HYPERFINE STRUCTURE OF THE RAYLEIGH LINE OF LIGHT SCATTERING IN A PARAMAGNET

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Abstract

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HYPERFINE STRUCTURE OF THE RAYLEIGH LINE OF LIGHT SCATTERING IN A PARAMAGNET

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1. Light scattering in a paramagnetic crystal must have features associated with the fact that the splittings of the spin levels of paramagnetic ions are of the same order of magnitude as the energy of the phonons participating in the scattering. Under resonant conditions the presence of spin-phonon interaction can appreciably change the spectrum of lattice vibrations, which will also lead to a change in light scattering. The question of the influence of spin-phonon interaction on the sound velocity was discussed earlier ([1]). In ([2]) the shift of the components of the fine structure of the Rayleigh scattering line in a paramagnet was considered. In the present article we shall show that, under certain conditions, an additional splitting of each component should arise.

2. The intensities of the Stokes and anti-Stokes lines of light scattering by phonons are determined by the Fourier transforms of the statistical averages $\langle b^+_{q\lambda}(t) b_{q\lambda}(0)\rangle$ and $\langle b_{q\lambda}(t) b^+_{q\lambda}(0)\rangle$, respectively; here $b_{q\lambda}(t)$ and $b^+_{q\lambda}(t)$ are the annihilation and creation operators of a phonon with wave vector $\mathbf{q}$ and polarization λ in the Heisenberg representation. Our problem is to calculate these quantities in the presence of interaction between the spin system and the phonons in a paramagnet. The Hamiltonian of the system may be written in the form

$$\mathcal{H} = \mathcal{H}_c + \mathcal{H}_\phi + \mathcal{H}_{c\phi};$$

$$\mathcal{H}_c = \sum_{jm} E_m P^j_{mm}, \qquad \mathcal{H}_\phi = \sum_{q\lambda} \hbar\omega^{(0)}_{q\lambda} b^+_{q\lambda} b_{q\lambda}, \tag{1}$$

$$\mathcal{H}_{c\phi} = i \sum_{jmn} \sum_{q\lambda} q \sqrt{\frac{\hbar}{2M\omega_{q\lambda}}}\, G^{\chi\lambda}_{mn} P^j_{mn} \left(b_{q\lambda} e^{i\mathbf{q}\mathbf{r}_j} - b^+_{q\lambda} e^{-i\mathbf{q}\mathbf{r}_j} \right),$$

where $\mathcal{H}_c$, $\mathcal{H}_\phi$, and $\mathcal{H}_{c\phi}$ are the Hamiltonians of the spin system, the phonons, and the spin-phonon interaction, respectively; E_m is the energy of the m-th spin level; P^j_{mn} is the one-particle static operator in the energy representation; $\mathbf{r}_j$ is the coordinate of the j-th paramagnetic ion; $\omega^{(0)}_{q\lambda}$ is the frequency of a free phonon; $\chi = \mathbf{q}/q$; M is the mass of the crystal; $G^{\chi\lambda}_{mn}$ is a constant characterizing the magnitude of the spin-phonon interaction. We shall find the correlation functions of the phonons through Green' s functions of the type

$$D_{q\lambda}(t-t') = i\theta(t-t')\langle b_{q\lambda}(t)b^+_{q\lambda}(t') - b^+_{q\lambda}(t')b_{q\lambda}(t)\rangle = \langle\langle b_{q\lambda} \mid b^+_{q\lambda}\rangle\rangle,$$

where $\theta(t-t') = 0$ for $t < t'$ and $\theta(t-t') = 1$ for $t > t'$. Writing out the equations for them in the usual way (see, for example, ([3])) and passing to Fourier transforms with respect to time, we truncate the chain of coupled equations on the basis of the smallness of the spin-phonon interaction:

$$\left\langle\!\!\left\langle \left(G^{\chi'\lambda'}_{nk} P^j_{mk} - G^{\chi'\lambda'}_{km} P^j_{kn}\right)\left(b_{q'\lambda'}e^{i\mathbf{q}'\mathbf{r}_j} - b^+_{q'\lambda'}e^{-i\mathbf{q}'\mathbf{r}_j}\right) \mid b^+_{q\lambda}\right\rangle\!\!\right\rangle_\omega \simeq$$

$$\simeq \delta_{q\mathbf{q}'}\delta_{\lambda\lambda'}G^{\chi\lambda}_{nm}\langle P_{mm}\delta_{km} - P_{nn}\delta_{kn}\rangle e^{i\mathbf{q}\mathbf{r}_j}D_{q\lambda}(\omega). \qquad (2)$$

As a result we obtain

$$\left\{\omega - \omega^{(0)}_{q\lambda} - q\sum_{mn}\frac{A^{\vec{\varkappa}\lambda}_{mn}}{\omega - \omega_{nm}}\right\} D_{q\lambda}(\omega) = -\frac{1}{2\pi},$$

$$A^{\vec{\varkappa}\lambda}_{mn} = \frac{N}{2\hbar\rho v_\lambda}\left|G^{\vec{\varkappa}\lambda}_{mn}\right|^2(\eta_m - \eta_n). \qquad (3)$$

Here N is the number of paramagnetic ions per unit volume; $\eta_m = \langle P_{mm}\rangle$ is the particle distribution function over spin levels; ρ is the density of the crystal; v_λ is the "seed" speed of sound. Let us consider the case in which $\omega^{(0)}_{q\lambda}$ is close to one of the frequencies $\omega_{nm} = \Omega$. Then in the sum (3) one may restrict oneself to a single term, after which the poles of the function $D_{q\lambda}(\omega)$ are readily found:

$$\omega^{(1,2)}_{q\lambda} = \frac{1}{2}\left\{\omega^{(0)}_{q\lambda} + \Omega \pm \sqrt{\left(\omega^{(0)}_{q\lambda} - \Omega\right)^2 + 4qA^{\vec{\varkappa}\lambda}_{\Omega}}\right\}. \qquad (4)$$

Next, by finding the spectral function from the imaginary part of $D_{q\lambda}(\omega)$, we obtain in the usual way for the desired quantities:

$$\langle b^+_{q\lambda}b_{q\lambda}\rangle_{\omega-\omega_0} = \frac{n(\omega-\omega_0)}{\omega^{(1)}_{q\lambda} - \omega^{(2)}_{q\lambda}}\left\{(\omega^{(1)}_{q\lambda} - \Omega)\,\delta(\omega - \omega_0 - \omega^{(1)}_{q\lambda}) - (\omega^{(2)}_{q\lambda} - \Omega)\,\delta(\omega - \omega_0 - \omega^{(2)}_{q\lambda})\right\},$$

$$\langle b_{q\lambda} b^{+}_{q\lambda} \rangle_{\omega-\omega_0} = \frac{n(\omega_0 - \omega) + 1}{\omega^{(1)}_{q\lambda} - \omega^{(2)}_{q\lambda}} \left\{ (\omega^{(1)}_{q\lambda} - \Omega)\, \delta(\omega - \omega_0 + \omega^{(1)}_{q\lambda}) - (\omega^{(2)}_{q\lambda} - \Omega)\, \delta(\omega - \omega_0 + \omega^{(2)}_{q\lambda}) \right\},$$

$$n(x) = \left(\exp \frac{\hbar x}{kT} - 1 \right)^{-1}, \tag{5}$$

where ω_0 is the frequency of the incident light. It follows from this that each component of the fine structure consists of two lines separated by the interval

$$\delta\omega = \sqrt{\left(\omega^{(0)}_{q\lambda} - \Omega\right)^2 + 4qA^{\vec{\varkappa}\lambda}_{\Omega}}. \tag{6}$$

The intensity of the lines corresponding to the region far from resonance decreases sharply (proportionally to $2qA^{\vec{\varkappa}\lambda}_{\Omega}/\Omega^2$ for $\omega^{(0)}_{q\lambda} \ll \Omega$). For $\omega^{(0)}_{q\lambda} = \Omega$, the two lines separated by the interval $\delta\omega = 2\sqrt{qA^{\vec{\varkappa}\lambda}_{\Omega}}$ have the same intensity.

The results obtained have the following meaning: owing to the spin-phonon interaction, oscillations arise in the system that are neither purely lattice nor spin oscillations. The greatest mixing occurs in the region where the frequencies of the free oscillations coincide. In the case when the width of the levels proves to be greater than $2\sqrt{qA^{\vec{\varkappa}\lambda}_{\Omega}}$, the ultrafine structure under resonance conditions remains unresolved, and we obtain only a broadening and shift of the fine-structure components, as was indicated in Ref. (2).

3. Let us estimate the magnitude of the splitting. For this purpose we use the results of experiments on acoustic paramagnetic resonance performed on an MgO crystal containing an admixture of Fe^{2+} with concentration $C = 7.5 \cdot 10^{-5}$ (4). The absorption coefficient of a longitudinal sound wave with frequency $\Omega/2\pi = 9.46 \cdot 10^9$ Hz along the (100) axis is $\alpha = 10^2$ cm^{-1} at the temperature of liquid helium, if the magnetic field is directed at right angles to the (100) axis. The constant $A^{\vec{\varkappa}\lambda}_{\Omega}$ can be estimated from the approximate relation $qA^{\vec{\varkappa}\lambda}_{\Omega}/\Delta \sim \pi v \alpha(\Omega)$, where Δ is the width of the acoustic line.

paramagnetic resonance. In the scattering of light from a mercury arc ($\omega_0/2\pi = 7 \cdot 10^{14}$ Hz) at a right angle, the frequency of the phonons participating in the scattering is $\omega^{(0)}/2\pi = 3 \cdot 10^{10}$ Hz, according to the Mandelstam-Brillouin formula (the speed of sound along (100) is $v = 9.25 \cdot 10^5$ cm/sec). Taking into account that $a(\Omega) \sim \Omega^2$ and $\Delta = 3 \cdot 10^8$ Hz [4], we obtain for the magnitude of the splitting $\delta\omega \approx 3 \cdot 10^9$ Hz. Since $\delta\omega > \Delta$, the hyperfine structure should be resolved. Information on theoretical estimates of the magnitude of the spin-phonon interaction for various paramagnetic ions is contained in the review [5].

In conclusion, we note that the results obtained may be applied in an analogous manner to the discussion of neutron scattering by phonons, since the scattering cross section is determined by the same correlation functions.

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REFERENCES

[1] B. I. Kochelaev, FTT, **4**, 1560 (1962); E. H. Jacobsen, K. W. H. Stevens, Phys. Rev., **129**, 2036 (1963).
[2] S. A. Al' tshuler, B. I. Kochelaev, ZhETF, **49**, No. 3 (1965).
[3] S. V. Tyablikov, V. L. Bonch-Bruevich, *Green' s Function Method in Statistical Mechanics*, Moscow, 1961.
[4] N. S. Shiren, Phys. Rev., **128**, 2103 (1962).
[5] S. A. Al' tshuler, B. I. Kochelaev, A. M. Leushin, UFN, **75**, 459 (1961).

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