Threshold phenomena in Raman scattering of light by polaritons

I. B. Levinson and I. L. Maksimov

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted November 21, 1975)
Pis'ma Zh. Eksp. Teor. Fiz. **23**, No. 1, 3–6 (5 January 1976)

We investigated the distinguishing features of Raman scattering of light by polaritons in the case when the decay of the transverse optical phonon has a threshold. The line shape is qualitatively altered with changing scattering angle.

PACS numbers: 78.30.–j

The line width and line shape in Raman scattering (RS) of light by polaritons[1] is due to the decay of a long-wave ($q=0$) transverse optical (TO) phonon contained in the polariton. This decay results usually in two short-wave acoustic phonons $\omega_1(\mathbf{q}_1)$ and $\omega_2(\mathbf{q}_2)$ at almost opposite points of the Brillouin zone ($\mathbf{q}_1 \approx -\mathbf{q}_2$) and is determined by the two-phonon density of states $\rho(\omega)$, $\omega=\omega_1+\omega_2$, near the TO-phonon frequency Ω_0.

The momentum $\mathbf{k}$ of the polariton that takes part in the scattering is connected with the scattering angle θ. Since a change of $\mathbf{k}$ leads to a change of the degree $S(k)$ of participation of the phonon in the polariton and to a change of the frequency $\Omega(k)$ of the polariton itself, the character of the decay changes, and consequently also the width and shape of the RS line. These changes are strongest when the TO phonon has a singularity, i.e., when Ω_0 is near a singularity of $\rho(\omega)$.

We consider below the case when $\mathbf{q}_1$ and $\mathbf{q}_2$ are near the edge of the Brillouin zone where $\max(\omega_1+\omega_2) \equiv \omega_0 < \Omega_0$ is reached. Then there exists a polariton threshold momentum k_0 such that $\Omega(k_0)=\omega_0$. Below the threshold, $k<k_0$, the decay $\Omega_0 \to \omega_1+\omega_2$ contributes to the width of the polariton; above the threshold, $k>k_0$, such a decay is impossible. From the general theory of threshold phenomena it follows that the structure of the polariton spectrum should become qualitatively altered at $k=k_0$. The purpose of this paper is to ascertain the effects of this alteration on the RS spectrum.

Omitting inessential factors, we can express the differential RS cross section in terms of the polariton retarded Green's function[3]:

$$\sigma(\theta, \nu') \sim -\operatorname{Im} D(k, \Omega), \tag{1}$$

$$\Omega = \nu' - \nu, \quad c^2k^2 = \nu^2 + \nu'^2 - 2\nu\nu'\cos\theta. \tag{2}$$

Here ν and ν' are the frequencies of the incident and scattered light, and c is the velocity of the light in the medium. Furthermore

$$D(k, \Omega) = (2\pi)^{-1}(\Omega^2 - c^2k^2)\{[\Omega^2 - \Omega_0^2 + 2\Omega_0\Pi(\Omega)][\Omega^2 - c^2k^2] - \Omega_p^2\Omega^2\}^{-1}, \tag{3}$$

where

$$\Omega_p^2 = \Omega_{LO}^2 - \Omega_{TO}^2 = \Omega_0^2(\epsilon_0/\epsilon_\infty - 1). \tag{4}$$

ϵ_0 and ϵ_∞ are the dielectric constants. $\Pi(\Omega)$ is the polarization operator of the TO phonon with $q=0$, and takes the phonon anharmonicity into account. Inasmuch as at a given k only values of Ω close to the polariton frequency $\Omega(k)$ calculated at $\Pi=0$ are significant, the cross section can be rewritten in the form

$$- \mathrm{Im} D(k, \Omega) = (4\pi)^{-1} S^2(k)\Gamma(\Omega)/\Omega_0 \times \{[\Omega - \Omega(k) + S(k)\Delta(\Omega)]^2 + [S(k)\Gamma(\Omega)]^2\}^{-1}, \quad (5)$$

where $\Delta = \mathrm{Re}\Pi$ and $\Gamma = \mathrm{Im}\Pi$. When calculating k in accord with (2) it can be assumed that $\nu' = \nu - \Omega(k)$. Then k turns out to be uniquely connected with θ and independent of ν', and at fixed θ the line shape is determined by the dependence of D on Ω.

If $\rho(\omega)$ has no singularities, then $\Pi(\Omega)$ is a smooth function. It can then be assumed in (5) that Γ and Δ are independent of Ω, and they can be calculated at $\Omega = \Omega(k)$. In this case the RS line has a Lorentz shape with width $S(k)\Gamma(\Omega(k))$ that changes smoothly with changing θ; it is difficult to observe the frequency shift of $S(k)\Delta(\Omega(k))$ experimentally.

For the threshold decay considered above, $\rho(\omega)$ has a singularity $(\omega_0 - \omega)^{1/2}$. In this case[2] we have at $\Omega > \omega_0$

$$\Gamma(\Omega) = \Gamma_0, \quad \Delta(\Omega) = \Delta_0 - [\gamma(\Omega - \omega_0)]^{1/2},$$

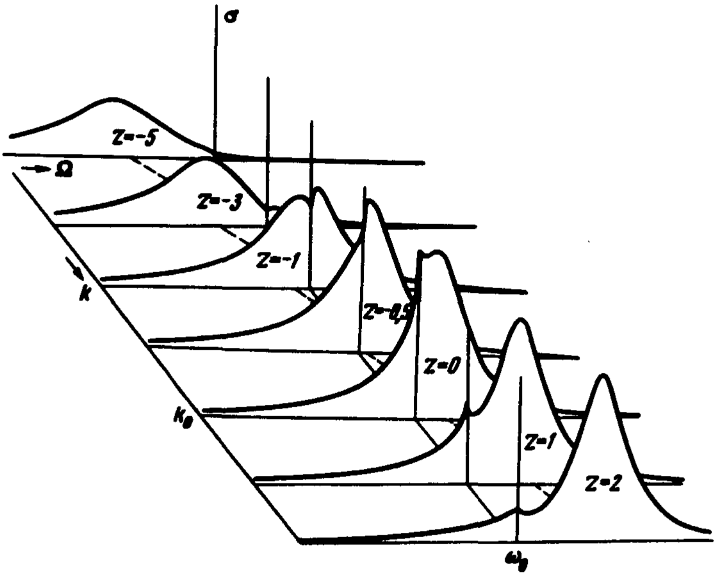
and at $\Omega < \omega_0$ (6)

$$\Gamma(\Omega) = \Gamma_0 + [\gamma(\omega_0 - \Omega)]^{1/2}, \quad \Delta(\Omega) = \Delta_0 .$$

Here Γ_0 and Δ_0 correspond to the smooth part of $\Pi(\Omega)$; near threshold they can be regarded as constant. Δ_0 can be included in the unrenormalized spectrum, and we therefore put henceforth $\Delta_0 = 0$. γ is a constant that accounts for the rate of the threshold decay. Substituting (6) in (5) we can find the RS line shape which, owing to dependence of Γ and Δ on Ω turns out to be quite complicated and strongly dependent on the ratio of the parameters $\delta(k) = \Omega(k) - \omega_0$, Γ_0, and γ. Therefore a change of θ when $\delta(k)$ changes causes a change in the line shape. The most significant change occurs at $\theta = \theta_0$, i.e., $k = k_0$, when $\delta(k)$ reverses sign. This is illustrated by the figure, which shows the shape of the RS line for different frequencies all the way to the threshold ($Z = \delta(k)/\Gamma_0$, $\gamma/\Gamma_0 = 0.8$).

A situation close to that considered here is realized in GaP, where the frequency $\Omega_0 = 366$ cm^{-1} is close to the sum of the frequencies of the acoustic phonons at the point X on the edge of the Brillouin zone: $LA(X) + TA(X) = 249$ cm^{-1} $+ 107$ cm^{-1} $= 356$ cm^{-1} $\equiv \omega_0$. Comparison of ω_0 with $\omega_{TA} + \omega_{LA}$ at the points K and L on the boundary of the zone allows us to assume that $\max(\omega_{TA} + \omega_{LA})$ is reached at the point X. On the other hand, temperature measurements of the line widths[4] indicate directly the presence of the decay TO $\to LA(X) + TA(X)$. Assuming for GaP the values $\Omega_{LA} = 403$ cm^{-1} and $\epsilon_\infty = 9.09$, we obtain $k_0 = 2\pi \cdot 2500$ cm^{-1} and $\theta_0 = 2°$.

Experiments on RS in GaP have indeed revealed anomalies. In scattering at $\theta = \pi/2$ the RS has a long-wave shoulder[5,6]; it is probably smoothed out (owing to the insufficient resolution) by an additional peak that can be seen in the figure at $k > k$ (i.e., $\theta > \theta_0$). In scattering at angles $\theta < 2°$ a sharp dependence of the



"width" of the RS line on the angle is observed.[7] It might be assumed that this is a masked indication of the fact that a line with complex shape cannot be described simply by means of the width. A careful measurement of the RS line shape at various angles θ near θ_0 would permit a very accurate determination of a large number of crystal parameters. Another material in which analogous RS anomalies were observed is ZnSe.[8]

The complex line shape leads to a nonexponential decay, which can be measured by using a picosecond probing pulse.[9] Unfortunately, the experiment realized in GaP[9] pertained to a polariton with $k \approx 2\pi \cdot 2770$ cm^{-1} and $\Omega(k) \approx 361$ cm^{-1}. Such a polariton lies far above the threshold, where the line has a shape close to Lorentzian, and it is therefore not surprising that the decay turned out to be exponential within the limits of the experimental accuracy. When polaritons with smaller k are excited one should expect a nonexponential decay, or at least a dependence of the lifetime on k.[10]

[1]D. L. Mills and E. Burstein, Rep. Prog. Phys. 37, 817 (1974).
[2]I. B. Levinson and É. I. Rashba, Usp. Fiz. Nauk **111**, 683 (1973) [Sov. Phys. Usp. 16, 892 (1974)]; Y. B. Levinson and E. I. Rashba, Rep. Prog. Phys. 36, 1499 (1973).
[3]H. S. Benson and D. L. Mills, Phys. Rev. **B1**, 4835 (1970).
[4]S. Ushioda, J. D. McMullen, and M. J. Delaney, Phys. Rev. **B8**, 4634 (1973).
[5]A. S. Barker, Jr., Phys. Rev. **165**, 917 (1968).
[6]B. Kh. Baĭramov, Yu. E. Kitaev, V. K. Negoduĭko, and É. M. Khashkhozhev, Fiz. Tverd. Tela 16, 1129, 2036 (1974) [Sov. Phys. Solid State 16, 1323 (1975)].
[7]S. Ushioda and J. D. McMullen, Solid State Commun. **11**, 299 (1972).

[8]J. H. Nicola and R. C. C. Leite, Phys. Rev. **B11**, 798 (1975).
[9]A. Laubereau, D. von der Linde, and W. Kaiser, Opt. Commun. 7, 173 (1973).
[10]P. G. Harper, Opt. Commun. 10, 68 (1974).