Raman scattering induced by strongly nonequilibrium phonons

S. E. Bulgadaev and I. B. Levinson

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted April 4, 1974)
ZhETF Pis. Red. **19**, 583–585 (May 5, 1974)

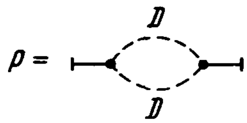
It is shown that a distribution of short-wave acoustic phonons with a small frequency spread leads to a distinctive renormalization of the spectrum of Raman scattering by long-wave optical phonons and to the appearance of a new branch of elementary excitations.

It can be regarded as experimentally proved, with a great degree of reliability, that it is possible to produce distributions of short-wave acoustic phonons with a small frequency spread $\Delta\omega$ about a certain central frequency ω_0.[1,2] The presence of such a distribution leads to a renormalization of the spectrum of the long-wave ($k \approx 0$) optical phonons, which can decay into two short-wave acoustic phonons (with opposite momenta q and $-q$).

We call attention in this paper to the fact that the restructuring of the spectrum due to the quasimonochromatic distribution of the acoustic phonons ($\Delta\omega$ is comparable with the width Γ_0 of the spontaneous decay of an optical phonon with $k=0$) differs qualitatively from the renormalization in the usual case of a broad distribution $\Delta\omega \gg \Gamma_0$. For a narrow distribution, a situation is possible wherein the presence of acoustic phonons does not stimulate decay, but to the contrary, hinders it. In addition, in the case of narrow distributions there appears a new branch of long-wave excitations of the phonon system. Such a picture, which contradicts the usual concepts, is possible because the usual representations are based on the kinetic equations for the occupation numbers; yet such equations do not hold in our case, since the quantum-mechanical widths are of the order of the statistical widths.[3]

To find the spectrum, we used a non-equilibrium diagram technique.[4] We calculated the retarded Green's function $G(t)$ for optical phonons with $k=0$. The polarization operator $G(t)$ of this Green's function can be obtained from the simplest diagram (see the figure), in which the Green's functions D of the acoustic phonons are assumed to be specified. The functions D have the same form as the thermodynamic-equilibrium functions, but the plank distribution is replaced by a narrow distribution N_ω centered about ω_0. It is assumed that $\omega_0 = \Omega_0/2$, where Ω_0 is the frequency of the optical phonon with $k=0$; obviously, the renormalization of the spectrum will be the largest in this case. In addition, it is precisely such distributions that arise in the experimental situation[1,5,6] when the acoustic phonons are parametrically excited.[7]

It should be emphasized that the discarding of the more complicated diagrams is not ensured in this case by the weakness of the anharmonicity, i.e., by the parameter $\Gamma_0/\Omega_0 \ll 1$. The point is that the presence of the scale $\Delta\omega$ in N_ω causes the parameter Γ_0/Ω_0 in the terms containing N_ω to be replaced by the parameter $\Gamma_0/\Delta\omega$, which generally speaking is not small. However, such terms always contain as a factor another characteristic parameters, the number of acoustic phonons



per unit cell, which is of the order of $N_0(\Delta\omega/\Omega_0)$, where N_0 is the occupation number of the central mode. Therefore, if we assume that $N_0 \ll \Omega_0/\Delta\omega$, then the discarding of the more complicated diagrams P becomes justified.

If we choose for N_ω a Lorentz distribution, then the Fourier transforms $P(\Omega)$ and $G(\Omega)$ can be calculated in explicit form. It is also possible to determine the spectrum accurately in this case. The function $G(\Omega)$ has two poles, Ω_1 and Ω_2, the positions of which depend on the concentration of the acoustic phonons. The critical concentration is the one corresponding to the occupation numbers

$$N_0^* = \frac{a}{8}\left(1 - \frac{1}{a}\right)^2 \qquad a = 2\frac{\Delta\omega}{\Gamma_0} \quad . \tag{1}$$

We introduce $\xi = N_0/N^*_0$. Then at $\xi < 1$ we have

$$\begin{aligned} &\mathrm{Re}\,\Omega_1 = \mathrm{Re}\,\Omega_2 = \Omega_0, \\ &\mathrm{Im}\,\Omega_{1,2} = \frac{1}{4}\Gamma_0[(a+1) \pm (a-1)\sqrt{1-\xi}]; \end{aligned} \tag{2}$$

and at $\xi > 1$

$$\begin{aligned} &\mathrm{Im}\,\Omega_1 = \mathrm{Im}\,\Omega_2 = \frac{1}{4}\Gamma_0(a+1), \\ &\mathrm{Re}\,\Omega_{1,2} = \Omega_0 \pm \frac{1}{4}\Gamma_0(a-1)\sqrt{\xi - 1}. \end{aligned} \tag{3}$$

As $N_0 \to 0$, one of the poles $\Omega_1 \to \Omega_0 + i\Gamma_0/2$, and the other $\Omega_2 \to \Omega_0 + i\Delta\omega$. Then Res $G(\Omega_1) \to 1$ and Res $G(\Omega_2) \to 0$. It is clear therefore that one of the poles is genetically connected with the usual pole of the optical phonon, while the other pole describes new elementary excitations with frequency Ω_0 and lifetime $(2\Delta\omega)^{-1}$. This time is large only for narrow acoustic-phonon distributions, and therefore such excitations cannot be observed in the usual situations when $\Delta\omega \approx \Omega_0$.

When N_0 increases, the poles Ω_1 and Ω_2 approach each other; they merge at $N_0 = N^*_0$. Starting with this value of N_0, the genesis of the poles becomes meaningless. At $N > N^*_0$ the elementary excitations of both types become mixed, and two types of excitations with identical damping $\Delta\omega + \frac{1}{2}\Gamma_0$ but with slightly different frequencies are produced; the frequency difference increases with increasing N_0.

It can be shown that the law governing the decay of the specified initial excitation of the optical mode with $k=0$ is determined by the low-frequency part of $G(t)^2$, i.e., by the displacement of the poles from the point Ω_0. It is seen from (2) and (3) that if $\Delta\omega > \Gamma_0/2$, then the rate of the exponential decay of both types of excitations exceeds the rate of the spontaneous decay, i.e., the decay is induced, just as in the classical case $\Delta\omega \gg \Gamma_0$. On the other hand, if $\Delta\omega < \Gamma_0/2$, then both types of excitations decay at a rate smaller than the spontaneous rate, i.e., destimulation of the decay takes place. In both cases, the decay is oscillatory at $N_0 < N_0^*$.

In stationary parametric generation of acoustic phonons by light, depending on the excitation conditions, the width $\Delta\omega$ is determined by the width Γ_0 or by the spectral width $\Delta\nu$ of the exciting light.[2,3] Typical values are $\Omega_0 \sim 1000\ \text{cm}^{-1}$, $\Gamma_0 \sim 1\ \text{cm}^{-1}$, and $\Delta\nu \sim 0.1$—$1\ \text{cm}^{-1}$. The occupation numbers of the produced acoustic phonons N depend on the excitation power; $N \sim 10^{-3}$—10 in different experiments.[1,5,6]

In connection with the possible destimulation of the decay, it is curious to note that in the experiments[5] where the largest values of N were reached, they observed not an increase in the decay rate in comparison with the spontaneous decay, but to the contrary, a slowing down. Unfortunately, there are no data on $\Delta\nu$ for this case.

The presence of new elementary excitations can affect the line shape of the Raman scattering of light by optical phonons, which is determined by Im $G(\nu - \nu')$, where ν and ν' are the frequencies of the incident and scattered light. With increasing pumping of the acoustic phonons, the line should first broaden or narrow down (depending on the relation between $\Delta\omega$ and $\Gamma_0/2$), and should split into two at a critical pumping corresponding to N^*_0.

The authors thank L.P. Gor'kov, É.I. Rashba, and L.P. Pitaevskiĭ for a discussion of the work.

[1]M.J. Colles and J.A. Giordmaine, Phys. Rev. Lett. **27**, 670 (1971).
[2]M.J. Colles, J. de Phys., Suppl. to No. 10, 41 (1972).
[3]I.B. Levinson, Zh. Eksp. Teor. Fiz. **65**, 331 (1973) [Sov. Phys.-JETP **38**, 162 (1974)].
[4]L.V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1515 (1964) [Sov. Phys.-JETP **20**, 1018 (1965)].
[5]R.R. Alfano and S.L. Shapiro, Phys. Rev. Lett. **26**, 1247 (1971).
[6]A. Laubereau, D. von der Linde, and W. Keiser, Phys. Rev. Lett. **27**, 802 (1971).
[7]R. Orbach, Phys. Rev. Lett. **16**, 15 (1966).