Properties of renormalized spectra of localized quasiparticles interacting with one- and two-mode phonons in Davydov’s model at T = 0K

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Motivation for the research

The rapid development of nanophysics has significantly actualized the theory of the interaction of systems of multi-band (multilevel) quasiparticles with multi-mode phonons. Such problems are important, in particular, for understanding the physical processes occurring in nanoheterostructures, which are the basic structural elements of quantum cascade lasers and infrared detectors.

Theory and analysis of results

I this paper we study the influence of dissipative mechanisms (taken into account by phenomenological decay) on the formation of properties of coefficient of the electromagnetic field absorption by localized quasiparticles interacting with one- and two-mode polarization phonons at T = 0K in Davydov’s model of the system.

From Kubo's theory [1,2] about the response of an isotropic system to external action it is known that the absorption coefficient of the electromagnetic field $(\chi(\tilde{\omega}))$ is related with the Fourier image of the retarded Green's function $(G(\tilde{\omega}))$ of quasiparticle by the expression

$$\chi(\tilde{\omega})=\frac{4\pi\hbar\omega_f^2 d^2}{v\omega v_g}\mathrm{I}(\tilde{\omega});\quad \mathrm{I}(\tilde{\omega})=-\mathrm{Im}\,G(\tilde{\omega});\quad (\tilde{\omega}=\omega+i\eta),\quad \eta\to+0. \qquad (1)$$

Here $\mathrm{I}(\tilde{\omega})$ is a function of the shape of the absorption band of the electromagnetic field, $d=er$ is a dipole moment, v is a unit cell volume, ω_f is a frequency of dipole transition, ω is an electromagnetic field frequency, v_g is a group velocity.

Further, studying only the shape of the field absorption band, we consider a system consisting of a localized quasiparticle (exciton, impurity, etc.) interacting with non-dispersive two-mode polarization phonons at T = 0K. The Hamiltonian of this system (excluding dissipative processes) in the representation of occupation numbers over all variables can be written as

$$\hat{\mathrm{H}}=\sum_{\vec{k}}E_0\hat{A}^+_{\vec{k}}\hat{A}_{\vec{k}}+\sum_{\lambda=1}^{2}\sum_{\vec{q}}\Omega_\lambda\left(\hat{B}^+_{\lambda\vec{q}}\hat{B}_{\lambda\vec{q}}+1/2\right)+\sum_{\vec{k}\lambda\vec{q}}\varphi_\lambda(\vec{q})\,\hat{A}^+_{\vec{k}}\hat{A}_{\vec{k}}\left(\hat{B}_{\lambda\vec{q}}+\hat{B}^+_{\lambda-\vec{q}}\right). \qquad (2)$$

Here E_0 is an energy of uncoupling quasiparticle, Ω_λ is an energy of λ- th phonon mode, $\varphi_\lambda(\vec{q})$ is a quasiparticle binding function with λ -th phonon mode. Quasiparticle $(\hat{A}^+_{\vec{k}},\hat{A}_{\vec{k}})$ and phonon $(\hat{B}^+_{\lambda\vec{q}},\hat{B}_{\lambda\vec{q}})$ operators of second quantization satisfy Bose commutative relationships. In Davydov’s model for the system under research, it is fulfilled the condition $\hat{n}^2=\hat{n}=\sum_{\vec{k}}\hat{A}^+_{\vec{k}}\hat{A}_{\vec{k}}$, which means that the eigenvalues of both of these operators ($\hat{n}$ i $\hat{n}^2$) can be either 1 or 0 , interpreted as a condition of presence (1) or absence (0) of "pure" quasiparticle state.

To calculate the Fourier image of the retarded Green's function without a decay, the Hamiltonian (4) is first diagonalized by the transition from the operators $\hat{A}_{\vec{k}},\hat{B}_{\lambda\vec{q}}$ to new ones $\hat{a}_{\vec{k}},\hat{b}_{\lambda\vec{q}}$ using a unitary operator [3]. As a result, the Hamiltonian (2) in the new operators gets a diagonal form

$$\hat{\mathrm{H}}=\sum_{\vec{k}}\mathscr{E}\hat{a}^+_{\vec{k}}\hat{a}_{\vec{k}}+\sum_{\lambda\vec{q}}\Omega_\lambda(\hat{b}^+_{\lambda\vec{q}}\hat{b}_{\lambda\vec{q}}+1/2);\quad \left(\mathscr{E}=E_0-\sum_{\lambda\vec{q}}\Omega_\lambda^{-1}\left|\varphi_\lambda(\vec{q})\right|^2\right), \qquad (3)$$

where $\mathscr{E}$ is an energy of new elementary excitations.

Now at T = 0K the two-time retarded Green's function

$$G(\vec{k},t)=-i\theta(t)\left\langle 0\right|[\hat{A}^+_{\vec{k}}(t),\hat{A}_{\vec{k}}(0)]\left|0\right\rangle, \qquad (4)$$

taking into account the Hamiltonian (3), the relationship between old and new operators and using Weyl operator identity [1,2], the exact expression is obtained

$$G(\vec{k},t)=-i\theta(t)\exp\left\{-\frac{i\mathscr{E}t}{\hbar}+\sum_{\lambda=1}^{2}\alpha_\lambda\left\{\exp\left(-\frac{i\Omega_\lambda t}{\hbar}\right)-1\right\}\right\};\quad \alpha_\lambda=\Omega_\lambda^{-2}\sum_{\vec{q}}\left|\varphi_\lambda(\vec{q})\right|^2, \qquad (5)$$

where α_λ is a dimensionless parameter characterizing the binding energy of a quasiparticle with λ -th mode of phonons.

The integration of expression (5) is performed exactly. Introducing phenomenological decay by replacing a small value ($\eta\to+0$) by a finite value $(\gamma>0)$ a representation of the Fourier image of Green's function, which is convenient for physical analysis, is obtained

$$G(\omega,\gamma)=\exp\left[-\sum_{\lambda=1}^{2}\alpha_\lambda\right]\left\{\frac{1}{\hbar\omega-\mathscr{E}+i\gamma}+\sum_{\lambda=1}^{2}\sum_{n=1}^{\infty}\frac{\alpha_\lambda^{n_\lambda}}{n_\lambda![\hbar\omega-\mathscr{E}-n_\lambda\Omega_\lambda+i\gamma]}+\right.$$
$$\left.+\sum_{n_1,n_2=1}^{\infty}\alpha_1^{n_1}\alpha_2^{n_2}/n_1!n_2![\hbar\omega-\mathscr{E}-n_1\Omega_1-n_2\Omega_2+i\gamma]\right\} \qquad (6)$$

The resulting expression for $G(\omega,\gamma)$ makes it possible to calculate and analyze the dependences of the components of the spectral parameters of the absorption coefficient of the electromagnetic field on its frequency and energy values that characterize the system. However, for simplicity, we will introduce dimensionless quantities

$$I(\xi)=\Omega_1\mathrm{I}(\omega);\ \xi=\Omega_1^{-1}(\hbar\omega-\mathscr{E});\quad \gamma=\frac{\gamma}{\Omega_1};\quad P_\lambda=\frac{\Omega_\lambda}{\Omega_1};\ (\lambda=1,2) \qquad (7)$$

Now we are going to calculate and analyze the function of the shape of the absorption band by a two-mode system

$$I(\alpha_1,\alpha_2;\xi)=\gamma\exp\left\{-\sum_{\lambda=1}^{2}\alpha_\lambda\right\}\left\{\frac{1}{(\xi^2+\gamma^2)}+\sum_{\lambda=1}^{2}\sum_{n=1}^{\infty}\alpha_\lambda^{n_\lambda}/n_\lambda![(\xi-n_\lambda P_\lambda)^2+\gamma^2]+\right.$$
$$\left.+\sum_{n_1,n_2=1}^{\infty}\alpha_1^{n_1}\alpha_2^{n_2}/n_1!n_2![(\xi-n_1-n_2P_2)^2+\gamma^2]\right\} \qquad (8)$$

Putting $\alpha_1=\alpha,\ \alpha_2=0,\ P_2=0$ from formula (8) the expression for the dimensionless function of the shape of the field absorption band by a single-mode system is obtained

$$I(\alpha,\xi)=\gamma e^{-\alpha}\left\{1/\xi^2+\gamma^2+\sum_{n=1}^{\infty}\alpha^n/n![(\xi-n)^2+\gamma^2]\right\}. \qquad (9)$$

From similar expressions (8) and (9) it is clear that both functions $(I(\alpha;\xi),\ I(\alpha_1,\alpha_2;\xi))$ are the superpositions of an infinite number of Lorentz functions (peaks) with very small decay $(\gamma<<1)$.

Lorentz peaks are characterized by obvious spectral parameters: heights (*I*) and half-widths (*Г*), which depend on the number of phonon modes of the system. Let us analyze the properties of $I(\alpha;\xi)$ and $I(\alpha_1,\alpha_2;\xi)$ functions for one- and two-mode system.

a) One-mode system. The properties of $I(\alpha;\xi)$ function are clearly visible from the analytical expression (9), as well as from Fig.1, which shows the results of calculations of its dependence on the dimensionless frequency (ξ) at different values α and γ. Function $I(\alpha;\xi)$ is a superposition of an infinite number of Lorentz peaks, the heights of their maxima $(I_\ell(\alpha))$ are located at $\xi_\ell=\ell=0,1,2,\dots,\infty$ and are determined by the values

$$I_\ell(\alpha)=\gamma e^{-\alpha}\sum_{n=0}^{\infty}\alpha^n/n![(\ell-n)^2+\gamma^2];\qquad \ell=0,1,2,\dots,\infty \qquad (10)$$

As a result, the half-width of $\ell-$ th peak (Γ_ℓ) is obtained as the difference of quantities which are the solutions of a nonlinear equation

$$\sum_{n=0}^{\infty}\alpha^n/n!\{1/[(\xi_\ell^{\lessgtr}-n)^2+\gamma^2]-1/[2(n^2+\gamma^2)]\}=0;\ \ell=0,1,2,\dots,\infty \qquad (11)$$

The lowest energy peak at $\ell=0$ corresponds to the main phonon-less state. The phonon satellite peaks at $\ell=1,2,\dots$ are formed by bound states of quasiparticles with one, two and the rest of phonons (the so-called phonon repeats). The maxima of the main and phonon satellite peaks are equidistant from each other (at a distance of one phonon energy), and the ratio of the $\ell-$th peak maximum to the main one is fixed by the expression

$$J_\ell(\alpha,\gamma)=\frac{I_\ell(\alpha,\gamma)}{I_0(\alpha,\gamma)}=\left[\sum_{n=0}^{\infty}\frac{\alpha^n}{n![n^2+\gamma^2]}\right]^{-1}\sum_{n=0}^{\infty}\frac{\alpha^n}{n![(\ell-n)^2+\gamma^2]};\ \ell=0,1,2,\dots,\infty \qquad (12)$$

Formulas (10) and (12) and Fig. 1 prove that the heights of $I_\ell(\alpha)$ peaks are more complicated functions than $\Gamma_\ell(\alpha)$ half-widths. First, the heights are sensitive to varying magnitudes γ,α and ℓ , that is especially clear from Fig. 1. In particular, the height, as a parameter of the functions that characterize the ground state $(I_{\ell=0}(\alpha;\xi))$, only decreases with increasing α in the interval $(0<\alpha<1)$, while the heights of the respective functions of the phonon satellite states ($I_{\ell\neq0}(\alpha;\xi)$) increase only. Herein, according to (12), if ℓ increases, the heights $I_{\ell\neq0}$ are rapidly decreasing. As a result, the peaks of high-energy satellite states "merge" with a continuous background.

It is worth to note that when $\alpha=1$ not just widths ($\Gamma_{\ell=0}\cong\Gamma_{\ell=1}$), but also the heights ($I_{\ell=0}\cong I_{\ell=1}$) of peaks of the main and the first phonon satellite (repeat) are almost the same (with accuracy $\leq1\%$) The revealed properties of $I(\alpha;\xi)$ functions in the interval $(0<\alpha<1)$ cause such a rapid "immersion" of satellite phonon repeats into a continuous background, that one can see only 3-4 bound states of the quasiparticle with phonons in addition to the main one, Fig. 1.

If $\alpha=1$ and the heights of the main and the first phonon repeats become equal ($I_0=I_1=7.38$), further increase of α magnitude leads only to a decrease of the first satellite peak height, as well as for the phonon-free one. Heights of other phonon satellite peaks continue to increase until constant α reaches the value of 2 when the heights of the first and second phonon peaks are the same. If α further increases, the heights of 0,1,2 peaks decrease, and the rest continue to increase. At $\alpha=3$ the heights of 2 and 3 peaks are equalized, and that of the rest – increase.

The established property leads to the fact that the curve, which is the envelope of the maxima of the absorption peaks, with increasing α , has a maximum in the field of phonon repeats the bigger is the interaction constant α.

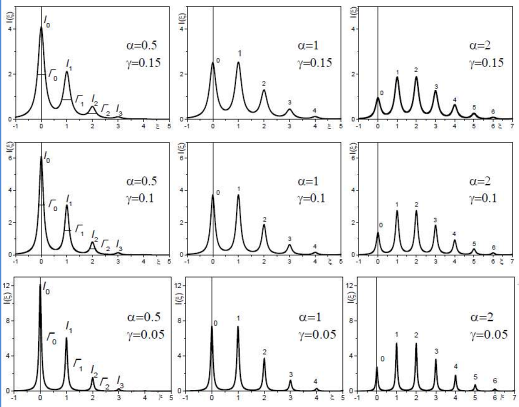

Fig.1. Evolution of the function of the shape of the absorption band $I(\alpha;\xi)$ at different values of decay (γ = 0.05; 0.1; 0.15). In the region of small and medium values of the interaction constant (α = 0.5; 1; 2) in one-mode system.

b) ***Two-mode system.*** The calculation and analysis of the properties of the spectral parameters of two-mode systems was performed for the example of the most typical physical systems in which the quasiparticle is weakly bound with one mode and the middle one with the other. Fig. 2 shows the results of the calculation of the form-function $I(\alpha_1,\alpha_2;\xi)$ of two-mode system in that frequency range (ξ), in which the main and satellite phonon peaks are fairly well identified without "merging" with the background.

In contrast to the one-mode, in the two-mode system the structure of the function bands $I(\alpha_1,\alpha_2;\xi)$ is more complicated because it is formed both by the bound states of the quasiparticle with the phonons of certain modes and with their combinations. Therefore, the function $I(\alpha_1,\alpha_2;\xi)$ is a superposition of the main peak (0,0) of the Lorentz type with height $I_{0,0}$ and half-width $\Gamma_{0,0}$ and three groups of infinite number of phonon satellite peaks, which are formed by bound states of quasiparticle with unmixed phonon modes $(I_{\ell_1\neq0,\ \ell_2=0};\ I_{\ell_1=0,\ \ell_2\neq0})$ and mixed phonon modes $(I_{\ell_1\neq0,\ \ell_2\neq0})$.

Fig. 2 shows that the picture of the formation of absorption bands $I(\alpha_1,\alpha_2;\xi)$ significantly depends both on the ratios (P_2) between the values of the energies of both phonon modes and the values of both constants (α_1,α_2).

The height $(I_{0,0})$ and half-width $(\Gamma_{0,0})$ of the main phonon-free peak is almost independent of the magnitude P_2 . If α_2 increases, only its height, which is mainly formed by the stronger interaction of the quasiparticle with the second phonon mode, gradually decreases.

The peaks corresponding to the phonon satellites of certain unmixed modes create a picture of structures that have the same properties as previously described for the corresponding one-mode structures. Herein, the heights of the peaks formed by a stronger interaction with the second mode significantly dominate over those formed by a weaker interaction with the first mode $(I_{0,\ell_2}>>I_{\ell_1,0})$.

As for the peaks which correspond to the combined frequencies of the satellite phonon states, at $P_2=1{,}25;\ 1{,}5;\ 1{,}75;\ 2;$, they appear either in the form of individual peaks or on the wings of peaks formed by the second phonon mode which stronger interacts with the quasiparticle. In the case of multiple frequencies $(P_2=2)$, as can be seen from Fig. 2, all combined peaks formed by the interaction of quasiparticles with superpositions of both phonon modes are "absorbed" into the satellite peaks of both separate modes. As a result, the spectrum looks like equidistant, with variable intensity and decay.

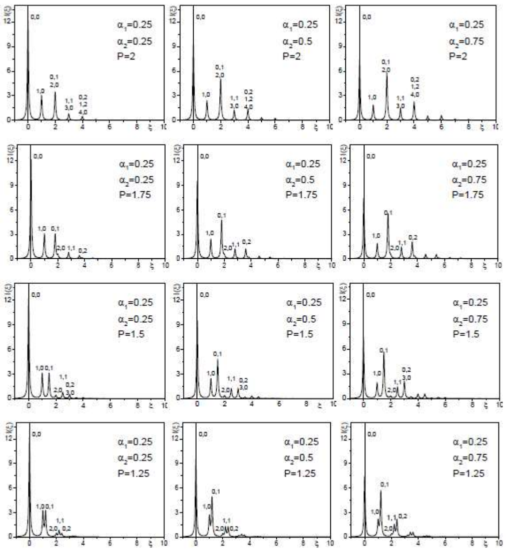

Fig.2. Function of the shape of the absorption band $I(\alpha_1,\alpha_2;\xi)$ in the region of small values of α_1 and medium α_2 at different ratios $P_2=1{,}25;\ 1{,}5;\ 1{,}75;\ 2;$ between their energies in two-mode system. The magnitude of the decay is $\gamma=0.05$.

Conclusion

1. On the basis of the Hamiltonian of the Frohlich type, which describes a localized quasiparticle interacting with two-mode non-dispersive phonons in Davydov’s model with the decay at T = 0K, an analytical calculation of Fourier image of retarded Green's function is performed. It allowed to obtain and analyze the frequency dependence of its imaginary part, which characterizes the function of the shape of the absorption band and its spectral parameters.

2. It is shown that in one-mode system the widths of the main and phonon satellite peaks vary slightly if α increases. The height of the main peak only decreases significantly, while the heights of the peaks of phonon repeats gradually increase, reach the height of the neighbor previous peak, then also decrease.

3. It is shown that a two-mode system has a complex structure of the function of the shape of the absorption band. It has both peaks corresponding to both separate phonon modes and their superpositions. It turned out that the pattern of peaks of phonon repeats of a larger phonon mode, with which the quasiparticle interacts stronger, is similar to the evolution of the corresponding one-mode system, and peaks formed by weaker mode or combinations of both modes are weak or even absorbed by stronger peaks and general continuous background.

References

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