ 10 keV the conversion-electron energy is completely absorbed by a beryllium rod of diameter ~ 1 μm. The results of calculations by formulas (6) and (7) for different values of the conversion coefficient α are listed in Table I. When (5) is taken into account, the solution of the heat-conduction equation yields the following expression for the rod temperature

$$T=\left\{T_0^4+\frac{4(Q_0^{(\gamma)}+Q_0^{(e)})}{a\rho\Gamma_0}[1-\exp(-\Gamma_0 t)]\right\}^{1/4}. \tag{8}$$

The values of T and T_t at the instant $t = 0.5\tau$, which is close to the time of maximum amplification,[10] are entered in the table. Estimates of the threshold line width Γ^* were taken from the paper of Gol'danskiĭ and Kagan,[2] and the critical value of T_t^* is obtained from formula (1) with allowance for the temperature depen-

TABLE I.

α	$Q_0^{(\gamma)}$, W/cm^3	$Q_0^{(e)}$, W/cm^3	T, K	T_t, K/sec	T_t^*, K/sec	Γ^*/Γ_0
10	$4\cdot 10^6$	$1.5\cdot 10^{12}$	$>T_{\rm melt}$	–	–	10
10^{-2}	$4\cdot 10^7$	$1.7\cdot 10^9$	295	10^9	$4\cdot 10^5$	10^2
0	$4\cdot 10^7$	0	78	$2\cdot 10^8$	$9\cdot 10^5$	10^2

dence of $\gamma(T) = 1.2 \times 10^{-12} T^3 K^{-1}$ (Ref. 8) at $T \ll \Theta_D$. In the case $\alpha = 10$ the sample temperature exceeds the melting temperature of the beryllium ($T_{\text{melt}} \approx 1560\,\text{K}$) and the procedure of calculating T_t becomes meaningless.

We note that solutions (4) and (8) do not contain any parameters characterizing heat exchange with the medium. The reason is that the intensity of the heat-releasing sources is in this case so large that even in heat exchange with liquid helium the heat is carried away too slowly to change noticeably the temperature of the body. Indeed, when the temperature differential changes from 0.4 to 600K, the coefficient of heat transfer 8 of a thin rod with $d \sim 10^{-3}$ cm changes from 0.6 to 1.7 W/cm^2 K (Ref. 11), leading to a heat-release intensity $4h\,(T - \Theta)/d \sim 10^6\,\text{W/cm}^3 \ll Q$. All that the helium thermostat does is maintain the sample temperature low up to the instant of the start of the activation.

It is seen from the table that at $\alpha > 10^{-2}$ the sample heating reaches or exceeds the temperature $T \sim 300\,\text{K}$ which is allowable from the point of view of the value of f. In addition, $T_t \gg T_t^*$, therefore the condition for γ amplification will not be satisfied. While a decrease of α down to zero does make it possible to maintain the sample at a temperature of low critical, the thermal expansion leads to the condition $T_t \gg T_t^*$ and consequently stops the lasing as a result of photoabsorption of the energy of the resonant γ quanta. Generally speaking, the heat-release rate can be decreased, but the requirement that the values of n_0, f, and Γ_0 in (6) be decreased contradicts the requirement that the gain be increased, and further decrease of the rod diameter or of its length [see (1), (6), and (7)] is useless, since it lowers the energy of the γ-radiation pulse. We emphasize also that the cases $\alpha = 0$ and $\alpha = 10^{-2}$ are far from realistic, since the electron conversion coefficients of Mössbauer isotopes range from 0.12 for Ni^{61} to 325 for Tm^{169}.

It must be noted, however, that one cannot rule out in principle the possibility of avoiding the harmful influence of thermal deformations and of heating in general, if one succeeds in satisfying the condition

$$\tau \ll \tau_{pe},\ \tau_{pp}, \tag{9}$$

at which the excitation and emission of the nuclei is faster than the heating and linear expansion of the sample. The condition (9) can be satisfied either by choosing an isotope with shorter lifetime τ, or by considerably decreasing the initial temperature at which the relaxation times are long. For example, for a beryllium matrix the relaxation time increases to 5×10^{-7} sec at $T = 10^{-5}\,\text{K}$. Unfortunately, this procedure encounters many difficulties. First, when τ is decreased the requirements with respect to the necessary pumping neutron fluxes become more stringent, and, second, it is not easy to obtain and use such low temperatures. We note that

$$\tau_{pe} \sim 1/T\Theta_D^2 v_s^2,$$

and it is therefore necessary to use crystals with as low values of Θ_D and v_s as possible.

We turn now to a recently proposed scheme of nothreshold γ amplification in a system of polarized nuclei. Vysotskiĭ[3] points out the possibility of amplification on one of the frequencies of the nuclear multiplet of polarized nuclei if the total density of the excited short-lived isotopes is lower by 3–5 orders of magnitude than that of the unexcited ones. This makes it possible to use as the pump the characteristic radiation of powerful pulsed x-ray installations[3] or coherent excitation by a modulated beam of relativistic electrons.[12]

We shall analyze the thermal processes in this model, choosing as the working element, just as in Ref. 3, the isotope Dy^{161}, for which $c_b = 4.3 \times 10^{-6} T^3$ J/g·K, $v_s \approx 10^5$ cm/sec, $\varkappa_{e\infty}$ 0.1 W/cm·K, $\Theta_D = 210$ K, and $\rho = 8.5$ g/cm^3.[7,8] To attain the required polarization of the ground state of the nuclei, the sample must be cooled to a temperature $T < 0.01\,\text{K}$. At $T < 10^{-3}\,\text{K}$ we then get from (2) and (3) the value $\tau_{pe} \gtrsim 1.7 \times 10^{-6}$ sec. The lifetime of the working transition of the considered four-level scheme is $\tau = 8.7 \times 10^{-9}$ sec, i.e., the condition (9) is satisfied, so that the influence of the temperature on the possibility of lasing can be neglected here. We indicate for comparison that if the initial temperature of the sample is of the order of 1K, which corresponds to $\tau_{pe} \sim 10^{-11}$ sec, then a reversal of inequality sign (9) leads to heating of the working medium to a temperature $T > T_{\text{melt}}$ at the following pump parameters: spectral density ~10^{11} quanta/sec·cm^2·Hz, quantum energy ~100 keV, spectral width of the characteristic line ~10 eV (Ref. 3); $\mu \approx 16.8$ cm^{-1} (Ref. 9), and $d \approx 10^{-3} - 10^{-4}$ cm.

We can conclude thus, that pumping and γ amplification in a system of polarized nuclei is realistic at infralow temperatures at which the times of the phonon-phonon and phonon-electron relaxations exceed the lifetime of the Mössbauer transition. In this case the processes of pumping and formation of the laser avalanche will be more rapid than the heating and thermal expansion of the working medium of the γ laser.

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General features of collision narrowing of spectral lines in gases

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A theoretical description is presented of the narrowing of the Doppler contour of spectral lines and of the narrowing of the *Q* branch of the Raman-scattering spectrum of molecules. The description is based on the exact properties of the collision integral. This reveals a number of new features of the phenomenon, some of which had been observed in experiment and found to have no theoretical explanation. The identity of the mathematical formalism used in the investigation of spectral structures of different origin emphasizes the common interference origin of the phenomenon.

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In the overwhelming majority of cases, collisions of a molecule with surrounding particles lead to a broadening of the spectral line. We consider below two deviations from this rule: the relatively rarely observed narrowing of the Doppler line contour by collisions, which was predicted by Dicke,[1] and the narrowing of the Q-branch of Raman scattering of simple molecules (see, e.g., Refs. 2–4 and the literature cited therein).

Model solutions were constructed for the theoretical description of these effects.[3,5–8] It is shown in the present article that the main qualitative features of the line-narrowing effects can be described without invoking model solutions, within the framework of a theory based only on the exact properties of the collision integral. In addition to an overall survey of the spectral-line narrowing process, this approach reveals new features, not contained in the model solutions, of the described phenomenon.

1. NARROWING OF THE DOPPLER CONTOUR OF A SPECTRAL LINE BY COLLISIONS

1. To facilitate the subsequent comparison with experiment, we consider the specific case of broadening of an isolated vibrational-rotational component of the Raman-scattering spectrum of a molecule. With simple and obvious change of the coefficients of the right-hand sides of the equations, all the formulas presented below are equally applicable to ordinary linear or two-photon absorption.

The line shape of an isolated vibrational-rotational component of the Raman-scattering spectrum of a molecule is described by the density-matrix element

$$\rho(q=0j_1m_1,\ q=1j_2m_2,\ \mathbf{v},\ t)\equiv e^{i(\omega_1-\omega_2)t}\rho(m_1m_2\mathbf{v}),$$

where q is the vibrational quantum number, j is the rotational number, m is the projection of the angular momentum on the z axis, and ω_1 and ω_2 are the frequencies of the incident and scattered waves. The matrix elements $\rho(m_1m_2v)$ satisfy the equation (see, e.g., Ref. 9)

$$\begin{gathered} i(\Delta\omega-\mathbf{pv})\rho(m_1m_2\mathbf{v})+\sum_{m_1'm_2'}\nu(m_1m_2,\, m_1'm_2',\, \mathbf{v})\rho(m_1'm_2'\mathbf{v}) \\ -\sum_{m_1'm_2'}\int A(m_1m_2\mathbf{v},\, m_1'm_2'\mathbf{v}')\rho(m_1'm_2'\mathbf{v}')\,d\mathbf{v}'=DQ(m_1m_2)W(\mathbf{v}), \\ D=\frac{i}{\hbar}(N_{m_1}-N_{m_2});\quad Q(m_1m_2)=\sum_{ks}E_{1k}E_{2s}^{*}(\alpha_{ks})_{m_1m_2}. \end{gathered} \tag{1.1}$$

Here $\Delta\omega=\omega_{12}-\omega_1+\omega_2$, $\mathbf{p}=\mathbf{p}_1-\mathbf{p}_2$, ω_{12} is the frequency of the molecular transition 1 → 2; $\mathbf{p}_{1,2}$ and $\mathbf{E}_{1,2}$ are the wave vectors and the complex amplitudes of the incident and scattered fields, $N_{mi}=n_i/(2j_i+1)$ is the equilibrium population, m_i are the components of the level with angular momentum j_i, $W(\mathbf{v})$ is the Maxwellian velocity distribution function, α_{ks} is the molecule scattering tensor, and $\hbar$ is Planck's constant.

The intensity of the Raman-scattering line is given by

$$I(\omega)=-2\,\mathrm{Re}\,i\omega_2\sum_{m_1m_2}Q^{*}(m_1m_2)\int\rho(m_1m_2\mathbf{v})\,d\mathbf{v}. \tag{1.2}$$

In the case of linear absorption, the transition density matrix is

$$\rho(q=0j_1m_1,\ q=1j_2m_2,\ \mathbf{v},\ t)\equiv e^{i\omega t}\rho(m_1m_2\mathbf{v}),$$

where ω is the frequency of the absorbed wave, and Eq. (1.1) remains unchanged if it is kept in mind that in this case $\Delta\omega=\omega_{12}-\omega$, $\mathbf{p}$ and $\mathbf{E}$ are the wave vector and amplitude of the absorbed wave, $Q=\mathbf{E}\cdot\mathbf{d}_{j_1m_1,j_2m_2}$, and $\mathbf{d}$ is the dipole moment of the transition. Expression (1.2) for the intensity likewise remains unchanged.

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