# WIDTHS OF PEAKS OF TWO-PHOTON RESONANT RAMAN SCATTERING IN CRYSTALS

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We determined the shift and broadening of the lines of two-photon resonant light scattering in CdS crystals excited with light of different wavelengths. The theory of resonant two-phonon scattering is considered and it is shown that these effects are a reflection of a fundamental distinguishing feature of resonant scattering.

An interesting distinguishing feature of resonant Raman scattering in crystals is the unusual narrowness of the two-phonon lines [1]. Their width is close to that of the one-phonon peak. This phenomenon is difficult to understand at first glance, since in two-phonon scattering, unlike in one-phonon scattering, the momentum conservation law permits participation of phonons with all possible momentum values, and the width of the two-phonon spectrum should seemingly be equal to double the width of the phonon branch. Figure 1a shows the spectrum obtained by us for resonant scattering by longitudinal optical (LO) phonons of a CdS crystal excited with light of $\lambda$ = 4880 Å. As seen from the figure the half-width of the 2LO line is $\sim$7 $cm^{-1}$. This width is greatly influenced by the anisotropy of the phonon branch [1]. When anisotropy is taken into account, the "true" value of the half-width of the 2LO line is $\sim$4 $cm^{-1}$, which is close to the half-width of the 1LO peak (3.6 $cm^{-1}$) and is much less than double the width of the optical branch in CdS (18 $cm^{-1}$) [2]. It is also remarkable that in two-phonon scattering there appear only phonons with small wave vectors, so that the frequency of the two-phonon maximum is equal to double the frequency of the one-phonon peak. This result is not explained by Born's semi-phenomenological theory [4] and differs considerably from the known experimental data on two-phonon scattering spectra in alkali-halide crystals [5, 6] far from resonance.

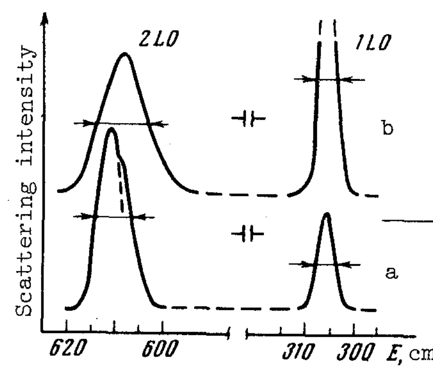


Fig. 1. Spectrum of resonant Raman scattering in CdS crystal excited with light $\lambda$ = 4880 Å (a) and 6328 Å (b) at 4.2°K. To exclude effects due to interaction with plasma oscillations [3], we investigated in our experiment only undoped insulating crystals.

In this paper, using longitudinal optical phonons as an example, we explain the observed features of resonant two-phonon scattering. We shall show that these features are a fundamental property of resonant scattering in crystals.

It was established earlier [7, 8] that to explain the experiments on one-phonon scattering by LO phonons in the resonance region it is necessary to take into account the intraband interaction of the LO phonons with the electrons. This interaction enhances the resonance in the dependence of the one-phonon scattering over the value obtained when only the interband interaction is taken into account [9]. In the case of two-phonon scattering, the intraband interaction, on the one hand, also enhances the resonance in the dependence of the scattering cross section on the incident-light frequency, and on the other hand, it leads to a very strong dependence of the scattering amplitude on the wave vectors of the produced phonons. Let us examine the case of resonant two-phonon scattering in greater detail.

The largest contribution to the scattering is made by the resonant terms of the amplitude ($P^{\alpha\beta}$). They have the following dependence on the frequencies of the incident and scattered light, and also on the frequency and momentum of the produced phonons:

$$P^{\alpha\beta} = \frac{2\pi e^4 \Omega_q^{\ell}}{\hbar^2 v_0 q^2}\left(\frac{\epsilon_0-\epsilon_\infty}{\epsilon_0\epsilon_\infty}\right)\int d^3p\left\{\frac{1}{\omega-\Omega^{cv}_{pp}}\left[\frac{1}{\omega-\Omega^{cv}_{pp}-\Omega^{\ell}_q+\Omega^{cc}_{p\,p-q}}+\right.\right.$$

$$\left.\left.+\frac{1}{\omega-\Omega^{cv}_{pp}-\Omega^{\ell}_q+\Omega^{vv}_{p\,p-q}}\right]\left[\frac{v^{\alpha}_{cv}(p)\,v^{\beta}_{vc}(p+q)}{\omega-\Omega^{cv}_{pp}-2\Omega^{\ell}_q+\Omega^{cc}_{p\,p-q}+\Omega^{vv}_{p\,p-q}}-\frac{v^{\alpha}_{cv}(p)\,v^{\beta}_{vc}(p)}{\omega-\Omega^{cv}_{pp}-2\Omega^{\ell}_q}\right]\right\}. \qquad (1)$$

Since the two phonons produced by the light scattering have practically equal and opposite momenta, we have written out here only one-half of the complete expression. The second half is obtained from (1) by replacing $\vec{q}$ with $-\vec{q}$. The following notation was used: $\Omega^{cv}_{pp'} = (\varepsilon_{cp} - \varepsilon_{vp'})/\hbar$, where $\varepsilon_{cp}$ and $\varepsilon_{vp'}$ are the electron energies in the c and v bands, $v_{cv}(\vec{p})$ is the matrix element of the electron velocity operator, $\Omega^{\ell}_q$ is the frequency of the longitudinal optical phonon with wave vector $\vec{q}$, $\varepsilon_0$ and $\varepsilon_\infty$ are the dielectric constants of the crystal, $\omega$ and $\omega' = \omega - 2\Omega^{\ell}_q$ are the frequencies of the incident and scattered light, and $v_0$ is the volume of the unit cell. Let us analyze the expression for the scattering amplitude in the limiting cases.

In the limit as $q \to 0$, Eq. (1) goes over, in the effective mass approximation, into

$$P^{\alpha\beta} = \frac{4\pi^3 e^4 \Omega^{\ell}_0}{\hbar^2 v_0 q^2}\left|v^{\alpha}_{cv}(0)\right|^2\delta_{\alpha\beta}\left(\frac{\epsilon_0-\epsilon_\infty}{\epsilon_0\epsilon_\infty}\right)\left(\frac{2m}{\hbar}\right)^4\left(\frac{\hbar q^2}{2m}\right)\frac{\delta+\delta'+2\delta''}{\delta''(\delta+\delta')(\delta'+\delta'')^2(\delta+\delta'')^2}. \qquad (2)$$

In the other limit $q \gg \sqrt{2m(\varepsilon_g - \omega')/\hbar}$ we obtain from (1)

$$P^{\alpha\beta} \sim \frac{4\pi^3 e^4 \Omega^{\ell}_0}{\hbar^2 v_0 q^2}\left|v^{\alpha}_{cv}(0)\right|^2\delta_{\alpha\beta}\left(\frac{\epsilon_0-\epsilon_\infty}{\epsilon_0\epsilon_\infty}\right)\left(\frac{2m}{\hbar}\right)^4\left(\frac{\hbar q^2}{2m}\right)\frac{(2m)^2}{(\hbar q^2)^2(\delta+\delta'')}. \qquad (3)$$

In formulas (2) and (3), m is the reduced effective mass of the electron and hole,

$$\delta = \sqrt{\frac{2m(\epsilon^{cv}_g-\omega)}{\hbar}}, \quad \delta' = \sqrt{\frac{2m(\epsilon^{cv}_g-\omega+\Omega^{\ell}_0)}{\hbar}}, \quad \delta'' = \sqrt{\frac{2m(\epsilon^{cv}_g-\omega')}{\hbar}},$$

and $\varepsilon^{cv}_g$ is the width of the forbidden band. Expressions (2) and (3) were obtained under the assumption that the extrema of the bands are located at the point p = 0.

Comparing (2) with (3) we see that at small values, $q \to 0$, the amplitude does not depend on q, whereas at $q \gg \delta''$ it is inversely proportional to $q^4$. Such a dependence of the amplitude on q causes phonons with momenta on the order of $\delta''$ to take the predominant part in the scattering. In resonant scattering, i.e., at small values of $(\varepsilon_g^{cv} - \omega')$, only phonons with small wave vectors will take part in the process. This means that a narrow peak should be observed in the spectrum of the two-phonon scattering, and the energy position of this maximum should be determined in practice by double the frequency of the optical branch at $q \sim 0$, as is indeed observed in the experiment. Figure 2 shows schematically the dependence of the cross section on the frequency of the scattered light. This dependence follows from (2) and (3), assuming for the phonons the simplest dispersion law $\Omega_q^{\ell} = \Omega_0^{\ell} - \beta(aq)^2$. With increasing difference between $\omega$ and $\varepsilon_g^{cv}$, there should occur not only an overall weakening of the scattering, but also a broadening of the range of values of the wave vectors of the phonons participating in the scattering. This should result in a shift of the scattering maximum (as a function of the dispersion $\beta$) and its broadening. Indeed, as seen from a comparison of curves a and b in Fig. 1, when the exciting frequency moves far from resonance ($\lambda$ = 6328 Å, curve b) one observes a noticeable broadening of the two-phonon peak and a certain shift (by 3 - 4 $cm^{-1}$) of the maximum towards lower phonon energies when compared with the more resonant excitation ($\lambda$ = 4880 Å, curve a). Similar regularities were observed also in other crystals (ZnO, ZnSe) at increasing deviations of the exciting-light frequency from resonance [1], and can be well explained by the proposed theory.

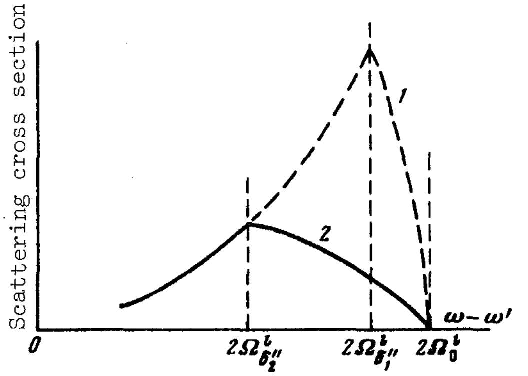


Fig. 2. Schematic dependence of the scattering cross section on the frequency of the scattering light $(\omega - \omega')$, corresponding to formulas (2) and (3) for two different values of $\varepsilon_g^{cv} - \omega$. Curve 1 corresponds to a case closer to resonance than curve 2; $\delta''_1$ and $\delta''_2$ are the wave vectors of the phonons that make the main contribution to the scattering in these two cases. The remaining symbols are explained in the text.

We have disregarded so far the exciton structure of the absorption edge. Its inclusion does not change the results. Moreover, formulas (2) and (3) remain valid also in the presence of an exciton, although the region of their applicability remains bounded by the inequality $\hbar(\varepsilon_g - \omega) \gg I_{ex}$, where $I_{ex}$ is the exciton ionization energy. At frequencies close enough to the exciton absorption line, this inequality does not hold. The dependence of the amplitude on the frequencies of the incident and scattered light is then altered, and the region of allowed values of phonon momenta becomes even narrower, and is determined in this case by the relation $q_{allow} \lesssim [2m(\hbar\varepsilon_g^{cv} - \hbar\omega' - I_{ex}]\hbar^{-2}$. We emphasize that the dependence of the amplitude on the phonon vector remains the same whether or not the exciton is taken into account, namely, the amplitude does not depend on q when $q \to 0$, and is inversely proportional to $q^4$ when $q \gg q_{allow}$. We note in conclusion that a similar phenomenon should be expected for other combinations of optical phonons that are allowed in the two-phonon spectrum, and also for multiphonon processes.

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# ACCELERATION OF LASER-PLASMA IONS IN A CYCLOTRON

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1. Experiments aimed at registering multiply charged ions in a laser plasma [1, 2] and at investigating such ion-emission characteristics as the maximum ionization multiplicity, the energy and spatial distribution of the ions, and the number of ions of given charge, have shown [3 - 5] that a laser plasma is an effective source of multiple charged ions (MCI) and can be used in accelerator injectors.

We report here experimental realization of acceleration of $D^+$ ions of a laser plasma in a cyclotron. We used internal ion injection, i.e., the laser plasma was produced in a magnetic field of the cyclotron inside the accelerating gap.

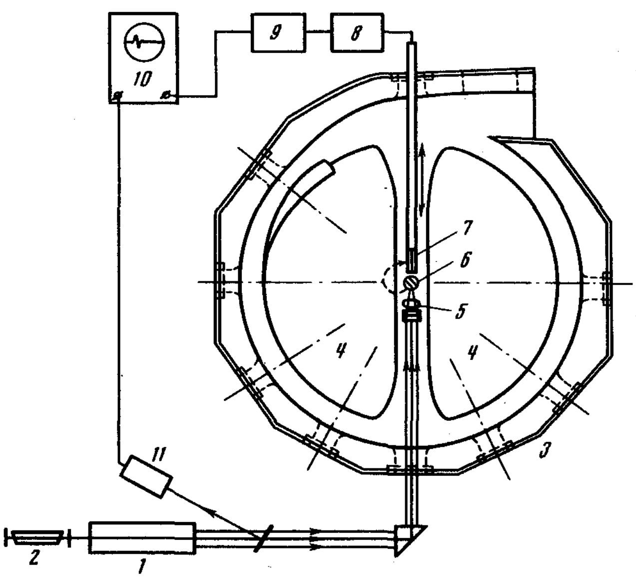


Fig. 1. Experimental setup: 1 - neodymium laser, 2 - adjusting He-Ne laser, 3 - cyclotron chamber, 4 - dees, 5 - optical system, 6 - target, 7 - probe with collectors, 8, 9 - amplifiers, 10 - oscilloscope, 11 - photocell.