ON THE THEORY OF PASSAGE OF γ QUANTA THROUGH A RESONANT MEDIUM

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The refractive index of a resonant medium is studied with account of coherent multiple scattering of γ quanta by nuclei located in the lattice. A general formula is derived for the refractive index of Mössbauer γ quanta in polycrystals. In the short-wavelength range ($\lambda \ll a$, λ is the γ quantum wavelength and a is the interatomic distance), coherent multiple γ-quantum scattering in a polycrystal yields a contribution which is proportional to $(\lambda/a)^2$. The time dependence of the probability of passage of γ quanta through a resonant filter is calculated. The characteristics of the Mössbauer spectra are considered as functions of the delay time and these spectra are compared with the ordinary Mössbauer spectra.

THE time dependence of the probability of passage of γ quanta through a resonant filter was first investigated by Lynch, Holland, and Hammermesh[1] and by Wu and co-workers[2]. The results of these investigations are in good agreement with the known classical theory of dispersion, according to which the reaction of the medium to the γ quantum can be described by a certain macroscopic refractive index with a frequency dependence in the form

$$n(\omega)=\left[1+\frac{\alpha}{\omega^2-\omega_0^2+i\gamma\omega}\right]^{1/2}\approx 1+\frac{1}{2}\frac{\alpha}{\omega^2-\omega_0^2+i\gamma\omega}, \quad (1)$$

where ω_0 and γ are the resonant frequency and the damping coefficient of the classical oscillators, and α is a certain factor that depends on the number of resonant oscillators per unit volume. As noted in[3], the "unexpected success" of such a simple model for the region of wavelengths much shorter than the interatomic distance ($\lambda \ll a$) is due to the fact that the contributions of all the nuclei to the coherent forward scattering of the γ quanta, which is responsible for the change in the primary wave inside the crystal, are characterized by the same phase. It is obvious here that the discrete structure of the arrangement of the nuclei does not play any role.

We show in this paper that it is precisely only when $\lambda \ll a$ that this simple model is valid. When $\lambda \sim a$, allowance for multiple coherent scattering of the γ quantum by an aggregate of resonant nuclei located in the periodic structure of the crystal can lead to a significant deviation of the refractive index from the value given by (1). Such a collective effect was first considered in[4,5], for regular systems of identical nuclei (single crystals). In the present paper we consider the case of a polycrystal and obtain general expressions for the refractive index of polycrystalline media. In the short-wave limit $\lambda \ll a$, formula (1) coincides, accurate to $(\lambda/a)^2$, with the general formula obtained by us.

The refractive index of a resonant medium was investigated in γ-resonant experiments. In ordinary experiments on γ resonance, however, one measures only the imaginary part, which is proportional to the cross section for the resonant absorption of the γ quantum by the nucleus:

$$\sigma(E) = -(4\pi/\lambda N)\,\mathrm{Im}\,n(E); \quad (2)$$

Here E is the energy of the γ quantum and N is the density of the resonant nuclei in the medium. The total refractive index can be investigated by measuring the time dependence of the probability of passage of a γ quantum through a resonant filter. The second part of this paper is devoted to the calculation of this time dependence. The results of the calculation of the Mössbauer spectra as functions of the delay time are compared with the corresponding results for ordinary Mössbauer spectra. This reveals certain advantages of the γ-resonance procedure in conjunction with the delayed-coincidence technique.

1. In quantum theory, the refractive index of a medium is directly connected with the amplitude of coherent forward scattering of a γ quantum by a nucleus $a^{(0)}_{coh}(E)$:

$$n(E) = 1 + (\lambda^2/2\pi)\,\mathrm{N}a^{(0)}_{coh}(E). \quad (3)$$

For the γ transition between degenerate levels with spin I of the excited state and the spin J of the ground state we have

$$a^{(0)}_{coh}(E) = \frac{\lambda f}{8\pi}\frac{2I+1}{2J+1}\frac{\Gamma_R}{E-E_0+i\Gamma/2}, \quad (4)$$

where E_0 is the resonance energy, Γ_R and Γ are the radiative and total widths of the excited level, and f is the probability of recoilless resonant γ-quantum absorption. It is easy to write similar formulas also for other cases (for example, for polarized nuclei or γ quanta[6]). Substitution of (4) in (3) yields for the refractive index an expression that coincides with (1).

However, formulas (1) and (3) are approximate, since they do not take into account the possibility of excitation transfer from nucleus to nucleus when the γ quantum passes through the resonant medium. Such a collective effect, which is connected with multiple scattering of the γ quantum by an aggregate of resonant nuclei, plays an important role both in single crystals and in polycrystals[4-7]. Let us generalize formulas (1) and (3) with allowance for this collective effect.

To simplify the derivation, we neglect first the influence of the thermal oscillations of the lattice and assume that the spin of the ground state of the nucleus is equal to zero. Allowance for the thermal oscillations of

the lattice and for the spin of the ground state of the nucleus has already been considered in detail in[7]. Nor shall we consider the Rayleigh scattering of the γ quantum by the electron shells. This assumption is fully justified for many Mössbauer γ quanta at a high concentration of the resonant nuclei.

Let us consider the motion of a γ quantum with wave vector $\mathbf{k}_0$ and polarization σ_0 in a crystalline medium. Passing through the medium, the γ quantum causes resonant transitions. Let C_{si_s} be the amplitude of the state corresponding to excitation of only a nucleus at the site s, with excited-state spin projection i_s, and let $C_{k\sigma}$ be the amplitude of the state corresponding to the situation in which all the nuclei are in the ground state and there exists a photon $(\mathbf{k}, \sigma)$ with energy E_k. The amplitude of the state $C_{k\sigma}$ is equivalent to the wave function of the photon $(\mathbf{k}, \sigma)$. These amplitudes satisfy the system of coupled equations

$$(E-E_k)C_{k\sigma}(E)=\sum_s H^{k\sigma}_{si_s}C_{si_s}(E), \qquad (5)$$

$$\left(E-E_0+\frac{i\Gamma_c}{2}\right)C_{si_s}(E)=\sum_{k\sigma}H^{si}_{k\sigma}\,C_{k\sigma}(E),$$

where Γ_c is the conversion width of the excited level, $\Gamma_c + \Gamma_R = \Gamma$, $H^{si_s}_{k\sigma}$ is the matrix element of the γ transition in the crystal, which in our case is given by

$$H^{si_s}_{k\sigma}=(H^{k\sigma}_{si_s})^*=e^{i\mathbf{k}\mathbf{r}_s}M^{i_s}_{k\sigma}, \qquad (6)$$

$\mathbf{r}_s$ are the coordinates of the s-th nucleus, and $M^{i_s}_{k\sigma}$ is the matrix element of the isolated nucleus. The matrix elements (6) are connected with the parameters of the excited state of the nucleus by the relation

$$\sum_{k\sigma}\frac{H^{si}_{k\sigma}H^{k\sigma}_{si'}}{E_0-E_k+i\delta}=\left(-i\frac{\Gamma_R}{2}+\Delta E_R\right)\delta_{ii'}, \qquad (7)$$

where δ is an infinitesimally small real number and $\Delta E_R = E_0 - \tilde{E}_0$ denotes renormalization of the energy of the excited state, which is connected with the presence of the self-field of the γ radiation. It is useful to note the connection between the matrix elements (6) and the amplitude of the resonant scattering of the γ quantum by a nucleus

$$a_s(\mathbf{k}\sigma,\mathbf{k}'\sigma')=\frac{1}{\lambda c}\sum_{i_s}\frac{H^{k\sigma}_{si_s}H^{si_s}_{k'\sigma'}}{E_k-E_0+i\Gamma/2}. \qquad (8)$$

The solution of the system of Eqs. (5) for the wave function of the photon $(\mathbf{k}_0, \sigma_0)$ leads to the equation

$$(E-E_{k_0}-g)C_{k_0\sigma_0}(E)=0. \qquad (9)$$

Here (see formula (9) of[7])

$$g=\sum_{\substack{ss'\\ i_s i_{s'}}}H^{k_0\sigma_0}_{si_s}\left[\frac{\delta_{ss'}}{E-E_0+i\Gamma/2}+\frac{L_{i_s i_{s'}}}{(E-E_0+i\Gamma/2)^2}\right. \qquad (10)$$

$$\left.+\sum_{s''i_{s''}}\frac{L_{i_s i_{s''}}L_{i_{s''}i_{s'}}}{(E-E_0+i\Gamma/2)^3}+\ldots\right]H^{s'i_{s'}}_{k_0\sigma_0},$$

where

$$L_{i_s i_{s'}}=\sum_{\sigma,\,k\neq k_0}\frac{H^{\sigma i_s}_{k\sigma}H^{k\sigma}_{s'i_{s'}}}{E-E_k+i\delta}. \qquad (11)$$

From equation (9) follows directly a formula for the refractive index:

$$n=1-\lambda g/2\pi c. \qquad (12)$$

Formulas (9) and (10) are general for the problem of the passage of a photon through a resonant medium with allowance for multiple coherent scattering by the nuclei (with the exception of the case of the single crystal when the Bragg condition is satisfied[4]). In the individual approximation when single scattering is neglected, only the first term remains in formula (10). Taking (8) and (12) into account, it is easily seen that

$$n=1-\frac{\lambda^2}{2\pi}\sum_s a^{(0)}_{s(\mathrm{coh})}, \qquad (13)$$

where $a^{(0)}_{s(\mathrm{coh})}$ is the amplitude of the coherent resonant forward scattering of the γ quantum by the s-th nucleus. (In (13), the normalization volume of the crystal is assumed to be equal to unity.)

Formula (13) coincides with (3) if all the resonant nuclei are identical. Actually the amplitudes for scattering by different nuclei can be different. By way of an example we point to the case when the nuclei are in chemically nonequivalent positions, and therefore their resonant energies experience different chemical shifts. If it is assumed that the distributions of these resonant energies have a Lorentz form with a center of gravity E_0' and a half width Γ', then the summation over the nuclei in formula (13) gives for the refractive index exactly the same expression (3), with

$$a^{(0)}_{\mathrm{coh}}(E)=\frac{\lambda f}{8\pi}\frac{2I+1}{2J+1}\frac{\Gamma_R}{E-E_0'+i(\Gamma+\Gamma')/2}. \qquad (14)$$

Although the example in question has a purely illustrative character, formulas (3) and (14), as well as the expressions that can be derived from them for the cross section of the resonant absorption, can be usefully employed for an experimental study of the mechanism of broadening of Mössbauer spectra.

Let us return to the general equations (9) and (12). In the general case, it is necessary to sum all the terms of the series (10), which represent the contributions made to the refractive indices by the different processes of multiple scattering of a γ quantum by an aggregate of nuclei. It is important to emphasize here that in final analysis the contributions are made not by all the multiple-scattering processes, but only by their coherent part. It is seen therefore that interference of these coherent radiations on the periodic arrangement of the nuclei should strongly influence the refractive index of the crystal. The summation of the series (10) gives a formula analogous to (3), except that in place of the amplitude for coherent forward scattering by an isolated nucleus we have the amplitude for coherent forward scattering of the γ quantum by an aggregate of resonant nuclei, $A^{(0)}_{\mathrm{coh}}(E)$. If we take into account the spin of the ground state of the nucleus and the influence of the thermal motions of the lattice, then the summation of the series (9) leads to the expression

$$n(E)=1+\frac{\lambda^2}{2\pi}NA^{(0)}_{\mathrm{coh}}(E),\quad A^{(0)}_{\mathrm{coh}}(E)=\frac{\lambda f}{8\pi}\frac{2I+1}{2J+1}\frac{\Gamma_R}{E-E_0+i\Gamma/2+R}. \qquad (15)$$

The contribution of multiple scattering of a γ quantum by an aggregate of nuclei is expressed by the complex quantity R. We are interested in its imaginary part, which gives the correction to the decay constant of the isolated nucleus. The real part of this complex quantity,

which corresponds to a correction to the resonant energy E_0, is apparently very small and general expressions for it are given in[4,7]. For a single crystal, when the Bragg condition is not satisfied, we have[4,7]

$$\operatorname{Im} R = -\eta \frac{2I+1}{(2J+1)(2L+1)} \varphi \Gamma_R, \tag{16}$$

and for the polycrystal[7]

$$\operatorname{Im} R = \eta \frac{2I+1}{2J+1} \Gamma_R \varphi \left[\sum_{\nu=0}^{L} C_2(LL2\nu, 1-1) \times \sum_{\tau=\tau_{min}}^{k_0/\pi} \frac{\pi \beta_\tau F_\tau}{2V_0 \tau k_0^2} P_{2\nu}\left(1 - \frac{2\pi^2\tau^2}{k_0^2}\right) - \frac{1}{2L+1} \right]. \tag{17}$$

In (16) and (17) L is the multipolarity of the γ transition, η the concentration of the resonant nuclei in the crystal, $2\pi\tau$ the reciprocal-lattice vector, F_τ and β_τ the structure factor and the periodicity factor for the vector τ, V_0 the unit-cell volume, and φ a factor characterizing the incoherence due to the thermal motions of the lattice[4,6]. At high temperatures $\varphi = 0$, and for a rigid lattice at zero temperature $\varphi = 1$. In the short-wave limit ($\lambda \ll a$) formula (7) simplifies to:

$$\operatorname{Im} R = \frac{3}{8\pi} \eta \Gamma_R \varphi \frac{2I+1}{2J+1} \left(\frac{\lambda}{a}\right)^2. \tag{18}$$

Formula (18) can be used with high degree of accuracy already at $\lambda/a \lesssim 10$. Thus, when $\lambda \ll a$ we can use for the refractive index of a polycrystal the usual formula (3) or the equivalent formula of classical dispersion theory. For a single crystal, however, and also for a polycrystal at $\lambda \sim a$, the collective effect plays an important role and formulas (1) and (3) are not valid.

2. Let us calculate in this section of the paper the time dependence of the probability of passage of a γ quantum through a resonant filter. On passing through the resonant filter, the intensity of the γ quanta emitted from the source does not decrease with time exponentially[1]. The Mössbauer spectrum also depends on the delay time and differs considerably from the ordinary one[2]. The indicated time dependences can be calculated with the aid of a Fourier transformation, using the corresponding expressions for the refractive indices. We write the general expressions for the probability W(t, v) of passage of a γ quantum through a resonant filter:

$$W(t, v) = |S(t, v)|^2, \tag{19}$$

$$S(t, v) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{e^{-iEt/\hbar}}{E - E_0 + v + i\Gamma/2} \exp\left(i \frac{2\pi n(E) d}{\lambda}\right) dE. \tag{20}$$

Here d is the thickness of the resonant filter, t the delay time reckoned from the instant of formation of the excited state in the source, and v the energy shift (including also the artificial Doppler shift). It is easy to verify that for large energy shifts ($v \to \infty$), and also in the absence of a resonant filter (d = 0), formulas (19) and (20) give the usual exponential decay law

$$W(t, \infty) = e^{-\Gamma t/\hbar}. \tag{21}$$

In addition, from (20) we can arrive at the well-known expression for the ordinary Mössbauer spectrum observed without delay:

$$W(v) = \int_0^\infty w(t, v)\, dt = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{dE}{(E - E_0 + v)^2 + \Gamma^2/4} \exp\left(-\frac{4\pi \operatorname{Im} n(E) d}{\lambda}\right). \tag{22}$$

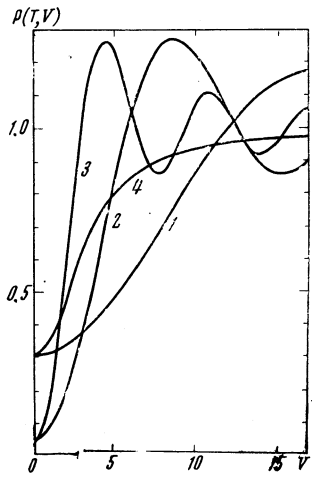

FIG. 1. Retarded Mossbauer spectra: 1–T = ½ (t = τ_n/2), 2–T = 1 (t = τ_n), 3–T = 2 (t = 2τ_n), 4–T = 0–∞ (ordinary Mossbauer spectrum). V–energy shift in units of Γ/2.

We shall calculate the function W(t, v) below for several concrete cases on the basis of formulas (19) and (20).

In the simplest case the refractive index takes the form (3). It is then convenient to rewrite (20) in the form

$$W(T, V) = |S(T, V)|^2, \tag{23}$$

$$S(T, V) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\exp(-ixT/2)}{x + V + i} \exp\left(\frac{iC/2}{x + i}\right) dx, \tag{24}$$

where

$$x = \frac{E - E_0}{\Gamma/2}, \quad V = \frac{v}{\Gamma/2}, \quad T = \frac{\Gamma}{\hbar} t = \frac{t}{\tau_n}, \quad C = \frac{\lambda^2}{2\pi} \frac{2I+1}{2J+1} \frac{\Gamma_R}{\Gamma} a$$

(τ_n is the lifetime of the excited state of the nucleus and C is the so-called effective thickness of the resonant filter). The integral (24) has been calculated in[1] and the results presented in the form of a series of Bessel functions.

Figure 1 shows the Mössbauer spectra for different delay times[1)] at C = 4. For convenience, the ordinates represent the relative transmission P(T, V) = W(T, V)/W(T, ∞), where in accordance with (21) we have $W(T, \infty) = e^{-T}$. (If we disregard the electron absorption, then W(T, ∞) is also the flux of γ quanta in the absence of the resonant filter.) As seen from Fig. 1, each Mössbauer spectrum breaks up into alternating intervals with normal and anomalous transmission. At energy shifts corresponding to the anomalous transmission, the flux of γ quanta emitted from the source after a certain time delay t is intensified by the presence of the resonant filter. This interesting fact was first observed by Wu and co-workers[2]. The modulation character of the Mössbauer is connected exclusively with the coherent property of the resonant γ quanta. Unlike the ordinary Mössbauer spectrum (22), the retarded Mössbauer spectra are constructed by coherent superposition of the wave functions of γ quanta passing through the resonant medium, and the modulation of the amplitude of the summary wave is connected here with the real part of the refractive index of the resonant medium.

[1)] Henceforth called the "retarded Mossbauer spectrum."

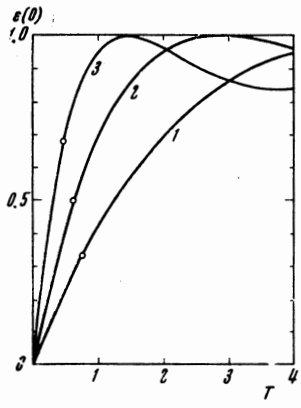

FIG. 2. Maximum absorption of γ-quantum flux as a function of the delay time: 1–C = 1, 2–C = 2, 3–C = 4 (C–effective thickness of filter); $T = t/\tau_n$.

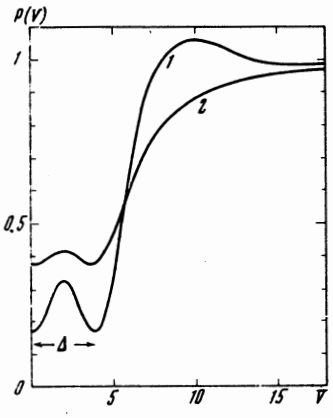

FIG. 3. Retarded (1) and ordinary (2) Mossbauer spectra in the presence of doublet splitting (C = 4, $\Delta = 4$): $1-1 < T < \infty$, $2-0 < T < \infty$.

Let us consider one of the characteristic parameters of the retarded Mössbauer spectrum—the value of the maximum absorption, determined by the relation

$$\varepsilon(0) = 1 - W(T, 0) / W(T, \infty) = 1 - P(T, 0). \qquad (25)$$

The integral (24) gives for the quantity $\epsilon(0)$ a simple analytic formula

$$\varepsilon(0) = 1 - J_0^2(\sqrt{\gamma CT}), \qquad (26)$$

where J_0 is a Bessel function of zero order. Figure 2 shows plots of $\epsilon(0)$ as functions of the delay time T at certain effective filter thicknesses. For comparison, the points of Fig. 2 show the corresponding values of $\epsilon(0)$ for the ordinary Mössbauer spectra, calculated in accordance with formula (23).

It follows from Figs. 1 and 2 that, starting with a certain value of the delay, the resonant line of the retarded Mössbauer spectrum becomes sharper than the resonant line of the ordinary Mössbauer spectrum. This means that with the aid of the technique of delayed coincidences it is possible to improve the resolution of the Mössbauer spectra[8]. To illustrate this conclusion, let us consider the case when doublet splitting is present in the Mössbauer spectrum. Starting from formula (13), we readily see that in place of formula (24) we have

$$S(T, V) = \int_{-\infty}^{\infty} \frac{e^{-ixT/2}}{x + V + i} \exp\left[i \frac{C}{4} \left(\frac{1}{x + i} + \frac{1}{x + \Delta + i} \right) \right] dx, \qquad (27)$$

where Δ is the splitting in units of $\Gamma/2$. Expression (27) is calculated by numerical integration[2)].

Figure 3 shows the results of a numerical calculation of the ordinary and retarded Mössbauer spectra for the delay interval $\tau_n < t < \infty$ ($1 < T < \infty$). From the experimental point of view, the choice of such a broad delay interval favors a simultaneous increase of both the resolving power of the doublet and the intensity of the incident γ-quantum flux[8].

The foregoing examples demonstrate the sensitivity of the retarded Mössbauer spectrum to the refractive index of the resonant medium. It is interesting to consider the extent to which the retarded Mössbauer spectra are sensitive to refractive-index changes due to the collective effect, especially in the case when such a change is small. In this case the retarded Mössbauer spectra can be calculated also from the general formulas (17) and (18), using expression (13) for the refractive index. The results of a numerical calculation show that the retarded Mössbauer spectra are similar to those shown in Fig. 1. Unfortunately, the sensitivity of these spectra to a small correction R in the expression for the refractive index (15) is approximately the same as the sensitivity of the ordinary Mössbauer spectra. Thus, investigations of the time dependence of the Mössbauer spectra offer no noticeable advantage for the observation of a certain small deviation of the refractive index from formula (1) in comparison with the ordinary γ-resonance method (without delay).

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[2)] Since the integrand in (27) converges poorly, in numerical integration it is convenient to replace the infinite limits of the integral by the sufficiently large quantities $\pm A (A \gg 1)$. The additional contribution of the two remaining integration intervals $(-\infty, -A)$ and $A, \infty)$ can easily be calculated analytically. A comparison of the values of the integral (27) calculated in this manner at $\Delta = V = 0$ with the analytic formula (26) reveals no discrepancies larger than 0.5%.

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