Size-effect resonance in Raman scattering of light

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(Submitted March 10, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **23**, No. 8, 444-448 (20 April 1976)

It is shown that a size-effect resonance takes place in resonant single-phonon
scattering and consists of an enhancement of the role of the Wannier-Mott exciton
with principal quantum number $n \approx \sqrt{\lambda/4a}$ (λ is the phonon wavelength and a is
the Bohr radius of the exciton).

An investigation of resonant scattering near the threshold of the intrinsic
absorption of a crystal having an exciton structure shows that this structure
determines the properties of the light-scattering amplitude in a wide frequency
interval. [1-4] In the calculation of the amplitude of the one-phonon scattering,
one usually limits himself to the nearest approximation in the parameter
$|k - k'|d$, where k and k' are the wave vectors of the incident and scattered
light, and d is the characteristic dimension of the scattering system. The am-
plitude of the allowed scattering in this case does not depend on the wavelength
of the light, and for the forbidden scattering it is proportional to $|k - k'|d$,
however, the coefficient of this factor is only a function of the frequency. [2-3]

It is shown in this paper that in the case when the main contribution to the
amplitude is made by states of hydrogen-like type (Wannier-Mott exciton), the
criterion for the applicability of the expansion for the exotonic states is only the
condition $|k - k'| \ll \pi/2n^2a$, where a is the Bohr radius of the exciton, and n is
the principal quantum number. In the region of the exotonic states, $|k - k'|a$
turns out to be not so small a quantity (for CdS in backscattering geometry,
$|k - k'|a \approx 0.24$ at a light wavelength 4880 Å), and for $n \approx 2-3$ this criterion is

not satisfied. Quantitatively this manifests itself in the fact that the contribu-
tions to the amplitude of the levels with $n > 1$ begin to increase, in spite of the
decreasing oscillator strength. This increase has a resonant character, i.e.,
starting with the number $n = n_0 \approx \sqrt{\pi/2a\,|\mathbf{k}-\mathbf{k}'|}$, the contributions of the levels
begin to decrease. This size effect takes place to an equal degree for allowed
and forbidden scattering. We note also that an analogous resonant contribution
is made also by the region of small positive energies in the continuous spec-
trum of the excitons at $pa = p_0 a \approx \sqrt{2a\,|\mathbf{k}-\mathbf{k}'|/\pi}$.

The amplitude of the single-phonon forbidden scattering by LA phonons can
be represented in the two-band approximation in the form

$$A_1(\mathbf{q}, \omega) \sim \frac{\Xi^{LO}}{qa} \sum_{\lambda\lambda'} \frac{(e^{i\mathbf{q}^c \mathbf{r}} - e^{-i\mathbf{q}^v \mathbf{r}})_{\lambda\lambda'} \, \psi_\lambda(0)\, \psi_{\lambda'}^*(0)}{\left[\Delta(\lambda) - \dfrac{\hbar k^2}{2M\Omega_{LO}} + i\gamma_\lambda\right]\left[\Delta_1(\lambda') - \dfrac{\hbar k'^2}{2M\Omega_{LO}} + i\gamma_{\lambda'}\right]}, \tag{(}$$

where

$$\Xi^{LO} = \left[\frac{2\pi e^2}{d}\,\hbar\Omega_{LO}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right)\right]^{1/2}$$

is the intraband matrix element of the Fröhlich electron-phonon interaction;
$\mathbf{q} = \mathbf{q} - \mathbf{k}'$; $\mathbf{q}^{c,v} = (\mu/m_{c,v})\mathbf{q}$; μ is the reduced effective carrier mass in the c and
v bands;

$$M = m_c + m_v, \quad \Delta(\lambda) = \frac{1}{\Omega_{LO}}(\omega - \epsilon_g - E_\lambda); \quad \Delta_1(\lambda') = \Delta(\lambda') - 1; \quad E_\lambda = -R/n^2)$$

or $E_\lambda = R(pa)^2$ for the discrete and continuous spectrum of the exciton, R is the
exciton binding energy; $(\cdots)_{\lambda\lambda'}$ is the matrix element in the hydrogenlike wave
functions $\psi_\lambda(\mathbf{r})$. The amplitude of the allowed scattering differs from (1) in the
substitution

$$\frac{\Xi^{LO}}{qa}(e^{i\mathbf{q}^c \mathbf{r}} - e^{-i\mathbf{q}^v \mathbf{r}}) \to (\Xi_{cc}e^{i\mathbf{q}^c \mathbf{r}} - \Xi_{vv}e^{-i\mathbf{q}^v \mathbf{r}}), \tag{(}$$

where $\Xi_{cc} \neq \Xi_{vv}$ are assumed to be constant and different from zero at the cente
of the Brillouin zone. As a result of the expansion of the exponentials in (1),
the expression for the amplitude begins to diverge like

$$\sum_n |\psi_n(0)|^2 < r^2 >_{nn} \sim \sum_n n \to \infty. \tag{(}$$

When the substitution (2) is made for the allowed scattering, the zeroth term
of the expansion does not diverge, but the first-order correction behaves like
(3).

The exact calculation of the matrix element $\langle\exp[i\mathbf{q}\cdot\mathbf{r}]\rangle_m$ yields

$$< e^{i\mathbf{q}\mathbf{r}} >_{nn} = (-1)^{n-1} \frac{z^{2n-2}}{(1+z^2)^n} \frac{\sin(2n \text{ arc tg} z)}{z} F\left(-n+1, -n+1; 2; -\frac{1}{z^2}\right), \tag{(4}$$

where $z = nqa/2$ and F is a hypergeometric function. Using the asymptotic form
of F at large negative arguments, we can represent (4) in the form

$$< e^{i\mathbf{q}\mathbf{r}} >_{nn} \approx \frac{1}{(1+z^2)^n} \frac{\sin(2n \text{ arc tg } z)}{nz}. \tag{5}$$

s seen from (5) and (1) a maximum contribution is made by those states
r which the argument of the sine is of the order of $\pi/2$. With further increase
n, when the argument of the sine in (5) becomes larger than π, the matrix
ement begins to oscillate rapidly and makes a negligible contribution to the
immation in (1). An analogous contribution is made also by the continuous
ıectrum of the exciton. Thus, in the calculation of the amplitude we can re-
ace the upper limit of the sum over the discrete state and the lower limit of
e integral over the continuous spectrum by finite quantities.

The thus-calculated cross section of the forbidden scattering by LL phonons,
ı a function of the frequency of the incident light, is shown in Fig. 1. The pa-
ımeters chosen were those of the CdS crystal, for which this cross section
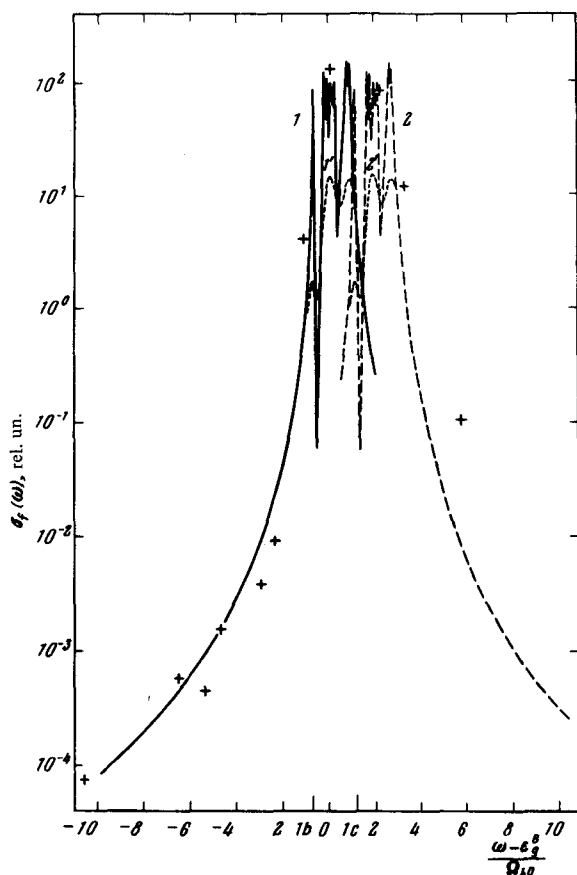as measured in[1]. The data of[1] are also shown in Fig. 1. At the experimen-

"IG. 1. Frequency dependence of the cross section of forbidden scattering by
,O phonons. We used for the calculations the parameters $m_c/m_v = 0.22$, R
$= 0.7\Omega_{LO}$, and $\gamma = 0.02\Omega_{LO}$ (curves 1 and 2) or $\gamma = 0.2\Omega_{LO}$ (curves 1' and 2'). The
ıositions of the experimental points of[1] were corrected in accord with the
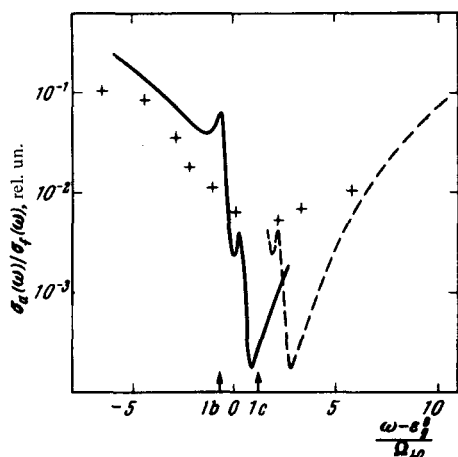ınown wavelengths of the lasers indicated in[1].

FIG. 2. Ratio of the cross sections ᴏ
the allowed and forbidden scattering
LO phonons. The experimental data
were taken from [1], and the positions
of the points were corrected as in Fi
1.

tal geometry used in [1], the contribution to the scattering amplitudes is made
the exciton series B and C. The experimental data are insufficient for a detai
comparison of the theory with experiment. For a qualitative comparison we
have calculated the cross sections obtained separately from the series B
(curves 1 and 1′) and the series C(2 and 2′). It is seen from the figure that go
agreement is obtained both in the region $\omega < \epsilon_g^B$ and in the region $\omega > \epsilon_g^B$. We
note that the size-effect resonance produces on each curve a clearly seen thir
(high-frequency) maximum. The cross-section structure connected with the
size-effect resonance should manifest itself more distinctly in the ratio of the
cross sections of the allowed and forbidden scattering. Figure 2 shows two
plots of the cross section ratio (separately for the series B and C) together w
the experimentally measured values of the ratio of the allowed and forbidden
scattering cross sections. [1] It is seen from the comparison that in this case
the theory explains qualitatively the observed effect.

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