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Stimulated gamma radiation of free isomer nuclei upon anti-Stokes transitions

A A Zadernovsky

Abstract. **The resonance anti-Stokes conversion of broadband X-ray radiation to stimulated gamma radiation of free isomer nuclei is considered. The conversion involves the two-quantum transition from the initial long-lived isomer state of a nucleus via an intermediate level to the final state located below the isomer state. The quantum-mechanical calculation of the cross section for resonance stimulated anti-Stokes scattering involving quanta of the different multipolarity yields the estimate of the gain of stimulated gamma radiation in a nuclear beam with the spectrally local inversion and the estimate of the threshold spectral density of the X-ray pump radiation flux.**

Keywords: *stimulated gamma radiation, anti-Stokes transitions, isomers.*

1. Introduction

The unique possibility of building a gamma-ray laser, which has attracted the attention of researchers for many years, is based on the use of isomer excited states of nuclei with the energy from tens of kiloelectronvolts to tens of megaelectronvolts and the lifetime from a few microseconds to a few tens and even hundreds of years.

An inverse medium prepared by the photochemical separation of long-lived excited nuclear isomers from unexcited isomers could be in principle used for amplification of stimulated gamma radiation with the resonance cross section $\sigma_0 = (\lambda^2/2\pi)\beta$, where λ is the transition wavelength and β is the ratio of the radiative transition width to the total width of the emission line.

However, the necessity of using nuclei with long lifetimes in the excited isomer state results in such a narrow radiative width of the operating transition that even small perturbations cause a significant broadening of the emission line and drastically reduce the coefficient β. First of all, this concerns the Doppler broadening, which proves to be so large in the gamma-ray range (the Doppler width is proportional to the gamma-ray transition energy) that the amplification of gamma quanta becomes impossible due to their nonresonance losses during propagation in the medium.

A A Zadernovsky Moscow State Institute of Radio Engineering, Electronics, and Automation (Technical University), prosp. Vernadskogo 78, 117454 Moscow, Russia

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Most of the schemes of a nuclear gamma laser assume the use of the Mössbauer effect for isomer nuclei in a crystal lattice to greatly increase the interaction of stimulated gamma radiation with resonance transitions. The use of Mössbauer nuclear transitions would allow one to eliminate the Doppler broadening and reduce the width of the gamma radiation line to the natural width. In this case, the coefficient β would achieve its maximum value equal to unity. Unfortunately, the Mössbauer effect is only possible for short-lived nuclear states with the lifetime shorter than ~ 10 μs, which is too small for separating excited nuclear isomers and preparing a crystal inverse medium from them.

The difficulties encountered in the creation of the self-consistent scheme of a solid-state gamma laser operating on Mössbauer nuclear transitions [1] stimulated the development of the alternative concept of a gamma laser [2] operating on cooled (monokinetic) beams of free nuclei. This concept is based on the use of the spectral splitting of the gamma radiation and absorption lines in a cooled nuclear ensemble.

It is known that the centres of the gamma radiation and absorption lines for a free nucleus are separated by a distance equal to the doubled recoil energy. Modern methods of the laser cooling of neutral atoms [3] allow one to reduce the Doppler width of the nuclear gamma-ray transition to such a degree that the gamma radiation and absorption lines are no longer overlapped. This facilitates the appearance of the spectrally local population inversion, i.e., the amplification of stimulated gamma radiation in a certain spectral region even when the number of excited nuclei does not exceed that of the unexcited ones.

Long-lived excited isomers, whose lifetime is sufficient for cooling a nuclear ensemble, are extremely attractive for building a free nucleus gamma-ray laser. In this case, there is still no point in using the isomer excited state as the upper level of the laser transition because the radiative width of this long-lived state is substantially smaller than the Doppler width even after cooling, so that the coefficient $\beta \ll 1$.

Instead, we consider the fast decay of a metastable isomer state by-passing a direct forbidden transition. Thus, anti-Stokes scattering of the intense electromagnetic radiation via a higher-lying, rapidly decaying level could accelerate the transition from the isomer level to the ground level. In the absence of resonance between the energy of incident photons and the transition energy between the isomer and intermediate levels, the decay rate increases very weakly even upon irradiation by high-power modern lasers [4 – 6]. The possibility of obtaining accelerated decay of a nuclear isomer state upon resonance anti-Stokes X-ray scattering was theoretically predicted in Ref. [7].

In [8–12], the accelerated release of the energy of a nuclear isomer state has been experimentally demonstrated upon giant resonance absorption of X-rays with the energy sufficient for reaching critical states of a nucleus caused by its deformation.

In this paper, we considered the resonance anti-Stokes conversion of broadband X-ray radiation to stimulated gamma radiation of free isomer nuclei. Anti-Stokes scattering is accompanied by the two-quantum transition of a nucleus from the initial long-lived isomer state via a higher-lying intermediate level to the final state, which is located below the isomer state. This process is accompanied by absorption of an X-ray quantum by a nucleus and simultaneous emission of spontaneous or stimulated gamma quantum. The reverse process is also possible, namely, resonance absorption of the emitted gamma quantum accompanied by the transition of the nucleus from the ground to the intermediate state. If the emission and absorption lines cease to overlap because of the recoil imparted to the nucleus, the stimulated radiation will be amplified.

The quantum-mechanical calculation of the cross section for resonance stimulated anti-Stokes scattering with quanta of different (sometimes large) multipolarity, which is typical for isomer nuclear transitions, yields the estimate of the gain of stimulated gamma radiation in a nuclear beam with the spectrally local inversion. The calculation was performed using the perturbation theory that requires some generalisation to take into account the recoil effect, which has been usually neglected in the studies of optical electronic transitions in an atom.

2. Radiative nuclear transitions with regard to the recoil

The Hamiltonian of a system consisting of a nucleus and a field can be represented as a sum of the Hamiltonian $H_0 = \boldsymbol{p}^2/2M$, which describes the motion of the nucleus as a free particle with the mass M and the momentum $\boldsymbol{p}$, the nuc-lear Hamiltonian $H_{\rm n}$, which describes the motion and inter-action between nucleons in the nucleus, the Hamiltonian $H_{\rm f} = \sum_{\boldsymbol{k}\sigma} \hbar\omega(a^+_{\boldsymbol{k}\sigma} a_{\boldsymbol{k}\sigma} + {}^1/_2)$ of a free electromagnetic field, which contains the creation ($a^+_{\boldsymbol{k}\sigma}$) and annihilation ($a_{\boldsymbol{k}\sigma}$) operators of a photon with energy $\hbar\omega$, momentum $\hbar\boldsymbol{k}$, and polarisation σ, and the Hamiltonian H_I of interaction between the nucleus and the electromagnetic radiation field.

The latter Hamiltonian can be in turn represented as a sum of three terms: $H_I = H_{\rm E} + H_{\rm M} + H_{\rm D}$. The first term $H_{\rm E}$ describes the electric multipole interaction of the nucleus with the radiation field and represents a series $H_{\rm E} = H_{\rm E1} + H_{\rm E2} + \ldots + H_{\rm E\mathit{L}} + \ldots = -(\boldsymbol{dE}) + \ldots$, where $\boldsymbol{d}$ is the dipole moment of the nucleus and

$$\boldsymbol{E} = {\rm i}\sum_{\boldsymbol{k}\sigma} (2\pi\hbar\omega)^{1/2} \boldsymbol{e}_{\boldsymbol{k}\sigma}\left(a_{\boldsymbol{k}\sigma}{\rm e}^{{\rm i}\boldsymbol{kR}} - a^+_{\boldsymbol{k}\sigma}{\rm e}^{-{\rm i}\boldsymbol{kR}}\right)$$

is the radiation electric field strength at the centre of mass of the nucleus with the radius vector $\boldsymbol{R}$.

The term $H_{\rm M}$ describes the magnetic multipole interaction of the nucleus and can be also written as a series $H_{\rm M} = H_{\rm M1} + H_{\rm M2} + \ldots + H_{\rm M\mathit{L}} + \ldots = -(\boldsymbol{\mu H}) + \ldots$, where $\boldsymbol{\mu}$ is the dipole magnetic moment of the nucleus and

$$\boldsymbol{H} = -{\rm i}\sum_{\boldsymbol{k}\sigma} (2\pi\hbar\omega)^{1/2} [\boldsymbol{e}_{\boldsymbol{k}\sigma} \times \boldsymbol{\kappa}]\left(a_{\boldsymbol{k}\sigma}{\rm e}^{{\rm i}\boldsymbol{kR}} - a^+_{\boldsymbol{k}\sigma}{\rm e}^{-{\rm i}\boldsymbol{kR}}\right)$$

is the magnetic field at the centre of mass of the nucleus ($\boldsymbol{\kappa} = \boldsymbol{k}/k$).

The third term $H_{\rm D}$ is nonlinear in the magnetic field and, as shown in [12, 13], $H_{\rm D} \sim (H^2_{\rm E}/mc^2) \sim (H^2_{\rm M}/mc^2)$, where m is the nucleon mass. Because $H_{\rm E,M}/mc^2 \ll 1$, we will neglect this term.

Series $H_{\rm E}$ and $H_{\rm M}$ are power series in the dimensionless parameter $r_0/\lambda \ll 1$, where $r_0 = 1.2 \times 10^{-13} A^{1/3}$ is the nuclear radius in centimetres; A is the number of nucleons; and λ is the characteristic radiation wavelength of the nucleus. In addition,

$$H_{\rm E\mathit{L}} \sim \left(\frac{r_0}{\lambda}\right)^{L-1/2}, \; H_{\rm M\mathit{L}} \sim \frac{v}{c}\left(\frac{r_0}{\lambda}\right)^{L-1/2},$$

where v/c is the ratio of the characteristic velocity of nucleons in the nucleus to the speed of light.

Finally, we can write the interaction Hamiltonian as a sum over multipole moments L:

$$H_I = \sum_L \sum_{\boldsymbol{k}\sigma} \Big[\Big(V^{({\rm E}L)}_{\boldsymbol{k}\sigma} + V^{({\rm M}L)}_{\boldsymbol{k}\sigma}\Big){\rm e}^{{\rm i}\boldsymbol{kR}} a_{\boldsymbol{k}\sigma}$$

$$+ \Big(V^{({\rm E}L)}_{\boldsymbol{k}\sigma} + V^{({\rm M}L)}_{\boldsymbol{k}\sigma}\Big)^* {\rm e}^{-{\rm i}\boldsymbol{kR}} a^+_{\boldsymbol{k}\sigma}\Big], \quad (1)$$

where both the electric $\Big(V^{({\rm E}L)}_{\boldsymbol{k}\sigma}\Big)$ and magnetic $\Big(V^{({\rm M}L)}_{\boldsymbol{k}\sigma}\Big)$ multiple interactions can be represented in the form $V^{(L)}_{\boldsymbol{k}\sigma} = (\omega/c)^{L-1/2} V^{(L)}_{\boldsymbol{\kappa}\sigma}$, where $V^{(L)}_{\boldsymbol{\kappa}\sigma}$ is independent of the photon frequency and depends only on the direction of its radiation specified by the unit vector $\boldsymbol{\kappa}$.

Let the nucleus has the momentum $\boldsymbol{p}_0$ and is in the excited state $|i\rangle$ with the energy $E_i = \hbar\omega_i$, while the radiation field contains $n_{\boldsymbol{k}\sigma}$ photons with the wave vector $\boldsymbol{k}$ and polarisation σ. In other words, the initial state of the system consisting of the nucleus and the radiation field has the form $|\boldsymbol{p}_0, i, n_{\boldsymbol{k}\sigma}\rangle$. The final state $|\boldsymbol{p}, f, n_{\boldsymbol{k}\sigma} + 1\rangle$ is represented by the nucleus with the momentum $\boldsymbol{p}$ in the lower-lying state $|f\rangle$ with the energy $E_f = \hbar\omega_f$ and $(n_{\boldsymbol{k}\sigma} + 1)$ photons in the radiation field.

The probability of the nuclear transition of multiplicity L between these states per unit time is

$$P^{(1)}_{fi} = \frac{2\pi}{\hbar}\left(\frac{\omega}{c}\right)^{2L-1} \left|\Big(V^{(L)*}_{\boldsymbol{\kappa}\sigma}\Big)_{fi}\right|^2 (n_{\boldsymbol{k}\sigma} + 1)\delta_{0,\boldsymbol{p}_0 - \boldsymbol{p} - \hbar\boldsymbol{k}}$$

$$\times\delta\left(\hbar\omega + E_f + \frac{p^2}{2M} - E_i - \frac{p_0^2}{2M}\right), \quad (2)$$

where we took into account that $\langle\boldsymbol{p}|{\rm e}^{-{\rm i}\boldsymbol{kR}}|\boldsymbol{p}_0\rangle = \delta_{0,\boldsymbol{p}_0 - \boldsymbol{p} - \hbar\boldsymbol{k}}$. The finite lifetime of the nucleus in the states with the energy E_i and E_f is taken into account using the Breit–Wigner procedure [14] by making the replacement $E_i \to E_i - {\rm i}\Gamma_i/2$ for the initial state of the transition and $E_f \to E_f + {\rm i}\Gamma_f/2$ for its final state and by replacing the delta function in (2) by a Lorentzian $g(\hbar\omega + E_f + p^2/2M - E_i - p_0^2/2M)$ with the FWHM equal to $\Gamma_i + \Gamma_f$, where $\Gamma_{i,f} = \hbar\gamma_{i,f}$; $\gamma_{i,f} = t^{-1}_{i,f} \ln 2$; and $t_{i,f}$, is the half-life of the state i and f, respectively, determined by radiative transitions and electron conversion. The factor $n_{\boldsymbol{k}\sigma} + 1$ takes into account the contribution of stimulated and spontaneous radiative transitions.

If we set $n_{\boldsymbol{k}\sigma} = 0$ in (2) and then sum up the obtained expression over the finite nuclear momentum $\boldsymbol{p}$, the momentum $\hbar\boldsymbol{k}$ and polarisation σ of the emitted photon, we obtain the probability $W_{\rm sp}^{(L)}$ of the radiative spontaneous decay of the nuclear transition per unit time (the radiative width $\gamma_{if}^{(\rm R)}$ of the transition):

$$W_{\rm sp}^{(L)} = \gamma_{if}^{(\rm R)} = \frac{1}{(2\pi\hbar)^2}\frac{1}{c}\left(\frac{\omega_{if}}{c}\right)^{2L+1} \times \sum_{\sigma}\int\left|\left(V_{\boldsymbol{\kappa}\sigma}^{(L)*}\right)_{fi}\right|^2 {\rm d}\Omega. \quad (3)$$

It follows from (3) that the transition matrix element averaged over angular variables and two possible types of polarisation of a photon has the form

$$\left|\tilde{V}_{fi}^{(L)*}\right|^2 = \frac{1}{2}\sum_{\sigma}\left(\frac{1}{4\pi}\int\left|\left(V_{\boldsymbol{\kappa}\sigma}^{(L)*}\right)_{fi}\right|^2 {\rm d}\Omega\right) = \frac{1}{8\pi}\frac{c(2\pi\hbar)^2\gamma_{if}^{(\rm R)}}{(\omega_{if}/c)^{2L+1}}. \quad (4)$$

The probability of emission of a spontaneous photon with some polarisation σ within the interval of frequencies ${\rm d}\omega$ and solid angles ${\rm d}\Omega$ is determined by the expression

$${\rm d}W_{\rm sp}^{(L)} = \frac{1}{(2\pi\hbar)^2}\frac{1}{c}\left(\frac{\omega}{c}\right)^{2L+1}\left|\left(V_{\boldsymbol{\kappa}\sigma}^{(L)*}\right)_{if}\right|^2 \times g\left[\omega - \omega_{if} - \frac{\omega_{if}}{c}(\boldsymbol{v}_0\boldsymbol{\kappa}) + \frac{E_{\rm R}}{\hbar}\right]{\rm d}\omega{\rm d}\Omega, \quad (5)$$

where the Lorentzian with the FWHM $\gamma_i + \gamma_f$ has a maximum at the frequency shifted relative to the transition frequency ω_{if} by $(\omega_{if}/c)(\boldsymbol{v}_0\boldsymbol{\kappa}) - E_{\rm R}/\hbar$ (here, the first term, which depends on the initial velocity $\boldsymbol{v}_0$ of the nucleus, describes the Doppler effect, the second term takes into account a decrease in the radiation frequency caused by the recoil upon emission of a gamma quantum, and $E_{\rm R} = (\hbar\omega_{if})^2/2Mc^2$ is the recoil energy imparted to the nucleus.)

Because of a random orientation of the nucleus relative to the radiation direction, we should replace the matrix element in (5) by its averaged value $\tilde{V}_{fi}^{(L)*}$ (4). The summation over two possible photon polarisations yields the probability of spontaneous emission of a photon in the frequency interval ${\rm d}\omega$ and the solid-angle interval ${\rm d}\Omega$ in the detuning range $\varDelta = |\omega - \omega_{if}| \ll \omega_{if}$ in the form

$${\rm d}W_{\rm sp}^{(L)} = \frac{\gamma_{if}^{(\rm R)}}{4\pi} g\left[\omega - \omega_{if} - \frac{\omega_{if}}{c}(\boldsymbol{v}_0\boldsymbol{\kappa}) + \frac{E_{\rm R}}{\hbar}\right]{\rm d}\omega{\rm d}\Omega, \quad (6)$$

which differs from the conventional expression [15] only by the presence of the Doppler term and the recoil term in the Lorentzian.

The cross section for stimulated radiation

$$\sigma_{\rm st} = \pi\lambda_{if}^2\frac{{\rm d}W_{\rm sp}^{(L)}}{{\rm d}\omega{\rm d}\Omega} = \frac{\lambda_{if}^2}{4}\gamma_{if}^{(\rm R)} g\left[\omega - \omega_{if} - \frac{\omega_{if}}{c}(\boldsymbol{v}_0\boldsymbol{\kappa}) + \frac{E_{\rm R}}{\hbar}\right] \quad (7)$$

and the absorption cross section

$$\sigma_{\rm ab} = \frac{2J_i + 1}{2J_f + 1}\frac{\lambda_{if}^2}{4}\gamma_{if}^{(\rm R)} g\left[\omega - \omega_{if} - \frac{\omega_{if}}{c}(\boldsymbol{v}_0\boldsymbol{\kappa}) - \frac{E_{\rm R}}{\hbar}\right] \quad (8)$$

in the same frequency region (where $\lambda_{if} = 2\pi c/\omega_{if}$ is the transition wavelength, and J_i and J_f are spins of the nucleus in the upper and lowers states) differ from conventional cross sections [15] only by the shift of the centres of emission and absorption lines relative to the transition frequency caused by the Doppler effect and recoil. One can see from (7) and (8) that the relative shift of the centres of absorption and emission lines equals $2E_{\rm R}/\hbar$.

3. Conversion of X-ray radiation to stimulated gamma radiation of free isomer nuclei

Consider a nucleus that initially occupies a long-lived isomer level with energy E_i (Fig. 1). Upon irradiation of the nucleus, it can experience the two-quantum transition to the lower-lying state with energy E_f via an intermediate level with energy E_s. This process is accompanied by absorption of a photon with energy $\hbar\omega_1$ by the nucleus and simultaneous emission of spontaneous or stimulated gamma quantum with energy $\hbar\omega_2$.

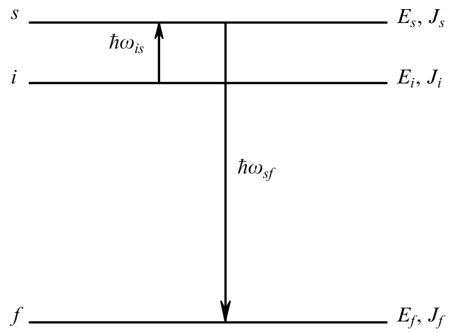

Figure 1. Anti-Stokes resonance two-quantum transition from the initial nuclear isomer state i via the intermediate state s to the final state f.

The initial state $|p_0, i, n_{\boldsymbol{k}_1\sigma_1}, n_{\boldsymbol{k}_2\sigma_2}\rangle$ of the system consisting of the nucleus and the field corresponds to the nucleus with the momentum p_0 in the isomer state i and $n_{\boldsymbol{k}_1\sigma_1}$ photons with the wave vector $\boldsymbol{k}_1$ and polarisation σ_1 in the mode of incident radiation, and also $n_{\boldsymbol{k}_2\sigma_2}$ photons with the wave vector $\boldsymbol{k}_2$ and polarisation σ_2 of scattered radiation. The intermediate states are represented by the set $|p_s, s, n_{\boldsymbol{k}_1\sigma_1} - 1, n_{\boldsymbol{k}_2\sigma_2}\rangle$ or $|p_s, s, n_{\boldsymbol{k}_1\sigma_1}, n_{\boldsymbol{k}_2\sigma_2} + 1\rangle$, while the final state has the form $|p, f, n_{\boldsymbol{k}_1\sigma_1} - 1, n_{\boldsymbol{k}_2\sigma_2} + 1\rangle$. The probability of this two-quantum transition per unit time is

$$P_{fi}^{(2)} = \frac{2\pi}{\hbar}\left(\frac{\omega_1}{c}\right)^{2L-1}\left(\frac{\omega_2}{c}\right)^{2K-1}\left|M_{fi}^{(L,K)}\right|^2 n_{\boldsymbol{k}_1\sigma_1}(n_{\boldsymbol{k}_2\sigma_2} + 1) \times g\left(\hbar\omega_2 + E_f + \frac{p^2}{2M} - \hbar\omega_1 - E_i - \frac{p_0^2}{2M}\right), (9)$$

where the matrix element has the form

$$M_{fi}^{(L,K)} = \sum_{\boldsymbol{p}_s}\left[\frac{\delta_{0,\boldsymbol{p}_s - \boldsymbol{p} - \hbar\boldsymbol{k}_2}\delta_{0,\hbar\boldsymbol{k}_1 + \boldsymbol{p}_0 - \boldsymbol{p}_s}\langle f|V_{\boldsymbol{\kappa}_2\sigma_2}^{(K)*}|s\rangle\langle s|V_{\boldsymbol{\kappa}_1\sigma_1}^{(L)}|i\rangle}{E_i + \hbar\omega_1 + p_0^2/2M - E_s - p_s^2/2M - i(\varGamma_i + \varGamma_s)/2}\right] + \sum_{\boldsymbol{p}_s}\left[\frac{\delta_{0,\boldsymbol{p}_s + \hbar\boldsymbol{k}_1 - \boldsymbol{p}}\delta_{0,\boldsymbol{p}_0 - \hbar\boldsymbol{k}_2 - \boldsymbol{p}_s}\langle f|V_{\boldsymbol{\kappa}_1\sigma_1}^{(L)}|s\rangle\langle s|V_{\boldsymbol{\kappa}_2\sigma_2}^{(K)*}|i\rangle}{E_i - \hbar\omega_2 + p_0^2/2M - E_s - p_s^2/2M - i(\varGamma_i + \varGamma_s)/2}\right]. \quad (10)$$

Note that both terms in (10) have a resonance nature. The first term is large when the incident-photon energy $\hbar\omega_1$ is close to the energy $E_s - E_i$ (this is possible if the level s lies above the level i). The second term is large if the energy of $\hbar\omega_2$ of the scattered photon is close to $E_i - E_s$ (this is possible when the level s lies below the final level f, if the latter is not the ground level). In our case (Fig. 1), the second term in (10) can be neglected.

If the radiation with the spectrally-angular photon flux density $F(\omega_1, \boldsymbol{\kappa}_1)$ is incident on a nucleus in the direction $\boldsymbol{\kappa}_1$, the probability of anti-Stokes scattering per unit time with emission of a spontaneous gamma quantum can be written in the form $\mathrm{d}W_{\rm sp} = \sigma_{\rm sp}(\omega_1, \boldsymbol{\kappa}_1)F(\omega_1, \boldsymbol{\kappa}_1)\mathrm{d}\omega_1\mathrm{d}\Omega_1$, where the cross section for spontaneous anti-Stokes scattering is determined by the expression

$$\sigma_{\rm sp}(\omega_1, \boldsymbol{\kappa}_1) = \frac{2J_s+1}{2J_i+1}\frac{\lambda_1^2}{4}\left(\frac{\omega_1}{\omega_{si}}\right)^{2L+1}\left(\frac{\omega_1+\omega_{if}}{\omega_{sf}}\right)^{2K+1}$$
$$\times\frac{\gamma_{si}^{(\rm R)}\gamma_{sf}^{(\rm R)}}{\gamma_i+\gamma_s}g\left(\omega_1-\omega_{si}-\frac{\omega_1}{c}(\boldsymbol{v}_0\boldsymbol{\kappa}_1)-\frac{1}{\hbar}\frac{(\hbar\omega_1)^2}{2Mc^2}\right), \quad (11)$$

which generalises the result obtained in Refs [5, 7] by taking into account in the Lorentzian with the FWHM $\gamma_i + \gamma_s$ the Doppler effect for a nucleus moving at the velocity $\boldsymbol{v}_0$ and the recoil experienced by the nucleus upon absorption of a photon with energy $\hbar\omega_1$.

If the function $F(\omega_1, \boldsymbol{\kappa}_1)$ has a broad spectrum (with the width greatly exceeding the width of each of the levels under study) and a narrow angular directivity ($\Delta\Omega_1 \ll 4\pi$) near $\boldsymbol{\kappa}_1$, then the cross section for stimulated anti-Stokes scattering can be represented in the form

$$\sigma_{\rm st} = \frac{2J_s+1}{2J_i+1}\frac{\lambda_{is}^2}{4}\frac{\lambda_{sf}^2}{4}\frac{\gamma_{si}^{(\rm R)}\gamma_{sf}^{(\rm R)}}{\gamma_i+\gamma_s}F(\omega_{si})$$
$$\times g\left[\omega_2-\omega_{sf}-\frac{\omega_{sf}}{c}(\boldsymbol{v}_0\boldsymbol{\kappa}_2)+\frac{E_{2\rm R}}{\hbar}\left(1-\frac{2E_{si}}{E_{sf}}(\boldsymbol{\kappa}_1\boldsymbol{\kappa}_2)\right)\right], \quad (12)$$

where the Lorentzian g has the width $\gamma_i + (\gamma_s + \gamma_f)/2$; the unit vector $\boldsymbol{\kappa}_2$ specifies the direction of emission of a stimulated gamma quantum; $E_{2\rm R} = (\hbar\omega_{sf})^2/2Mc^2$ is the recoil energy upon emission of this gamma quantum; and $F(\omega_{si})$ is the spectral density of the incident X-ray flux at the transition frequency ω_{si}.

4. Amplification of stimulated gamma radiation

To estimate the gain of stimulated gamma radiation by an ensemble of free isomer nuclei, it is necessary to consider two competing processes: anti-Stokes conversion of broad-band X-ray radiation to stimulated gamma radiation of nuclei with the cross section (12) and absorption of emitted gamma quanta accompanied by the transition of the nucleus from the state f to the state s (Fig. 1) with the cross section (8), where the subscript i should be replaced by the index s. The cross sections of these processes should be averaged over all possible velocities of emitting nuclei. For this purpose, one should multiply them by the probability $f(u)\mathrm{d}u = (M/2\pi kT)^{1/2}\exp(-Mu^2/2kT)\mathrm{d}u$ of falling the projection of the nucleus velocity on the direction of radiation $u = (\boldsymbol{v}_0\boldsymbol{\kappa}_2)$ within the interval from u to $u + \mathrm{d}u$ and integrate the expression obtained over all possible projections. The result depends on the degree of scatter in the velocities of nuclei, i.e., on the temperature T of the nuclear ensemble.

For $(E_{2\rm R}kT)^{1/2} \ll \hbar(\gamma_s + \gamma_f)$ (the width of the level being measured is very small, $\gamma_i \ll \gamma_s, \gamma_f$, and we neglect it hereafter), the Doppler term $(\omega_{sf}/c)(\boldsymbol{v}_0\boldsymbol{\kappa}_2)$ in (12) and (8) can be neglected. In this case, a homogeneous broadening dominates, and the gain of stimulated gamma radiation contains contributions from all nuclei with the coefficient $G = \sigma_{\rm st}n_i - \sigma_{\rm ab}n_f$ (n_i and n_f are the concentrations of nuclei in the isomer state i and the final state f, respectively). The gain at the maximum of the emission line $\omega_2 = \omega_{sf} - (E_{2\rm R}/\hbar)[1 - (2E_{si}/E_{sf})(\boldsymbol{\kappa}_1\boldsymbol{\kappa}_2)]$ is equal to

$$G = \frac{\lambda_{sf}^2}{2\pi}\frac{\gamma_{sf}^{(\rm R)}}{\gamma_s+\gamma_f}\left\{\frac{2J_s+1}{2J_i+1}\frac{\lambda_{si}^2}{4}\frac{\gamma_{si}^{(\rm R)}}{\gamma_s}F(\omega_{si})n_i\right.$$
$$\left.-\frac{2J_s+1}{2J_f+1}n_f\left[\frac{\hbar(\gamma_s+\gamma_f)}{4E_{2\rm R}[1-(E_{si}/E_{sf})(\boldsymbol{\kappa}_1\boldsymbol{\kappa}_2)]}\right]^2\right\}. \quad (13)$$

One can see that for $E_{2\rm R} \gg \hbar(\gamma_s + \gamma_f)$, when the emission and absorption lines are no longer overlapped, and for a sufficiently high spectral density $F(\omega_{si})$ of the X-ray flux, the gain at the centre of the emission line becomes possible even in the absence of inversion.

In the opposite case of $(E_{2\rm R}kT)^{1/2} \gg \hbar(\gamma_s + \gamma_f)$, when the Doppler broadening exceeds the homogeneous broadening, the gain at the maximum of the emission line is described by the expression

$$G = \frac{\lambda_{sf}^2}{2\pi}\frac{\gamma_{sf}^{(\rm R)}}{\gamma_{\rm D}}(\pi\ln 2)^{1/2}\left\{\frac{2J_s+1}{2J_i+1}\frac{\lambda_{si}^2}{4}\frac{\gamma_{si}^{(\rm R)}}{\gamma_s}F(\omega_{si})n_i\right.$$
$$\left.-\frac{2J_s+1}{2J_f+1}n_f\exp\left[-\frac{E_{2\rm R}}{kT}\left(1-\frac{E_{si}}{E_{sf}}(\boldsymbol{\kappa}_1\boldsymbol{\kappa}_2)\right)^2\right]\right\}, \quad (14)$$

where $\hbar\gamma_{\rm D} = 4(E_{2\rm R}kT\ln 2)^{1/2}$ is the Doppler FWHM of the emission line. For $E_{2\rm R} \gg \hbar\gamma_{\rm D}$, i.e., when the emission and absorption lines are no longer overlapped, the gain can be positive at sufficiently large $F(\omega_{si})$ even in the absence of inversion.

The numerical calculation performed for the nucleus $^{242}_{95}$Am (the energy of the isomer state is $E_i = 48.63$ keV, the half-life is 141 year, the spin and parity are 5^-; the energy of the intermediate level is $E_s = 52.9$ keV, the spin and parity are 3^-; the ground-state half-life is 16.02 h, the spin and parity are 1^-) gives (see (14)) $G = \sigma_{\rm st}n_i = 7.3\times10^{-48}\times T^{-1/2}F(\omega_{si})n_i$, where we took into account that $n_f = 0$. For the rates of radiative transitions $\gamma_{si}^{(\rm R)}$ and $\gamma_{sf}^{(\rm R)}$ (both transitions are electric quadrupole $E2$ transitions), we used the Weisskopf estimate $\gamma^{(\rm R)} = 7.28\times10^{-8}E^5A^{4/3}$ [16] ($\gamma^{(\rm R)}$ is measured in reciprocal seconds and the transition energy is measured in kiloelectronvolts) and the Doppler width was estimated from the formula $\gamma_{\rm D} = 1.08\times10^{12}\times E(T/A)^{1/2}$ (E was measured in kiloelectronvolts and T was measured in kelvins). The nonresonance losses of gamma quanta with energy ~ 50 keV were mainly caused by photo-ionisation of atoms with the cross section $\sigma_i = 5\times10^{-22}$ cm^2, so that the threshold spectral density of the photon flux required to achieve the gain is estimated at $T = 10^{-8}$ K as $F(\nu_{si}) = 2\pi F(\omega_{si}) \approx 4.3\times10^{22}$ photon cm^{-2} s^{-1} Hz^{-1}.

A similar calculation for the nucleus $^{102}_{45}$Rh (the isomer-state energy $E_i = 140.75$ keV, the half-life is ~ 2.9 years, the spin and parity is 6^+; the intermediate-level energy is $E_s = 154.43$ keV, the spin and parity is 5^+; the lower-level energy is $E_f = 105.22$ keV, the half-life is 0.9 ns, and the spin and parity are represented by the superposition from 1^+, 2^+, and 3^+) gives $G = \sigma_{\rm st} n_i = 4.6 \times 10^{-44} T^{-1/2} F(\omega_{si}) n_i$ for $T \geqslant 10^{-7}$ K. At lower temperatures, the homogeneous broadening dominates, and the gain $G = \sigma_{\rm st} n_i$ becomes equal to $2.4 \times 10^{-40} F(\omega_{si}) n_i$ (see (13)) and no longer depends on temperature. The $s \to f$ transition is an electric quadrupole transition, while the rate of the magnetic dipole transition $s \to i$ was estimated as $\gamma_{si}^{(\rm R)} = 3.15 \times 10^4 E_{si}^3$ [16] ($\gamma_{si}^{(\rm R)}$ is measured in reciprocal seconds and the transition energy E_{si} is measured in kiloelectronvolts). As a result, we obtain for the threshold spectral density of the X-ray radiation $F(\nu_{si}) = 2\pi F(\omega_{si}) \approx 1.3 \cdot 10^{19}$ photon cm^{-2} s^{-1} Hz^{-1} at $T = 10^{-8}$ K.

The accuracy of the performed numerical calculations is determined by the reliability of estimates of the rates of radiative nuclear transitions, which is approximately within two-three orders of magnitude.

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