LONG RANGE ORDER AND SUPERFLUIDITY FOR BOSE CONDENSED EXCITONS

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ABSTRACT

The problem whether a Bose-Einstein condensed system of Wannier excitons will exhibit superfluid properties is briefly reviewed. The long range order of such a system is shown to lead to a two-fluid model, which describes the superfluid flow of excitation energy. Qualitative remarks are given about the influence of perturbations, which will cause the exciton superfluidity to decay in time.

About 15 years ago the possibility of a Bose-Einstein condensation of excitons in highly excited semiconductors at low temperatures has been considered for the first time[1,2]. The analogy with He-II soon gave rise to speculations about the superfluid properties of the Bose condensed exciton gas. Moskalenko[2] argued on the basis of a two-fluid model that a crystal with a Bose condensed exciton gas would act as a thermal superconductor. However, the direct observations of these superfluid effects are rather difficult and it was suggested to search for evidence of a change in the excitation spectrum of the exciton system, which should alter at the inset of condensation from a quadratic to a linear spectrum for small momentum values. The consequences of superfluidity for the exciton transport has further been treated in terms of a Ginzburg-Landau equation.[3] The penetration depth of excitons is expected to increase considerably if the condensation takes place. Keldysh and Kozlov[4] as well as Hanamura[5] stressed the importance of the fact that excitons are not perfect Bosons. The deviations from the Bose nature leads to a repulsive interaction between two excitons if the spin is not taken into account. Both theories, the pairing theory of ref.4 and the Boson treatment of ref.5 confirmed the existence of a condensate and of a linear excitation spectrum, which have been assumed in the earlier treatments. All existing microsopic theories of the condensed exciton phase are limited to the low density limit $na_o^3 \ll 1$, where n is the exciton concentration and a_o the exciton Bohr radius. The region in which the exciton density is close to the ionization limit $(na_o^3 \approx 1)$ is up to now not accessible to theory. If the spins of the electrons and holes are taken into account, the exciton molecule can be shown to be stable. The interaction between excitation molecules is repulsive, so that a Bose-Einstein condensation of excitonic molecules rather than of excitons is to be expected.[6] In the following we disregard the formation of molecules (by omitting the spin variables), because the question about the possibility of superfluidity is fundamentally the same for a gas of excitons or excitonic molecules. The possibility of superfluidity has been questioned by Kohn and Sherrington[7] for all composite Bosons, which are built up from particles and holes (*e.g.* excitons or excitonic molecules). We reinvestigate this problem, following essentially the approach by Hanamura and the author[8]. We treat a system of Wannier excitons which is supposed to be in thermal quasi-equilibrium; *i.e.* we assume that the exciton relaxation time is much smaller than the exciton life time. Under these conditions one can describe a quasi-stationary state, in which the generation rate is balanced by the decay rate, by the introduction of a quasi-chemical potential. We do not treat the interactions of the electron-hole system with other perturbing fields such as photons, phonons and impurities, assuming that these interactions are weak as compared to the interactions within the system. Furthermore, a direct semiconductor is treated, but the generalization to an indirect one is straightforward.[8] The Hamiltonian for this system in the effective mass approximation is

$$H = \int d^3x_e \; \psi^+(\vec{x}_e) \; (-\frac{\hbar^2}{2m} \nabla^2_{x_e} - \mu_e + E_g) \; \psi(\vec{x}_e)$$

$$+ \int d^3x_h \; \phi^+(\vec{x}_h) \; (-\frac{\hbar^2}{2m_h} \nabla^2_{x_h} - \mu_h) \; \phi(\vec{x}_h) \qquad (1)$$

$$+ \tfrac{1}{2} \int d^3x_e \int d^3x'_e \; V(|\vec{x}_e - \vec{x}'_e|) \; \psi^+(\vec{x}_e) \; \psi^+(\vec{x}'_e) \; \psi(\vec{x}'_e) \; \psi(\vec{x}_e)$$

$$+ \tfrac{1}{2} \int d^3x_h \int d^3x'_h \; V(|\vec{x}_h - \vec{x}'_h|) \; \phi^+(\vec{x}_h) \; \phi^+(\vec{x}'_h) \; \phi(\vec{x}'_h) \; \phi(\vec{x}_h)$$

$$- \int d^3x_e \int d^3x_h \; V(|\vec{x}_e - \vec{x}_h|) \; \psi^+(\vec{x}_e) \; \phi^+(\vec{x}_h) \; \phi(\vec{x}_h) \; \psi(\vec{x}_e),$$

where ψ, ϕ and μ_e, μ_h are the field operators and the chemical potentials of the electrons and holes, respectively. $V(r)$ is the Coulomb potential. The condensed state of the exciton system (eq.1) has been analysed in ref.4 and ref.5. Using the results of these treatments, one can show that the second reduced electron-hole density matrix is of the form

$$< \psi^+(\vec{x}'_e) \; \phi^+(\vec{x}'_h) \; \phi(\vec{x}_h) \; \psi(\vec{x}_e) > = \varphi^*(r') \, \varphi(r) \; \rho_1(\vec{R};\vec{R}'),$$

$$\rho_1(\vec{R};\vec{R}') = \Psi^*(\vec{R}')\Psi(\vec{R}) + \tilde{\rho}_1(\vec{R};\vec{R}'), \qquad (2)$$

where $\vec{r}$ and $\vec{R}$ are the relative coordinate and the center of mass coordinate of an exciton, respectively. φ is the wave function of the internal exciton motion in the lowest state; we call $\rho_1(\vec{R};\vec{R}')$ the first reduced exciton density matrix. ρ_1 shows the well-known off-diagonal long range order (ODLRO) with the order parameter $\Psi(\vec{R})$ and the non-condensate part $\tilde{\rho}_1$ which decays as $|\vec{R}-\vec{R}'| \to \infty$. The question now is,[7] whether ODLRO of the electron-hole density matrix (which corresponds to DLRO for a particle-particle density matrix) does lead to a superfluid behaviour of the exciton system. In order to investigate this question, we derive the equation of motion for the second electron-hole density matrix by using the Heisenberg equations of the various field operators. Because we confine ourselves to the region of the phase diagram in which only excitons exist, we pair each unpaired field operator which appears in the resulting equation. The product of two unpaired electron operators, *e.g.*, would be transformed into

$$<\psi^+(\vec{x}_e') \; \psi(\vec{x}_e) > \to \int d^3r' \int d^3r < \psi^+(\vec{x}_e') \; \phi^+(\vec{x}_h') \; \phi(\vec{x}_h) \; \psi(\vec{x}_e) > \varphi(r') \; \varphi^*(r) \, . \tag{3}$$

Finally, we eliminate the internal exciton motion and obtain an equation for the first reduced exciton density matrix

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \rho_1(\vec{R};\vec{R}') &= - \frac{\hbar^2}{2M} (\nabla_R^2 - \nabla_{R'}^2) \; \rho_1(\vec{R};\vec{R}') \\ &+ \int d^3R'' \; \{W(|\vec{R}-\vec{R}''|) - W(|\vec{R}-\vec{R}'|)\} \; \rho_2(\vec{R},\vec{R}'';\vec{R}'';\vec{R}'',\vec{R}') , \end{aligned} \tag{4}$$

where the exciton-exciton interaction is given by

$$\begin{aligned} W(|\vec{R}-\vec{R}'|) &= \int d^3r \int d^3r' \; \{V(|\vec{R}-\vec{R}'|) + V(|\vec{R}-\vec{R}' + \vec{r}-\vec{r}'|) \\ &- V(|\vec{R}-\vec{R}'-\vec{r}'|) - V(|\vec{R}-\vec{R}'+\vec{r}|)\}\{|\varphi(r)|^2 \, |\varphi(r')|^2 \\ &- \varphi(|\vec{r}+\vec{R}-\vec{R}'|) \; \varphi^+(r) \; \varphi(|\vec{r}'+\vec{R}'-\vec{R}|) \; \varphi^+(r')\} \\ &\approx \delta^3(\vec{R}-\vec{R}') \; \frac{26}{3} \, \pi \, E_{ex}^b \; a_o^3 \; , \end{aligned} \tag{5}$$

where E_{ex}^b is the exciton binding energy and a_o the exciton Bohr radius. This result is valid in the low density limit $na_o^3 \ll 1$ and for $m_h \gg m_e$. The interaction potential is repulsive due to exchange effects. Using the same techniques as above, we further derive an equation of motion for the exciton order parameter

$$\begin{aligned} i\hbar \, \dot{\Psi}(\vec{R}) &= [- \frac{\hbar^2}{2M} \nabla_R^2 + E_g - E_{ex}^b - (\mu_e + \mu_h)] \Psi(\vec{R}) \\ &+ \int d^3R' \; W(|\vec{R}-\vec{R}'|) \; \rho_{3/2}(\vec{R},\vec{R}';\vec{R}') \; ; \end{aligned} \tag{6}$$

where $\rho_{3/2}$ is the three-leg function.

Both equations (4) and (6) are of the same form as the corresponding ones for a system of interacting elementary Bosons.[9] As is known from the theory of He-II, eqs.(4) and (6) are sufficient to derive rigorously the following conservation laws

$$\frac{\partial}{\partial t} n + \vec{\nabla}\cdot\vec{j} = 0 \qquad \frac{\partial}{\partial t} E + \vec{\nabla}\cdot\vec{Q} = 0$$
$$\frac{\partial}{\partial t} \vec{j} + \vec{\nabla}\cdot\overleftrightarrow{\Pi} = 0 \qquad \frac{\partial}{\partial t} \vec{v}_s + \vec{\nabla}\left(\tfrac{1}{2} \vec{v}_s^{\,2} + \frac{\mu}{M}\right) = 0, \tag{7}$$

where n, $\vec{j}$ and E are the densities of the particles, the particle current and the energy, respectively. $\overleftrightarrow{\Pi}$ is the stress tensor, $\vec{Q}$ the energy current. The superfluid velocity is defined as $\vec{v}_s=\vec{\nabla}\theta h/M$ where θ is the phase of the order parameter Ψ. In order to derive a hydrodynamic two-fluid model from these equations, we have to assume that the system is in local equilibrium, *i.e.* we must be in a regime which is dominated by exciton-exciton collisions (the local equilibrium should not be established primarily by exciton-phonon collisions). If these conditions are fulfilled, we obtain a two-fluid model, which describes the superfluid flow of excitation energy (not of mass and charge). Because the interaction (5) is weak, one can use the Bogolubov theory to calculate all thermo-dynamic quantities explicitly.

The finite lifetime of excitons will make superfluidity a transient phenomena. Recently, Nagaoka[10] and Nakajima[11] stressed that in a real system the phase θ of the order parameter is most likely pinned by number-nonconserving processes. In order to see the essence of this mechanism, it is sufficient to add to a Hamiltonian of weakly interacting Bosons the following perturbation

$$\sum_k (g_k\, b_k^+ + g_k^+\, b_k) \tag{8}$$

where b_k is a Boson operator, and g_k is assumed to be a classical amplitude. In the condensed state we get in addition to the Bogolubov Hamiltonian a term which depends explicitly on the phase. Writing $g_o = |g_o| \exp(i\alpha)$, we get $\Delta E = 2|g_o| n_o^{1/2} \cos(\alpha-\theta)$. The ground state is no longer continuously degenerate with respect to Θ, thus $\vec{v}_s$ is no longer a hydrodynamic variable. Perturbations like eq.(8) could arise mainly from interactions of the excitons with the light field and impurities. Thus it remains an open question, whether real systems can be found and prepared in which number-nonconserving processes are so weak, that effects of a superfluid flow of excitation energy can be observed.

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