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К теории параметрического взаимодействия волн в экситон-фононных системах пьезоэлектрических кристаллов

Рассматривается параметрическое возбуждение собственных волн в экситон-фононных системах с учетом эластопьезоэлектрических и электрострикционных свойств кристалла. Получены аналитические выражения для инкрементов неустойчивости продольных акустических и экситонных мод в определенных резонансных условиях относительно частот. Показано, что наличие дрейфа электронов может значительно уменьшить пороговое значение внешнего поля излучения в случае усиления акустических волн.

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On the Theory of Parametric Wave Interaction in Exciton-Phonon Systems of Piezoelectric Crystals

The problem of parametric excitation of eigenwaves in exciton-phonon systems is solved taking into account elastopiezoelectric and electrostrictive effects of the crystal. Analytical expressions for the instability growth rates are obtained for longitudinal acoustical and excitonic modes of the system in certain resonant frequency conditions. It is shown that the presence of the electron drift currents may considerably lower the threshold value of the driving radiation field for the instability of acoustical modes.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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1. INTRODUCTION

The parametric interactions in solids, particularly the excitation of eigenwaves under the action of strong electromagnetic radiation fields, have been the subject of many theoretical and experimental studies in recent years.

A quantum approach to the problem of parametric excitation in electron-phonon systems was developed in several works /1-5/. For the acoustical waves the study has been carried out preferently in the aspect of the stimulated Brillouin scattering phenomena (see, for example, /6/ and references therein).

In the recent paper /7/ we have considered the problem of parametric excitation of acoustical modes in crystals where the electron-phonon interaction via the deformation potential as well as the piezoelectric and electrostrictive effects are taken into account. It was also shown that the presence of electron drift currents in such systems may considerably influence the process, and the threshold driving field values for the instability of the acoustical modes may be considerably lowered. This is important in the sence of the possibility of experimental observation of this phenomenon and of the perspective of its application in solid electronics.

It is of certain interest to consider the analogous problem for piezocrystals in excitonic region of spectra where light is coupled to sound not only through electrostrictive and piezoelectric interactions but also through the mechanism of exciton-phonon interaction. This is the purpose of the present paper.

In section 2 the basic equations of the problem are presented. Section 3 is devoted to the detailed analysis of the dispersion equations. The analytical expressions for the instability growth rates of acoustical and exci -

tonic modes of the system are obtained in some resonant conditions concerned the wave frequencies that shows an explicit dependence on parameters characterizing the elastopiezoelectric and electrostrictive properties of the crystal. It is also shown that the influence of the electron drift current on the process of wave amplification is realized in the same way as in the case of parametric excitation of the only acoustical modes.

2. BASIC EQUATIONS

Considering an exciton-acoustical phonon system in piezoelectric crystals we shall use the phenomenological approach to the problem that proves to be appropriate in this case since we are interested in the common features of quasiparticles of all components (of excitons especially) that are independent upon their models.

Thus we start from the following equations:

1) The equation of motion of an elastic medium including the terms quadratic in electromagnetic field,

$$\rho_0 \frac{\partial^2 U_i}{\partial t^2} = \lambda_{ik\ell m} \frac{\partial U_{\ell m}}{\partial x_k} - e_{\ell,ik} \frac{\partial E_\ell}{\partial x_k} + \\ + \mathcal{A}_{ik\ell m} \frac{\partial}{\partial x_k} E_\ell E_m + \frac{1}{8\pi} \frac{\partial}{\partial x_k} (E_i D_k + E_k D_i), \tag{1}$$

which is followed from the equation of state of the crystal,

$$\sigma_{ik} = \lambda_{ik\ell m} U_{\ell m} - e_{\ell,ik} E_\ell + \mathcal{A}_{ik\ell m} E_\ell E_m + \frac{1}{8\pi}(E_i D_k + E_k D_i) \tag{2.1}$$

$$D_i = (\vec{E} + 4\pi \vec{P})_i + e_{i,jk} U_{jk} - \mathcal{A}_{ijk\ell} U_{k\ell} E_j . \tag{2.2}$$

Here ρ_0 is the mass density of the medium; U_i the i-component of the elastic displacement vector $\vec{U}$;

$$U_{\ell m} = \frac{1}{2}\left(\frac{\partial U_\ell}{\partial x_m} + \frac{\partial U_m}{\partial x_\ell}\right)$$

is the strain tensor; E_i the i-component of the electric field that is assumed to consist of the self-consistent perturbation field $\delta \vec{E}(\vec{r},t)$ and the external pumping field $\vec{E}_0(t)$,

$$\vec{E}(\vec{r},t) = \vec{E}_0(t) + \delta \vec{E}(\vec{r},t), \tag{3}$$

where the pumping electromagnetic field is taken in the known dipole approximation in the form

$$\vec{E}_0(t) = \vec{E}_0 \sin\omega_0 t \tag{4}$$

(The condition for this approximation to be valid were discussed in detail in many works (see, for example,/5,8/)); D_i is the i-component of the electric displacement vector $\vec{D}$;

$$\lambda_{ik\ell m} = \lambda'_{ik\ell m} + i\lambda''_{ik\ell m} \tag{5}$$

is the generalized complex elastic stiffness tensor including the effects of electron damping of sound (see /9/) as well as possible gains due to electron drift currents if such are present /7/ ;

$e_{\ell,ik}$ and $\mathcal{A}_{ik\ell m}$ are the tensors of piezoelectric constants and electrostrictive coefficients, respectively; σ_{ik} - the stress tensor of the crystal and $\vec{P}$ the polarization vector.

2) The linearized equation of motion for the nonequilibrium part $\vec{P}(\vec{r},t)$ of the polarization vector of the crystal including the effects of space dispersion:

$$\frac{\partial^2 \vec{P}}{\partial t^2} + \omega_r^2 \vec{P} - \alpha \,\mathrm{grad\,div}\, \vec{P} - \beta \,\mathrm{rot\,rot}\, \vec{P} = \\ = \chi\rho_0 \delta\vec{E}(\vec{r},t) + \chi[\delta\rho \cdot \vec{E}_0(t) - \frac{2}{\omega_0^2 - \omega_r^2} \frac{\partial \vec{E}_0(t)}{\partial t} \frac{\partial \delta\rho}{\partial t}], \tag{6.1}$$

where ω_r is the linear exciton frequency in the absence of electric fields, (mechanical exciton frequency), α and β are phenomenological parameters characterizing the effects of space dispersion in the system, χ the linear susceptibility and $\delta\rho$ the deviation of the elastic medium mass density from its equilibrium value ($\rho = \rho_0 + \delta\rho$). In obtaining (6.1) the total polarization vector $\vec{P}$ of the crystal was written in the form

$$\vec{P} = \vec{P}_0 + \vec{P}(\vec{r},t) \tag{6.2}$$

with $\vec{P}_0$ determined by equation

$$\frac{\partial^2 \vec{P}_0}{\partial t^2} + \omega_r^2 \vec{P}_0 = \chi\rho_0 \vec{E}_0(t) \tag{6.3}$$

and so

$$\vec{P}_0(t) = -\frac{\chi\rho_0}{\omega_0^2 - \omega_r^2} \vec{E}_0(t). \tag{6.4}$$

3) The system of Maxwell equations,

$$\text{rot rot } E + \frac{1}{c^2}\frac{\partial^2 \vec{D}}{\partial t^2} = 0, \qquad (7.1)$$

$$\text{div}\,\vec{D} = 0. \qquad (7.2)$$

4) Finally, the mass continuity equation:

$$\frac{\partial \rho}{\partial t} + \text{div}\,\rho\vec{V} = 0 \qquad (8)$$

with $\vec{V} = \frac{\partial \dot{U}}{\partial t}$.

Restricting the consideration by only longitudinal vibrations with the wave vector $\vec{k} \| \vec{z}$, i.e., from the field equations only (7.2) is taken which with the use of (2.2) will take the following linearized form:

$$[e_3 - (\vec{\mathcal{A}} \cdot \vec{E}_0(t))]\frac{\partial^2 \vec{U}}{\partial z^2} + \frac{\partial}{\partial z}[\delta\vec{E} + 4\pi\vec{P}] = 0,$$

where $e_3 \equiv e_{z,zz}$ and $\mathcal{A} = (\mathcal{A}_{zzzz}, \mathcal{A}_{zyzz}, \mathcal{A}_{zzzz})$, performing the linearization procedure with respect to the nonequilibrium parts of all quantities considered and introducing the space-time Fourier transformation one obtains from the system of basic equations, (1), (6)-(8), the following equations for the harmonics of $\vec{P}$ and $\vec{U}$;

$$\vec{P}(\vec{k},\omega) = \frac{\chi\rho_0}{\omega^2 - \omega_r^2(\vec{k})} \cdot \frac{1}{\epsilon(\vec{k},\omega)}\{ike_3\vec{U}(\vec{k},\omega) -$$

$$-\frac{kE_0}{2}(\mathcal{A} - 1)[\vec{U}(\vec{k}, \omega + \omega_0) - \vec{U}(\vec{k}, \omega - \omega_0)] -$$

$$- kE_0 \frac{\omega\omega_0}{\omega_0^2 - \omega_r^2}[\vec{U}(\vec{k}, \omega + \omega_0) + \vec{U}(\vec{k}, \omega - \omega_0)]\}, \qquad (9)$$

$$\{[\frac{k^2(\lambda_3 + e_3^2)}{\rho_0} - \omega^2] + \Omega_{\mathcal{A}}^2[\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi}]\}\vec{U}(\vec{k},\omega) =$$

$$= -\frac{i\Omega_e^2}{2}[3\mathcal{A} + \frac{\epsilon(0,\omega_0)}{4\pi}][\vec{U}(\vec{k},\omega+\omega_0) - \vec{U}(\vec{k},\omega-\omega_0)] +$$

$$+\frac{\Omega_{\mathcal{A}}^2}{2}[\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi}][\vec{U}(\vec{k},\omega+2\omega_0) + \vec{U}(\vec{k},\omega-2\omega_0)] +$$

$$+\frac{4\pi k}{\rho_0}\{ie_3\vec{P}(\vec{k},\omega) - E_0[\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi}][\vec{P}(\vec{k},\omega+\omega_0) - \vec{P}(\vec{k},\omega-\omega_0)]\}. \qquad (10)$$

In (9)-(10) the notations are as follows:

$$\epsilon(\vec{k},\omega) = 1 - \frac{4\pi\chi\rho_0}{\omega^2 - \omega_r^2(\vec{k})} \qquad (11)$$

is the exciton dielectric function $(\omega_r^2(\vec{k}) = \omega_r^2 + \alpha k^2)$; $\Omega_{\mathcal{A}}^2 = \frac{\mathcal{A}(kE_0)^2}{\rho_0}$ and $\Omega_e^2 = \frac{k^2 e_3 E_0}{\rho_0}$ are the quantities characterizing the coupling between the driving field and the crystal lattice through electrostictive and piezoelectric effects. We note also that this is the case of $\vec{E}_0 \| \vec{k} \| \vec{z}$ and then in (9)-(10) $\mathcal{A} \equiv \mathcal{A}_{zzzz}$, $\lambda_3 \equiv \lambda_{zzzz} = \lambda_3' + i\lambda_3''$. Equations (9)-(10) are the basic ones for the further analysis of the process of propagation and possible amplifications of the eigenmodes of our system.

3. ANALYSIS OF DISPERSION EQUATIONS

The instability analysis in this section we shall start with the determination of the eigenmodes of the system in the absence of the external radiation field.

Thus, setting $E_0 = 0$ in (9)-(10) one obtains after some algebra the dispersion equation for eigenmodes in the form

$$\omega^4 - \omega^2[\frac{k^2(\lambda_3 + e_3^2)}{\rho_0} + \omega_r^2(\vec{k}) + 4\pi\chi\rho_0] +$$

$$+ \frac{k^2}{\rho_0}[\lambda_3(\omega_r^2(\vec{k}) + 4\pi\chi\rho_0) + e_3^2(\omega_r^2(\vec{k}) + 8\pi\chi\rho_0)] = 0. \qquad (12)$$

Now, writing ω in the form

$$\omega = \omega' + i\omega'' \qquad (13)$$

with $\omega'' \ll \omega'$, one finds the frequencies ω_s', ω_e' and the dampings ω_s'', ω_e'' of the acoustical and exciton modes, respectively, in the form:

$$\omega_s'^2 = (kV_s)^2(1 + \frac{e_3^2}{\lambda_3'}), \qquad (14.1)$$

$$\omega_s'' = \frac{1}{2\omega_s'}\frac{k^2\lambda_3''}{\rho_0}, \qquad (14.2)$$

where $V_s = (\lambda_s'/\rho_0)^{1/2}$ is the sound velocity in the absence of any external fields and without an account of the piezo-effect;

$$\omega_e'^2 = \omega_r^2 + a k^2 + 4\pi\chi\rho_0 \equiv \omega_r^2(\vec{k}) + 4\pi\chi\rho_0 , \tag{15.1}$$

$$\omega_e'' = 0. \tag{15.2}$$

Thus we see that in this treatment the exciton mode is undumped one.

Now, when the pumping field is turned on, $E_0 \neq 0$, we shall consider the situation when from the harmonics of lattice vibrations $\vec{U}(\vec{k}, \omega \pm S\omega_0)$(S is integer) only one mode with $\omega = \omega_s' \ll \omega_0$ is excited. This means that in equation (10) only $\vec{U}(\vec{k},\omega)$ must be retained, so that we have

$$[(\omega^2 - \frac{k^2}{\rho_0}(\lambda_s + e_s^2)) - \Omega_{\mathcal{A}}^2(\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi})]\vec{U}(\vec{k},\omega) =$$
$$= \frac{4\pi k}{\rho_0}\{-ie_s\vec{P}(\vec{k},\omega) + E_0(\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi})[\vec{P}(\vec{k},\omega+\omega_0) - \vec{P}(\vec{k},\omega-\omega_0)]\}. \tag{16}$$

Equation (9) in these conditions yields:

$$\vec{P}(\vec{k},\omega) = \frac{\chi\rho_0}{\omega^2 - \omega_r^2(\vec{k})} \cdot \frac{ike_s}{\epsilon(\vec{k},\omega)}\vec{U}(\vec{k},\omega), \tag{17.1}$$

$$\vec{P}(\vec{k},\omega+\omega_0) = \frac{\chi\rho_0}{(\omega+\omega_0)^2 - \omega_r^2(\vec{k})} \cdot \frac{kE_0}{\epsilon(\vec{k},\omega+\omega_0)} \times$$

$$\times [\frac{\mathcal{A}-1}{2} - \frac{(\omega+\omega_0)\omega_0}{\omega_0^2 - \omega_r^2}]\vec{U}(\vec{k},\omega), \tag{17.2}$$

$$\vec{P}(\vec{k},\omega-\omega_0) = -\frac{\chi\rho_0}{(\omega-\omega_0)^2 - \omega_r^2(\vec{k})} \cdot \frac{kE_0}{\epsilon(\vec{k},\omega-\omega_0)} \times$$

$$\times [\frac{\mathcal{A}-1}{2} + \frac{(\omega-\omega_0)\omega_0}{\omega_0^2 - \omega_r^2}]\vec{U}(\vec{k},\omega). \tag{17.3}$$

Inserting (17) into (16) we find the dispersion relation in the form

$$[\omega_s^2 - \omega^2 + \Omega_{\mathcal{A}}^2(\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi}) + \frac{4\pi\chi(ke_s)^2}{\omega^2 - \omega_e^2}] =$$

$$= -4\pi\chi(kE_0)^2(\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi})\{\frac{1}{(\omega+\omega_0)^2 - \omega_e^2}[\frac{\mathcal{A}-1}{2} - \frac{(\omega+\omega_0)\omega_0}{\omega_0^2 - \omega_r^2}] + \frac{1}{(\omega-\omega_0)^2 - \omega_e^2}[\frac{\mathcal{A}-1}{2} + \frac{(\omega-\omega_0)\omega_0}{\omega_0^2 - \omega_r^2}]\}. \tag{18}$$

Since we are interested in the possible amplification of the waves, small shifts in frequencies will be neglected in further analysis. Now, if the frequency ω_0 of the external radiation field satisfies the resonant condition

$$\omega_0 - \omega = \omega_e' , \tag{19}$$

then the growth rate γ due to the driving field E_0 for both exciton ω_e' and acoustical phonon ω_s' modes will be expressed by the formula

$$\gamma_- = kE_0[\frac{\pi\chi}{\omega_s'\omega_e'}(\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi})(\frac{\mathcal{A}-1}{2} - \frac{\omega_e\omega_0}{\omega_0^2 - \omega_r^2})]^{1/2}. \tag{20}$$

Formally, in the same way, one can obtain the expression for γ when the other resonant condition,

$$\omega + \omega_0 = \omega_e' \tag{21}$$

is fulfilled. Namely, one has:

$$\gamma_+ = kE_0[\frac{\pi\chi}{\omega_s'\omega_e'}(\mathcal{A} + \frac{\epsilon(0,\omega_0)}{8\pi})(\frac{\omega_e\omega_0}{\omega_0^2 - \omega_r^2} - \frac{\mathcal{A}-1}{2})]^{1/2}. \tag{22}$$

Taking into account the fact that $\omega_0 \gg \omega_r$ and $\mathcal{A} \gg 1$ for almost all crystals used in optical investigations (see, for example,/10/) we see that only the case (19)-(20) can be realized. We can write now the complete expressions for the instability growth rates of exciton and phonon modes as follows:

$$\gamma_e = \gamma_- , \tag{23.1}$$

$$\gamma_S = \omega_S'' + \gamma_- . \qquad (23\text{-}2)$$

To clarify the influence of the electron absorption process and of the electron drift currents on the development of the wave instability we write down here the formula determining λ'' obtained in /7/ for the case of longitudinal field,

$$k_j k_m \lambda_{ij\ell m}'' = \pi\omega < \delta(\vec{k}\vec{v}_0)(\tilde{L}_i \tilde{L}_\ell - \tilde{\Lambda}_i \tilde{\Lambda}_\ell + \tilde{\Lambda}_{ij\ell m} k_j k_m) > -$$

$$- \pi\omega_d < \delta(\vec{k}\vec{v}_0) L_i'' L_\ell'' > , \qquad (24)$$

where

$$\tilde{L}_i = \tilde{L}_{ij} k_j , \text{ etc....,}$$

$\tilde{L}_{ij}$, L_{ij}'' , $\tilde{\Lambda}_{ij}$, $\tilde{\Lambda}_{ij\ell m}$ are functions of the conduction electron density Ne , the relative part and velocity of drifting electrons and the constant Λ_{ij} of the electron-phonon interaction via the deformation potential in the presence of electron drift currents; $\omega_d = (1-\delta_0) k V_d$,$(1-\delta_0)$ being the part of all condition electrons that is moving with the velocity $\vec{V}_d$ in the wave propagation direction $\vec{k}$ relatively to the crystal lattice; the bracket <...> means averaging by the formula

$$<A> \equiv \int A f_0'(\vec{p}) d\vec{p} ,$$

where $f_0'(\vec{p})$ is the derivative of the electron distribution function $f_0(\vec{p})$ with respect to the electron energy, $\vec{V}_0$ is the thermal motion velocity of an electron. For the crystal model "jellium" where $\Lambda_{ij}(\vec{p})=0$ equation (24) leads to a simple expression for λ_S'' :

$$\lambda_S'' = - \frac{(\pi\hbar)^3 N_e^2}{2kP_F^2} (\omega_S' - \omega_d), \qquad (25)$$

where P_F is the Fermi momentum of an electron. From (20), (23) and (25) one can see that the growth rate of the acoustical mode increases rapidly with the increasing of the velocity and relative part of moving electrons and then the threshold value of the field $\vec{E}_0$ for the occurrence of growing waves may be considerably lowered. It is obvious also that when $\omega_d \geq \omega_S'$ the external field will

amplify the arising drift instability if the temperature is low enough so that the viscous dissipation in the crystal can be neglected.

To calculate the instability threshold field for the exciton mode one must take into account the exciton damping Γ_e determined independently in optical experiments. Thus the threshold field value E_{0th} must satisfy the condition

$$\gamma_e \geq \Gamma_e .$$

A more detailed analysis of the instability growth rates as functions of the wave number k and of other parameters of the crystal as well as some numerical estimations for concrete experimental conditions will be performed separately in the next work.

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