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POLARITON EFFECTS IN THE ONE-PHONON RESONANT SCATTERING OF MÖSSBAUER RADIATION

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Abstract. Polariton effects are considered in the case of one-phonon scattering of Mössbauer radiation in a resonant medium. It is shown that the polariton renormalization of group velocity of the initial and the scattered beam causes a gap in the spectrum of coherently scattered radiation. Such gap is centered at the resonant frequency ω_r and its width and shape depend on the acoustical properties of the scatterer. The properties of the vibrational modes, active in one-phonon coherent processes, and some aspects of the experimental investigation of the gap are also discussed.

Key words: Mössbauer effect, one-phonon scattering, polariton gap.

1. INTRODUCTION

Propagation of electromagnetic waves in resonant media is connected with the creation of polaritons, i.e. real elementary excitations of the system consisting of a transverse electromagnetic field and a resonant subsystem of the matter [1]. In the case of γ-radiation the corresponding renormalization of the phase velocity is very small ($|v_{ph}-c_0|/c_0 \ll 10^{-5}$, c_0 is the velocity of light in the vacuum). Hence, polariton effects are usually neglected in Mössbauer spectroscopy.

Nevertheless, as has been shown in [2, 3], the group velocity of nuclear polariton is strongly diminished in the resonance region ($v_g \lesssim 10^{-5}c_0$) due to the increase of the inert nuclear component of excitation. This phenomenon enables the observation of the formation and the propagation of polariton packets directly in time-of-flight Mössbauer experiments [2, 3].

In this paper, a polariton effect of another type is pointed out. To that end one-phonon resonant scattering of Mössbauer radiation in forward directions is considered. It is shown that polariton renormalization induces a gap in the spectrum of coherently scattered radiation in the spectral region where group velocity of polaritons becomes comparable to or is less than the velocity of sound in the scatterer medium ($v_g \lesssim c^s$). This polariton gap (PG) is centered on the scatterer resonant frequency ω_r and its width and shape depend on the frequency of the initial radiation, ω_i, and the acoustic properties of the sample. Also, the vibrational modes active in coherent one-phonon processes are different in the cases of resonant and non-resonant scattering. Thereby polariton effects appear in the spectral region, being available to common Mössbauer spectroscopy.

2. ONE-PHONON COHERENT SCATTERING

Let us consider the one-phonon scattering, i.e. the case where one phonon is created or absorbed during the scattering of a γ-quantum in condensed matter. Then in coherent processes conservation of both energy and quasi-momentum must take place so that

$$\vec{k}_f = \vec{k}_i \pm \vec{Q}_m, \qquad (1)$$
$$\omega_f = \omega_i \pm \Omega_m.$$

Here $\omega_{i(f)}$ is the frequency of the initial (scattered) radiation, $\vec{k}_{i(f)}$ is the wave vector of the initial (scattered) polariton, $\vec{Q}_m$ and Ω_m are the wave vector and the frequency, resp., of the created or the absorbed phonon in the m-th vibrational branch.

Formulae (1) fix a relationship between the scattering angle Θ ($\Theta = \widehat{\vec{k}_i, \vec{k}_f}$) and the frequency change Δ ($\Delta = \omega_f - \omega_i$). They also determine the wave vector and the frequency of the phonons involved in coherent scattering and corresponding to some part of the scattering spectrum. In this paper, a part of the spectrum is of interest which is available to the common methods of Mössbauer spectroscopy. Such "quasielastic" region ($\hbar|\Delta| \lesssim 10^{-7}$—$10^{-6}$ eV) corresponds to scattering in forward directions ($\Theta \lesssim 10''$), and acoustic long-wave vibrations ($\Omega \sim 10^2$—10^3 MHz) are the only active modes in one-phonon processes [4]. For these modes a simple dispersion relation is valid:

$$\Omega_m = c^s_m (\vec{Q}/Q) Q, \qquad (2)$$

where the sound velocity c^s_m varies with the direction of wave propagation rather slowly.

Conservation laws (1) enable the establishment of some features of the frequency spectrum of scattered radiation. A trivial example may be a gap in an one-phonon spectrum at the frequencies

$$|\omega_i - \omega_f| = |\Delta| \leqslant \Delta^D, \qquad (3)$$

where $\Delta^D = 2\pi c^s/L$, L is the characteristic dimension of the scatterer. Indeed, in a real scatterer the wavelength of acoustic modes is limited by the dimensions of the sample. Hence, the possible change of the radiation frequency cannot be arbitrarily small as fixed by inequality (3).

In this paper, another gap-type spectral peculiarity is considered, which arises in resonant coherent scattering only. For this purpose we will discuss the non-resonant and resonant cases separately in the following sections.

3. NON-RESONANT SCATTERING

In non-resonant cases the phase velocity of Mössbauer polaritons differs very slightly from c_0, so that it may be assumed that

$$k(\omega) \approx \omega / c_0 \qquad (4)$$

in formulae (1). Then, according to conservation laws (1) and dispersion relations (2) and (4), a continuous one-phonon spectrum of the scattered γ-quanta arises in the region $\hbar|\Delta| \lesssim 10^{-7}$ eV (if only the above-mentioned gap, induced by finite dimensions of the sample, is ignored).

Thereby, a one-to-one relationship exists between the scattering angle Θ and the frequency change Δ for each branch of acoustic vibrations (Fig. 1):

$$\Theta_m = 2 \arcsin[(|\Delta|/2\alpha_m\omega_i)(1-\alpha^2_m)^{1/2}], \tag{5}$$

where

$$\alpha_m = c^s_m(\vec{n})/c_0, \tag{6}$$

and the unit vector $\vec{n}$ fixes the direction of propagation of the acoustic wave involved in the scattering process $(\vec{n} = \vec{Q}_m/Q_m = (\vec{k}_f - \vec{k}_i)/|\vec{k}_f - \vec{k}_i|)$. Naturally, the vector $\vec{n}$ lies in the same plane as $\vec{k}_i$ and $\vec{k}_f$ whereby

$$\vartheta_m = \widehat{\vec{k}_i, \vec{Q}_m} \approx \pi/2 \tag{7}$$

is valid for the whole spectral region considered. In contrast to ϑ_m the values of the scattering angles Θ_m, determined by formula (5) are extremely small ($\ll 10''$).

As to vibrational branches, the longitudinal waves are the only vibrational modes which can be active in coherent one-phonon scattering processes in a non-resonant case. Indeed, as it is known, the amplitude of the one-phonon non-resonant scattering is proportional to $(\vec{k}_f - \vec{k}_i)\cdot\vec{e}_m$ where $\vec{e}_m$ is the polarization vector of the created or the absorbed phonon. For coherent processes it means that $(\vec{k}_i - \vec{k}_f)\cdot\vec{e}_m = \vec{Q}_m\cdot\vec{e}_m$ and hence the conclusion about active longitudinal modes is straightforward.

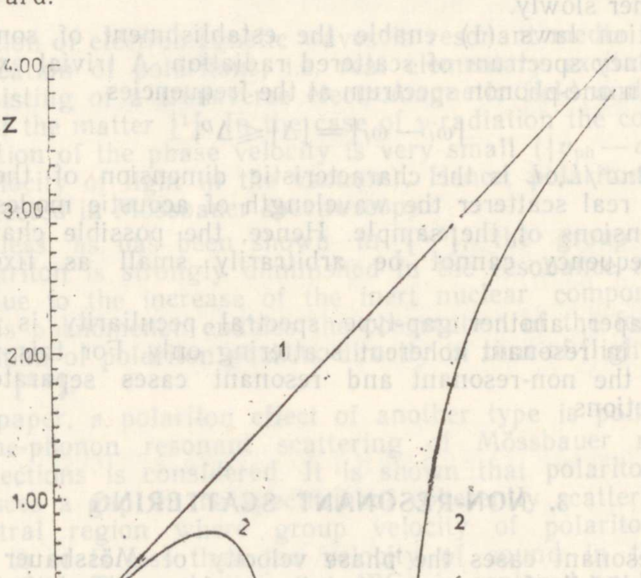

Fig. 1. Scattering angle as a function of frequency change, $Z = \Theta\alpha^{1/2}\omega_i/\omega_0$, $X = \Delta/\alpha^{1/2}\omega_0$, $\omega_r - \omega_i = 2\alpha^{1/2}\omega_0$; *1* — non-resonant case, *2* — resonant case. The frequency ω_0 is defined by formula (9).

4. RESONANT SCATTERING

As it was mentioned above, in a resonance case polaritons with a large nuclear component arise in the scatterer. Hence, the dispersion relation between its wave vector and frequency becomes more complicated [2, 5]:

$$k(\omega)=(\omega/c_0)\left[1+4\omega_0^2/[(\omega_r^2-\omega^2)+i\omega\Gamma]\right]^{1/2}. \quad (8)$$

Here ω_r and Γ are the frequency and the width of the Mössbauer absorption line in the scatterer and

$$\omega_0^2=(\pi c^3/\omega_r^2)\Gamma_0 n_c \exp(-2W)(2J+1)/2(2J_0+1), \quad (9)$$

where Γ_0 is the radiative width of the Mössbauer level, n_c is the density of resonant nuclei in the scatterer, J and J_0 are the spins of the excited and ground states of such nuclei and the Debye-Waller factor W determines the probability of zero-phonon transitions.

In formula (8) the resonant term is rather small [6]. Nevertheless, in the frequency region $|\omega-\omega_r|\leqslant\omega_0$ this term becomes important inasmuch as the polariton group velocity $d\omega/dk$ is considered. Thereby a remarkable diminishing of the group velocity at $|\omega-\omega_r|\leqslant\omega_0$ is caused by the increase of the inert nuclear component of polariton excitations. At the same time absorption effects can be neglected and the real wave vectors $\vec{k}$ can be assumed in the region $\Gamma\ll|\omega-\omega_r|<\omega_0$:

$$k\approx[\omega+\omega_0^2/(\omega_r-\omega)]/c_0. \quad (8a)$$

The polariton renormalization (8, 8a) of the wave vectors $\vec{k}_i$ and $\vec{k}_f$ essentially modifies the spectrum of coherently scattered radiation in the spectral region $\hbar|\omega_f-\omega_r|<\alpha_m^{1/2}\omega_0\sim10^{-7}$ eV. Indeed, let us consider one of the branches of acoustic vibrations in the scatterer. According to formula (1) the scattering angles Θ_m are given by the following expression:

$$\Theta_m=2\arcsin\{(|\Delta|/2\alpha_m\omega_i)[1-[\alpha_m\omega_0^2/(\omega_f-\omega_r)(\omega_i-\omega_r)]^2\}, \quad (10)$$

which now includes the resonant term. In contrast to non-resonant cases formula (10) does not fix one-to-one relationship of Θ_m and Δ (Fig. 1). Furthermore, in some spectral region around the resonant frequency ω_r any suitable solution for the scattering angle is absent altogether. It means that no phonons of appropriate frequency $(\Omega_m=|\Delta|)$ exist in the scatterer which could be active in coherent processes.

In the spectral distribution of coherently scattered radiation the lack of active phonons causes a gap at the frequencies

$$|\omega_f-\omega_r|\leqslant(\alpha_m\omega_0^2)/|\omega_i-\omega_r|=\Delta_m^P, \quad (11)$$

thus being a pure polariton effect. Such polariton gap (PG) is centered at the resonance frequency ω_r and its width Δ_m^P depends on both the initial frequency ω_i and the acoustical properties of the scatterer [7]. The most effective formation of PG takes place if the initial frequency has a moderate shift from the exact resonance ($\Gamma<|\omega_i-\omega_r|<\alpha_m^{1/2}\omega_0$, this aspect will be discussed in more detail in the next section). In this case the group velocity of initial polaritons is comparable to or is less than the corresponding sound velocity c_m^s.

Some remarks about the phonons active in coherent scattering can also be made on the basis of formulae (1). First, in the resonant case the direction of an active acoustic wave is given by the following expression:

$$\vartheta_m = \widehat{\vec{k}_i,\ \vec{Q}_m} \approx \arccos[\alpha_m + (\alpha_m \omega_0^2)/(\omega_r - \omega_i)(\omega_r - \omega_f)]. \qquad (12)$$

As can be seen in Fig. 2., in the spectral region where polariton effects become negligible $(|\omega_r - \omega_f| \gg (\alpha_m \omega_0^2)/|\omega_i - \omega_r|)$, $\vartheta_m \approx \pi/2$ and the active acoustic wave propagates perpendicularly to the initial beam of radiation, i.e. in the same way as in non-resonant cases. However, in the gap region $(|\omega_f - \omega_r| \sim (\alpha_m \omega_0^2)/|\omega_i - \omega_r|)\, \vartheta_m$ varies rapidly from zero to $\pi/2$ and from π to $\pi/2$.

Further, considering that in the resonant case the one-phonon scattering amplitude is proportional to $\vec{k}_i \cdot \vec{e}_m$ or $\vec{k}_f \cdot \vec{e}_m$, then outside the PG region, according to (12), only transverse acoustic modes are effectively involved in coherent scattering processes. On the contrary, in the vicinity of the PG region ($|\omega_f - \omega_r| \geqslant \Delta_l$, Δ_l is the width of the particular PG connected with the longitudinal branch of vibrations) longitudinal acoustic waves become active.

Thus in real situations, when all three acoustic branches participate in coherent processes, a complete spectral gap arises which is composed by a particular PG given by formula (11). As before, such spectral peculiarity is centered at ω_r but its shape can be rather complicated, especially in acoustically anisotropic media.

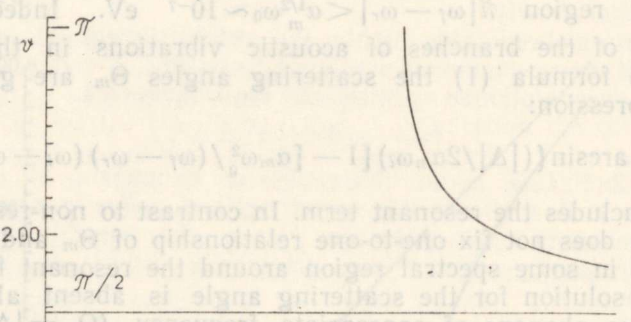

Fig. 2. Direction of propagation of the active acoustic wave as a function of frequency change, $\vartheta = \widehat{\vec{k}_i,\ \vec{Q}}$, $X = \Delta/\alpha^{1/2}\omega_0$, $\omega_r - \omega_i = 2\alpha^{1/2}\omega_0$.

5. POLARITON GAP

In this section, some general characteristics of the polariton gap will be considered and some aspects of its experimental investigation will be discussed. First it can be pointed out that according to formula (11) the width of the PG Δ^P increases with the decrease of $|\omega_i - \omega_r|$ and becomes infinite at exact resonance. Such unrealistic increase is a result of the assumption about negligible absorption and entirely real wave vectors $\vec{k}_i$ and $\vec{k}_f$. In reality, the complex polariton wave vectors mean an approximate character of quasi-momentum conservation law, especially in the exact resonance case. Then, as a result, the polariton gap turns out to be totally smeared and a smooth spectrum of coherently scattered radiation arises if $|\omega_i - \omega_r| \lesssim \Gamma$.

On the contrary, if the excitation of resonant nuclei takes place outside the Mössbauer absorption line, ω_i being sufficiently close to ω_r $(\Gamma \ll |\omega_i - \omega_r| \sim \alpha_m^{1/2}\omega_0 \ll \alpha_m^{1/2}\omega_0^2/\Gamma)$, absorption has rather moderate influence on the PG. The most significant effect is the smearing of the sharp edges of the gap, whereby the width of the smooth edges will be $\sim\Gamma$ $(\Gamma \ll \Delta^P \sim \alpha_m^{1/2}\omega_0)$. Analogous changes in the scattering spectrum are caused by the final spectral width of the initial Mössbauer radiation.

Thus it may be concluded that the most convenient spectral region to observe polariton effects is $\Gamma < |\omega_{i(f)} - \omega_r| < \alpha^{1/2}\omega_0$. This frequency region $(\sim 10^{-7}$—10^{-6} eV in ^{57}Fe case) is quite available for contemporary Mössbauer spectrometry. Thereby the demands on spectral resolution are much lower than in common Mössbauer experiments. It must also be pointed out that extremely small values of Θ_m exclude any methods based on the analysis of angular characteristics of scattered radiation.

The greatest obstacle in detecting PG is the low intensity of the scattered radiation, caused by a small number of low-frequency phonons in matter in usual conditions. Therefore, the problem of the background becomes of great importance. A part of this background, connected with incoherent and multi-phonon scattering, may be relatively weak if one--phonon scattering dominates and collimated beams of radiation are used. Then in the actual spectral region $(\Gamma < |\omega_f - \omega_r| < \alpha^{1/2}\omega_0)$ the background consists mainly of the γ-quanta directly transmitted through the scatterer. Although the PG usually does not coincide with the zero--phonon line of the source $(|\omega_i - \omega_r| \gg \Gamma$ in the PG region), in real cases the intensity of the initial Mössbauer radiation is rather significant at the frequencies correponding to the PG. In addition, the separation of the transmitted and the scattered radiation by their directions is practically impossible here.

Nevertheless, in sufficiently thick scatterers the signal/background ratio may become reasonable. Indeed, if the zero-phonon line of the source lies in the region $|\omega_r - \omega_i| > \alpha^{1/2}\omega_0$, then the PG arises at the frequencies $|\omega_r - \omega_f| < \alpha^{1/2}\omega_0$, i.e. in the region where absorption is rather strong. In these conditions the distance passed by the scattered polaritons in the medium is significantly shorter than the sample thickness and the probability of γ-quanta being absorbed in the scatterer is much less than in the case of direct transmission. As a result, in a thick scatterer the background is supressed by the scatterer itself. However, the strong non-resonant absorption limits the maximal thicknesses which could be used in the experiment. In addition, a significant distortion of spectral distribution of the scattered γ-quanta, caused by resonant absorption, must be taken into account. In this aspect the registration of the excitation spectrum instead of the scattering spectrum may be more convenient.

It may be concluded that the discovery and the investigation of the PG in the spectrum of the scattered Mössbauer radiation require rigid experimental conditions. The situation may become much better if ultrasonic waves of the frequency ~ 100 MHz and of a considerable intensity will be excited in the scatterer by some external source.

COMMENTS AND REFERENCES

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4. For this reason possible localization effects of vibrational modes are ignored in this paper. Also the Umklapp processes are not analyzed here.
5. In this paper a negligible hyperfine structure of Mössbauer levels and an isotropic tensor of dielectric permittivity are assumed.
6. In the case of ^{57}Fe nucleus $|c_0k(\omega)-\omega|/\omega<10^{-5}$ even at $|\omega_r-\omega|\sim\Gamma$.
7. Hence PG differs strictly from the spectral gap discussed in section 2. The latter is centered at the frequency ω_i whereby its width depends on the dimensions of the scatterer only. Thereby the reason for such spectral peculiarity is the absence of phonons of an appropriate frequency in the sample. Therefore this gap can arise in both coherent and incoherent scattering in resonant as well as in non-resonant cases. On the contrary, in the case of PG the phonons of an appropriate frequency are present in the scatterer, but they cannot be active in coherent processes.

POLARITONEFEKTID MÖSSBAUERI KIIRGUSE ÜHEFONOONSEL RESONANTSHAJUMISEL

Mati HAAS

On vaadeldud polaritonefekte Mössbaueri kiirguse ühefonoonse hajumise korral resonantses keskkonnas. On näidatud, et polaritonide grupikiiruse renormeerimine tekitab koherentselt hajunud kiirguse spektris pilu, mille keskpunktiks on resonantssagedus ω_r ja mille laius ning kuju sõltuvad hajutaja akustilistest omadustest. On vaadeldud ka aktiivsete võnkemoodide omadusi koherentsetes protsessides ja polaritonpilu eksperimentaalseks uurimiseks vajalikke tingimusi.

ПОЛЯРИТОННЫЕ ЭФФЕКТЫ В ОДНОФОНОННОМ РЕЗОНАНСНОМ РАССЕЯНИИ МЁССБАУЕРОВСКОГО ИЗЛУЧЕНИЯ

Мати ХААС

Рассмотрены поляритонные эффекты, возникающие при однофононном рассеянии Мёссбауеровского излучения в резонансной среде. Показано, что поляритонная перенормировка групповой скорости падающего и рассеянного пучка приводит к образованию щели в спектре когерентно рассеянного излучения. Центром такой щели является резонансная частота ω_r, а ее ширина и форма зависят от акустических свойств рассеивателя. Обсуждены также свойства колебательных мод, активных в однофононных когерентных процессах, и условия для экспериментального изучения поляритонной щели.