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Corresponding Member of the Academy of Sciences of the USSR E. F. GROSS, S. A. PERMOGOROV, B. S. RAZBIRIN

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Abstract

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AN OPTICAL ANALOG OF THE MÖSSBAUER EFFECT

In the spectra of a number of crystals (CdS, CdSe, ZnS, ZnTe, CdTe, GaP, HgJ_2, etc.) near the edge of the fundamental absorption at 4° K, narrow absorption and emission lines are observed ([1–5]). The width of these lines is of the order of 10^{-4} eV, i.e., close to the width of atomic lines. A characteristic feature of the spectrum of narrow lines is the existence among them of ultranarrow resonance lines (the emission lines coincide with the absorption lines to an accuracy of 10^{-5} eV). Each resonance line in absorption and emission is accompanied by several "phonon repetitions,"arising upon the emission or absorption of a photon and the simultaneous excitation of phonons, and together with them forms vibrational series. The vibrational series in absorption and emission are mirror-symmetric with respect to the resonance lines ([4,5]). Two types of electron-vibrational spectra are observed, which are presented for the case of luminescence of a crystal at 4° K in Fig. 1. One of the spectra (Fig. 1*a*) arises as a result of the interaction of the electronic transition only with the optical phonons of the crystal; the other (Fig. 1*b*)—with both optical and acoustic phonons of the lattice.

A number of works have established ([3,6,7,4]) that the appearance of ultranarrow resonance lines must be associated with the formation of exciton states near defects of the crystal. Some authors ([6,7]) consider such exciton states as complexes formed from an exciton and a defect (bound excitons). Such "immobile" exciton complexes are a kind of impurity center in the lattice, and, apparently, the treatment used in the theories of absorption and luminescence spectra of impurity centers ([8,9]) can be applied to them. However, these theories give a good explanation of broad electron-vibrational bands and of Levinson' s law of mirror symmetry, and do not explain the appearance and properties of ultranarrow resonance lines corresponding to a purely electronic transition, without interaction with lattice phonons.

The phononless transition, unusual within the framework of the former theories of absorption and luminescence of crystals and until recently unknown in the optical spectra of crystals, is very similar in its external aspect to the appearance of narrow lines in γ-spectra—the Mössbauer phenomenon ([10]). As E. D. Trifonov ([11]) first showed, this similarity is not accidental and purely external, but has a deeper basis. The similarity between the theory of the Mössbauer effect and

Fig. 1. Schematic representation of the electron-vibrational emission spectra of a CdS crystal at $T = 4.2^\circ$ K for the case of interaction only with optical phonons (a) and for the case of interaction with optical and acoustic phonons (b)

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the theory of the optical spectra of an impurity center is a consequence of the invariance of the oscillator energy $\frac{1}{2}\hbar\omega(p^2 + q^2)$ with respect to the replacement of momentum by coordinate. But the physical processes of interaction with the lattice in γ-spectra and optical spectra are, of course, different.

Transitions between nuclear levels upon the emission or absorption of a γ quantum hardly affect the outer electronic shell of the atom, do not change its configuration, and for this reason displacements of the equilibrium positions of nuclei in the crystal lattice during such transitions may be neglected. The displacement of the center of mass of the nucleus and the associated change in the vibrational state of the crystal are here caused by recoil—the transfer to the nucleus of the momentum of the γ quantum upon its emission. Conversely, in the optical spectra of impurity centers the recoil effect $\Delta p = E/c$ is very small owing to the smallness of the energy E of optical transitions,

and it may be neglected. But, in contrast to γ transitions in the nucleus, optical transitions are associated with a substantial change in the configuration of the outer electron shell. Therefore they can lead to appreciable changes in the equilibrium positions of the nuclei and, as a consequence, to interaction with vibrations of the crystal lattice.

Both phenomena—the optical spectra of impurity centers and the γ spectra of nuclei embedded in a crystal lattice—have a common feature, namely: the existence, alongside a spectrum caused by interaction with lattice vibrations, of a very narrow resonant "phononless" line. It is interesting to note that the shape of a mirror-symmetric optical spectrum is very similar to the shape that the γ spectrum of a nucleus in a crystal should have. All the above prompted us to consider the low-temperature spectrum of ultranarrow resonant lines in a crystal as an optical analogue of the Mössbauer effect (4). It was of interest to determine how far and how deep this analogy extends.

Fig. 1. Schematic representation of the electron-vibrational emission spectra of a CdS crystal at $T = 4.2^\circ$ K for the case of interaction only with optical phonons (a) and for the case of interaction with optical and acoustic phonons (b).

As is known from experiments on the absorption of γ quanta, the Mössbauer resonance line is very sensitive to an increase in temperature. Its intensity

decreases sharply, but the line width does not change appreciably. At present there are a number of theoretical works describing the temperature dependence of the Mössbauer line (14, 15) and agreeing with experiment (10, 16).

On the other hand, theoretical works devoted to the optical spectra of crystals (11-13) have shown that, with increasing temperature, the "phononless" line, analogous to the Mössbauer line, likewise should not broaden, while its intensity decreases, and the faster the smaller the mean energy of the phonons with which the electronic transition interacts—a result coinciding with the theoretical description of the temperature dependence of the Mössbauer line.

It was therefore of interest to study experimentally the temperature dependence of the intensity of ultranarrow "phononless"lines in optical spectra, to compare it with the results of theory, and to compare it with the temperature dependence of the Mössbauer line. To this end we investigated the luminescence of CdS single crystals in the region of narrow lines of bound excitons ($\lambda\lambda$ 4860-5100 Å) and studied the temperature dependence of the ratio of the intensity of the "phononless" lines to the intensity of their "phonon repetitions" *, as is done in theoretical works and in experiments with

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* When the temperature of a CdS crystal is raised from 4 to 77° K, in addition to the redistribution of intensity between the lines studied by us, there is also an overall decrease in the intensity of the emission, the same both for the "phononless" lines and for their "phonon repetitions." Such a decrease is apparently connected both with an increase in the probability of nonradiative transitions and with dissociation of the luminescing complexes as a result of thermal detachment of the exciton from the defect, and has no place in the Mössbauer effect, where the lifetime of nuclei in an excited state does not depend on temperature. In the case of measuring the relative intensities of the lines, the overall decrease in intensity is automatically excluded, and we are able to make a comparison with the Mössbauer effect.

phenomenon. We selected for investigation the ultra-narrow intense lines λ4888.6 Å and λ4869 Å and their "phonon repetitions."

The line λ4888.6 Å corresponds to a purely electronic transition, alongside which its combinations with both optical and acoustic phonons are clearly observed (see Fig. 1b). Interaction with acoustic phonons appears in the emission spectrum as a narrow continuous band located next to the line λ4888.6 Å, on the long-wavelength side of it. The width of this "acoustic band" is 5 Å, and its maximum is shifted relative to the "phononless" line by 1.6 Å (Fig. 2). The small width of the "acoustic band" shows that the electronic transition here interacts with the long-wavelength portion of the acoustic branch.

Fig. 2. Change with temperature of the line λ4888.6 Å and the band caused by interaction with acoustic phonons

The second line, λ4869 Å, also corresponds to a phononless transition. However,

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Figure 2: Fig. 2. Change with temperature of the line λ4888.6 Å and the band caused by interaction with acoustic phonons

this transition, unlike the preceding one, is not accompanied by combinations with acoustic phonons, but combines only with optical phonons (Fig. [[unclear: figure number]], *a*).

The temperature behavior of the lines under interaction with acoustic phonons turned out to be different from that under interaction with optical phonons.

1. **Case of interaction with acoustic phonons.** Figure 2 shows the change with temperature of the "phononless" emission line λ4888.6 Å and of the band caused by interaction with acoustic vibrations. As is evident from the figure, the relative intensity of the line λ4888.6 Å is very sensitive to temperature changes. The line, without changing its width, rapidly weakens as the temperature is raised from 4°K, and already at 26°K it is barely distinguishable against the background of the "acoustic band." Changes also occur in the "acoustic band." The part of the band situated on the short-wavelength side of the "phononless" line increases considerably. This part is associated with electron-vibrational transitions in which the energy of a phonon from the lattice is added to the photon energy. Such temperature behavior of the "phononless" resonance line and of the "acoustic band" is in complete agreement with the predictions of theo-

by E. D. Trifonov and K. K. Rebane [11,12], and agrees with the temperature dependence of the resonance line in the Mössbauer effect.

Fig. 3 graphically represents the change with temperature of the ratio of the intensity of the "phononless"line to the total intensity of the line and the "acoustic band." The form of the curve of the decrease in relative intensity is similar to that of the curve for the temperature decrease of the relative intensity of the Mössbauer line [10], with the difference that in the case of the optical spectrum the sharp decrease occurs in a range of much lower temperatures than in the Mössbauer effect. This difference can be understood if one takes into account that a sharp decrease in intensity should occur at the temperature at which the energy of thermal vibrations (kT) reaches the mean energy of the phonon interacting with the transition. In our case the optical transition interacts with the long-wavelength region of the acoustic branch (phonons of small energies). In the case of the Mössbauer experiment [10], however, it follows from Visscher' s theoretical consideration [14] that the nuclear transition apparently interacts with the entire acoustic branch.

2. The case of interaction with optical phonons.
When the temperature is raised from 4 to 77°K, the relative intensity of the line

Fig. 3

Figure 3: Fig. 3

λ4869 Å does not change. At 77°K this line is still rather intense and sharp. This insensitivity of the relative intensity of the line λ4869 Å to an increase in temperature is connected with the fact that the corresponding electronic transition interacts only with optical lattice vibrations, whose characteristic temperature is high. A similar insensitivity of the Mössbauer line to an increase in temperature, in the case when the γ-transition interacts with optical phonons, was observed in the γ-spectra of radioactive tin [16]. Thus, also in the case of interaction with optical phonons an analogy is observed between the optical spectrum and the Mössbauer effect.

Fig. 3. Graph of the change in the relative intensity of the line λ4888.6 Å with increasing temperature. S is the ratio of the intensity of the line to the total intensity of the line and the "acoustic band."

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Physical-Technical Institute
named after A. F. Ioffe
Academy of Sciences of the USSR

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Note: Figure translations are in progress. See original paper for figures.