Resonant scattering of neutrons and γ quanta by crystals at large recoil energy

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The influence of vibrational relaxation on incoherent resonant scattering of neutrons and γ quanta by crystals is considered theoretically. It is assumed that the recoil energy received by the nucleus upon absorption of the primary particle greatly exceeds the characteristic vibrational quantum, the lifetime of the excited state of the nucleus is much longer than the characteristic time of the phase vibrational relaxation, and the excitation takes place in the phonon wing of the absorption spectrum of the nucleus. It is shown that under these conditions the principal components (in intensity) of the smoothed spectra of the scattered neutrons or γ quanta are of the type of ordinary and hot-luminescence components, the former corresponding to emission of a secondary particle after complete vibrational relaxation, and the latter corresponding to emission after the phase relaxation but during the energy relaxation. If the lifetimes of the excited nuclear state are comparable with or larger than the characteristic period of the vibrations of the nucleus, the form of the spectrum of the hot-luminescence type is determined essentially by the detailed law governing the damping of the excited oscillations of the nucleus.

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Many substantial singularities of the energy and angular distributions of neutrons and γ quanta of moderate energy, scattered by crystals, are due to the vibrational motion of the scattering nuclei. In the nonresonant case, these singularities have been investigated in considerable detail (see, e.g., [1,2]). For resonant scattering, some properties, governed by the nuclear vibrations, of the spectra of the secondary neutrons and γ quanta were considered in [3-6]. In general, however, the resonance case has remained little investigated to this day. Yet the role of the vibrational motion of nuclei in resonant and nonresonant processes is substantially different, and the spectra of the scattered neutrons or γ quanta can differ qualitatively from each other in the two cases.

The reason for this difference is the delay that occurs when the scattered particle is emitted in the resonant case. This delay is due to the transition of the nucleus that has absorbed the primary particle into an excited quasistationary state (compound nucleus), and the delay time can be comparable with the characteristic relaxation time of the vibrational state of the excited (compound) nucleus, or may even exceed the relaxation time. These relaxation processes disturb strongly the phase correlation between the incident and scattered waves and this, as we shall show, greatly affects all the characteristics of the scattering.

The role of vibrational relaxation in the formation of the spectra of resonantly scattered γ quanta and neutrons was considered in [7,8,11]. There, however, the only investigated case was that when the recoil energy acquired by the nucleus after absorbing the primary particle, R_{ii}, is small ($R_{ii} \lesssim \hbar\bar{\omega}$, where $\bar{\omega}$ is the characteristic vibration frequency of the scattering nucleus). The form of the spectrum at large recoil energy ($R_{ii} \gg \hbar\bar{\omega}$) was not established in [7,8]. Yet it is precisely in this case that the motion of the excited nucleus is strongly perturbed by excitation in the phonon wing of the absorption spectrum, and the effects connected with vibrational relaxation should be most noticeable. The present paper is devoted to a theoretical investigation of such effects.

1. GENERAL FORMULAS

We consider the scattering of an unpolarized and fully collimated beam of γ quanta or neutrons by a crystal scatterer. We assume that the crystal contains nuclei whose excitation energy is at resonance with the energy of the incident particle (resonant nuclei). Such nuclei can be located either at the sites of an ideal lattice, or be present as impurities in the crystal. For simplicity we assume the crystal to have cubic symmetry and that the nuclei occupy in it equivalent crystallographic positions. The last assumption is not fundamental, and the results obtained below can be easily generalized to the case of a crystal having a different symmetry or containing resonant nuclei in nonequivalent positions.

Under the foregoing conditions, for a monochromatic incident beam, the spectral distribution of the resonantly and incoherently scattered neutrons or γ quanta is given by the formula[3-7]

$$I(\omega_f;\omega_i,\theta)=\frac{\gamma^2}{4\pi}\int_{-\infty}^{\infty} ds\int_0^{\infty} d\tau\int_{-\tau}^{\tau} ds'\, e^{-\gamma\tau}\exp[i(\omega_f-\omega_i)s]\times\exp[2i(\omega_{10}-\omega_i)s']K_{if}(\tau,s,s'), \quad (1)$$

where

$$K_{if}(\tau,s,s')=\langle\exp[i\mathbf{k}_i\hat{\mathbf{r}}(0)]\exp[-i\mathbf{k}_f\hat{\mathbf{r}}(\tau+s')]\times\exp[i\mathbf{k}_f\hat{\mathbf{r}}(\tau+s+s')]\exp[-i\mathbf{k}_i\hat{\mathbf{r}}(s+2s')]\rangle. \quad (2)$$

Here $\omega_i, \mathbf{k}_i$ and $\omega_f, \mathbf{k}_f$ are the frequencies and the wave vectors of the incident and scattered neutron or γ quantum, θ is the scattering angle ($\theta = \mathbf{k}_i, \mathbf{k}_f$), ω_{10} is the excitation frequency of the immobile resonant nucleus, γ is the natural width of the excited level, $\hat{\mathbf{r}}(x) = \exp\times(ixH/\hbar)\mathbf{r}\exp(-ixH/\hbar)$, $\mathbf{r}$ is the radius vector of the scat-

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tering nucleus, H is the Hamiltonian of the vibrational motion of the nuclei in the crystal, and the symbol $\langle\ldots\rangle$ denotes temperature averaging over the vibrational states of the lattice.

In the pair-correlation approximation (the Baym approximation), the correlator $K_{if}(\tau, s, s')$ is equal to [7,8]

$$K_{if}(\tau, s, s')=\exp\{2[-W_{ii}(0)-W_{ff}(0)+W_{if}(\tau+s')+W_{fi}(s'-\tau)-W_{if}(\tau+s'+s)-W_{fi}(s+s'-\tau)+W_{ii}(s+2s')+W_{ff}(s)]\}, \quad (3)$$

where

$$W_{\lambda\lambda'}(x)=\frac{M_r}{3\hbar^2}R_{\lambda\lambda'}\langle\hat{\mathbf{r}}(0)\hat{\mathbf{r}}(x)\rangle. \quad (4)$$

Here

$$R_{\lambda\lambda'}=\frac{\hbar^2\mathbf{k}_\lambda\mathbf{k}_{\lambda'}}{2M_r}, \quad (5)$$

and M_r is the mass of the resonant nucleus. The quantities R_{ii} and $R_{ff}(\lambda, \lambda'=i$ and $\lambda, \lambda'=f)$ are the recoil energies acquired by the nucleus upon absorption and emission of the primary and secondary γ quantum or neutron.

In the derivation of (1) we neglected the hyperfine interaction of the nuclear electric and magnetic moments with internal electric and magnetic fields in the crystal, as well as the increase of the exciting-nucleus mass. The diffuse motion of the nuclei is not taken into account in (1), and the vibrational motion is considered in the adiabatic approximation.

At a finite spectral width of the incident flux, and with allowance for the finite spectral resolution of the instruments that record the scattered particles, the observed spectrum of the secondary neutrons or γ quanta is described by the formula

$$I(\omega_f; \omega_i, \theta)=\int d\bar{\omega}_i\, d\bar{\omega}_f I_i(\omega_i-\bar{\omega}_i)I(\bar{\omega}_f; \bar{\omega}_i, \theta)I_f(\omega_f-\bar{\omega}_f). \quad (6)$$

The function $I_i(\omega_i-\bar{\omega}_i)$ determines here the frequency distribution of the incident neutrons or γ quanta, while $I_f(\omega_f-\bar{\omega}_f)$ characterizes the resolving power of a detector tuned to register quanta of frequency ω_f. The functions I_i and I_f are normalized to unity. Assume that they are bell-shaped with maxima at the frequencies ω_i and ω_f and with widths equal to Δ_i and Δ_f.

We consider henceforth the case when: 1) the recoil energy R_{ii} is much larger than the energy of vibrational quantum characteristic of the resonant nucleus $\hbar\bar{\omega}(R_{ii}\gg\hbar\bar{\omega})$, 2) the nucleus is excited in the region of the phonon wing of the spectrum of the resonant absorption of the neutrons or γ quanta ($|\omega_i-\omega_{10}-R_{ii}/\hbar|\lesssim\Gamma$, where $\Gamma\sim[\hbar^{-1}\bar{\omega}R_{ii}(2n(\bar{\omega})+1)]^{1/2}$ is the width of the phonon wing of the absorption spectrum and $n(\omega)=[\exp(\hbar\omega/k_BT)-1]^{-1}$), 3) Δ_i and Δ_f satisfy the condition $\bar{\omega}\ll\Delta_i, \Delta_f\ll\Gamma$. By the same token, the observed spectrum $\bar{I}(\omega_f; \omega_i, \theta)$ is a smoothed spectrum, having neither narrow lines (e.g., a Mössbauer line or a quasi-elastic scattering line) nor a phonon structure. However, since the spectrum $I(\omega_f; \omega_i, \theta)$ is of the multiphonon type at $R_{ii}\gg\hbar\bar{\omega}$, the smoothed spectrum does not differ in its principal (multiphonon) part in practice from the exact spectrum.[2)]

The condition $\bar{\omega}\ll\Delta_i, \Delta_f\ll\Gamma$ makes it possible to confine oneself in (1) to the region of small values of the variables s and s' (for the smoothed spectrum, the significant values of s and s' are $|s'\lesssim\Delta_i^{-1}\ll\bar{\omega}^{-1}$ and $|s|\sim\Delta_f^{-1}\ll\bar{\omega}^{-1}$). This makes it possible in turn to expand the expression under the exponential sign in (3) in powers of s and s' and retain terms up to second order inclusive.[3)] As a result we obtain for the spectrum $\bar{I}(\omega_f; \omega_i, \theta)$ the following approximate expression, which is valid at scattering angles $\theta\gg(\hbar\omega/R_{ii})^{1/2}$:

$$I(\omega_f; \omega_i, \theta)\approx\frac{\gamma^2}{4\pi}\int_{-\infty}^{\infty}ds\int_0^{\infty}d\tau\int_{-\tau}^{\tau}ds'\, e^{-\gamma\tau}\exp[i(\omega_f-\omega_i)s]$$
$$\times\exp[i(\omega_{10}-\omega_i)2s']\exp\left\{\frac{i}{\hbar}[(R_{ff}-2R_{if}(\tau)+R_{ii})s+2R_{ii}s']-\Gamma_{ff}^2s^2-\Gamma_{ii}^2(s+2s')^2+2[\Gamma_{if}(\tau)]^2s(s+2s')\right\}, \quad (7)$$

where

$$R_{if}(\tau)=-\frac{\hbar^2(\mathbf{k}_i\mathbf{k}_f)}{2M_r}\frac{1}{6i\hbar}\sum_\alpha\{\langle[\hat{p}_\alpha(\tau), \hat{r}_\alpha(0)]\rangle+\langle[p_\alpha(0), \hat{r}_\alpha(\tau)]\rangle\},$$
$$\Gamma_{ii(ff)}^2=R_{ii(ff)}\frac{\langle\hat{p}^2\rangle}{3\hbar^2M_r}, \quad (8)$$
$$\Gamma_{if}^2(\tau)=R_{if}(0)\frac{1}{6\hbar^2M_r}[\langle\hat{\mathbf{p}}(\tau)\hat{\mathbf{p}}(0)\rangle+\langle\mathbf{p}(0)\hat{\mathbf{p}}(\tau)\rangle],$$

$\hat{\mathbf{p}}$ is the momentum operator of the scattering nucleus.

In the case of weak anharmonicity, the functions Γ_{ii}^2, $\Gamma_{ff}^2, \Gamma_{if}^2(\tau)$ and $R_{if}(\tau)$ are approximately of the form

$$\Gamma_{ii(ff)}^2=R_{ii(ff)}\frac{1}{6\hbar}\sum_p\omega_p|e(p)|^2[2\bar{n}(\omega_p)+1],$$
$$[\Gamma_{if}(\tau)]^2=R_{if}(0)\frac{1}{6\hbar}\sum_p\omega_p|e(p)|^2[2\bar{n}(\omega_p)+1]\exp[-\gamma_p|\tau|]\cos\omega_p\tau, \quad (9)$$
$$R_{if}(\tau)=\frac{\hbar^2(\mathbf{k}_i\mathbf{k}_f)}{2M_r}\frac{1}{3}\sum_p|e(p)|^2\exp(-\gamma_p|\tau|)\cos\omega_p\tau.$$

Here ω_p, $\mathbf{e}(p)$ and $\bar{n}(\omega_p)$ are the frequency, polarization vector, and occupation number of the pth normal mode of the resonant-nucleus oscillations. The factor $\exp(-\gamma_p|\tau|)$ determines the p-mode damping due to the interaction between the modes. It is seen from (9) that $\Gamma_{ii(ff)}\sim[R_{ii(ff)}\bar{\omega}/\hbar]^{1/2}$ and $\hbar\Gamma_{ii(ff)}\ll R_{ii(ff)}$ at $R_{ii(ff)}\ll\hbar\bar{\omega}$. In addition, inasmuch as in the significant cases we have $R_{ii}\ll\hbar\omega_i$ and $R_{ff}\ll\hbar\omega_f$, while $\hbar|\omega_i-\omega_f|\lesssim2(R_{ii}+R_{ff})$, it follows that $R_{ii}\approx R_{ff}$ and $\Gamma_{ii}\approx\Gamma_{ff}$.

We note that according to formula (7) the smoothed spectrum $\bar{I}(\omega_f; \omega_i, \theta)$ at $\theta\gg(\hbar\bar{\omega}/R_{ii})^{1/2}$ does not depend on the concrete form of the functions I_i and I_f. This dependence appears in the case of small scattering angles ($\theta\lesssim(\hbar\bar{\omega}/R_{ii})^{1/2}$), at which formula (7) is not directly applicable. We shall, however, not consider henceforth scattering through angles $\theta\lesssim(\hbar\bar{\omega}/R_{ii})^{1/2}$.

2. SPECTRA OF THE LUMINESCENCE AND HOT-LUMINESCENCE TYPE

The variables τ, s, and s' in (1)–(8) have the dimension of time and describe the time evolution of the state of the scattering nucleus,[12] namely: τ is the average (over two amplitudes of the transition probability) time during which the excited state of the nucleus (compound nucleus) exists, while s and s' determine the phase difference between the two probability amplitudes of the transitions to the final and intermediate state, respectively.

The significant values of s and s' in (7) lie in the range $|s'|, |s| \lesssim \Gamma_{ii}^{-1} \ll \bar{\omega}^{-1}$. (The exceptions are the already noted scattering through small angles $\theta \lesssim (\hbar\bar{\omega}/R_{ii})^{1/2}$ and the scattering through angles $\pi - \theta \lesssim (\hbar\bar{\omega}/R_{ii})^{1/2}$, which, however, will not be considered below.) On the other hand, the significant values of τ do not exceed the lifetime of the excited nuclear state $(\tau \lesssim \gamma^{-1})$.

In most cases of resonant scattering of γ quanta with moderate energies, the condition $\gamma \ll \Gamma_{ii}$ is satisfied. In a large number of cases of resonant scattering on neutrons with energy $\hbar\omega_i \gtrsim 50$ eV, the condition $\gamma \ll \Gamma_{ii}$ is also satisfied. Under these conditions the part of the spectrum with highest intensity $\bar{I}(\omega_f; \omega_i, \theta)$ is determined by the region $|\tau| \gg |s'|$, and the limits of integration with respect to τ and s' in (7) can be replaced respectively by 0, $+\infty$ and $\pm\infty$. Then the integrals with respect to s and s' in (7) can be directly evaluated and we get

$$\bar{I}(\omega_f; \omega_i, \theta) \approx \bar{I}_a(\omega_i) \int_0^\infty d\tau e^{-\gamma\tau} \bar{i}(\omega_f; \omega_i, \theta, \tau)\gamma, \tag{10}$$

where

$$\bar{I}_a(\omega_i) = \frac{\gamma\pi^{1/2}}{4\Gamma_{ii}} \exp\left\{-\frac{(\omega_{10} - \omega_i + R_{ii}/\hbar)^2}{4\Gamma_{ii}^2}\right\}, \tag{11}$$

$$\bar{i}(\omega_f; \omega_i, \theta, \tau) = [2\pi^{1/2}\Gamma(\tau, \theta)]^{-1} \exp\left\{-\frac{[\omega_f - \omega(\tau, \theta)]^2}{4[\Gamma(\tau, \theta)]^2}\right\}, \tag{12}$$

$$\Gamma(\tau, \theta) = \{\Gamma_{ff}^2 - [\Gamma_{if}(\tau)]^4/\Gamma_{ii}^2\}^{1/2},$$
$$\omega(\tau, \theta) = \omega_{10} - \frac{1}{\hbar}(R_{ff} - 2R_{if}(\tau)) - \frac{\Gamma_{if}^2(\tau)}{\Gamma_{ii}^2}\left(\omega_{10} - \omega_i + \frac{R_{ii}}{\hbar}\right). \tag{13}$$

In formula (10), $\bar{I}_a(\omega_i)$ is the smoothed spectrum of the resonant absorption of neutrons or γ quanta in an equilibrium vibrational state of the resonant nucleus at $R_{ii} \gg \hbar\bar{\omega}$.[13] On the other hand, the function $\bar{i}(\omega_f; \omega_i, \theta, \tau)$ is the instantaneous spectrum of the secondary particles emitted after phase relaxation of the vibrational state of the excited nucleus.[4)] The energy relaxation, however, may not yet be completed at the instant of their emission, and this state differs in general from equilibrium. Thus, with respect to the mechanism of the relaxation processes, formula (10) describes a spectrum analogous to the luminescence spectrum in secondary emission of the impurity centers of the crystal.[9-11] In analogy with the theory of the latter phenomenon, we shall call the components of the spectrum $\bar{I}(\omega_f; \omega_i, \theta)$, corresponding to emission of a secondary particle before or after the completion of the energy relaxation, spectra of the type of hot luminescence (HL) and ordinary luminescence (OL).

The instantaneous spectra in (10) have a Gaussian shape and their widths and positions depend on the instant of time τ. This dependence is determined in expressions (13) by the functions $R_{if}(\tau)$ and $\Gamma_{if}^2(\tau)$. The qualitative behavior of the functions $R_{if}(\tau)$ and $\Gamma_{if}^2(\tau)$ is known: they are determined by the law governing the variation of the velocity of the nucleus (see formulas (8) and (9)), and the dependence on the variable τ has the character of damped oscillations about a zero value. After the lapse of the characteristic time t_0, the functions $R_{if}(\tau)$ and $\Gamma_{if}(\tau)$ become in fact equal to zero, and this corresponds to establishment of an equilibrium vibrational state of the scattering nucleus.[5)]

In the case of resonant scattering of γ quanta, the condition $\gamma t_0 \ll 1$ is satisfied in a number of actual cases. Then the principal intensity component of the spectrum $\bar{I}(\omega_f; \omega_i, \theta)$ is a spectrum of the OL type—$\bar{I}_L(\omega_f, \omega_i)$.[7,8] This spectrum is determined by the times $\tau \gg t_0$ at which $R_{if}(\tau) \approx 0$ and $\Gamma_{if}(\tau) \approx 0$. Accurate to corrections $\sim\gamma t_0$, the spectrum $\bar{I}_L(\omega_f, \omega_i)$ is given by [7,8]

$$\bar{I}_L(\omega_f, \omega_i) = \bar{I}_a(\omega_i)\bar{I}_e(\omega_f), \tag{14}$$

where

$$\bar{I}_e(\omega_f) = \frac{1}{2\pi^{1/2}\Gamma_{ff}} \exp\left\{-\frac{[\omega_f - \omega_{10} + R_{ff}/\hbar]^2}{4\Gamma_{ff}^2}\right\}. \tag{15}$$

The spectrum $\bar{I}_e(\omega_f)$ is the smoothed spectrum of the γ quanta emitted by the resonant nucleus in an equilibrium vibrational state at a high recoil energy.

As follows from formula (14), a spectrum of the OL type does not depend on the characteristics of the incident flux, except via the absorption spectrum. In particular, its position and width do not depend on the scattering angle θ (Fig. 1).

If the excited nuclear state has a shorter lifetime $\gamma t_0 \gtrsim 1$ but $\gamma \ll \Gamma_{ii}$), the intensity of the OL-type spectrum decreases rapidly and the spectrum $\bar{I}(\omega_f; \omega_i, \theta)$ is in fact a spectrum of the HL type $(\bar{I}_{HL}(\omega_f; \omega_i, \theta)$. Moreover, in the region $|\omega_f - \omega_{10} + R_{ff}/\hbar| \gg \Gamma_{ff}$, where the spectrum $\bar{I}_L(\omega_f, \omega_i)$ becomes weak, $\bar{I}(\omega_f; \omega_i, \theta)$ coincides with $\bar{I}_{HL}(\omega_f; \omega_i, \theta)$ even if $\gamma t_0 \ll 1$.

A spectrum of the HL type corresponds in formula (10) to times $\tau \lesssim t_0$, when the functions $\Gamma_{if}(\tau)$ and $R_{if}(\tau)$

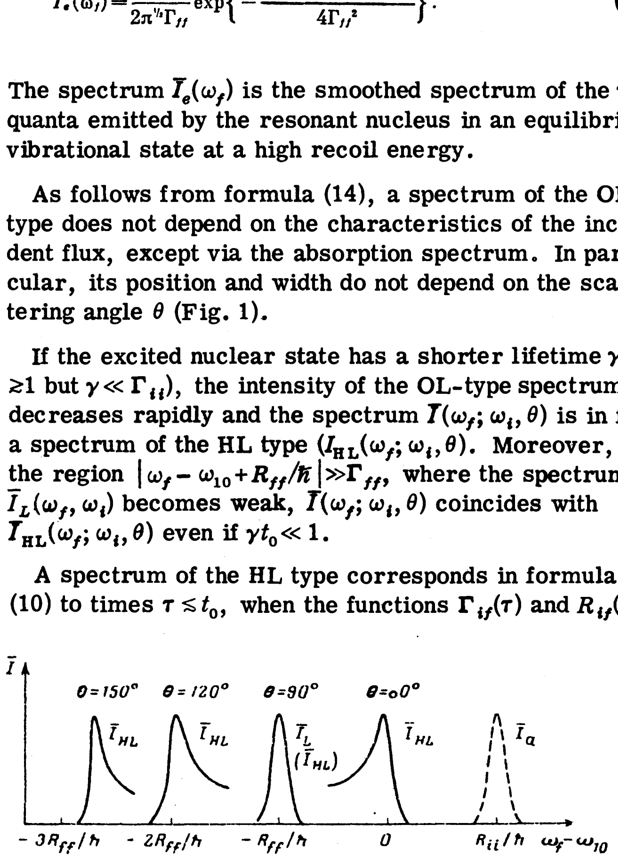

FIG. 1. Position and shape of maximum $n = 0$ of the type-HL spectrum at various scattering angles $\theta = 150°$, 120°, 90°, 60° ($R_{ff}/\hbar = 20\Gamma_{ff}$, $\gamma t_0 \ll 1$, $\omega_i = \omega_{10} + R_{ii}/\hbar + 0$, $1\Gamma_{ii}$, $|\omega(0, \theta) - \omega(\tau_2, \theta)| > |R_{if}(0)|$, the spectra are normalized to unity at the maximum point).

differ substantially from zero. Thus, the form of the spectrum $\bar{I}_{HL}(\omega_f;\omega_i,\theta)$ is determined by the detailed law that governs the damping of the excited vibrations of the resonant nucleus and can serve as a source of information on the vibrational relaxation processes.

3. FORM OF THE SPECTRUM OF THE HOT-LUMINESCENCE TYPE

In the general case of the HL-type spectrum defined by formula (10) admits of no exact analytic description. It turns out, however, that the form of $\bar{I}_{HL}(\omega_f;\omega_i,\theta)$ in individual sections of this spectrum is given approximately by expressions simpler than (10).

Indeed, according to (12) and (13) the width and position of the instantaneous spectra of the γ quanta or neutrons $\bar{i}(\omega_f;\omega_i,\theta,\tau)$ are determined by the oscillating functions $R_{if}(\tau)$ and $\Gamma^2_{if}(\tau)$, and therefore also oscillate with the time τ. Let us examine these oscillation in somewhat greater detail.

We designate the instants of time at which the derivative $dR_{if}(\tau)/d\tau$ vanishes by $\tau_n(n=0,1,2,\ldots)$, and let $\tau_{n+1}>\tau_n$ and $\tau_0=0$. We note that at $R_{ii}\gg\hbar\varpi$ the vibrational motion of the excited nucleus is described in the quasiclassical approximation. In this case the instants τ_n are the instants of the zero acceleration of a classical particle.

During the time interval between τ_n and $\tau_{n+1}(\tau_{n+1}-\tau_n\sim\pi/\varpi)$, the position of the maximum of the instantaneous spectrum $\omega(\tau,\theta)$ shifts by an amount $\sim2(|R_{if}(\tau_n|+|R_{if}\times(\tau_{n+1})|)/\hbar$. (We recall that $R_{if}(\tau_n)$ and $R_{if}(\tau_{n+1})$ are of opposite sign, and as a result of the process of energy relaxation we have $|R_{if}(\tau_n)|>|R_{if}(\tau_{n+1})|$.) Inasmuch as at $\tau_n\lesssim t_0$ we have $|R_{if}(\tau_n)|\gg\hbar\Gamma_{ff}$, the indicated shift greatly exceeds at small n the maximum possible width of the instantaneous spectrum (equal to Γ_{ff}). It follows therefore directly that the width of a spectrum of the HL type at $\gamma/\varpi\lesssim1$ should be $\sim2(|R_{if}(0)|+|R_{if}(\tau_1)|)/\hbar$, i.e., it should greatly exceed the width of the OL-type spectrum, and the HL spectrum should have a shape greatly different from Gaussian.[6)]

Let us investigate the spectral distribution of the secondary particles emitted by the nucleus in the time interval between τ_n and τ_{n+1}. According to (10), this distribution is given by the expression

$$\bar{i}_n(\omega_f;\omega_i,\theta)=\bar{I}_a(\omega_i)\int\limits_{\tau_n}^{\tau_{n+1}}d\tau\gamma e^{-\gamma\tau}\bar{i}(\omega_f;\omega_i,\theta,\tau). \tag{16}$$

In the calculation of the integral with respect to τ in (16) it is convenient to introduce a new integration variable, R_{if}. Since the function R_{if} is monotonic and single-valued in the region $\tau_n\leqslant\tau\leqslant\tau_{n+1}$, it follows that

$$\bar{i}_n(\omega_f;\omega_i,\theta)=\bar{I}_a(\omega_i)\int\limits_{R_{if}(\tau_n)}^{R_{if}(\tau_{n+1})}dR_{if}\frac{\gamma e^{-\gamma\tau}}{2\pi\Gamma(\tau,\theta)}\left(\frac{dR_{if}}{d\tau}\right)^{-1}$$
$$\times\exp\left\{-\frac{[\hbar(\omega_f-\omega(0,\theta))+2(R_{if}(0)-R_{if})]^2}{4\hbar^2\Gamma^2(\tau,\theta)}\right\}. \tag{17}$$

(The time τ in (17) is a function of R_{if}.)

In the case $\gamma\ll\Gamma_{ii}$, the significant values of R_{if} in (17) lie in the region

$$|\hbar(\omega_f-\omega(0,\theta))+2(R_{if}(0)-R_{if})|\lesssim\hbar\Gamma_{ff}.$$

This frequency region corresponds to a very narrow interval of the time ($|\tau-\tau_n|\lesssim\Gamma_{ii}^{-1}$), where τ_{fn} is the solution of the equation $\omega_f=\omega(\tau,\theta)$ and lies in the interval $\tau_n\leqslant\tau\leqslant\tau_{n+1}$. At such small changes of τ, the functions $e^{-\gamma\tau}$ and $\Gamma(\tau,\theta)$ in (17) can be regarded as constants equal to their values at the point τ_{fn}. On the other hand, the behavior of the function $(dR_{if}/d\tau)^{-1}$ depends substantially on the instant of time τ_{fn}.

If the acceleration of the nucleus at the instant τ_{fn} is large ($|\tau_n-\tau_{fn}|\varpi\sim1$ and $|\tau_{n+1}-\tau_{fn}|\varpi\sim1$), then $(dR_{if}/d\tau)^{-1}$ in the region of τ indicated above is in fact a constant. Conversely, in the case $|\tau_{fn}-\tau_n|\varpi\ll1$ or $|\tau_{fn}-\tau_{n+1}|\varpi\ll1$ this function changes appreciably after a time interval Γ_{ii}^{-1}, a fact that must be taken into account when calculating the integral in (17).

It is expedient to continue the analysis of the function $\bar{i}_n(\omega_f;\omega_i,\theta)$ separately of the spectrum sections where

$$\hbar|\omega_f-\omega(\tau_l,\theta)|\ll|R_{if}(\tau_l)|,\quad l=n,\ n+1 \tag{18a}$$

and for the region contained between the frequencies $\omega(\tau_n,\theta)$ and $\omega(\tau_{n+1},\theta)$ and defined by the inequalities

$$|R_{if}(\tau_n)|\lesssim\hbar|\omega_f-\omega(\tau_n,\theta)|\lesssim2|R_{if}(\tau_n)|-|R_{if}(\tau_{n+1})|,$$
$$(-1)^n\cos\theta[\omega(\tau_n,\theta)-\omega_f]>0. \tag{18b}$$

We consider first the last of these regions. The frequencies ω_f satisfying the inequalities (18b) correspond to instants of time τ_{fn} at which the acceleration of the nucleus is large. In this case, in accord with the reasoning presented above, the function $(dR_{if}/d\tau)^{-1}$ in (17) can be regarded as a constant. The integral in (17) is then easy to evaluate and we obtain

$$\bar{i}_n(\omega_f;\omega_i,\theta)\approx\bar{I}_a(\omega_i)\Phi_n(\omega_f;\omega_i,\theta), \tag{19}$$

where

$$\Phi_n(\omega_f;\omega_i,\theta)=\gamma e^{-\gamma\tau_{fn}}\left(\frac{dR_{if}}{d\tau}\bigg|_{\tau=\tau_{fn}}\right)^{-1}. \tag{20}$$

The function $\Phi_n(\omega_f;\omega_i,\theta)$ that describes the section of the spectrum $\bar{i}_n(\omega_f;\omega_i,\theta)$ defined by the inequalities (18b) is a smooth function. At $\gamma\lesssim\varpi$ it has near the frequency $\omega((\tau_{n+1}-\tau_n)/2,\theta)$ a relatively gently sloping minimum, and it increases monotonically towards the frequencies $\omega(\tau_n,\theta)$ and $\omega(\tau_{n+1},\theta)$.

The frequency regions defined by condition (18a) in the spectrum $\bar{i}_n(\omega_f;\omega_i,\theta)$ correspond to secondary particles emitted by the nucleus near the equilibrium position ($|\tau_{fn}-\tau_n|\varpi\ll1$ or $|\tau_{fn}-\tau_{n+1}|\varpi\ll1$). Using for R_{if} an approximate expression valid at $\varpi|\tau-\tau_n|\ll1$,

$$R_{if}(\tau)\approx R_{if}(\tau_n)-\tfrac{1}{2}A_n^{if}(\tau-\tau_n)^2, \tag{21}$$

we represent $\bar{i}_n(\omega_f;\omega_i,\theta)$ in these regions (at $\gamma\lesssim\varpi$) in the form

$$\bar{i}_n(\omega_f;\omega_i,\theta)\approx\bar{I}_a(\omega_i)F_n^{(-)}(\omega_f;\omega_i,\theta)\quad\text{at}\quad|\omega_f-\omega(\tau_n,\theta)|\ll|R_{if}(\tau_n)|/\hbar; \tag{22a}$$

$$\bar{i}_n(\omega_f;\omega_i,\theta)\approx \bar{I}_a(\omega_i)F_{n+1}^{(+)}(\omega_f;\omega_i,\theta)\quad \text{at}\quad |\omega_f-\omega(\tau_{n+1},\theta)|\ll|R_{if}(\tau_{n+1})|/\hbar. \tag{22b}$$

In formulas (21), (22a), and (22b) we have

$$A_n^{if}=-\frac{d^2R_{if}(\tau)}{d\tau^2}\bigg|_{\tau=\tau_n}, \tag{23}$$

$$F_n^{(\mp)}(\omega_f;\omega_i,\theta)=\frac{e^{-\gamma\tau_n}}{2a_n}\int_0^\infty\frac{d\xi}{\xi^{1/2}}\exp[-(\xi+c_n)^2]\exp(\mp b_n\xi^{1/2}), \tag{24}$$

$$a_n=\gamma^{-1}[2\pi|A_n^{if}|\Gamma(\tau_n,\theta)]^{1/2},\quad b_n=\gamma[2\Gamma(\tau_n,\theta)]^{1/2}|A_n^{if}|^{-1/2},\quad c_n=\frac{1}{2}\frac{(-1)^n\cos\theta}{|\cos\theta|}\frac{\omega_f-\omega(\tau_n,\theta)}{\Gamma(\tau_n,\theta)} \tag{25}$$

We note that in the case of weak anharmonicity

$$A_n^{if}=R_{if}(0)\frac{1}{3}\sum_p\omega_p^2|e(p)|^2\exp(-\gamma_p|\tau_n|)\cos\omega_p\tau_n, \tag{26}$$

so that in order of magnitude we have $|A_n^{if}|\sim\bar{\omega}^2|R_{if}(\tau_n)|$.

The functions $F_n^{(\mp)}(\omega_f;\omega_i,\theta)$ at $\gamma/\bar{\omega}\lesssim 1(b_n\ll 1)$ have a sharp maximum in the frequency region $|\omega_f-\omega(\tau_n,\theta)|\lesssim\Gamma(\tau_n,\theta)$ and have an appreciable asymmetry.[7] Namely, at $|c_n|\gg 1$ they attenuate exponentially towards $c_n>0$ like $\exp(-c_n^2)|c_n|^{-1/2}$ and towards $c_n<0$ more slowly, like $|c_n|^{-1/2}\exp(\mp b_n|c_n|^{1/2})$. (We note that far from the region $\hbar|\omega_f-\omega(\tau_n,\theta)|\ll|R_{if}(\tau_n)|$ of interest to us the function $F_n^{(+)}$ again increases at $c_n<0$.)

The general spectrum of the HL type is expressed in terms of the sum of the spectral functions $\bar{i}_n(\omega_f;\omega_i,\theta)$ considered above:

$$\bar{I}_{HL}(\omega_f;\omega_i,\theta)=\sum_n\bar{i}_n(\omega_f;\omega_i,\theta)\qquad(\tau_n\leqslant t_0). \tag{27}$$

According to the preceding arguments and formulas (19), (22a), (22b), and (27), the spectrum $\bar{I}_{HL}(\omega_f;\omega_i,\theta)$ takes at $\gamma t_0\ll 1$ the form of a series of sharp maxima against a smoother continuous background. The individual sections of the background are determined by the functions $\Phi_n(\omega_f;\omega_i,\theta)$, and each maximum is described by the function $F_n^{(-)}+\delta_{n0}F_n^{(+)}$ at a definite value of n.

The first maximum ($n=0$) of the spectrum $\bar{I}_{HL}(\omega_f;\omega_i,\theta)$ is located in the region $|\omega_f-\omega_{10}+(R_{ff}-2R_{if}(0))/\hbar|\lesssim\Gamma_{ff}$ and corresponds to secondary γ quanta or neutrons emitted directly after excitation of the nucleus (formation of the compound nucleus), when the acceleration of this nucleus is still relatively small. The remaining maxima correspond to later instants of the passage of the nucleus through the equilibrium position. The number and position of the maxima with $n\geqslant 1$ are determined by the detailed course of the vibrational relaxation, and their largest number should be expected if slowly damped localized oscillations are present in the crystal when $t_0>\bar{\omega}^{-1}$.

It should be noted that the position, width, and sign of the asymmetry of the individual maxima, as well as the width of the entire spectrum $\bar{I}_{HL}$ depend strongly on the scattering angle θ (Fig. 1). The dependence of the form of a spectrum of the HL type on the excitation frequency ω_i is quite weak. We point out here that the spectra of the secondary particles scattered through angles θ and $\pi-\theta$ are mirror images of each other relative to the point $\omega_f-R_{ff}/\hbar$. When the scattering angle tends to $\pi/2(\theta\to\pi/2)$ an appreciable narrowing of the spectrum $\bar{I}_{HL}$ is also observed.

In the case $t_0^{-1}\sim\gamma\lesssim\bar{\omega}$ the intensity of the maxima with $n\geqslant 1$ decreases exponentially. However, they remain asymmetrical in shape. The total width of the spectrum $\bar{I}_{HL}(\omega_f;\omega_i,\theta)$ also remains essentially constant, although there is some decrease in the intensity of the continuous background.

We point out that resonances with $\gamma\sim\bar{\omega}$ and $R_{ii}\gg\hbar\bar{\omega}$ are encountered both in scattering of γ quanta and among neutron resonances with energies $\hbar\omega_{10}\gtrsim 50$ eV in isotopes of the actinide group (U, Th, Pu, and others). By the same token, the spectra of the resonant scattering of neutrons by crystals containing tracer isotopes could be used to investigate the vibrational relaxation in such crystals.

In the case $\gamma\gg\bar{\omega}$ all that remains in the $\bar{I}_{HL}$ spectrum is the first maximum with $n=0$, and its width and asymmetry decrease rapidly with increasing γ (Fig. 2). In the limit $\gamma\gg(\Gamma_{ii}\bar{\omega})^{1/2}$ the spectrum $\bar{I}(\omega_f;\omega_i,\theta)$ assumes a Gaussian shape[5,8] and in the case $\gamma\gtrsim\Gamma_{ii}$ an important role is assumed in the formation of the spectrum by processes of emission prior to the damping of the phase correlation (it is no longer permissible to replace the finite limits of integration with respect to s' in (7) by $\pm\infty$). These processes are similar to Raman scattering in the secondary emission of the impurity center. On the whole, however, the spectrum $\bar{I}(\omega_f;\omega_i,\theta)$ at $\gamma\gg(\Gamma_{ii}\bar{\omega})^{1/2}$ practically coincides with the analogous spectrum for an ideal gas at a certain effective temperature.[8] This result is easily understood if it is recognized that at short lifetimes of the excited state of the nucleus its velocity has in fact no time to change.

In conclusion, we call attention to the fact that the above-noted properties of the spectrum $\bar{I}(\omega_f;\omega_i,\theta)$ are essentially preserved also in the case of excitation by a beam of neutrons or γ quanta with a wider frequency distribution than $\Delta_i\ll\Gamma_{ii}$. Even in the case $\Delta_i\gg\Gamma_{ii}$ (but $|\omega_{10}-\omega_i+R_{ii}/\hbar|\ll\Delta_i$) the spectrum $\bar{I}(\omega_f;\omega_i,\theta)$ in the entire region of the frequencies, with the exception of the far wings, is given at $\gamma\ll\Gamma_{ii}$ by an expression similar to (10):

$$\bar{I}(\omega_f,\theta)\approx I_i(0)\frac{\gamma^2}{2}\int_0^\infty d\tau\,e^{-\gamma\tau}\frac{\pi^{1/2}}{\Gamma_{ff}}\exp\left\{-\frac{[\omega_f-\omega_{10}+(R_{ff}-2R_{if}(\tau))]^2}{4\Gamma_{ff}^2}\right\}. \tag{28}$$

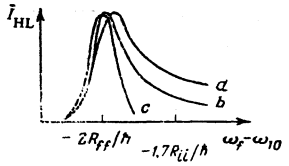

FIG. 2. Shape of the $n=0$ maximum of the HL spectrum at different lifetimes of the excited state of the nucleus: a) $b_0\ll 1$; b) $b_0=0.4$; c) $(R_{ii}/\hbar\omega)^{1/4}\gg b_0\gg 1$; $R_{ff}/\hbar=20\Gamma_{ff}$; the spectra are normalized to unity at the point of the maximum.

We emphasize also that an investigation of the resonant scattering of neutrons in the case of narrow resonances of width $\gamma \lesssim 0.05$ eV can yield a rather detailed information on the law governing the motion of the scattering nucleus. Similar investigations are of interest also for the study of radiation damage in a crystal as a result of knocking out a nucleus from the lattice site by the recoil momentum acquired by the nucleus. The resonant scattering spectrum is in this case also a spectrum of the HL type and can yield information on the stopping of the knocked-out nucleus. Strictly speaking, however, formulas (10)–(28) are not valid for this case.

1)In [7,8], use was made of the results of the theory of resonant secondary emission of impurity centers in crystals, developed in [9-11]. The use of the results of [9-11] is justified because of the analogy between vibrational relaxation processes that occur in the course of secondary emission of impurity centers, on the one hand, and incoherent resonant scattering of neutrons or γ quanta by crystals, on the other.

2)Generally speaking, expressions (1) and (6) describe the spectral distribution of the particles scattered only in incoherent fashion. However, since the multiphonon scattering is in fact completely incoherent, the function $\bar{I}(\omega_f;\ \omega_i,\ \theta)$ determines at $R_{ii} \gg \hbar\bar{\omega}$ with good accuracy the entire smoothed spectrum of the resonantly scattered neutrons or γ quanta.

It should also be noted that allowance for a number of factors which we are neglecting, such as the hyperfine interaction, the diffuse motion of the nuclei, or the increase of the nuclear mass on going to the excited state, can be significant only if the aforementioned narrow spectral lines are investigated. When the smoothed spectrum is considered, the influence of these factors becomes insignificant.

3)The pair-correlation approximation used to derive formula (3) is valid in general in the case of weak anharmonicity of the crystal-lattice vibrations. However, a direct analysis of expression (2) shows that in the case of small s and s' this approximation is applicable with respect to the correlator $K_{if}(\tau,\ s,\ s')$ at any value of the anharmonicity. In fact, the corrections to the pair-correlation approximation in the expression for $\ln K_{if}(\tau,\ s,\ s')$ turn out to be proportional at least to the third powers of s and s', and can be discarded at small s and s'.

4)We emphasize that formula (10) describes approximately the considered spectrum $\bar{I}(\omega_f;\ \omega_i, \theta)$ only when the main contribution to the integral with respect to τ is made by the region $\gamma^{-1} \gtrsim \tau \gg |s|,\ |s'|$, where the significant values are $|s|,\ |s'| \sim \Gamma_{ii}^{-1}$. Thus, the significant times τ greatly exceed the characteristic phase correlation times, which amount to $\sim\Gamma_{ii}^{-1}$.

5)We note that in the case of large recoil energy the characteristic time $t_0 (t_0 \geq \bar{\omega}^{-1})$ greatly exceeds the characteristic time $\sim\Gamma_{ii}^{-1} \sim (R_{ii}\bar{\omega}/\hbar)^{-1/2}$ of the damping of the phase correlation.

6)It is necessary to consider separately scattering through angles close to $\pi/2(|\theta - \pi/2 \lesssim (\hbar\bar{\omega}/R_{ii})^{1/2}$. Inasmuch as at $\theta \approx \pi/2$ we have $R_{if}(\tau) \approx 0$ and $\Gamma_{if}(\tau) \approx 0$ at all values of τ, in this case the difference between the processes of type OL and HL disappears and the entire spectrum $\bar{I}(\omega_f;\ \omega_i, \theta)$ is described, apart from corrections $\sim\gamma/\Gamma_{ii}$, by formula (14).

7)A plot of the function $2a_0F_0$ at $\gamma = 0$ is given in [12].

8)The similarity of the neutron or γ-quantum spectra resonantly scattered by an ideal gas and by crystals at large recoil energy was first pointed out in [6].

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