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THEORY OF EXCITATION OF EXCITONS AND PHONONS BY A STRONG ELECTROMAGNETIC FIELD

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A theory of second-order absorption involving the excitation of two quasiparticles in a strong electromagnetic field has been developed. The dependence of the susceptibility of such a process on the electric field has been obtained. Such a dependence appears in fields which are, generally speaking, much weaker than atomic fields. The characteristic critical field is estimated for different cases: excitation of two phonons, of a Frenkel exciton and an intramolecular phonon, of a Wannier-Mott exciton and of optical and acoustic phonons.

1. INTRODUCTION

THE most frequently occurring process of absorption of light in a medium can be described in the following manner. A photon excites an atom, a molecule or some quasiparticle in a solid (electron, exciton, phonon etc.), and this quasiparticle in its turn distributes its energy over the numerous degrees of freedom of a solid (for example, excites phonons with different propagation vectors), and this leads to heating of the medium. At the same time it follows from the conservation laws that the photon energy $\hbar\omega$ coincides (within the limits of the width of the absorption line) with the energy of the quasiparticle

$$\hbar\omega = \hbar\omega(\mathbf{k}j), \tag{1}$$

while the propagation vector of the photon $\mathbf{q}$ coincides with the propagation vector of the quasiparticle

$$\mathbf{q} = \mathbf{k}. \tag{2}$$

If we take into account the circumstance that the propagation vector of the photon (in optics and at lower frequencies) is much smaller than $1/a$, where a is the period of the lattice, then it follows from this that the usual process of absorption involves quasiparticles in a very narrow long wavelength part of the Brillouin zone ($\mathbf{k} \approx 0$).

At the same time one can demonstrate many examples of absorption processes of a different type (in view of the lack of a unified terminology we shall refer to them as second order absorption processes). In the case of this type of absorption the photon excites two quasiparticles at once: phonon-phonon ([1], p. 419), magnon-magnon[2], exciton-phonon (indirect transitions in semiconductors, cf.,[3], p. 166). The conservation laws in this case have the form

$$\hbar\omega = \hbar\omega(\mathbf{k}_1 j_1) \pm \hbar\omega(\mathbf{k}_2 j_2), \qquad \mathbf{q} = \mathbf{k}_1 \pm \mathbf{k}_2, \tag{3}$$

where we have denoted by $\hbar\omega(\mathbf{k}j)$ the energy of the quasiparticle of the j-th kind with propagation vector $\mathbf{k}$.

The study of second order absorption enables us to obtain information on quasiparticles with propagation vectors over the whole Brillouin zone (as follows from relations (3)).

In this paper we consider second order absorption in a strong electromagnetic field when the change in the absorption coefficient under the action of this field is significant. Before constructing a rigorous theory in subsequent sections we present simple considerations indicating the existence of such a dependence. For the sake of definiteness we consider the case when the photon frequency is equal to the sum of the frequencies ω_1 and ω_2 of quasiparticles 1 and 2: $\omega = \omega_1 + \omega_2$. The number of photons n_{ph} absorbed per unit time, as can be seen to follow from simple balance equations, is equal to

$$-\dot{n}_{\mathrm{ph}} = w[n_{\mathrm{ph}}(n_1+1)(n_2+1) - n_{\mathrm{ph}}n_1n_2] \approx wn_{\mathrm{ph}}(n_1+n_2+1). \tag{4}$$

Here we have neglected the term wn_1n_2 since according to our assumption $n_{\mathrm{ph}} > n_1, n_2$. (This term essentially describes the spontaneous emission of two excited quasiparticles $n_1 \neq 0$ and $n_2 \neq 0$.)

Usually the experimentally observed second order absorption is described by the right hand side of (4) with n_1 and n_2 equal to their thermodynamic average values n_1^0 and n_2^0. In a sufficiently strong photon field $n_{\mathrm{ph}} \gg 1$ the values of n_1 and n_2 themselves begin to depend on n_{ph}. Indeed, the balance equations for n_1 and n_2 in the same approximation have the form

$$\begin{aligned} n_1 &= -\gamma_1(n_1 - n_1^0) + wn_{\mathrm{ph}}(n_1+n_2+1), \\ n_2 &= -\gamma_2(n_2 - n_2^0) + wn_{\mathrm{ph}}(n_1+n_2+1), \end{aligned} \tag{5}$$

where γ_1 and γ_2 are phenomenologically introduced damping constants. A straightforward analysis of equations (5) (for a given value of n_{ph} shows that in the case

$$wn_{\mathrm{ph}} < \gamma_1\gamma_2 / (\gamma_1 + \gamma_2)$$

there exists a stable stationary solution of these equations:

$$n_1 + n_2 + 1 = \frac{n_1^0 + n_2^0 + 1}{1 - wn_{\mathrm{ph}}(\gamma_1+\gamma_2)/\gamma_1\gamma_2}. \tag{6}$$

From (4) and (6) it follows that for values of wn_{ph} comparable to $\gamma_1\gamma_2/(\gamma_1 + \gamma_2)$ the absorption coefficient depends in an essential manner on the number of photons (or on the square of the amplitude of the field) and increases sharply. (For sufficiently large values of n_1 and n_2 formula (6) ceases to be valid since one must take into account the omitted nonlinear terms $n_1 n_2$.) For $wn_{\mathrm{ph}} > \gamma_1\gamma_2/(\gamma_1 + \gamma_2)$ the stationary solution (6) is unstable and generation of quasiparticles of frequencies ω_1 and ω_2 occurs. This is essentially parametric excitation of oscillations of frequency ω_1 and ω_2 under the action of a pumping signal of frequency $\omega = \omega_1 + \omega_2$.

2. GENERAL CONSIDERATION OF PROCESSES INVOLVING THE EXCITATION OF TWO QUASIPARTICLES

Before we proceed to consider specific second order absorption processes we consider a general scheme which we shall then apply to different cases.

Let the Hamiltonian for a system of quasiparticles interacting with the electric field **E** have the form

$$H = \sum_{\mathbf{k}j} \hbar\omega(\mathbf{k}j)\, a_j^+(\mathbf{k})\, a_j(\mathbf{k}) - \frac{\hbar}{2} \sum_{b,\mathbf{q},\mathbf{k},\mathbf{k}',j,j'} E_b(\mathbf{q})$$

$$\times\Big\{A_b\begin{pmatrix} -\mathbf{k} & -\mathbf{k}' \\ j & j' \end{pmatrix} a_j^+(-\mathbf{k})\, a_{j'}^+(-\mathbf{k}') + A_b^*\begin{pmatrix} \mathbf{k} & \mathbf{k}' \\ j & j' \end{pmatrix} a_j(\mathbf{k})\, a_{j'}(\mathbf{k})$$

$$+ B_b\begin{pmatrix} -\mathbf{k} & \mathbf{k}' \\ j & j' \end{pmatrix} a_j^+(-\mathbf{k})\, a_{j'}(\mathbf{k}')$$

$$+ B_b^*\begin{pmatrix} \mathbf{k} & -\mathbf{k}' \\ j & j' \end{pmatrix} a_j(\mathbf{k})\, a_{j'}^+(-\mathbf{k}) \Big\}\Delta(\mathbf{k} + \mathbf{k}' + \mathbf{q}) \tag{7}$$

and the polarization have the corresponding form

$$P_a = \frac{\hbar}{2} \sum_{\mathbf{k}j,\mathbf{k}'j'} \Big\{\Big[A_a\begin{pmatrix} -\mathbf{k} & -\mathbf{k}' \\ j & j' \end{pmatrix} a_j^+(-\mathbf{k})\, a_{j'}^+(-\mathbf{k}')$$

$$+ B_b\begin{pmatrix} -\mathbf{k} & \mathbf{k}' \\ j & j' \end{pmatrix} a_j^+(-\mathbf{k})\, a_{j'}(\mathbf{k}')\Big] e^{i(\mathbf{k}+\mathbf{k}')\mathbf{r}} + \text{herm. conj.}\Big\} \tag{8}$$

Here A_b and B_b are functions of **k** and j the form of which is determined in each specific case; a_j^+ and a_j are the creation and annihilation operators for particles of the j-th kind. They satisfy the commutation relations for Bose-particles;

$$[a_j(\mathbf{k}),\ a_{j'}^+(\mathbf{k}')] = \delta_{jj'}\Delta(\mathbf{k} - \mathbf{k}').$$

The function $\Delta(\mathbf{x})$ differs from zero and is equal to unity for **x** equal to zero or to one of the reciprocal lattice vectors. It is assumed that all quantities are normalized per unit volume, and $E_b(\mathbf{q})$ is the spatial Fourier component of the electric field.

The Hamiltonian (7) can describe second-order absorption; the terms which are responsible for the usual absorption in (7) have been omitted. From the Hamiltonian (7) follow the equations of motion for the quantities $\langle a_{j_1}(\mathbf{k}_1) a_{j_2}(\mathbf{k}_2)\rangle$ and others which determine the average polarization (8). Solving these equations under the condition that the frequencies of the electric field satisfy

$$\omega \approx \omega_{j_1}(\mathbf{k}_1) + \omega_{j_2}(\mathbf{k}_2), \tag{9}$$

and assuming that this condition is not satisfied for any other values of j (for a given ω) and neglecting small nonresonant terms we obtain

$$\langle a_{j_1}(\mathbf{k}_1)\, a_{j_2}(\mathbf{k}_2)\rangle = -A_b\begin{pmatrix} \mathbf{k}_1 & \mathbf{k}_2 \\ j_1 & j_2 \end{pmatrix} E_b(\mathbf{q}, \omega)\, e^{-i\omega t}$$

$$\times \frac{[n_{j_1}(\mathbf{k}_1) + n_{j_2}(\mathbf{k}_2) + 1]\,\Delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{q})}{\omega - \omega(\mathbf{k}_1 j_1) - \omega(\mathbf{k}_2 j_2) + i[\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{k}_2)]}, \tag{9'}$$

where $\gamma_j(\mathbf{k})$ is a phenomenologically introduced damping constant. Here $n_j(\mathbf{k}) = \langle a_j^+(\mathbf{k}) a_j(\mathbf{k})\rangle$ is the average number of quasiparticles for which in turn one can write equations the solution of which in the same approximation has the form

$$n_{j_1}(\mathbf{k}_1) + n_{j_2}(\mathbf{q} - \mathbf{k}_1) + 1 = \frac{n_{j_1}{}^0(\mathbf{k}_1) + n_{j_2}{}^0(\mathbf{q} - \mathbf{k}_1) + 1}{1 - \eta}, \tag{10}$$

where

$$\eta = \frac{\left|A_b\begin{pmatrix} \mathbf{k}_1 & \mathbf{q} - \mathbf{k}_1 \\ j_1 & j_2 \end{pmatrix}\right|^2 |E_b(\mathbf{q}, \omega)|^2 [\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2}{\gamma_{j_1}(\mathbf{k}_1)\gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)\{[\omega - \omega_{j_1}(\mathbf{k}_1) - \omega_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2 + [\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2\}}, \tag{11}$$

while $n_j^0(\mathbf{k}) = [\exp(\hbar\omega_j(\mathbf{k})/kT) - 1]^{-1}$ is the equilibrium value of the number of quasiparticles.

We further introduce the susceptibility describing the second order process:

$$P_a = \chi_{ab}(\mathbf{q}, \omega)\, E_b(\mathbf{q}, \omega)\, e^{i(\mathbf{qr} - \omega t)}.$$

Then from (8)–(11) we obtain

$$\chi_{ab}(\mathbf{q}, \omega) = -\frac{\hbar}{2} \sum_{\mathbf{k}_1} A_a^*\begin{pmatrix} \mathbf{k}_1 & \mathbf{q} - \mathbf{k}_1 \\ j_1 & j_2 \end{pmatrix} A_b\begin{pmatrix} \mathbf{k}_1 & \mathbf{q} - \mathbf{k}_1 \\ j_1 & j_2 \end{pmatrix}$$

$$\times \frac{\omega - \omega_{j_1}(\mathbf{k}_1) - \omega_{j_2}(\mathbf{q} - \mathbf{k}_1) - i[\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]}{[\omega - \omega_{j_1}(\mathbf{k}_1) - \omega_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2 + [\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2}$$

$$\times \frac{n_{j_1}{}^0(\mathbf{k}_1) + n_{j_2}{}^0(\mathbf{q} - \mathbf{k}_1) + 1}{1 - \eta}. \tag{12}$$

The imaginary part of this expression describes second order absorption which in accordance with the elementary considerations stated in the Introduction increases as η approaches unity.

In an analogous manner we obtain the expression for $\chi_{ab}(\mathbf{q}, \omega)$ for $\omega \approx \omega_{j_1}(\mathbf{k}_1) - \omega_{j_2}(\mathbf{q} - \mathbf{k}_1)$:

$$\chi_{ab}(\mathbf{q}, \omega) = -\frac{\hbar}{4} \sum_{\mathbf{k}_1} \Big[B_a\begin{pmatrix} \mathbf{q} - \mathbf{k}_1 & \mathbf{k}_1 \\ j_2 & j_1 \end{pmatrix} + B_a^*\begin{pmatrix} \mathbf{k}_1 & \mathbf{q} - \mathbf{k}_1 \\ j_1 & j_2 \end{pmatrix}\Big]$$

$$\times\Big[B_b\begin{pmatrix} \mathbf{k}_1 & \mathbf{q} - \mathbf{k}_1 \\ j_1 & j_2 \end{pmatrix} + B_b^*\begin{pmatrix} \mathbf{q} - \mathbf{k}_1 & \mathbf{k}_1 \\ j_2 & j_1 \end{pmatrix}\Big]$$

$$\times \frac{[\omega - \omega(\mathbf{k}_1 j_1) + \omega(\mathbf{q} - \mathbf{k}_1 j_2)] - i\{\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]}{[\omega - \omega(\mathbf{k}_1 j_1) + \omega(\mathbf{q} - \mathbf{k}_1 j_2)]^2 + [\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2}$$

$$\times \frac{n_{j_2}{}^0(\mathbf{q} - \mathbf{k}_1) - n_{j_1}{}^0(\mathbf{k}_1)}{1 \pm \eta_1}, \tag{13}$$

where $\omega(\mathbf{k}_1 j_1) > \omega(\mathbf{q} - \mathbf{k}_1 j_2)$ and the plus sign refers to the usual equilibrium situation when $n_{j_1}^0 > n_{j_2}^0$, while the minus sign occurs in the case of the "inverted" situation when $n_{j_1}^0 < n_{j_2}^0$. In the latter case "negative absorption" or induced emission occurs at a frequency ω. The quantity η_1 has the form

$$\eta_1 = \left|B_b\begin{pmatrix} \mathbf{q} - \mathbf{k}_1 & \mathbf{k}_1 \\ j_2 & j_1 \end{pmatrix} + B_b^*\begin{pmatrix} \mathbf{k}_1 & \mathbf{q} - \mathbf{k}_1 \\ j_1 & j_2 \end{pmatrix}\right|^2$$

$$\times |E_b(\mathbf{q}, \omega)|^2 [\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2$$

$$\times \{4\gamma_{j_1}(\mathbf{k}_1)\gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)\}^{-1}\{[\omega - \omega(\mathbf{k}_1 j_1) + \omega(\mathbf{q} - \mathbf{k}_1 j_2)]^2$$

$$+ [\gamma_{j_1}(\mathbf{k}_1) + \gamma_{j_2}(\mathbf{q} - \mathbf{k}_1)]^2\}^{-1}. \tag{14}$$

In the usual equilibrium case an increase in the field $|\mathrm{E}|^2$ leads to a decrease in absorption—i.e. to saturation: the absorbed power in a sufficiently strong field $\eta_1 \gg 1$ ceases to depend on the field, while the imaginary part $\chi''_{ab}(\mathbf{q}, \omega)$ is inversely proportional to the square of the absolute value $|E_b(\mathbf{q}, \omega)|^2$. Here and subsequently we take $E(\mathbf{q}, \omega)$ to denote the external macroscopic field of the electromagnetic wave in a crystal. This means that in calculating the energy spectrum of the quasiparticles we should not take into account the long wavelength electromagnetic field (i.e., we are dealing with ''mechanical excitons'' if we utilize the terminology of Agranovich and Ginzburg[4]). This also means that q and ω are independent variables, and the relation between these two quantities arises in the course of the solution of the problem of the characteristic waves in the crystal.

However, it should be kept in mind that the essential difference between real and mechanical excitons will be apparent only in the narrow long-wavelength part of the Brillouin zone and, correspondingly, only in a narrow part of the zone of two-particle absorption. Moreover, since, generally speaking, second-order absorption is not associated with the points of intersection of the dispersion curves for photons and quasiparticles the macroscopic field of the wave in the crystal at a frequency (3) may be not significantly different from the field in vacuo.

We further note that the occurrence of absorption at this frequency leads to the complex nature of the vector $q = q_1 + q_2$. The investigation carried out above, strictly speaking, is valid for $q_2 \ll q_1$ (i.e., when absorption over one wavelength is small). However, in the practically interesting cases when $q \ll 1/a$ and we neglect spatial dispersion, q can be set equal to zero and we can keep in mind the fact that the solutions so obtained are valid subject to a very weak restriction on the magnitude of the absorption $q_2 \ll a^{-1}$.

In an analogous manner we can carry out a general investigation of the second order Raman effect—the Raman-induced scattering involving the excitation of two quasiparticles. In this case in the Hamiltonian (7) one must replace $E_b(q)$ by $E_b(q)E_c(q')$ and, correspondingly, instead of A_b one must introduce the quantity A_{bc}. The theory of the second order induced Raman effect involving phonons has been given by Genkin, Faĭn, and Yashchin[5].

We now proceed to a discussion of different specific cases of second order absorption.

3. EXCITATION OF PHONONS

The phonon Hamiltonian describing the processes in which we are interested can be written in the adiabatic approximation in the form (cf., for example,[1], Secs. 17 and 21, and[5])

$$\begin{aligned} H = \frac{1}{2} \sum_{\mathbf{k}j} [P(\mathbf{k}j)P(-\mathbf{k}j) + \omega^2(\mathbf{k}j)Q(\mathbf{k}j)Q(-\mathbf{k}j)] \\ + \frac{1}{3} \sum_{\mathbf{k}_1 j_1,\, \mathbf{k}_2 j_2,\, \mathbf{k}_3 j_3} \Phi(\mathbf{k}_1 j_1, \mathbf{k}_2 j_2, \mathbf{k}_3 j_3) Q(\mathbf{k}_1 j_1) Q(\mathbf{k}_2 j_2) \\ \times Q(\mathbf{k}_3 j_3) \Delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3) - \sum_{\mathbf{k}j,\, \mathbf{q}} M_a(\mathbf{k}j) Q(\mathbf{k}j) E_b(\mathbf{q}) \Delta(\mathbf{q} + \mathbf{k}) \\ - \frac{1}{2} \sum_{\mathbf{k}_1 j_1,\, \mathbf{k}_2 j_2,\, \mathbf{q}} M_a(\mathbf{k}_1 j_1, \mathbf{k}_2 j_2) Q(\mathbf{k}_1 j_1) Q(\mathbf{k}_2 j_2) E_a(\mathbf{q}) \Delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{q}), \end{aligned} \tag{15}$$

where Q(**k**j) and P(**k**j) are the normal coordinates and momenta describing the j-th branch of oscillation; Q(**k**j) is expressed in terms of the creation and annihilation operators:

$$Q(\mathbf{k}j) = \sqrt{\frac{\hbar}{2\omega(\mathbf{k}j)}} [a(\mathbf{k}j) + a^+(-\mathbf{k}j)]. \tag{16}$$

Treating the field **E** as given and considering for the sake of simplicity the most important case **q** = 0 we express Q(0j) (for the optical branches) in terms of $E_b(\omega)e^{-i\omega t}$ and substitute it into the energy terms describing anharmonism (the second sum in (15)). Further utilizing formula (16) we reduce the energy of interaction with the field $E_b(\omega)e^{-i\omega t}$ to the form (7) where now we have

$$\begin{aligned} A_b \begin{pmatrix} \mathbf{k} & \mathbf{k}' \\ j & j' \end{pmatrix} = \frac{M_b(\mathbf{k}j, \mathbf{k}'j')}{2\sqrt{\omega(\mathbf{k}j)\omega(\mathbf{k}'j')}} \\ - \sum_{j_1} \frac{\Phi(0j_1, \mathbf{k}j, \mathbf{k}'j') M_b(0j')}{[\omega^2(0j_1) - \omega^2 - i\omega\gamma_{j_1}(0)]\sqrt{\omega(\mathbf{k}j)\omega(\mathbf{k}'j')}}. \end{aligned} \tag{17}$$

The second order absorption associated with the first term in (17) was discussed in[5], and that associated with the second term (involving acoustic phonons) was discussed in[6]. Here we give only the estimates for the critical field E_{cr} for which η in (11) and η_1 in (14) become close to unity, while the absorption coefficient depends in an essential manner on the field. In accordance with[5,7] the estimates for the coefficients Φ and M_a have the following form:

$$\begin{aligned} M_a(\mathbf{k}j) \sim \omega(\mathbf{k}j), \qquad M_a(\mathbf{k}j, \mathbf{k}'j') \sim \omega(\mathbf{k}j)\omega(\mathbf{k}'j')E_{at}^{-1}, \\ \Phi(\mathbf{k}j, \mathbf{k}'j', \mathbf{k}''j'') \sim \omega(\mathbf{k}j)\omega(\mathbf{k}'j')\omega(\mathbf{k}''j'')E_{at}^{-1}, \end{aligned} \tag{18}$$

where $E_{at} \sim e/a^2$, e is the electron charge, a is the lattice period. From this and from (11) and (14) (for $\eta \approx 1$) we obtain

$$\begin{aligned} E_{cr} \sim E_{at} \left[\frac{\gamma_j(\mathbf{k})\gamma_{j'}(-\mathbf{k})}{\omega(\mathbf{k}j)\omega(-\mathbf{k}j')} \right]^{1/2} \quad \text{for} \quad \omega^2 < \omega^2(0j), \\ E_{cr} \sim E_{at} \left[\frac{\gamma_j(\mathbf{k})\gamma_{j'}(-\mathbf{k})}{\omega(\mathbf{k}j)\omega(-\mathbf{k}j')} \right]^{1/2} \frac{\gamma_j(0)}{\omega(0j')} \quad \text{for} \quad \omega^2 \sim \omega^2(0j). \end{aligned} \tag{19}$$

4. EXCITATION OF FRENKEL EXCITONS AND INTRAMOLECULAR PHONONS IN MOLECULAR CRYSTALS

In the papers by Rashba, Broude, and Sheka[8,9] in order to explain the spectra of a number of molecular crystals a theory of second order absorption has been developed involving the excitation of electron and intramolecular vibrational excitations. Roughly speaking, this case corresponds to a transformation of a photon into an intramolecular electron-vibrational excitation which dissociates into an exciton and a phonon which propagate through the crystal independently of each other. (Under certain conditions a bound electron-vibrational excitation appears which propagates through the crystal as a single particle.) In the theory of Rashba[8] the energy of interaction with the electric field is determined mainly by the dipole moment operator which corresponds to a transition in an isolated molecule

$$\mathbf{d}_n = \mathbf{d}\psi_n\varphi_n + \mathbf{d}^*\psi_n^+\varphi_n^+,$$

where **d** is the matrix element for the electron-vibrational transition within the molecule n; ψ_n^+ is the creation operator for an electronic excitation and φ_n^+ is the creation operator for a vibrational excitation.

Making the transition to the momentum representation[1]

$$\psi_{\mathbf{n}} = N^{-1/2}\sum_{\mathbf{k}}{}' \psi_{\mathbf{k}} e^{i\mathbf{kn}}, \qquad \varphi_{\mathbf{n}} = N^{-1/2}\sum_{\mathbf{k}} \varphi_{\mathbf{k}} e^{i\mathbf{kn}},$$

we introduce the polarization operator

$$\mathbf{P}(\mathbf{r}, t) = \sum_{\mathbf{kk'}}{}' \mathbf{d}\psi_{\mathbf{k}}\varphi_{\mathbf{k'}} e^{i(\mathbf{k}+\mathbf{k'})\mathbf{r}} + \text{herm. conj.} \qquad (20)$$

Thus, comparing (20) with (8) we arrive at the conclusion that the role of A_b in this case is played by

$$A_b\begin{pmatrix} \mathbf{k} & \mathbf{k'} \\ j & j' \end{pmatrix} = \frac{2d_b}{\hbar}$$

and, in accordance with (11), we obtain for the critical field introduced in the preceding sections

$$E_{\text{cr}} \sim \hbar\sqrt{\gamma_e(\mathbf{k})\gamma_f(-\mathbf{k})} / |d|, \qquad (21)$$

where $\gamma_e(\mathbf{k})$ and $\gamma_f(\mathbf{k})$ are the damping constants for the exciton and the phonon. For a matrix element for the intramolecular transition of order a the critical field is of order

$$E_{\text{cr}} \sim E_{at}\sqrt{\gamma_e(\mathbf{k})\gamma_f(-\mathbf{k})} / \omega_e, \qquad (22)$$

where ω_e is the characteristic frequency for an electronic intramolecular transition.

5. EXCITATION OF WANNIER-MOTT EXCITONS AND PHONONS

We now proceed to a discussion of the possible dependence of the absorption coefficient on the excitation of Wannier-Mott excitons and phonons. We shall be dealing with the so-called indirect transitions in semiconductors. The theory of such transitions for weak electric fields has been developed by Elliot[10] (cf., also[3], p. 153).

The Hamiltonian describing excitons and phonons interacting with one another in an external electric field has the form

$$\begin{aligned} H = {} & \sum_{\mathbf{k}\nu} \hbar\omega(\mathbf{k}\nu) b_\nu^+(\mathbf{k}) b_\nu(\mathbf{k}) + \sum_{\mathbf{f}\sigma} \hbar\omega(\mathbf{f}\sigma)[\alpha_\sigma^+(\mathbf{f})\alpha_\sigma(\mathbf{f}) + 1/2] \\ & + \sum_{\mathbf{k}\nu,\,\mathbf{k'}\nu',\,\sigma\mathbf{f}} i\Delta(\mathbf{f}-\mathbf{k}+\mathbf{k'}) G_{\sigma\mathbf{f}}(\nu\mathbf{k}, \nu'\mathbf{k'}) b_\nu^+(\mathbf{k}) b_{\nu'}(\mathbf{k'})[\alpha_\sigma(\mathbf{f}) - \alpha_\sigma^+(-\mathbf{f})] \\ & - \sum_{\nu b} [M_b(\nu) b_\nu + M_b^+(\nu) b_\nu^+] E_b, \end{aligned} \qquad (23)$$

where $b_\nu^+(\mathbf{k})$ is the creation operator for an exciton in the ν-th exciton band with propagation vector $\mathbf{k}$, $\alpha_\sigma^+(\mathbf{f})$ is the creation operator for a phonon of the σ-th branch with the propagation vector $\mathbf{f}$; the constant $M_b(\nu) = \sqrt{N} d_{b\nu_0}$, where N is the number of cells per unit volume, $d_{b\nu_0}$ is the matrix element of the dipole moment for the transition $0 \rightarrow \nu$ for one exciton.

The discussion then proceeds in complete analogy with Sec. 3. The electric field with propagation vector $\mathbf{q} \sim 0$ excites excitons with $k \sim 0$:

$$b_{\nu_-} = \frac{M_b^*(\nu)}{\hbar(\omega_\nu - \omega)} E_b(\omega) e^{-i\omega t}, \qquad b_{\nu_-}^+ = \frac{M_b(\nu)}{\hbar(\omega_\nu + \omega)} E_b(\omega) e^{-i\omega t}, \qquad (24)$$

where the index minus denotes the part of b_ν proportional to $e^{-i\omega t}$ and $\omega_\nu \equiv \omega(\nu 0)$. Substituting (24) into (23) we bring the energy of interaction with the field $E_b(\omega)e^{-i\omega t}$ into the form (7), where now we have

$$A_b^*\begin{pmatrix} -\mathbf{k} & -\mathbf{f} \\ \nu & \sigma \end{pmatrix} = -i\sum_{\nu'} G_{\sigma f}(\nu-\mathbf{k}, \nu' 0)\frac{M_b(\nu')}{\hbar(\omega_{\nu'} - \omega)}.$$

From here it is easy in this case to determine the parameter η (11). We quote the estimate for the critical field (for which $\eta \sim 1$):

$$E_{\text{cr}} \sim \sqrt{\gamma_\nu(\mathbf{k})\gamma_\sigma(-\mathbf{k})} \Big/ \left| \sum_{\nu'} \frac{G_{\sigma f}(\nu-\mathbf{k}, \nu' 0) M_b(\nu')}{\hbar(\omega_{\nu'} - \omega)} \right|. \qquad (24')$$

In order to make estimates we consider the case of the interaction of a Wannier-Mott exciton with acoustic vibrations (cf.,[3], p. 151):

$$G_{\text{acoust}\,\mathbf{k}}(\nu-\mathbf{k}, \nu' 0) = \sqrt{\frac{\hbar}{2MNu}}|\mathbf{k}|^{1/2}[q_h E_{mm} - q_e E_{nn}], \qquad (25)$$

where M is the mass of an elementary cell, u is the velocity of sound, E_{ll} is the deformation potential of the l-th zone, q_h and q_e are overlap coefficients of order unity. For the case of the interaction with optical phonons we have (per unit volume) ([3], p. 151)

$$G_{\text{opt}\,\mathbf{k}}(\nu-\mathbf{k}, \nu' 0) = \sqrt{2\pi\hbar\omega_\sigma e^2}\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right)^{1/2}\frac{|q_e(\mathbf{k}) - q_h(\mathbf{k})|}{|\mathbf{k}|}, \qquad (26)$$

where ϵ_∞ and ϵ_0 are the dielectric constants at high and low frequencies. Estimates obtained utilizing relations (24)—(26) yield for the interaction with acoustic phonons

$$E_{\text{cr}} \sim E_{at}\frac{ea}{d_{\nu 0}}\left[\frac{\gamma_\nu(\mathbf{k})\gamma_\sigma(-\mathbf{k})}{\omega_\nu(\mathbf{k})\omega_\sigma(-\mathbf{k})}\right]^{1/2}\frac{\hbar|(\omega - \omega_\nu)|}{E_{ll}} \qquad (27)$$

and for the interaction with optical phonons

$$E_{\text{cr}} \sim E_{at}\frac{ea}{d_{\nu 0}}\left[\frac{\gamma_\nu(\mathbf{k})\gamma_\sigma(-\mathbf{k})}{\omega_\nu(\mathbf{k})\omega_\sigma(-\mathbf{k})}\right]^{1/2}\frac{a|\mathbf{k}|}{|q_e(\mathbf{k}) - q_h(\mathbf{k})|}\left|\frac{\omega - \omega_\nu}{\omega_\nu}\right|. \qquad (28)$$

From this it can be seen that when ω and $\omega(\nu 0)$ are sufficiently close (but when the whole discussion is still valid) and for $d_{\nu_0} \sim ea$ the critical field may be sufficiently small. The most significant discriminating factor is the square root of the product of the ratios of the damping constants to the frequency. This quantity can be sufficiently small at low temperatures. We also note that the denominator $|q_e - q_h|$ in the right hand side of (28) for small values of $|\mathbf{k}|$ (less than the reciprocal of the exciton dimensions) tends towards zero in the same manner as the numerator $\sim |\mathbf{k}|$.

6. CONCLUSION

We have examined the dependence on the electric field of the absorption coefficient in a second order process when two quasiparticles participate. Such a dependence manifests itself in fields which are, generally speaking, considerably weaker than the atomic fields. The characteristic critical field E_{cr} is of the order

$$E_{at}\left[\frac{\gamma_1(\mathbf{k})\gamma_2(-\mathbf{k})}{\omega_1(\mathbf{k})\omega_2(-\mathbf{k})}\right]^{1/2},$$

where γ_1, ω_1 and γ_2, ω_2 are the damping constants and the frequencies of the corresponding quasiparticles. It is of interest to note that the condition $E > E_{\text{cr}}$ is essentially the condition for the parametric excitation of oscillations at frequencies ω_1 and ω_2 under the action of pumping of frequency $\omega \approx \omega_1 + \omega_2$. In this case oscilla-

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[1] In contrast to[8] we consider for the sake of simplicity the case of a single sublattice.

tions of frequency ω_1 and ω_2 must be generated. An investigation of such generation (which has not been carried out in this paper) is considerably more complicated and requires the inclusion of nonlinear interactions between the oscillations ω_1 and ω_2. In contrast to the theory presented in this paper in this case it is essential to consider not only the interaction with the oscillations $\omega_j(\mathbf{k})$ and $\omega_{j'}(-\mathbf{k})$, but also with oscillations involving a number of other propagation vectors $\mathbf{k}' \neq \mathbf{k}$ which satisfy the resonance condition.

It should be noted that the parametric generation of quasiparticles of the type described above was first experimentally observed in the well known experiments of Damon, Bloembergen and Wang on the saturation of ferromagnetic resonance accompanied by the appearance of an instability of the spin waves-magnons[11]. Utilizing the results of Suhl's theory[12] it is not difficult to make an estimate that the critical magnetic field in this case is equal to

$$H_{\mathrm{cr}} \sim M\gamma_{\mathbf{k}} / \omega_{\mathbf{k}},$$

where M is the saturation magnetization, $N\mu \sim Neav/c \approx E_{at}v/c$; $\gamma_{\mathbf{k}}$ and $\omega_{\mathbf{k}}$ are the damping constant and the frequency of the spin wave; μ is the Bohr magneton, while v/c is the ratio of the velocity of the orbital electron to the velocity of light. Because the condition for ferromagnetic resonance was simultaneously satisfied this field was in fact still smaller by a factor γ/ω_H where γ is the line width for the ferromagnetic resonance, while ω_H is its frequency.

Thus, in ferromagnetic systems the situation is most favorable due to the presence of the factor $v\gamma/c\omega_H$ which is an additional one compared to the processes discussed above. Nevertheless, it should be emphasized that it is quite possible to attain the fields E_{cr} with the aid of lasers. The field E_{cr}, as follows from the estimates given above, is always smaller than the fields E_{at}, while the latter, as is well known, can be attained in a focused laser beam. It is possible in principle to observe the effect described above in all crystals in which second order absorption is observed (in particular, in direct transitions). Difficulties can be associated with the necessity of an exact coincidence of the band of two-particle absorption with the laser frequency, and this sharply reduces the choice of suitable crystals. Experimental difficulties can also arise because of processes leading to a destruction of the crystal in such fields.

In conclusion we note that generation in a Raman-laser is essentially also a process analogous to those considered above. Indeed, here a photon of frequency ω also excites two quasiparticles, one of which is a photon of frequency ω_1, while the other one is a phonon of frequency ω_j: $\omega = \omega_1 + \omega_j$. Here induced emission occurs at the frequency ω_1. From the same point of view we can also discuss the processes described above and consider that at frequencies ω_j and $\omega_{j'}$ $(\omega \approx \omega_j + \omega_{j'})$ induced oscillations (arising in the process of excitation by the fields of oscillations ω_j and $\omega_{j'}$) of frequencies ω_j and $\omega_{j'}$ occur. This same remark also applies to processes with $\omega = \omega_j - \omega_{j'}$.

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