EXCITON SPECTRA AND THEIR RENORMALIZATION BY THE EXCITON-PHONON INTERACTION IN LAYERED CRYSTALS

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We consider exciton states for the layered semiconductor GaSe with electron and hole spectra described by the Fivaz dispersion law. It is shown that the problem of the calculation of Wannier—Mott exciton spectra for this crystal is reduced to solving a differential equation with operator chain fractions. The exciton dispersion law obtained from this equation is used in the calculations of the real and imaginary parts of the mass operator describing the exciton-phonon interaction.

1. Introduction

The peculiarities of the exciton states in strongly anisotropic, in particular layered crystals (LCs) were considered in [1—5]. This interest is caused by the supposition about the possibility to realize high-temperature superconductivity in such media due to the exciton mechanism [6, 7]. The problem of the calculation of Wannier—Mott exciton spectra for LCs is investigated in a number of theoretical works (see, e.g., survey [8]). However, the question of the influence of the dispersion law of charge carriers on the exciton states in LCs is not clarified enough. Here, the exciton spectrum of LCs is constructed by using the equation derived in [9]. We will show that this equation allows one to obtain, in certain approximations, the analytical expression for the dependence between exciton energy and quasimomentum. This dispersion law is more general than the one commonly used for the analysis of the excitonic processes in LCs [7]. The obtained dispersion law describes both closed and open isoenergetic surfaces in the Brillouin zone and allows one to determine the condition for a change in the exciton isoenergetic surface topology. The obtained dispersion is used to investigate the influence of the exciton-phonon interaction on the exciton spectrum parameters in layered semiconductor GaSe at various temperatures. The frequency dependence of the absorption coefficient in the vicinity of the own absorption edge is determined as well. The results of calculations are compared with the experimental data.

2. The Exciton Spectrum for the Crystals with the Fivaz Dispersion of Charge Carriers

As known, the exciton spectrum and the wave functions are determined from the Wannier equation [10]. Here and below, we suppose that the energy spectra of electrons and holes in LCs are described by the Fivaz dispersion law [11, 12]

$$\varepsilon_{c,v}\left(\vec{k}\right) = E_{c,v} + \frac{\hbar^2}{2m^*_{c,v\perp}}\left(K_x^2 + K_y^2\right) + \\ +4I_{c,v}\sin^2\left(\frac{K_z d}{2}\right), \quad (1)$$

where d — the lattice parameter along the c axis, and $I_{c,v}$ – the layer parameters in the electron (c) and hole (v) spectra, respectively, equal to the quarter of the band width along [001] and determined as matrix elements of the Hamiltonian constructed on the wave functions of the neighboring layers.

In this case, the equation describing the exciton states may be written as [9]

$$\hat{A}F\left(\beta_\perp, \beta_\parallel\right) + \left(a\left(K_z\right) + ib\left(K_z\right)\right)F\left(\beta_\perp, \beta_\parallel + d\right) + \\ + \left(a\left(K_z\right) - ib\left(K_z\right)\right)F\left(\beta_\perp, \beta_\parallel - d\right) = \varepsilon F\left(\beta_\perp, \beta_\parallel\right), \quad (2)$$

where

$$\hat{A} = -\frac{\hbar^2}{2\mu_\perp}\nabla_\perp^2 - \frac{e^2}{\sqrt{\varepsilon_\perp\varepsilon_\parallel\beta_\perp^2 + \varepsilon_\perp^2\beta_\parallel^2}},$$

$$F\left(\beta_\perp, \beta_\parallel\right) = e^{-i\alpha K_\perp\beta_\perp}U\left(\vec{\beta}\right), \quad \alpha = \frac{1}{2}\frac{m^*_{c\perp} - m^*_{v\perp}}{m^*_{c\perp} + m^*_{v\perp}},$$

$$a\left(K_z\right) = -\left(I_c + I_v\right)\cos\frac{K_z d}{2},$$

$$b\left(K_z\right) = -\left(I_c - I_v\right)\sin\frac{K_z d}{2},$$

$$\varepsilon = E - E_g - 2\left(I_c + I_v\right) - \frac{\hbar^2 K_\perp^2}{2\left(m_{c\perp}^* + m_{v\perp}^*\right)},$$

and $E_g = E_c - E_v$ is the forbidden band width.

Let us assume that the distance between the electron and the hole in an exciton along the c axis, [001], is not greater than the lattice parameter d, i.e. $F\left(\beta_\perp, \beta_\parallel \pm md\right) = 0$ if $m{>}0$. In this case, the terms with displacements in the arguments in (2) are equal to zero and (2) is reduced to the equation

$$(\hat{A} - \varepsilon)F\left(\beta_\perp, \beta_\parallel\right) = 0. \tag{3}$$

Further, we suppose that the electron and the hole are separated by a distance not greater than $2d$. By substituting $\beta_\parallel \to \beta_\parallel \pm d$ in (2), taking into account that $F\left(\beta_\perp, \beta_\parallel \pm 2d\right) = 0$, and designating $\hat{A}_{\pm d} = -\frac{\hbar^2}{2\mu_\perp}\nabla_\perp^2 - \frac{e^2}{\sqrt{\varepsilon_\perp \varepsilon_\parallel \beta_\perp^2 + \varepsilon_\perp^2 \left(\beta_\parallel \pm d\right)^2}}$, we get

$$\hat{A}_{\pm d}F\left(\beta_\perp, \beta_\parallel \pm d\right) + \left(a\left(K_z\right) \mp ib\left(K_z\right)\right)F\left(\beta_\perp, \beta_\parallel\right) =$$

$$= \varepsilon F\left(\beta_\perp, \beta_\parallel \pm d\right). \tag{4}$$

By excluding the functions with displacements in arguments in the system of equations (3), (4) for the functions $F\left(\beta_\perp, \beta_\parallel\right)$ and $F\left(\beta_\perp, \beta_\parallel \pm d\right)$, we obtain the equation

$$\left\{\left(\hat{A} - \varepsilon\right) + \frac{c^2}{\varepsilon - \hat{A}_{+d}} + \frac{c^2}{\varepsilon - \hat{A}_{-d}}\right\}F\left(\beta_\perp, \beta_\parallel\right) = 0, \tag{5}$$

where $c\left(K_z\right)^2 = a^2 + b^2 = I_c^2 + I_v^2 + 2I_c I_v \cos\left(K_z d\right)$.

Taking m = 2,3..., we may transform Eq. (2) by induction to a differential equation with operator chain fractions and without displacements in arguments

$$\left\{\hat{\varepsilon}_0 + \cfrac{c^2}{\hat{\varepsilon}_{+1} - \cfrac{c^2}{\hat{\varepsilon}_{+2} - \cfrac{c^2}{\hat{\varepsilon}_{+3} - \ldots}}} + \cfrac{c^2}{\hat{\varepsilon}_{-1} - \cfrac{c^2}{\hat{\varepsilon}_{-2} - \cfrac{c^2}{\hat{\varepsilon}_{-3} - \ldots}}}\right\} \times$$

$$\times F\left(\beta_\perp, \beta_\parallel\right) = 0, \tag{6}$$

where $\hat{\varepsilon}_{\pm n} = \varepsilon - \hat{A}_{\pm nd}$. Equation (6) can be solved, in principle, in the following way: the fractions in (6) are broken on the first, second, etc. steps, the obtained equations are solved, and the convergence of the results is analyzed. It should be mentioned that the transformation of Eq. (2) to (6) is justified by the fact that, as a rule, the convergence of chain fractions is better than that of the corresponding series.

Further, we consider the case where the distance between the electron and the hole in a layer plane is much larger than along the c axis, $\beta_\perp \gg \frac{\varepsilon_\perp}{\varepsilon_\parallel}\beta_\parallel$. Then the term with $\beta_\parallel^2$ in the operator $\hat{A}$ may be neglected. In this case, Eq. (2) may be separated as

$$\left\{-\frac{\hbar^2}{2\mu_\perp}\nabla_\perp^2 - \frac{e^2}{\sqrt{\varepsilon_\perp \varepsilon_\parallel}\beta_\perp}\right\}F_1\left(\beta_\perp\right) = \varepsilon_1 F_1\left(\beta_\perp\right), \tag{7}$$

$$(a + ib)\,\Omega\left(\beta_z + d\right) + (a - ib)\,\Omega\left(\beta_z - d\right) = \varepsilon_2 \Omega\left(\beta_z\right), \tag{8}$$

where $F_1\left(\beta_\perp\right)\Omega\left(\beta_\parallel\right) = F\left(\beta_\perp, \beta_\parallel\right)$, $\varepsilon = \varepsilon_1 + \varepsilon_2$. Equation (8) is analogously reduced to the one with algebraic chain fraction:

$$\varepsilon_2 = \cfrac{2c\left(K_z\right)^2}{\varepsilon_2 - \cfrac{c\left(K_z\right)^2}{\varepsilon_2 - \cfrac{c\left(K_z\right)^2}{\varepsilon_2 - \ldots}}}. \tag{9}$$

In accordance with the Vorpitski theorem [7], the fraction on the right-hand side of (9) is convergent if $\varepsilon_2^2 \geq 4c\left(K_z\right)^2$. Let the value of this fraction be t. So, (9) may be written as $\frac{\varepsilon_2}{2} = t = \frac{c\left(K_z\right)^2}{\varepsilon_2 - t}$, which yields $t_{1,2} = 0.5\left(\varepsilon_2 \pm \sqrt{\varepsilon_2^2 - 4c\left(K_z\right)^2}\right)$ and

$$\varepsilon_2 = \pm 2c = \pm 2\sqrt{I_c^2 + I_v^2 + 2I_c I_v \cos\left(K_z d\right)}. \tag{10}$$

Equation (7) coincides with one obtained in the approximation of a two-dimensional crystal [14]. Its solutions are expressed in terms of a degenerate hypergeometric function, and the quantity ε_1 has got the discrete spectrum of values $\varepsilon_1 = -\frac{R}{\left(n + \frac{1}{2}\right)^2}$, n = 0, 1,... , where $R = \frac{\mu_\perp e^4}{2\hbar^2 \varepsilon_\perp \varepsilon_\parallel}$ is the effective Rydberg constant.

Taking into account (10), we may write the exciton energy E as

$$E = E_0 + E\left(\vec{K}\right), \tag{11}$$

where E_0 is the exciton formation energy, $E_0 = E_g - \frac{R}{\left(n + \frac{1}{2}\right)^2}$, and

$$E\left(\vec{K}\right) = \frac{\hbar^2 K_\perp^2}{2\left(m_{c\perp}^* + m_{v\perp}^*\right)} +$$

$$+2\left\{I_c + I_v \pm \sqrt{I_c^2 + I_v^2 + 2I_c I_v \cos\left(K_z d\right)}\right\} \tag{12}$$

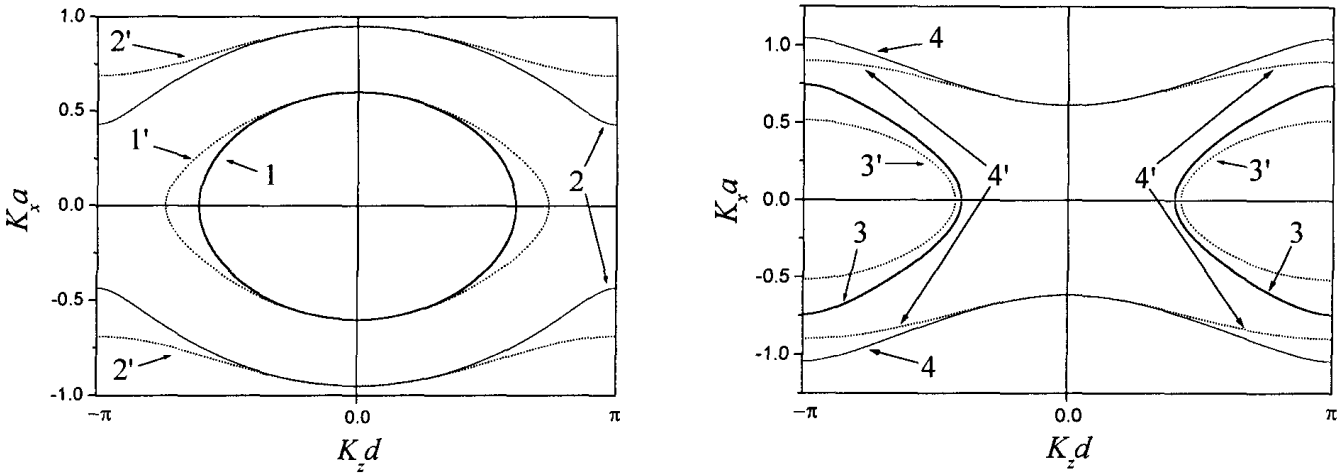

Fig. 1. Cross-sections of the isoenergetic surfaces in GaSe for the lower (a) and upper (b) exciton bands by (010) plane. *1*, *1′* — $E = 0.1$ eV (the region $E < 4I_c$), *2*, *2′* — $E = 0.25$ eV ($E > 4I_c$), *3*, *3′* — $E = 0.45$ eV ($E < 4\,(I_v + I_c)$), *4*, *4′* — $E = 0.60$ eV ($E > 4\,(I_v + I_c)$). Surfaces *1* — *4* correspond to the dispersion law (12), *1′* — *4′* — to the Fivaz dispersion laws (13) and (15)

is the dispersion law. Relation (12) describes two exciton bands symmetric in respect to (001) plane. The cross-sections of isoenergetic surfaces by (010) plane for both bands are presented in Fig. 1. At $K_\perp = 0$, the interval of energies for the lower band corresponding to minus in (12) is $0 \div 4I_c$ if K_z is changed from 0 to π/d. If $I_c > I_v$, this interval is $0 \div 4I_v$; here and below, we suppose that $I_v > I_c$ as it takes place for GaSe. For the upper band, the energy is changed from $4\,(I_c + I_v)$ at $K_z = 0$ to $4I_v$ at $K_z = \pi/d$. Thus, the minima of bands are displaced by π/d along [001]. The bands are separated by the distance $4\,(I_c + I_v)$ at Γ point and join at A point. Isoenergetic surfaces, being closed or open along [001], may be realized for both bands (Fig. 1). The isoenergetic surface topology changes at $E = 4I_c$ in the lower band and at $E = 4\,(I_v + I_c)$ in the upper one.

The dispersion law (12) may be reduced to the Fivaz one using the expansion $\sqrt{1-2x} \approx 1 - x$ if $x^2 \ll 1$. For the lower and upper bands, we obtain, respectively,

$$E\left(\vec{K}\right) = \frac{\hbar^2 K_\perp^2}{2\,(m_{c\perp}^* + m_{v\perp}^*)} + 4\frac{I_c I_v}{I_c + I_v}\sin^2\left(\frac{K_z d}{2}\right), \quad (13)$$

$$E\left(\vec{K}\right) = \frac{\hbar^2 K_\perp^2}{2\,(m_{c\perp}^* + m_{v\perp}^*)} + 4\,(I_c + I_v) - 4\frac{I_c I_v}{I_c + I_v}\sin^2\left(\frac{K_z d}{2}\right). \quad (14)$$

Thus, in a certain approximation, the exciton spectra in both bands are described by the Fivaz dispersion with the layer parameter $I_e = I_c I_v/(I_c + I_v)$. The points of the Lifshits transition are, respectively, $E = 4I_e$ for the lower band and $E = 4\,(I_c + I_v) + 4I_e$ for the upper one. That is, the transition occurs at the points not the same as for the dispersion law (12). In Fig. 1, we also present the cross-sections of isoenergetic surfaces for the Fivaz dispersion laws (13), (14). As seen, the forms of the isoenergetic surfaces are identical at Γ point for both types of dispersion, whereas they are essentially different at the boundaries of the Brillouin zone.

Because $4I_c$ is equal to 0.198 eV for GaSe, the minimum of the upper exciton band lies sufficiently far in the region of the own absorption, and this band cannot be practically observed. That is why, we consider only the lower band in the following calculations.

3. Exciton Spectrum Renormalization by the Exciton-phonon Interaction in LCs

The information about the exciton dispersion is important for the analysis of the shapes of absorption and dispersion lines [15]. Here, we use the dispersion laws (12), (13) in this analysis, as well as the anisotropic parabolic dispersion law

$$E\left(\vec{K}\right) = \frac{\hbar^2 K_\perp^2}{2\,(m_{c\perp}^* + m_{v\perp}^*)} + I_{\rm ex} d^2 K_z^2 \quad (15)$$

obtained from the Fivaz law by expanding the cosine in (13) in a series.

The analysis of the shapes of absorption and dispersion lines is realized by the calculation of the frequency dependences of the real and imaginary parts of the mass operator which describes the exciton spectrum renormalization by the exciton-phonon interaction. In

the one-phonon approximation, the mass operator looks as [9]

$$M(\vec{q},\omega)=\sum_{s\vec{p}}|\varphi_s(\vec{p})|^2\left(\frac{\nu_{s\vec{p}}}{\hbar\omega-E(\vec{q}+\vec{p})+\hbar\omega_s(\vec{p})}+\right.$$

$$\left.+\frac{1+\nu_{s\vec{p}}}{\hbar\omega-E(\vec{q}+\vec{p})-\hbar\omega_s(\vec{p})}\right), \qquad (16)$$

where δ — a certain small value, ω — the light frequency, $\vec{p}$ — the phonon quasimomentum, $\nu_{s\vec{p}}=\{\exp(\hbar\omega_s(\vec{p})/kT)-1\}^{-1}$ — the phonon distribution function, and $\omega_s(\vec{p})$ — the phonon dispersion law; for longitudinal optical (LO) phonons, $\omega_s(\vec{p})=\Omega$, Ω — the frequency of optical oscillations, $\Omega=300$ cm^{-1} [8]; for longitudinal acoustic (LA) phonons, $\omega^2(p_\perp,p_\parallel)=v_\perp^2p_\perp^2+\lambda p_\perp^4+\gamma(p_\parallel)$ [16], $\gamma(p_\parallel)=4\sin^2\left(\frac{p_\parallel d}{2}\right)$, $v_\perp, v_\parallel$ — the velocities of the sound in the layer plane and perpendicularly to it, $v_\perp=4515$ m/s, $v_\parallel=2482$ m/s [13], λ — the constant of bending oscillations, for GaSe $\lambda=0$ [16], and $\varphi_s(\vec{p})$ – the function describing the interaction between phonons and excitons that, in the cases of LO and LA phohons, is the same as in [15]. The values of the deformational potentials of the conduction C_c and valence bands C_v are defined in accordance with [17] and are equal to –10.6 eV and 16.3 eV, respectively. Here, we use the values of GaSe effective masses $m^*_{\perp c}=0.17m_0$, $m^*_{\parallel c}=\frac{\hbar^2}{2I_cd^2}=0.3m_0$ ($I_c=0.050$ eV), $m^*_{\perp v}=0.8m_0$, $m^*_{\parallel v}=\frac{\hbar^2}{2I_vd^2}=0.2m_0$ ($I_v=0.074$ eV), because these values obtained from the conductivity measurements [8] ensure the deformational potentials of the valence and conduction bands to be close to ones obtained for other LCs [8].

The real and imaginary parts of (16) may be written as

$$M(\vec{q},\omega)=\Delta(\vec{q},\omega)-\frac{i\hbar}{2}\Gamma(\vec{q},\omega). \qquad (17)$$

Further in accordance with [15], we will neglect the dependence of the mass operator on the exciton quasimomentum $\vec{q}$ because, for the visible spectral range, $|\vec{q}|\approx10^4$ cm^{-1} which is significantly lower than the values of the wave vectors in the Brillouin zone ($\sim10^8$cm^{-1}). The obtained expressions are

$$\Delta_{\rm ac}(\omega)=\frac{\hbar}{9\pi^2\rho\bar{v}}\lim_{\eta\to0}\left[\int_0^{\pi/d}dp_\parallel\int_0^{\pi/a}p_\perp dp_\perp\sqrt{p_\parallel^2+p_\perp^2}\times\right.$$

$$\times\left\{\frac{C_c}{\left[1+\alpha_1^2(p_\parallel^2+p_\perp^{+2})^2\right]}-\frac{C_v}{\left[1+\alpha_2^2(p_\parallel^2+p_\perp^{+2})^2\right]}\right\}^2\times$$

$$\left.\times\left\{\nu_{\rm ac}\frac{\hbar\omega-E(\vec{p})+\hbar\omega_{\rm ac}(\vec{p})}{\left[\hbar\omega-E(\vec{p})+\hbar\omega_{\rm ac}(\vec{p})\right]^2+\eta^2}+\right.\right.$$

$$\left.\left.+(1+\nu_{\rm ac})\frac{\hbar\omega-E(\vec{p})-\hbar\omega_{\rm ac}(\vec{p})}{\left[\hbar\omega-E(\vec{p})-\hbar\omega_{\rm ac}(\vec{p})\right]^2+\eta^2}\right\}\right], \qquad (18)$$

$$\Delta_{\rm op}(\omega)=\frac{e^2\hbar\Omega}{\pi}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\lim_{\eta\to0}\left[\int_0^{\pi/d}dp_\parallel\int_0^{\pi/a}p_\perp dp_\perp\times\right.$$

$$\times\left\{\frac{1}{[1+\alpha_1^2(p_\parallel^2+p_\perp^{+2})^2]}-\frac{1}{[1+\alpha_2^2(p_\parallel^2+p_\perp^{+2})^2]}\right\}^2\times$$

$$\left.\times\left\{\nu_{\rm ac}\frac{(p_\parallel^2+p_\perp^2)[\hbar(\omega+\Omega)-E(\vec{p})]}{(p_\parallel^2+p_\perp^2)^2[\hbar(\omega+\Omega)-E(\vec{p})]^2+\eta^2}+\right.\right.$$

$$\left.\left.+(1+\nu_{\rm ac})\frac{(p_\parallel^2+p_\perp^2)[\hbar(\omega-\Omega)-E(\vec{p})]}{(p_\parallel^2+p_\perp^2)^2[\hbar(\omega-\Omega)-E(\vec{p})]^2+\eta^2}\right\}\right], \qquad (19)$$

$$\Gamma_{\rm ac}(\omega)=\frac{2(m^*_{c\perp}+m^*_{v\perp})}{9\pi\rho v\hbar^2}\int_0^{\pi/d}dp_\parallel\left[\sqrt{p_\parallel^2+p_\perp^{+2}}\times\right.$$

$$\times\left\{\frac{C_c}{[1+\alpha_1^2(p_\parallel^2+p_\perp^{+2})^2]}-\frac{C_v}{[1+\alpha_2^2(p_\parallel^2+p_\perp^{+2})^2]}\right\}^2\times$$

$$\times\{F_+^+ + F_-^+\}+\sqrt{p_\parallel^2+p_\perp^{-2}}\left\{\frac{C_c}{[1+\alpha_1^2(p_\parallel^2+p_\perp^{-2})^2]}-\right.$$

$$\left.-\frac{C_v}{[1+\alpha_2^2(p_\parallel^2+p_\perp^{-2})^2]}\right\}^2\{F_+^- + F_-^-\}\Bigg], \qquad (20)$$

$$\Gamma_{\rm op}(\omega)=\frac{2e^2(m^*_{c\perp}+m^*_{v\perp})\Omega}{\hbar^2}\left(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right)\times$$

$$\times\left[\nu\int_0^{\pi/d}\frac{f_a}{p_\parallel^2+p_a^2}\left\{\frac{1}{[1+\alpha_1^2(p_\parallel^2+p_a^2)^2]}-\right.\right.$$

$$\left.-\frac{1}{[1+\alpha_2^2(p_\parallel^2+p_a^2)^2]}\right\}dp_\parallel+(1+\nu)\int_0^{\pi/d}\frac{f_u}{p_\parallel^2+p_b^2}\times$$

$$\left.\times\left\{\frac{1}{[1+\alpha_1^2(p_\parallel^2+p_b^2)^2]}-\frac{1}{[1+\alpha_2^2(p_\parallel^2+p_b^2)^2]}\right\}^2dp_\parallel\right], \qquad (21)$$

where

$$F_+^+ = \frac{\nu_{\rm ac}^+\kappa_+^+}{\left|1-\frac{(m_{c\perp}^*+m_{v\perp}^*)}{\hbar\sqrt{p_\perp^{+2}+\frac{\gamma(p_\parallel)}{\bar v}}}\right|}, \quad F_-^+ = \frac{(1+\nu_{\rm ac}^+)\kappa_-^+}{\left|1+\frac{(m_{c\perp}^*+m_{v\perp}^*)}{\hbar\sqrt{p_\perp^{+2}+\frac{\gamma(p_\parallel)}{\bar v}}}\right|},$$

$$F_+^- = \frac{\nu_{\rm ac}^-\kappa_+^-}{\left|1-\frac{(m_{c\perp}^*+m_{v\perp}^*)}{\hbar\sqrt{p_\perp^{-2}+\frac{\gamma(p_\parallel)}{\bar v}}}\right|}, \quad F_-^- = \frac{\nu_{\rm ac}^-\kappa_-^-}{\left|1+\frac{(m_{c\perp}^*+m_{v\perp}^*)}{\hbar\sqrt{p_\perp^{+2}+\frac{\gamma(p_\parallel)}{\bar v}}}\right|},$$

$\bar v$ — the sound velocity averaged over directions,

$$p_{a,b} = \sqrt{\frac{2\left(m_{c\perp}^*+m_{v\perp}^*\right)}{\hbar^2}\left[\hbar\left(\omega\pm\Omega\right)-\beta\left(p_\parallel\right)\right]}, \quad \beta\left(p_\parallel\right) = E\left(\vec p\right) - \frac{\hbar^2 p_\perp^2}{2\left(m_{c\perp}^*+m_{v\perp}^*\right)}$$ — the part of the exciton dispersion law dependent on $p_\parallel$, $f_{a,b} = \begin{cases} 0, p_{a,b}\notin[0;\pi/a] \\ 1, p_{a,b}\in[0;\pi/a] \end{cases}$,

$$p_\perp^{\pm 2} = \frac{2(m_{c\perp}^*+m_{v\perp}^*)}{\hbar^2}\left\{\hbar\omega + \frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{d^2} - \beta\left(p_\parallel\right)\pm \right.$$

$$\left. \pm\sqrt{\frac{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)^2}{d^4}+\frac{2v_\parallel^2\left(m_{c\perp}^*+m_{v\perp}^*\right)\left(\hbar\omega-\beta\left(p_\parallel\right)\right)}{d^2}+\hbar^2\gamma\left(p_\parallel\right)}\right\},$$

$$\kappa_+^+ = \begin{cases} 0, p_\perp^+\notin\left[0;\frac{\pi}{a}\right] & \text{or} \quad \hbar\omega\le\beta\left(p_\parallel\right)-\frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{2d^2}-\frac{\hbar^2d^4\gamma(p_\parallel)}{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)}, \\ 1, p_\perp^+\in\left[0;\frac{\pi}{a}\right] & \text{and} \quad \hbar\omega\ge\beta\left(p_\parallel\right)-\frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{2d^2}-\frac{\hbar^2d^4\gamma(p_\parallel)}{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)}, \end{cases} \qquad \kappa_-^+ = 0,$$

$$\kappa_+^- = \begin{cases} 0, p_\perp^-\notin\left[0;\frac{\pi}{a}\right] & \text{or} \quad \hbar\omega\le\beta\left(p_\parallel\right)-\frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{2d^2}-\frac{\hbar^2d^4\gamma(p_\parallel)}{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)} \quad \text{or} \quad \hbar\omega\ge\beta\left(p_\parallel\right)-\frac{\hbar^2d^4\gamma(p_\parallel)}{2v_{II}^2\left(m_{c\perp}^*+m_{v\perp}^*\right)}, \\ 1, p_\perp^-\in\left[0;\frac{\pi}{a}\right] & \text{and} \quad \hbar\omega\ge\beta\left(p_\parallel\right)-\frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{2d^2}-\frac{\hbar^2d^4\gamma(p_\parallel)}{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)} \quad \text{and} \quad \hbar\omega\le\beta\left(p_\parallel\right)-\frac{\hbar^2d^4\gamma(p_\parallel)}{2v_{II}^2\left(m_{c\perp}^*+m_{v\perp}^*\right)}, \end{cases}$$

$$\kappa_-^- = \begin{cases} 0, p_\perp^-\notin\left[0;\frac{\pi}{a}\right] & \text{or} \quad \hbar\omega\le\beta\left(p_\parallel\right)-\frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{2d^2}-\frac{\hbar^2d^4\gamma(p_\parallel)}{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)} \quad \text{or} \quad \hbar\omega\le\beta\left(p_\parallel\right)-\frac{\hbar^2d^4\gamma(p_\parallel)}{2v_\parallel^2\left(m_{c\perp}^*+m_{v\perp}^*\right)}, \\ 1, p_\perp^-\in\left[0;\frac{\pi}{a}\right] & \text{and} \quad \hbar\omega\ge\beta\left(p_\parallel\right)-\frac{v_\parallel^2(m_{c\perp}^*+m_{v\perp}^*)}{2d^2}-\frac{\hbar^2d^4\gamma(p_\parallel)}{v_\parallel^4\left(m_{c\perp}^*+m_{v\perp}^*\right)} \quad \text{and} \quad \hbar\omega\ge\beta\left(p_\parallel\right)-\frac{\hbar^2d^4\gamma(p_\parallel)}{2v_\parallel^2\left(m_{c\perp}^*+m_{v\perp}^*\right)}. \end{cases}$$

The numerical integration of expressions (18)—(21) for the anisotropic parabolic dispersion law (15), Fivaz one (13), and (12) obtained here shows that, in all the considered cases, the real and imaginary parts of the mass operator do not differ significantly. The results obtained for the dispersion law (12) are slightly near the ones obtained for the anisotropic parabolic dispersion law: the relative differences between the real and imaginary parts are 0.87 and 0.35%, respectively. Whereas the same values obtained for the dispersion law (12) and the Fivaz one are 2.32 and 5.15%. Thus, the exciton dispersion law (12) obtained from the idea of a strong crystal anisotropy leads to the results practically identical for the ones obtained for the parabolic dispersion which is a usual model of the exciton dispersion in semiconductors [8, 10]. This peculiarity is in agreement with the well-known fact that the crystal anisotropy of GaSe does not manifest itself in the exciton spectrum structure that may be described well enough by the usual hydrogen-like model [8].

Tne frequency dependences of the real and imaginary parts at different temperatures are shown in Fig. 2,*a*, *b*, where the energy is counted from the exciton band bottom E_0. As seen from Fig. 2, both the real and imaginary parts of the mass operator are significantly differ from zero in the region near the bottom of the exciton band and increase in the absolute values with temperature. Such a behavior affects the optical properties of the crystal near the own absorption edge. Particularly, the absorption coefficient is determined by the expression [15]

$$\kappa\left(\omega\right) = \frac{\omega}{c_0}\sqrt{2\left[-\varepsilon'\left(\omega\right)+\sqrt{\varepsilon'\left(\omega\right)^2+\varepsilon''\left(\omega\right)^2}\right]}, \qquad (22)$$

where

$$\varepsilon'\left(\omega\right) = \varepsilon_\infty - \frac{E_p^2 f}{2\hbar\omega}\frac{\hbar\omega-E_0-\Delta\left(\omega\right)}{\left[\hbar\omega-E_0-\Delta\left(\omega\right)\right]^2+\frac{1}{4}\hbar^2\Gamma\left(\omega\right)^2},$$

$$\varepsilon''\left(\omega\right) = \frac{E_p^2 f}{2\hbar\omega}\frac{\frac{1}{2}\hbar\Gamma\left(\omega\right)}{\left[\hbar\omega-E_0-\Delta\left(\omega\right)\right]^2+\frac{1}{4}\hbar^2\Gamma\left(\omega\right)^2},$$

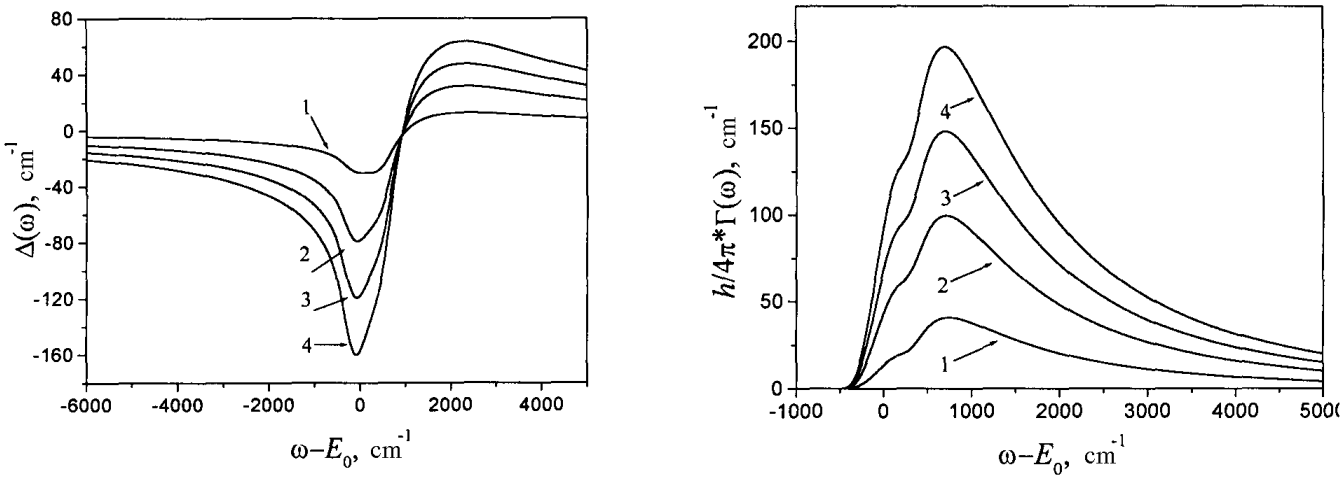

Fig. 2. Frequency dependences of the real (*a*) and imaginary (*b*) parts of the mass operator at $T = 77$ (curve *1*), 200 (*2*), 300 (*3*), and 400 K (*4*) for the dispersion law (12)

c_0 — the velocity of light, E_p — the energy of plasma oscillations, $E_p = \sqrt{\frac{\hbar^2 e^2}{\varepsilon_0 V_0 m_{\rm ex}}}$, $m_{\rm ex}$ — the exciton mass, $m_{\rm ex} = m_c^* + m_v^*$, V_0 — the elementary cell volume, f — the oscillator strength of the transition to an excited state. The oscillator strength was calculated from the data on the integral absorption and the exciton absorption line halfwidth [18]. Our calculations give the values of the average oscillator strength $f = 9{\times}10^{-4}$ at 300 K for the first exciton peak. The calculated absorption coefficient is only in the qualitative agreement with the experimental value [2]. In particular, the value of the calculated absorption coefficient at the maximum is twice lower than the experimentally obtained value. Such a discrepancy may be caused by the insufficient precision of the approximations used in the derivation of (12) and (18)—(21), as well as by the possible presence of different polytypes in the crystal used in the absorption measurements [2].

4. Conclusions

Here, we have considered the differential equation describing exciton states in the crystal with the Fivaz dispersion law for charge carriers. This equation contains the displacements in arguments and may be reduced to the one containing the operator chain fractions. In a particular case, the variables in the equation may be separated. One of the obtained equations defines the energy levels of an exciton (this equation is similar to the one obtained in the two-dimensional model of exciton) and the other does its dispersion law. The obtained dispersion law allows us to describe the isoenergetic surfaces closed or open in the [001] direction and, in addition, is used in the calculation of the real and imaginary parts of the mass operator describing the exciton-phonon interaction. We have shown that the real and imaginary parts of the mass operator are close to ones obtained in the cases of the Fivaz and anisotropic parabolic exciton dispersions. The results obtained with the dispersion law derived here are close to those obtained for the anisotropic parabolic dispersion law. This is in agreement with the fact that the crystal anisotropy of GaSe does not manifest itself in the exciton spectrum structure which is described well enough by the usual hydrogen-like model. Both the real and imaginary parts of the mass operator are significantly differ from zero in the region near the bottom of the exciton band and increase in the absolute values with temperature. The calculations of the absorption coefficient near the own absorption edge show a qualitative agreement with the experimental data.

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ЕКСИТОННІ СПЕКТРИ ТА ЇХ ПЕРЕНОРМУВАННЯ ЕКСИТОН-ФОНОННОЮ ВЗАЄМОДІЄЮ У ШАРУВАТИХ КРИСТАЛАХ

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Р е з ю м е

На прикладі кристала GaSe розглядаються екситонні стани у шаруватому напівпровіднику, електронний та дірковий спектри якого описуються законами дисперсії Фіваза. Показано, що задача розрахунку спектра екситонів Ваньє—Мотта у такому кристалі зводиться до розв'язання диференціального рівняння, яке містить операторні ланцюгові дроби. Отримано вираз для закону дисперсії екситонів, за допомогою якого розраховано дійсну та уявну частини масового оператора, що описує екситон-фононну взаємодію.