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PHONON INFLUENCE ON KINEMATIC EXCITON LEVELS

ВЛИЯНИЕ ФОНОНОВ НА УРОВНИ КИНЕМАТИЧЕСКИХ ЭКСИТОНОВ

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In the paper presented we shall analyse the kinematic interaction of Frenkel excitons in the presence of phonons and determine the life-time of the kinematic exciton level at room temperature.

Kinematic exciton levels represent additional excitations in the exciton system, which occur due to the kinematic exciton interaction. Taking into account that they appear in consequence of the correct decoupling of the Green functions of the form $\langle\langle B^+BB|B^+B^+B\rangle\rangle$, it is clear that they occur in three-particles exciton processes and in such where two excitons, previously fused in a new, unstable exciton with an approximately twice as high energy, disintegrate into two simple excitons after a certain period of time. The energy quantum, being released in this fusion-disintegration process, represents a kinematic excitation. An introduction to the kinematic exciton levels analysis is given in [1], where the multilevel exciton scheme is analysed. It is shown that such levels exist for all wave-vector values. It is shown in [2] that the influence of kinematic excitations on luminescent and light absorption processes is a considerable one, and that two levels having a final life-time of the order of 10^{-13} to 10^{-14} s are obtained. An exciton system with a simple lattice in a two-level scheme at low concentrations has been discussed here, while in [3] we have analysed the exciton system at high concentrations and we obtained one kinematic exciton level whose lifetime is of order 10^{-14} to 10^{-15} s. A molecular crystal with a complex lattice at low and high concentrations has been analysed in [4]. The number of kinematic levels is equal to the number of normal exciton levels, and both are equal to the number of sublattices. Nonconservation effects have not been taken into account in the analysis up to now. This has been done in [5] showing that four kinematic levels, having a final lifetime of the order of 10^{-15} s correspond to each of the normal exciton levels. Our purpose is to examine the influence of phonons on kinematic exciton levels. We shall give for $T=300$ K the final results during the life-time of those levels.

The analysis of the combined effect of the exciton-exciton and the exciton-phonon interaction on the dielectrical properties of the crystal [6] proceeded from the total Hamiltonian in the form:

$$\mathcal{H}=\mathcal{H}_{ex}+\mathcal{H}_{ph}+\mathcal{H}_{int} \tag{1}$$

containing optical and mechanical excitations, as well as their interaction. Further analysis required to consider the exciton Green function

$$L_{a-b}(t)=\langle\langle P_a(t)|P_b^+(0)\rangle\rangle \tag{2}$$

i.e. its Fourier transform $L_k(\omega)$.

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Using standard two-time temperature Green functions formalism given in detail in [7] we obtained the final expression for the exciton Green function

$$L_k(\omega)=\frac{i}{2\pi}\,\frac{1+A_1(k,\omega)+A_2(k,\omega)}{\omega-A(k,\omega)} \tag{3}$$

$$A(k,\omega)=\lambda_k+N^{-1}\sum_q a_1^2(k,q)\,[(1+n_q)(\omega-\lambda_{k-q}-\omega_q)^{-1}+$$
$$+n_q(\omega-\lambda_{k-q}+\omega_q)^{-1}]$$

$$A_1(k,\omega)=\frac{8\pi}{iN^2}\sum_{q_1,q_2}[a_2(k,q_1,q_2)+a_3(k,q_1,q_2,\omega)]\int_{-\infty}^{+\infty}d\omega_1\,d\omega_2\times$$
$$\times G_{q_1}(\omega_1)G_{q_2}(\omega_2)G_{q_3}(\omega_3)$$

$$A_2(k,\omega)=-\frac{8\pi}{iN^3}\sum_{q_1,q_2,q_3}[a_4(k,q_1,q_2,q_3)+a_5(k,q_1,q_2,q_3,\omega)]\times$$
$$\times\int_{-\infty}^{+\infty}d\omega_1\,d\omega_2\,G_{q_1}(\omega_1)G_{q_2}(\omega_2)G_{q_3}(\omega_3)$$

$$\lambda_k=\omega_\Delta+\omega_X(k);\quad \omega_\Delta=\hbar^{-1}\Delta;\quad \omega_X(k)=\hbar^{-1}X_k;\quad n_q=\left[\exp\left(\frac{\hbar vq}{\Theta}\right)-1\right]^{-1}$$

$$a_1(k,\omega)=\left(\frac{\hbar}{2M\omega_q}\right)^{-1}[k l_q\lambda_k-(k-q)l_q\lambda_{k-q}];\qquad \Theta=k_BT$$

$$a_2(k,q_1,q_2)=\omega_X(k+q_1-q_2)-\omega_Y(q_1-q_2);$$

$$a_3(k,q_1,q_2,\omega)=\frac{1}{4}a_1(k,q_1)a_1(q_2,q_1)\,[(\omega-\lambda_{k-q_1}-\omega_{q_1})^{-1}-(\omega-\lambda_{k-q}+\omega_{q_1})^{-1}]$$

$$q_3=k+q_1-q_2;\quad \omega_3=\omega+\omega_1-\omega_2\,.$$

Here we wish to emphasize that the Wick theorem for the Bose operators has been strictly applied in decoupling boson Green functions, and that contributions proportional to the exciton concentration have been neglected. If we make in (2) a transition from the Pauli operators P and P^+ to the Bose operators B and B^+ according to formulas from [8] taken in the approximation

$$P=B-B^+BB,\quad P^+=B^+-B^+B^+B, \tag{4}$$

we obtain the following expression

$$L_{ab}(t)\approx G_{ab}(t)+2D_{ab}(t)G_{ab}^2(t) \tag{5}$$

where

$$L_{ab}(t)=\langle\langle P_a(t)|P_b^+(0)\rangle\rangle \tag{6}$$

$$G_{ab}(t)=\langle\langle B_a(t)|B_b^+(0)\rangle\rangle;\quad D_{ab}(t)=\langle\langle B_a^+(t)|B_b(0)\rangle\rangle.$$

Here we have also neglected the terms proportional to the exciton concentration. After Fourier transformations of the type

$$F_{ab}(t)=\frac{1}{N}\sum_k\int_{-\infty}^{+\infty}d\omega F_k(\omega)\exp\,[ik(a-b)-i\omega t] \tag{7}$$

expression (5) takes the form

$$L_k(\omega)=G_k(\omega)+\frac{1}{N^2}\sum_{q_1,q_2}\int_{-\infty}^{+\infty}d\omega_1\,d\omega_2\,G_{q_1}(\omega_1)G_{q_2}(\omega_2)G_{q_3}(\omega_3). \tag{8}$$

As for the boson Green functions G, they should be replaced by the value of the function L in the zero approximation according to the exciton-exciton interaction. This value is obtained from (3) for $A_1 = A_2 = 0$. Instead of that we shall take both functions G in the zero approximation, according to the exciton-exciton and the exciton-phonon interaction, i.e. in the form

$$G = \frac{1}{\omega - \lambda} - i\pi\delta(\omega - \lambda). \tag{9}$$

Using the expression (9) and the approximation

$$1 + A_1 + A_2 \approx (1 - A_1 - A_2)^{-1} + \sigma(A_1^2, A_2^2, A_1A_0),$$

we can express the function L in the following way:

$$L_k(\omega) = G_k(\omega)\,[1 + A_0(\boldsymbol{k}, \omega)] \tag{10}$$

where

$$A_0(\boldsymbol{k}, \omega) = \frac{2\pi(\omega - \lambda)}{i} \frac{1}{N^2} \sum_{q_1 q_2} \int_{-\infty}^{+\infty} d\omega_1\, d\omega_2\, G_{q_1}^{(0)}(\omega_1)\, G_{q_2}^{(0)}(\omega_2) \times \times G_{k-q_1+q_2}^{(0)}(\omega + \omega_1 - \omega_2). \tag{11}$$

If we equalize now the right-hand sides of the expression (3) and (10), we obtain the final expression for the boson Green function $G_k(\omega)$

$$G_k(\omega) = \frac{i}{2\pi} \frac{1}{\omega - A(\boldsymbol{k}, \omega)} \frac{1}{1 + A_0(\boldsymbol{k}, \omega) - A_1(\boldsymbol{k}, \omega) - A_2(\boldsymbol{k}, \omega)}. \tag{12}$$

According to the expression (12) we can determine normal exciton levels as well as kinematic exciton levels in the presence of phonons. We obtain the normal exciton levels from the equation

$$\omega - A(\boldsymbol{k}, \omega) = 0, \tag{13}$$

where kinematic exciton levels are obtained from the equation

$$1 + A_0(\boldsymbol{k}, \omega) - A_1(\boldsymbol{k}, \omega) - A_2(\boldsymbol{k}, \omega) = 0. \tag{14}$$

Further analysis requires a series of simplifications in the expression (14). Those simplifications are mainly obtained by neglecting the dependence of A_0, A_1 and A_2 on the wave vector, since summarizing according to vectors in (3) leads to multiple singular integrals, whose theory has not been fully developed up to now. Hence each phonon frequency is replaced by a Debye frequency and all "non-parallel interactions", i.e. all terms which are proportional to the product $\boldsymbol{q}\boldsymbol{l}_\kappa$ are neglected. After those simplifications the equation (14) obtains the form

$$1 - (\omega - \omega_\Delta)^{-1}[\omega_X(0) - \omega_Y(0)] + i\frac{3\pi}{32}\,\omega_X^{-2}(0)\,(\omega - \omega_\Delta)\,[\omega_X(0) - \omega_Y(0)] - \frac{\hbar\omega_D\omega_\Delta^2}{4Mv^2}(\omega - \omega_\Delta)^{-2}\left\{\frac{\omega_D}{(\omega - \omega_\Delta)^2 - \omega_D^2} - \frac{(n_D + 2)\,[\omega_X(0) - \omega_Y(0)]}{(\omega - \omega_\Delta - \omega_D)^2} - \frac{(n_D - 1)\,[\omega_X(0) - \omega_Y(0)]}{(\omega - \omega_\Delta - \omega_D)^2}\right\} = 0 \tag{15}$$

$$\omega = \omega_1 + i\omega_2\,.$$

From the imaginary part of the kinematic exciton level frequency we can determine the lifetime of the kinematic exciton level. We solve the equation (15) in the approximation $|\omega_1 - \omega_\Delta| \ll \omega_2$ and take the usual values from crystallooptics

$$M = 2 \times 10^{-22}\ \text{g}, \quad v = 10^8\ \text{cm/s}, \quad \omega_D = 1.1 \times 10^{13}\ \text{Hz}, \quad \omega_\Delta = 4.4 \times 10^{15}\ \text{Hz}, \quad \omega_X(0) = 0.9 \times 10^{14}\ \text{Hz}, \quad \omega_Y(0) = 1.4 \times 10^{14}\ \text{Hz}. \tag{16}$$

According to (15) and (16) at $T = 300$ K we obtain $\tau = 1.1 \times 10^{-15}$ s. As we can see, we have a good agreement with experimental results [9], but it should be mentioned that we think that even a better agreement with experiment can be reached through a more sophisticated exciton-phonon interaction analysis, which will be the aim of our further research. This analysis, as well as previous ones ([1] to [5]), contributes to the assertion that kinematic exciton levels, rather than normal exciton levels, appear and are observed in the experiment.

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