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MULTIPHONON RAMAN SCATTERING OF LIGHT

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Multiphonon Raman scattering of light by longitudinal optical phonons is considered for the case when the photon frequency is close to the width of the forbidden band. Either exciton levels or impurity electron levels (donor impurity) are considered as intermediate states. The scattering cross section is calculated. It is shown that if the exciton levels are regarded as the intermediate states the Raman scattering cross section depends on the parity of the scattering order. If the impurity levels are regarded as the intermediate states the Raman cross section does not depend on parity of the scattering order.

As is well known, the cross section of multiphonon Raman scattering of light is, generally speaking, much smaller than the cross section of first-order scattering. Therefore Raman scattering of light of order higher than the second was not investigated until most recently either experimentally or theoretically. However, if the frequency of the incident or scattered light is closer to the natural frequency of a transition of the medium, for example to the exciton-absorption line[1,2], then the cross section of Raman scattering of light of any order increases, and it becomes possible to observe Raman scattering of higher orders. It is precisely under these conditions that the experiment in[1-3] was performed. However, there is apparently no more or less detailed theoretical analysis of such effects.

The present paper is devoted to a theoretical analysis of multiphonon Raman scattering of light in the case when the frequencies of the photons are close to the exciton or impurity absorption lines. The scattering cross section is determined. It is shown that the cross section for scattering with participation of the exciton levels as intermediate states can be essentially different for different approximations, values, depending on the parity of the number of excited phonons. This feature is simply the consequence of the momentum conservation law, i.e., the requirement that the total quasimomentum of the phonons produced in scattering (the Stokes process) be of the order of the photon momentum, accurate to the reciprocal lattice vector. In the case of Raman scattering with participation of impurity levels as the intermediate levels, the total quasimomentum is not conserved, and the scattering cross section is independent of the parity of the number of resultant phonons.

It is well known that the Raman scattering cross section is given by

$$\frac{\partial\sigma}{\partial o}=\frac{e^4\omega_2}{v_0 m^4 c^4\omega_1}\sum|M_{lk}|^2, \tag{1}$$

$$M_{lk}=\sum_m\left\{\frac{(\mathbf{P}\mathbf{e}_2^*)_{lm}(\mathbf{P}\mathbf{e}_1)_{mk}}{\hbar(\omega_{km}+\omega_1)}+\frac{(\mathbf{P}\mathbf{e}_1)_{lm}(\mathbf{P}\mathbf{e}_2^*)_{mk}}{\hbar(\omega_{km}-\omega_2)}\right\}, \tag{2}$$

where ω_1 and ω_2 are the frequencies of the incident and scattered light with polarizations $\mathbf{e}_1$ and $\mathbf{e}_2$, $\mathbf{P}$ is the summary momentum of the electrons, v_0 is the volume, ω_{km} is the difference of the terms, k is the initial state of the system (which we shall henceforth assume to be the ground state, i.e., we consider the case of zero temperature), l is the final state of the system, and the summation in (1) is over all the final states with equal energy. Since $\omega_{km} < 0$, it follows that if ω_1 is close to any frequency $\omega_{m'k}$, one term in (2) becomes the largest, and the remaining terms can be neglected. This is precisely the case considered in this paper.

The interaction of the electrons with longitudinal optical phonons will be described by a Froelich Hamiltonian. The matrix element of the operator of the electron-phonon interaction contains as a factor the matrix element $\exp(i\mathbf{q}\cdot\mathbf{r})$. To obtain an estimate of the magnitude of the interaction, we expand the exponential in powers $(\mathbf{q}\cdot\mathbf{r})$ up to the first nonvanishing term, and find that for interband transitions the electron-phonon interaction operator is proportional to the lattice constant a_0, whereas for transitions between exciton levels or levels of the weakly-bound impurity electron, the electron-phonon interaction operator is proportional either to the exciton radius a_1, or to the radius of the first Bohr orbit of the impurity electron a_2. For a Wannier-Mott exciton, or for shallow impurities, we have $a_1 \gg a_0$ and $a_2 \gg a_0$. We shall henceforth consider therefore only the interaction of the phonons either with excitons or with impurity electrons (a donor impurity is assumed throughout).

By calculating the wave functions of the system in the $(n-\nu)$-th and ν-th approximations of perturbation theory, we obtain for M_{lk} the following expression:

$$M_{lk}=\sum_{\nu=0}^{n}\sum_{m_0\ldots m_n}\left[\hbar^{n+1}(\omega_1+\omega_{m_0k})\,\omega_{m_0m_1}\ldots\omega_{m_0m_n}\right]^{-1}\cdot \tag{3}$$

$$\times(\mathbf{P}\mathbf{e}_2^*)_{lm_\nu}V_{m_\nu m_{\nu-1}}\ldots V_{m_1m_0}V_{m_0m_{\nu+1}}\ldots V_{m_{n-1}m_n}(\mathbf{P}\mathbf{e}_1)_{m_nk},$$

where V is the operator of the electron-phonon interaction and the matrix elements are taken over the unperturbed wave functions of the non-interacting electrons and phonons; in addition it is assumed that $|\omega_1+\omega_{k,m_0}| \ll \omega_1$.

Let us consider first Raman scattering of light with participation of exciton states as the intermediate states. We shall assume the excitons to be of the

Wannier-Mott type and use the effective-mass approximation. Introducing the coordinate of the center of gravity of the exciton R and the relative distance $\rho = \mathbf{r}_1 - \mathbf{r}_2$, where $\mathbf{r}_1$ and $\mathbf{r}_2$ are the radius vectors of the electron and of the hole with effective masses m_1 and m_2, we represent the exciton-phonon interaction Hamiltonian in the form[4]

$$V=\sum_{\mathbf{q}} 2C_{\mathbf{q}} \sin\left(\frac{\mathbf{q}\rho}{2}\right) \left[a_{\mathbf{q}}^{+} \exp(-i\mathbf{q}\mathbf{R}-i\alpha\mathbf{q}\rho)+a_{\mathbf{q}} \exp(i\mathbf{q}\mathbf{R}+i\alpha\mathbf{q}\rho)\right], \tag{4}$$

$$\alpha=\frac{m_1-m_2}{2(m_1+m_2)} \qquad C_{\mathbf{q}}^2=\frac{2\pi e^2\hbar\omega_{\mathbf{q}}}{v_0 q^2}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right), \tag{5}$$

where ω_q is the frequency of the phonon with wave vector q, and ϵ_∞ and ϵ_0 are the dielectric constants for high and low frequencies. An obvious consequence of (4) is the fact that the longitudinal optical phonons, in contradiction to the results of[5], contribute to the cross section of the Raman scattering even in the simple semiconductor two-band model.

In a number of semiconductors, the frequency of the optical phonon is larger than the frequencies of the transitions between the exciton levels, so that we put approximately in (3) $|\omega_{mm_1}| \sim \omega_q$, and also neglect the dispersion of the optical phonons, obtaining as a result[1)]

$$M_{lk}=\sum_{m,t}\left\{\frac{1}{\hbar^{n+1}\omega_0^n(\omega_2-\omega_{m0})}(\mathbf{P}\mathbf{e}_2^*)_{0m}\left(\prod_{i=1}^{n}2C_{\mathbf{q}_i}\sin\left(\frac{\mathbf{q}_i\rho}{2}\right)\right)_{mt}(\mathbf{P}\mathbf{e}_1)_{t0}\right.$$
$$\left.+\frac{(-1)^n}{\hbar^{n+1}\omega_0^n(\omega_1-\omega_{m0})}(\mathbf{P}\mathbf{e}_2^*)_{0t}\left(\prod_{i=1}^{n}2C_{\mathbf{q}_i}\sin\left(\frac{\mathbf{q}_i\rho}{2}\right)\right)_{tm}(\mathbf{P}\mathbf{e}_1)_{m0}\right\}, \tag{6}$$

where $\omega_0 \approx \omega_g$ and the indices 0, m, and t number the electron wave functions; in particular, Φ_0 is the wave function of the system of electrons in the ground state, In (6) we took into account the momentum conservation law, i.e., in the dipole approximation

$$\sum_{i=1}^{n}\mathbf{q}_i=0. \tag{7}$$

The summation over all the final states in (1) reduces to summation over all the directions of the emitted phonons when the condition (7) is satisfied. We replace the summation by integration up to q_M, where q_M is the maximum wave vector for which it is still possible to neglect the dispersion of the optical phonons, and we take (7) into account, approximately, by integrating only with respect to $\mathbf{q}_2\ldots\mathbf{q}_n$. We get

$$\frac{\partial\sigma}{\partial o}=\frac{e^4\omega_2}{m^4c^4\omega_1}\left[\frac{2e^2}{\hbar\omega_0}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\right]\frac{\pi}{\hbar^2v_0^2}$$
$$\times\sum_{mm'tt'}\left\{(\mathbf{P}\mathbf{e}_2^*)_{0m}(\mathbf{P}\mathbf{e}_1)_{m'0}(\mathbf{P}\mathbf{e}_2^*)_{0t}(\mathbf{P}\mathbf{e}_1)_{t'0}\right.$$
$$\times W_{mt'm't}\left[\frac{1}{(\omega_1-\omega_{m'0})(\omega_1-\omega_{t'0})}+\frac{1}{(\omega_2-\omega_{m0})(\omega_2-\omega_{t0})}\right. \tag{8}$$
$$\left.\left.+\frac{(-1)^n}{(\omega_1-\omega_{m'0})(\omega_2-\omega_{t0})}+\frac{(-1)^n}{(\omega_2-\omega_{m0})(\omega_1-\omega_{t'0})}\right]\right\},$$

$$W_{mt'm't}=\left\langle v_m(\mathbf{r}_1)v_t'(\mathbf{r}_2)\right|\left(\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}-\frac{1}{|\mathbf{r}_1+\mathbf{r}_2|}\right)^{n-1}\frac{1}{q_1^2}(\mathbf{q}_1\mathbf{r}_1)(\mathbf{q}_1\mathbf{r}_2)\Big|$$
$$\times v_m(\mathbf{r}_1)v_t(\mathbf{r}_2)\Big\rangle. \tag{9}$$

Here $v_m(\mathbf{r})$ are hydrogenlike wave functions.

At first glance expression (9) diverges, but when the integral in (9) is calculated it is necessary to replace the divergent terms $|\mathbf{r}_1+\mathbf{r}_2|^{-1}$ by $\pi^{-}q_M$, and then (9) becomes finite. The matrix elements of the operator P between the ground state and the exciton levels have different values for transitions to the s- and p-states of the exciton, and also decrease strongly with increasing principal quantum number (see, for example,[6]). In particular, the matrix elements for the transition to the s-state are larger by $a_0^{-1}a_1$ times than for the transitions to p-states. Since two of the four states m, m′, t, and t′ in (8) and (9) should be p-states in the case of odd n, this leads to alternation of the scattering cross section as a function of the order of the Raman process, namely

$$\frac{\partial\sigma^{(2k-1)}/\partial o}{\partial\sigma^{(2k)}/\partial o}\approx\frac{a_0^4\partial\sigma^{(2k)}/\partial o}{a_1^4\partial\sigma^{(2k+1)}/\partial o}, \tag{10}$$

if it is assumed that the resonant denominator (8) does not change appreciably when the order of the scattering changes. The superior indices denote here the order of the scattering (the number of appearing phonons).

Let us consider the case of even n. In this case one can take the 1s state of the exciton as the intermediate state m, m′, t, or t′. Calculating the integral in (9) by expanding the integrand in spherical harmonics and using the explicit expressions for the matrix elements of the operator P, we obtain

$$\frac{\partial\sigma}{\partial o}=\frac{Ne^4\omega_2}{v_0m^4c^4\omega_1}|(\mathbf{p}\mathbf{e}_2)_{cv}(\mathbf{p}\mathbf{e}_1)_{cv}|^2\left[\frac{2e^2q_M}{\pi\hbar\omega_0}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\right]^n$$
$$\times\frac{\pi}{2(n-3)q_M^3a_1^3}\frac{\Omega_0^2N}{a_1^3v_0}\left[\frac{1}{(\hbar\omega_1-E_g+E_1)^2}+\frac{1}{(\hbar\omega_2-E_g+E_1)^2}\right.$$
$$\left.+\frac{2}{(\hbar\omega_1-E_g+E_1)(\hbar\omega_2-E_g+E_1)}\right] \tag{11}$$

when n > 2. When n = 2 we have

$$\frac{\partial\sigma}{\partial o}=\frac{Ne^4\omega_2}{v_0m^4c^4\omega_1}|(\mathbf{p}\mathbf{e}_2)_{cv}(\mathbf{p}\mathbf{e}_1)_{cv}|^2\left[\frac{2e^2}{\pi\hbar\omega_0a_1}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\right]^2$$
$$\times\frac{\pi\Omega_0^2N}{2a_1^3v_0}\left[\frac{1}{(\hbar\omega_1-E_g+E_1)^2}+\frac{1}{(\hbar\omega_2-E_g+E_1)^2}\right.$$
$$\left.+\frac{2}{(\hbar\omega_1-E_g+E_1)(\hbar\omega_2-E_g+E_1)}\right]. \tag{12}$$

Here p_{cv} is the matrix element of the momentum operator for transitions between valence and conduction bands at k = 0, N is the total number of electrons, Ω_0 is the volume of the unit cell, E_g is the width of the forbidden band, and E_1 is the energy of the exciton in state 1s, reckoned from the bottom of the conduction band. When n is odd there is an additional small factor of the order of $a_1^{-2}a_0^2$, and therefore we do not present the explicit expression for the Raman-scattering cross section for this case. However, relation (10) is valid only if $\omega_0 \gg \omega_{exc}$, where ω_{exc} is the frequency of the transition between the exciton levels; this condition apparently is rarely satisfied. In the case when

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[1)] We have retained in (6) only terms with $\nu = 0$ and $\nu = n$, which give the largest contribution to the scattering cross section. As a result of the momentum conservation law, the phonon momentum should be equal to the exciton momentum, and it is necessary to sum over all the wave vectors of the exciton. In this case there appears in the denominators of the terms with $\nu \neq 0$, n a large frequency, equal, accurate to Planck's constant, to the kinetic energy of the exciton on the boundary of the Brillouin zone. In the terms with $\nu = 0$ and n, only transitions to the intermediate exciton states with zero wave vector are possible, and there is no need for summing over the Brillouin zone.

$\omega_0 > \omega_{exc}$ we can obtain for the scattering cross section a much larger value at odd $n > 1$, by expanding $\omega_{mm_1}^{-1}$ in (3) in powers of $\omega_0^{-1}\omega_{exc}$ and confining ourselves to terms proportional to $\omega_0^{-1}\omega_{exc}$,

$$M_{lk} = M_{lk}{}^0 + \frac{i}{\hbar^{n+1}\omega_0{}^n n!}\sum_{m,m'}\left\{\frac{(\mathbf{P}\mathbf{e}_2{}^*)_{lm}(\mathbf{P}\mathbf{e}_1)_{m'k}}{\omega_2+\omega_{lm}}\right.$$

$$\times\sum_{\nu=1}^{n}\frac{1}{\nu}\left(\frac{d(V_1\ldots V_\nu)}{dt}V_{\nu+1}\ldots V_n\right)_{mm'} -$$

$$\left. - \frac{(\mathbf{P}\mathbf{e}_2{}^*)_{lm'}(\mathbf{P}\mathbf{e}_1)_{mk}}{\omega_1+\omega_{km}}\sum_{\nu=1}^{n}\frac{1}{\nu}\left(\frac{d(V_1\ldots V_\nu)}{dt}V_{\nu+1}\ldots V_n\right)_{m'm}\right\}, \tag{13}$$

where

$$V_\nu = 2C_{\mathbf{q}_\nu}\sin\left(\frac{\mathbf{q}_\nu\rho}{2}\right)\exp(-i\mathbf{q}_\nu\mathbf{R} - i\alpha\mathbf{q}_\nu\rho),$$

M_{ik}^0 is given by formula (6).

Calculation of (13) for arbitrary n is a complicated matter, and we therefore consider only the case n = 3. Using the explicit expression for the derivative of the operator with respect to the time, and also summing over the final states in (1), we obtain the following expression for the scattering cross section:

$$\frac{\partial\sigma}{\partial o} = \frac{Ne^4\omega_2}{v_0 m^4 c^4\omega_1}|(\mathbf{p}\mathbf{e}_2)_{cv}(\mathbf{p}\mathbf{e}_1)_{cv}|^2\left[\frac{2e^2 q_M}{\pi\hbar\omega_0}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\right]^3$$

$$\times\frac{\Omega_0{}^2 q_M N}{a_1{}^3 v_0}\frac{\hbar^2\alpha^2}{4\mu^2 a_1{}^4\omega_0{}^2}\left[\frac{1}{(\hbar\omega_2-E_g+E_1)^2}+\frac{1}{(\hbar\omega_1-E_g+E_1)^2}\right.$$

$$\left.-\frac{2}{(\hbar\omega_1-E_g+E_1)(\hbar\omega_2-E_g+E_1)}\right], \tag{14}$$

where $\mu^{-1} = m_1^{-1} + m_2^{-1}$. We see that compared with (11), there have appeared additional factors of the order of $\hbar^2/4\mu^2 a_1^4\omega_0^2 \approx \omega_{exc}^2/\omega_0^2$; in addition, there is an additional factor proportional to the mass difference of the electron and hole.

Let us consider now scattering with participation of impurity electron states as intermediate states. In the interaction between the phonons and weakly bound impurity electrons, the momentum conservation law is not satisfied, since the impurity atom is rigidly bound to the main matrix. This makes Eq. (7) no longer valid, and the number of final states in the summation of (1) increases. It turns out as a result that the scattering cross section has an appreciable magnitude even at not very large impurity concentrations. The scattering cross section is likewise independent of the parity of the order of the Raman process.

Under the same assumptions as made in the derivation of (6)[2)], we get

$$M'_{lk} = \frac{C_{\mathbf{q}_1}\ldots C_{\mathbf{q}_n}}{\hbar^{n+1}\omega_0{}^n}\sum_{\nu=0}^{n}\left\{(-1)^\nu\binom{n}{\nu}\sum_{t,m,p}(\mathbf{P}\mathbf{e}_2{}^*)_{0t}\left(\exp\left[i(\mathbf{R}_i-\mathbf{r})(1\right.\right.\right.$$

$$\left.\left.-\delta_{\nu 0})\sum_{j=1}^{\nu}\mathbf{q}_j\right]\right)_{tm}\left(\exp\left[i(\mathbf{R}_i-\mathbf{r})(1-\delta_{\nu n})\sum_{j=\nu+1}^{n}q_j\right]\right)_{mp}(\mathbf{P}\mathbf{e}_1)_{p0}$$

$$\left.\times\frac{1}{\omega_2+\nu\omega_0-\omega_{m0}}\exp\left[-i\mathbf{R}_i\sum_{j=1}^{n}q_j\right]\right\}, \tag{15}$$

where $\mathbf{R}_i$ is the coordinate of the impurity atom and δ_{ji} is the Kronecker symbol. Substituting (15) in (1) and summing over all the impurity atoms, and also over all the final states, we obtain

$$\frac{\partial\sigma}{\partial o} = \frac{N_d e^4\omega_2}{v_0 m^4 c^4\omega_1}\left[\frac{e^2}{\pi\hbar\omega_0}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\right]^n\frac{\pi^4}{2^4 a_2{}^4 q_M{}^4}$$

$$\times\sum_{m,p,t,m',p',t'}\sum_{\nu\geq\nu'=0}^{n}(\mathbf{P}\mathbf{e}_2{}^*)_{0t}(\mathbf{P}\mathbf{e}_1)_{p0}(\mathbf{P}\mathbf{e}_2{}^*)_{0t'}^*(\mathbf{P}\mathbf{e}_1)_{p0}^*$$

$$\times R_{m\,t'\,p\,m'}^{t\,m'\,m\,p'}(\nu,\nu')(-1)^{\nu+\nu'}\,.$$

$$\times(2-\delta_{\nu\nu'})\binom{n}{\nu}\binom{n}{\nu'}\frac{1}{\omega_2+\nu\omega_0-\omega_{m0}}\frac{1}{\omega_2+\nu'\omega_0-\omega_{m'0}}, \tag{16}$$

where N_d is the total number of impurity atoms, and

$$R_{mt'pm'}^{tm'mp'}(\nu,\nu') = a_2{}^4\left(\frac{\pi}{2}\frac{1}{q_M}\right)^{n-4}\left\langle v_t(\mathbf{r}_1)v_{m'}(\mathbf{r}_2)v_m(\mathbf{r}_3)v_{p'}(\mathbf{r}_4)\right.$$

$$\left.\times\left|\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|^{\nu'}}\frac{1}{|\mathbf{r}_3-\mathbf{r}_4|^{n-\nu}}\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|^{\nu-\nu'}}\right|v_m(\mathbf{r}_1)v_{t'}(\mathbf{r}_2)v_p(\mathbf{r}_3)v_{m'}(\mathbf{r}_4)\right\rangle. \tag{17}$$

The matrix element of the transition of the momentum operator between the Bloch wave function of the valence band with quasimomentum k and the wave function of the localized state is equal to

$$\mathbf{p}_{cv}v_0^{-1/2}\int v(\mathbf{r})\,d\mathbf{r} \tag{18}$$

when $ka_2 \ll 1$. In the opposite case when $ka_2 \gg 1$ the matrix elements become small, since the wave function of the localized state is constructed of wave functions of the conduction band with $k < a_2^{-1}$. We shall therefore consider only transition to the 1s state of the impurity atom in accordance with (18), and the summation over the Brillouin zone will be carried out only up to $k_M = a_2^{-1}$. After performing other calculations, we obtain

$$\frac{\partial\sigma}{\partial o} = \frac{N_d e^4\omega_2}{v_0 m^4 c^4\omega_1}\left[\frac{e^2 q_M}{\pi\hbar\omega_0}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\right]^n|(\mathbf{p}\mathbf{e}_2)_{cv}(\mathbf{p}\mathbf{e}_1)_{cv}|^2\frac{a_2{}^4 m_1{}^2}{\hbar^4}\frac{m_2{}^2}{m_1{}^2}$$

$$\times\frac{8^4\pi^2}{2^4}\sum_{\nu\geq\nu'=0}^{n}(-1)^{\nu+\nu'}\binom{n}{\nu}\binom{n}{\nu'}(2-\delta_{\nu\nu'})R_{m,1s,1s,m'}^{1s,m',m,1s}(\nu,\nu')$$

$$\times F(\hbar\omega_2+\hbar\nu\omega_0-E_g+E_m')F(\hbar\omega_2+\hbar\nu'\omega_0-E_g+E_m'), \tag{19}$$

where

$$F(x) = 1-\left|\frac{2m_2a_2{}^2x}{\hbar^2}\right|^{1/2}\operatorname{arctg}\left|\frac{\hbar^2}{2m_2a_2{}^2x}\right|^{1/2}, \tag{20}$$

E_m' is the energy of the impurity level, reckoned from the bottom of the conduction band. The large numerical coefficient $8^4\pi^2$ is a consequence of the fact that

$$\int v_{1s}(r)\,dr = 8(\pi a_2{}^3)^{1/2}.$$

The expression $R_{m,1s,1s,m'}^{1s,m',m,1s}(\nu,\nu')$ is normalized in such a way that if m and m′ are 1s states, then $R_{m,1s,1s,m'}^{1s,m',m,1s}(\nu,\nu) \approx 1/4$ when $\nu > 0$. We note in this case that the divergent expression $|\mathbf{r}_1 = \mathbf{r}_2|^{-1}$ must be replaced in the calculation of (17) by $2\pi^{-1}q_M$.

It follows from the uncertainty principle that $\hbar^2 a_2^{-2} m_1^{-1} \approx E_1'$. Putting in (11) $\hbar\omega_2 - E_g + E_1 \approx E_1'$ and comparing with (19), we find that the ratio of the Raman-scattering cross section with allowance for the exciton states as intermediate states to the cross section of Raman scattering with allowance for the impurities has at n = 4 the following order of magnitude

$$\gamma = \frac{1}{8\pi}\frac{N}{N_d}\frac{\Omega_0}{a_1{}^3}; \tag{21}$$

It is assumed that $q_M a_1 \approx 1$ and $a_1 \approx a_2$.

In[1,2] there was observed multiphonon Raman scat-

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2) However, all the terms in (3) with $\nu \neq 0$ or n are taken into account.

tering of light up to ninth order in CdS, and furthermore under conditions when the frequency ω_2 fell in the region of exciton absorption. For CdS the calculations performed in[7] show that one can choose for q_M^{-1} a value on the order of $a \approx 30$ A. In this case we get

$$\frac{2e^2}{\pi\hbar\omega_0 a_1}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\approx 1, \qquad (22)$$

so that the dependence of the scattering cross section on the frequency should be determined by the Lorentz curve $(\hbar\omega_2 - E_g + E_1)^{-2}$ for even n. For odd n there is an additional small factor of the order of 10^{-1}. This peculiarity is apparently not observed in the experiment. We note that in the case of scattering with impurities taking part we have

$$\frac{e^2}{\pi\hbar\omega_0 a_2}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\approx \frac{1}{2}$$

when $a_1 \approx a_2$, and the scattering cross section decreases with increasing n more rapidly than in the case of scattering with excitons taking part. However, for impurities the scattering cross section does not depend on the parity of the number of resultant phonons.

For CdS we have $\gamma = 3 \times 10^{-5} N_d^{-1} N$ at $n = 4$, so that in order for the scattering cross section with participation of impurities to be comparable with the scattering cross section with participation of excitons, the impurity concentration must be of the order of 3×10^{17} cm^{-3}.

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