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The Frenkel Exciton Spectrum under the Condition of Bose Condensation

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Abstract_ Bose condensation of Frenkel excitons is studied and corrections of the superfluid spectrum of excitations in the system of noncondensed bosons due to the nonlinear effects are obtained

The problem of exciton Bose condensation has been studied by many authors[1-4], and recent experimental evidences in favour of it are available[5]. The question unresolved yet, is, to what ex - tent does the fact that excitons are not really bosons, influence their collective properties.

The collective properties of Frenkel excitons had been studied in some detail in [3,6]. If one uses the exact bosonic representa - tion of Pauli operators describing excitons, as a consequence of different commutation relations, there arises so called "kinematic" interaction. It leads to the repulsion between excitons at small distances (due to the scattering on a $\hat{\delta}$ potential). In this way we can treat the system as a nonideal Bose gas in the approximation of hard spheres. In the presence of Bose condensation, we start with the following Hamiltonian, applying standard Bogolyubov procedure[7]

$$H=\sum_{\vec{k}\neq 0}\left[\alpha(\vec{k})+b\right]B^{+}_{\vec{k}}B_{\vec{k}}+\frac{1}{2}\sum_{\vec{k}\neq 0} b\left(B^{+}_{\vec{k}}B^{+}_{-\vec{k}}+B_{\vec{k}}B_{-\vec{k}}\right)+$$

$$+\frac{u\,\bar{n}^{2}\hbar^{2}a^{2}}{N\nu_{0}m^{*}}+\frac{\tilde{\gamma}_{0}}{N}\sum_{\vec{k}_1,\vec{k}_2,\vec{k}_3\neq 0}B^{+}_{\vec{k}_1}B^{+}_{\vec{k}_2}B_{\vec{k}_3}B_{\vec{k}_1+\vec{k}_2-\vec{k}_3} \qquad (1)$$

where $\alpha(\vec{k})=\frac{\hbar^2 k^2}{2m^*}$ represents the exciton energy in the effective mass approximation, m^* - the exciton effective mass and

$$\beta=n_0\gamma_0\ ,\qquad \gamma_0=\frac{2\pi\hbar^2 a}{m^*\Omega_0}\ ,\qquad n_0=\frac{\mathcal{N}_0}{N} \tag{2}$$

$\mathcal{N}$ is the total number of the excitons, $\mathcal{N}_0$ the number of condensed excitons, N the number of lattice sites and Ω_0 the elementary cell volume. $B_{\vec{k}}$ are Bose operators in momentum representation.

If there were no fourth order term, the result would be the standard superfluid spectrum[7]. Following theese ideas, we introduce new Bose operators $C_{\vec{k}}$ defined as

$$B_{\vec{k}}=u_{\vec{k}}C_{\vec{k}}+v_{\vec{k}}C^+_{-\vec{k}}\ ;\qquad u^2_{\vec{k}}-v^2_{\vec{k}}=1 \tag{3}$$

Substituting (3) into (1) and reducing all operator forms to normal products, we retain in the Hamiltonian the part quadratic in , and also the forms of the type $C^+C^+C^+C$ + h.c., which contribute to the spectrum in the second order perturbation theory. We eliminate the terms of the type CC + h.c. from the quadratic part, and the contribution of fourth order terms we calculate applying the Green's functions method[8,9]. As t e result of such procedure, we obtained the following spectrum:

$$\mathcal{E}(\vec{k})=E(\vec{k})+\delta E(\vec{k}) \tag{4}$$

where $E(\vec{k})=\left\{[\alpha(\vec{k})+\beta+\hat{\delta}_1]^2-(\beta+\hat{\delta}_2)^2\right\}^{1/2}$ with $\hat{\delta}_1$ and $\hat{\delta}_2$ satisfying the following nonlinear integral relations

$$\hat{\delta}_1=\frac{4\gamma_0}{N}\sum_{\vec{q}}v^2_{\vec{q}}\ ;\qquad \hat{\delta}_2=\frac{2\gamma_0}{N}\sum_{\vec{q}}u_{\vec{q}}v_{\vec{q}} \tag{5}$$

$$v^2_{\vec{q}}=\frac{1}{2}\left(\frac{\alpha(\vec{q})+\beta+\hat{\delta}_1}{E(\vec{q})}-1\right);\qquad u_{\vec{q}}v_{\vec{q}}=-\frac{\beta+\hat{\delta}_2}{2E(\vec{q})} \tag{6}$$

and $\delta E(\vec{k})$ - the correction of the second order in γ_0 is given by the following relation:

$$\delta E(\vec{k})=\left(\frac{\gamma_0}{N}\right)^2\sum_{\vec{k}_1\vec{k}_2}\frac{\Pi_{\vec{k}_1\vec{k}_2\vec{k}}}{E(\vec{k})-E(\vec{k}_1)-E(\vec{k}_2)-E(\vec{k}-\vec{k}_1-\vec{k}_2)} \tag{7}$$

where: $\Pi_{\vec{k}_1\vec{k}_2\vec{k}} = L_{\vec{k}_1,\vec{k}_2,\vec{k}-\vec{k}_1-\vec{k}_2}\big(L_{\vec{k}_1,\vec{k}_2,\vec{k}-\vec{k}_1-\vec{k}_2} + L_{\vec{k}_2,\vec{k}_1,\vec{k}-\vec{k}_1-\vec{k}_2} +$

$$+ L_{\vec{k}_1,\vec{k}-\vec{k}_1-\vec{k}_2,\vec{k}_2} + L_{\vec{k}_2,\vec{k}-\vec{k}_1-\vec{k}_2} + L_{\vec{k}-\vec{k}_1-\vec{k}_2,\vec{k}_1,\vec{k}_2} + L_{\vec{k}-\vec{k}_1-\vec{k}_2,\vec{k}_2,\vec{k}_1}\big) \quad (8)$$

and $$L_{\vec{k}_1,\vec{k}_2,\vec{k}_3} = 2\left(u_{\vec{k}_1} u_{\vec{k}_2} v_{\vec{k}_2} u_{\vec{k}_1+\vec{k}_2+\vec{k}_3} + v_{\vec{k}_1} v_{\vec{k}_2} u_{\vec{k}_3} v_{\vec{k}_1+\vec{k}_2+\vec{k}_3}\right) \quad (9)$$

Solving (5) approximately, we find $\delta_1 = \phi_0 \gamma_0 n_0^{3/2} + O(n_0^{3/2})$ and $\delta_2 = -\phi_2 \gamma_0 n_0 + O(n_0^{3/2})$ where $0<\phi_0 \lesssim 1$ and $0<\phi_2 \leq 1$. The further analysis of the spectrum (4) was performed numerically for the set of parameters: $m^* = 50 m_e$ and $n_0 = 10^{-2}$. The results are presented on the Figure 1. The values of the wavevector are given in cm^{-1}.

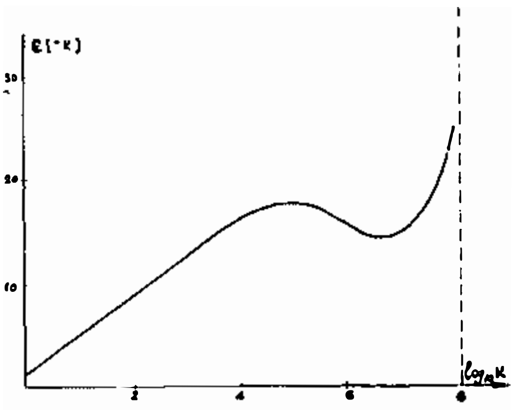

Fig. 1 Spectrum described by (4)

The spectrum (4) differs essentially from those obtained by Bogolybov[7]. It posesses the rotonic minimum for high momenta. The small gap appearing in the spectrum can be considered as the result of the crude hard sphere model for the scattering potential. The more realistic choice of the scattering potential would lead to the absence of the gap, analogously to the result of Sunakawa et al[10], where the problem og liquid He^4 was studied.

If the exciton system during the exciton lifetime ($\tilde{\tau}$) can achieve quasiequilibrium state, (which is possible for $\tilde{\tau} > \tau_e$, where τ_e is the relaxation time of the system lattice + excitons), one can examine reversible optical hydrodynamical phenomena, in the same way as it was done for Wannier-Mott excitons[1,2].(The condition $\tau > \tau_e$ iscertainly fullfiled for triplet excitons[6] because $\tau \sim 10^{-3}$ sec, and $\tau_e < 10^{-10}$ sec.)

The most interesting result is the possibility of the super - fluid energy transfer through the crystal. If N_0 condensate excitons move superfluidly with the velocity v_s ($v_s < v_{cr} = \frac{\hbar}{m^* d} \ln \frac{d}{r}$ where d is the crystal thickness and r the radius of a vortex[1]) then during the lifetime they will pass the distance $l = v_s \tau$ (which is much longer than the mean free path of the excitons during the difusion process[6]) and during the deexcitation they release the energy $N_0 \Delta$, where Δ is the excitation energy of a molecule. This kind of movement of the condensate excitons one can achieve by keeping the temperature difference on the edges of the crystals, which presents an analogy with the thermomechanical effect in superfluid He^4.

R E F E R E N C E S

1)S.A.Moskalenko, Fiz. tverd. tela 4 (1962) 276

2)V.A.Gergel, R.F.Kazarinov, R.A.Suris, Zh.eksp. teor. Fiz. 53 (1967) 544; 54 (1968) 298

3)V.M.Agranovič, B.S.Tošić, Zh.eksp. teor. Fiz. 53 (1967) 149

4)L.V.Keldiš in "Problems of Theoretical Physics", Izd. Nauka, Moskvs 1972 (in Russian)

5)M.Grosmann, S.G.Ekomoss and J.Ringeisen, eds: "Molecular Spectroscopy of Dense Phases", Elsevier Publ. Comp., Amsterdam (1976)

6)V.M.Agranovič, "Theory of Excitons", Izd. Nauka. Moscow (1968) (in Russian)

7)N.N.Bogolybov, J.Phys. 9 (1947) 23