DIELECTRIC CONSTANT OF CRYSTAL IN FUNCTION OF THE WAVE VECTOR

D. Lj. Mirjanić*, B. S. Tošić** and J. P. Šetrajčić**
*) Faculty of Technology, University „Đuro Pucar Stari" of Banja Luka
**) Institute of Physics, Faculty of Science, University of Novi Sad

The aim of this paper is to investigate the effects of phonons on the dielectric characteristics of molecular crystals. There are number of papers that treat this problem but all take the Hamiltonian of exciton-phonon interaction to be of the cubic form that consists of pair of excitonic operators and one phononic operator. Here we will examine different approach which is based on an idea of hybridization of excitonic and vibrational excitations. As known, hybridization of two different types of excitations can occur only if their Hamiltonian of interactions takes quadratic form where the operator pairs contain operator products one or the other type of quasi-particle.

There is a question about the creation of quadratic form on excitonic and phononic operators which will represent interaction term of the lowest order. One must say that this form can appear only in crystals where the centar of inversion of the molecule does not coincide with the centar of inversion of the molecule does not coincide with the centar of inversion of the crystal. Then the matrix elements of the operator dipol-dipol [1] interaction $V_{\vec{n}\vec{m}}$ (fooo), $V_{\vec{n}\vec{m}}$ (ofoo), $V_{\vec{n}\vec{m}}$ (oofo) and $V_{\vec{n}\vec{m}}$ (ooof), where „o" denotes lowest state of the molecule, and „f" denotes his excited state, are different from zero and their presence in excitonic Hamiltonian causes bilinear form of excitonic and phononic operators. Besides mentioned matrix elements excitonic Hamiltonian contains also linear terms in excitonic creation and annihilation operators. Before elimination of these terms, which can always be done using a well known Bogolyubov procedure [2], where one expands the matrix elements on phononic displacements using scheme: $V_{\vec{n}\vec{m}} P_{\vec{n}} \rightarrow V_{\vec{n}-\vec{m}} + U_{\vec{n}} - U_{\vec{m}} P_{\vec{n}} = V_{\vec{n}-\vec{m}} P_{\vec{n}} - P_{\vec{n}} (U_{\vec{n}} - U_{\vec{m}}) \nabla V_{\vec{n}-\vec{m}}$ These first term is eliminated from Hamiltonian, and second leads to the bilinear form in excitonic operators P and on phononic operators b which express the displacements. If to such Hamiltonian of exciton-phonon interaction, we add quadratic excitonic Hamiltonian and quadratic phononic Hamiltonian we get the bilinear form which by „u – v" Bogolyubov's transformations [3] can be diagonalized. Energies of hybrid excitations are gotten as a solution of secular equation of the system which defines functions u and v (details of this calculation can be seen in [4]):

$$H_{eff} = H_o + \sum_{k,\sigma} \lambda_\sigma(\vec{k})\; a_\sigma^+(\vec{k})\; a_\sigma(\vec{k}) \qquad (1)$$

where $\lambda_\sigma(\vec{k})$ are energies of hybrid excitations, and:

$$\lambda_1(\vec{k}) = \Delta + \frac{\hbar^2 k^2}{2m^*} \; ; \quad m^* \approx m_{ex}\left(1 - \frac{2|V|^2 m_{ex}}{M\Delta^2}\right); m_{ex} = \frac{\hbar^2}{2X_{a^2}}$$

$$\lambda_2(\vec{k}) = \hbar\omega k \; ; \quad \omega = v - \frac{|V|^2}{Mv\Delta} \qquad (2)$$

As can be seen the energy of first branch of hybrid excitations λ_1 is close to excitonic energy Δ. The effect of hybridization is manifested on lowering the excitonic effective mass $m < m_{ex}$, which in the last consequence means that these hybrid excitations are somewhat faster than exciton. As for as the energie of the second branch of hybrid excitations λ_2, it is somewhat lower than the phonon energy, due to the decrease in the speed of sound $\omega < v$. Using Hamiltonian (1) and the energy relations (2) we can calculate dielectric constant of the system. In calculation we will use only onedimensional molecular structure.

Expression for dielectric constant of the system, which one gets using famous Djaloshinskii and Pitajevskii procedure [5], in isotropic approximation has a form:

$$\epsilon^{-1}(k,\omega) = 1 - i\,\frac{\tau_o S_o^2}{4\hbar} \sum_{\sigma=1}^{2} \left[G_\sigma(k,\omega) + G_\sigma(k,-\omega)\right]. \qquad (3)$$

where S_o is the intensity of the local electric field of the elementary crystal cell and τ_o is the volume of the cell. $G_\sigma(k,\omega)$ are retarted comutator Green's functions formed by operators a^+ and a which create and annihilate hybrid excitations. On the basis of paper [6] this functions have the form:

$$G_\sigma(k,\omega) \equiv \langle\langle a_\sigma(k) \,|\, a_\sigma^+(k) \rangle\rangle_\omega = \frac{i}{2\Pi}\,\frac{1}{\omega - \Omega_\sigma(k)}; \qquad (4)$$

$$\Omega_\sigma(k) = \frac{\lambda_o(k)}{\hbar}; \quad \sigma = 1,2$$

Substituting (4) into (3) we get:

$$\epsilon^{-1}(k,\omega) = 1 + \frac{\tau_o S_o^2}{4\Pi\hbar}\left[\frac{\Omega_2(k)}{\omega^2 - \Omega_1^2(k)} + \frac{\Omega_2(k)}{\omega^2 - \Omega_2^2(k)}\right]; \qquad (5)$$

$$\Omega_1(k) = \Omega_\Delta + \frac{\hbar k^2}{2m^*} \; ; \quad \Omega_2(k) = \omega k; \; \Omega_\Delta = \frac{\Delta}{\hbar}$$

Numerical analysis of dielectric constant ϵ (k, ω) is done using the following set of numerical data: $\mu = 5 \cdot 10^{-3}$, $m^* = 10^{-30}$ kg, $M = 10^{-25}$ kg; $\omega = 3 \cdot 10^3$ ms^{-1}; $\frac{\Omega_X}{\Omega_\Delta} = \frac{1}{50}$; $\Omega_\Delta = 5 \cdot 10^{15}$ s^{-1} ; $\frac{|V|}{\Delta} = 10^{-1}$; $v = 3 \cdot 1 \cdot 10^3$ ms^{-1} for frequencies $\frac{\omega}{\Omega_\Delta}$ = 0.8; 0.95; 1.1 and it is shown in Fig. 1.

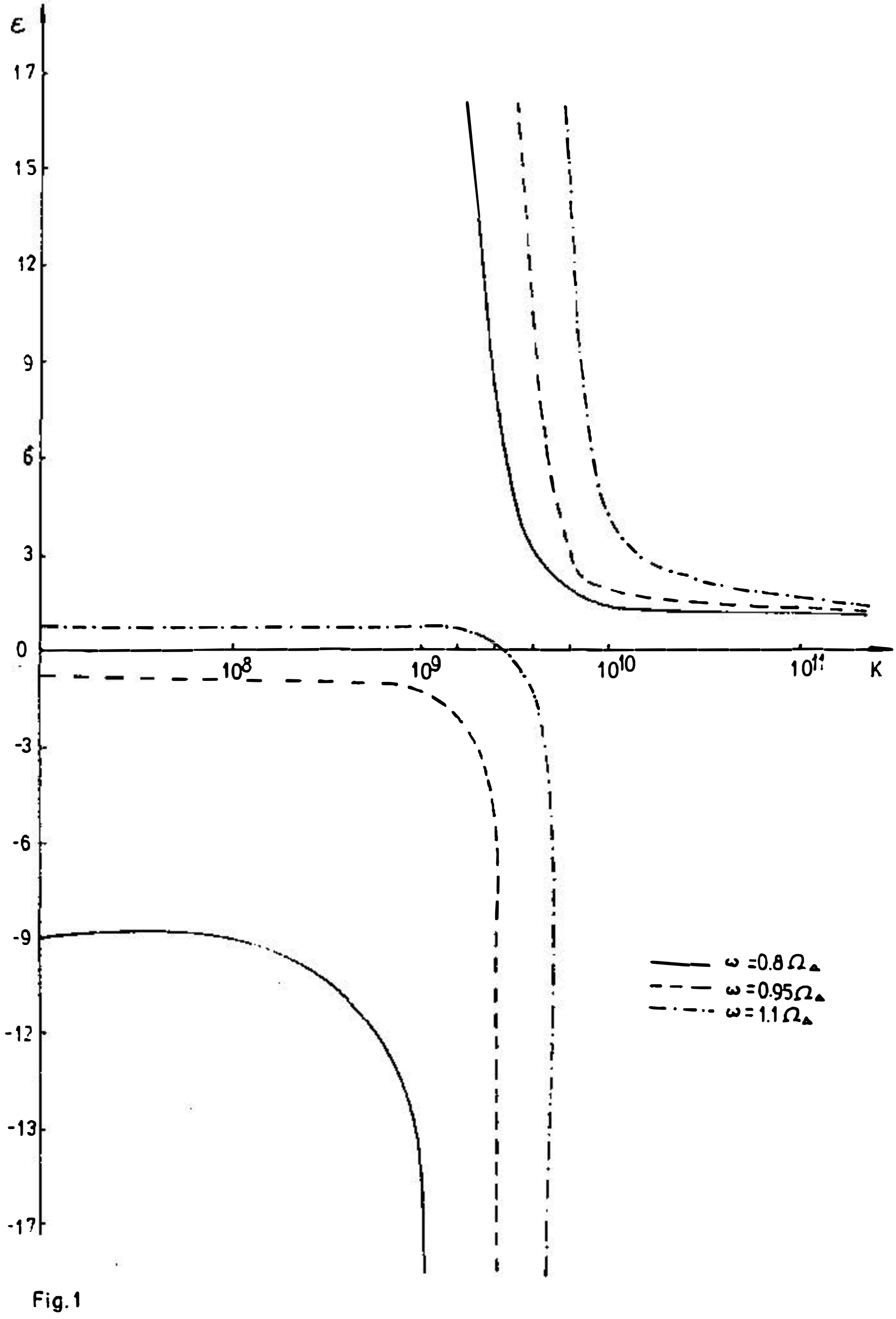

Fig. 1

On the basis of procedure analysis we can conclude that dielectric constant has a singularity in point k_S which has in the interval $10^9 - 10^{10}$ m^{-1}. For $k < k_S$ $\epsilon < 1$, but its negative for $\omega < \Omega_\Delta$ and for $\omega > \Omega_\Delta$ it is positive. In the region $k > k_S$ $\epsilon > 1$ and $\epsilon \rightarrow 1$ when $k \rightarrow +\infty$.

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