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Derivation of the two-fluid model for Bose-condensed excitons[*]

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The second reduced electron-hole density matrix is shown to have the property of off-diagonal long-range order for a Bose-condensed system of Wannier excitons. From the equations of motion for this density matrix the conservation laws for the density, the particle current density, and the energy density of the excitons are derived, as well as the equation of motion for the velocity of the condensate. These equations are obtained by projecting the electron-hole density matrices into the exciton space. From the above-mentioned conservation laws the two-fluid model is deduced, which describes the superfluid flow of excitation energy. In the low-density limit all hydrodynamical quantities are calculated explicitly. Extensions to the case of condensation in the presence of static homogeneous electromagnetic fields and to the case of indirect semiconductors are given.

I. INTRODUCTION

The idea of a Bose condensation of Wannier excitons was first discussed by Blatt *et al.*[1] in terms of an ideal Bose gas. They noted that due to the small effective masses in semiconductors the critical exciton concentration for a Bose condensation at helium temperatures is of the order of 10^{16} cm^{-3}. Casella[2] investigated more closely the following problem: in which types of semiconductors these concentrations of excitons could be obtained in a quasiequilibrium situation. The excitons system is in a quasiequilibrium if the thermal relaxation time of the exciton gas is much smaller than the exciton lifetime. Subsequently, several authors discussed, in analogy with superfluid helium, aspects of the superfluidity of such a condensed system of exciton bosons, both in terms of a Landau two-fluid model[3] and in terms of a Landau-Ginzburg theory.[4] In recent years, theoretical interest has turned to the statistical properties of excitons at high densities. Owing to the Fermi statistics of the electrons and holes, the exciton statistics deviate from the simple Bose statistics. This is evident at exciton concentrations n, for which $na_0^{*3} \approx 1$, where a_0^* is the exciton Bohr radius. At these concentrations, the bound exciton states merge into the ionization continuum, i.e., the exciton system goes over into a electron-hole plasma. But even at concentrations far below the Mott transition, where $na_0^{*3} \ll 1$ and where the Bose condensation is supposed to occur at sufficiently low temperatures, the influence of the underlying Fermi statistics is important. The problem is closely related to that of composite particles in nuclear physics. It is therefore not surprising that the same methods which have been proven to be successful in nuclear physics found application in the exciton problem. Keldysh and Kozlov[5] used a BCS-type pairing theory for the electrons and holes and found a Bogolubov spectrum for the elementary excitations in a condensed system. Hanamura[6] made use of a boson description by Usui[7] and Marumori,[8] in which the real excitons are projected into an ideal boson space. Deviations of the exciton statistics from that of bosons results in effective interactions between the excitons. The methods of Refs. 5 and 6 have been shown to be equivalent in the lowest order of exciton concentration.[9] Kohn and Sherrington[10] pointed out that only composite bosons, which are built from Fermi particles, show off-diagonal long-range order (ODLRO) in the reduced particle density matrices. Composite bosons consisting of particles and holes, such as excitons, show diagonal long-range order (DLRO) in the reduced particle density matrices. Because ODLRO in this sense is a necessary and sufficient condition for superfluid flow of charge and mass, Kohn and Sherrington concluded that an exciton system can never become superfluid.

It is the purpose of this paper[11] to investigate the connection between the long-range order in the reduced density matrices and the possibility of superfluidity in a condensed exciton system again. The characteristic long-range order in an exciton system is the correlation between an annihilation of an exciton and the generation of another one at distant locations. Such a correlation is described by the second reduced electron-hole density matrix and not by the second reduced particle density matrix which has been considered by Kohn and Sherrington. The reduced electron-hole density matrices will be shown (Sec. II) to have ODLRO for a condensed exciton system (this corresponds to the DLRO found by Kohn and Sherrington for the reduced particle density matrices). Subsequently, it will be shown that the property of ODLRO of the reduced electron-hole density matrices leads directly to a two-fluid model, which describes the superfluid flow of excitation energy in contrast to a superfluid flow of charge or mass. For this purpose, we project the electron-hole reduced density matrices into the exciton space to obtain reduced exciton density matrices, which obey the equations of motion which are typical for an interacting Bose system (Sec. III).[12]

The resulting exciton-exciton interaction is repulsive for spinless electrons and holes owing to the Pauli principle. From the equations of motion for the exciton density matrices we deduce (Sec. IV) the conservation laws of the density, the particle current density, and the energy density of the excitons, together with an equation of motion for the velocity of the condensate component (which is equivalent with the velocity of the superfluid component). The equations form the basis of the two-fluid model. In the low-density limit $na_0^{*3} \ll 1$, to which we confine ourselves, all hydrodynamic quantities of the two-fluid model can be evaluated within the Bogolubov approximation (Sec. V). This derivation provides us with the dependences of all hydrodynamic quantities on the temperature, the exciton concentration, and the relative velocity between the super and the normal component of the exciton system. In Sec. VI we will extend the treatment to the case of excitons in static, homogeneous electromagnetic fields and to the case of excitons in indirect semiconductors.

II. ODLRO OF THE REDUCED ELECTRON-HOLE DENSITY MATRICES

The correlation of the annihilation of one electron-hole pair and the generation of another one at a different location is described by the second reduced electron-hole density matrix

$$\rho_2^{e-h}(\vec{x}_h, \vec{x}_e; \vec{x}_h', \vec{x}_e') = \langle \psi^\dagger(\vec{x}_e')\phi^\dagger(\vec{x}_h')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle . \quad (2.1)$$

Here, $\psi(\vec{x}_e)$ and $\phi(\vec{x}_h)$ are the electron and hole field operators, respectively, which obey Fermi statistics. The reduced density matrix can be calculated for a low-density exciton gas by making use of the BCS-type theory of Ref. 5. The leading term of the result is

$$\rho_2^{e-h}(\vec{x}_h, \vec{x}_e; \vec{x}_h', \vec{x}_e') = \varphi^*(r')\varphi(r) n e^{i\vec{K}\cdot(\vec{R}-\vec{R}')} , \quad (2.2)$$

where $\varphi(r)$ is the wave function of the internal exciton motion in the lowest state. The coordinates $\vec{r}$ and $\vec{R}$ are given by

$$\vec{r} = \vec{x}_e - \vec{x}_h \quad \text{and} \quad \vec{R} = \alpha\vec{x}_h + \beta\vec{x}_e , \quad (2.3)$$

with

$$\alpha = m_h/M \quad \text{and} \quad \beta = m_e/M ,$$

where $M = m_e + m_h$ is the translational mass of an exciton. $\vec{K}$ is the center-of-mass wave vector, n is the exciton density. The same result can be derived within the boson formalism of Ref. 8, by assuming a coherent antisymmetrized Glauber state for the condensed exciton system. The form (2.2) shows complete ODLRO in the sense, that ρ_2^{e-h} does not decay as the pair $(\vec{x}_e, \vec{x}_h)$ is taken far apart from the pair $(\vec{x}_e', \vec{x}_h')$, i.e., $\vec{r} \approx \vec{r}' \approx 0$ and $|\vec{R} - \vec{R}'| \to \infty$. If only a fraction of all excitons is condensed, ρ_2^{e-h} will be of the form

$$\begin{aligned} \rho_2^{e-h}(\vec{x}_h, \vec{x}_e; \vec{x}_h', \vec{x}_e') &= \varphi^*(r')\varphi(r)\rho_1(\vec{R}; \vec{R}') , \\ \rho_1(\vec{R}; \vec{R}') &= \Psi^*(\vec{R}')\Psi(\vec{R}) + \tilde{\rho}_1(\vec{R}; \vec{R}') , \end{aligned} \quad (2.4)$$

where $\rho_1(\vec{R}; \vec{R}')$, which is only a function of the center-of-mass coordinates of the excitons, will be called the first reduced exciton density matrix. $\Psi(\vec{R})$ is the exciton order parameter, which in a fully condensed and homogeneous system [Eq. (2.1)] is of the form $\Psi(\vec{R}) = n^{1/2}e^{i\vec{K}\cdot\vec{R}}$. The function $\tilde{\rho}_1(\vec{R}, \vec{R}')$ stems from the noncondensate and decays as $\vec{R} - \vec{R}'$ gets large. The product of the pair wave functions $\varphi^*(r')\varphi(r)$ appears as a factor in front of both the condensate and the noncondensate of the excitons, reflecting the fact that both fractions consist of bound electron-hole pairs. This has to be contrasted with the case of a superconductor, in which the second reduced particle density matrix ρ_2 has ODLRO in the form

$$\begin{aligned} \rho_2(\vec{x}\uparrow, \vec{y}\downarrow; \vec{y}'\downarrow, \vec{x}'\uparrow) = {} & \langle \psi^\dagger(\vec{x}'\uparrow)\psi^\dagger(\vec{y}'\downarrow)\rangle \\ & \times \langle \psi(\vec{y}\downarrow)\psi(\vec{x}\uparrow)\rangle + \tilde{\rho}_2 . \end{aligned} \quad (2.5)$$

Here the Cooper pair functions $\langle\psi(\vec{y}\downarrow)\psi(\vec{x}\uparrow)\rangle$ appear only in the condensate part, because these bound pairs are formed not until the process of condensation.

The first reduced exciton density matrix can be obtained from Eq. (2.4) as

$$\rho_1(\vec{R}; \vec{R}') = \int d^3r \int d^3r' \, \varphi^*(r)\varphi(r')\rho_2^{e-h}(\vec{x}_h, \vec{x}_e; \vec{x}_h', \vec{x}_e') \quad (2.6)$$

and the exciton order parameter as

$$\Psi(\vec{R}) = \int d^3r \, \varphi^*(r)\langle\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle . \quad (2.7)$$

Equations (2.6) and (2.7) can be considered to be slightly more general definitions of ρ_1 and Ψ than the ones given in Eq. (2.4). The definitions [(2.6) and (2.7)] are motivated by the fact that we are not interested in the internal motion of excitons, but in the long-range correlation of the center-of-mass motion of excitons. Therefore, we introduce the projection of the electron-hole density matrix and of the electron-hole order parameter $\langle\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle$ into the exciton space. This is done by multiplying these functions with the wave functions of the internal motion of the excitons and by integrating with respect to the internal coordinates.

For the fourth reduced electron-hole density matrix we generalize Eq. (2.4) to

$$\rho_4^{e\text{-}h}(\vec{x}_h''',\vec{x}_e''',\ \vec{x}_h,\ \vec{x}_e;\ \vec{x}_h',\ \vec{x}_e',\ \vec{x}_h'',\ \vec{x}_e'') = \varphi^*(r'')\varphi^*(r')\varphi(r)\varphi(r''')\rho_2(\vec{R}''',\ \vec{R};\ \vec{R}',\ \vec{R}'')$$
$$-\varphi^*(\vec{x}_e' - \vec{x}_h'')\varphi^*(\vec{x}_e'' - \vec{x}_h')\varphi(r)\varphi(r''')\rho_2(\vec{R}''',\ \vec{R};\ \vec{R}_{x_e'',x_h'},\ \vec{R}_{x_e',x_h''})\,. \quad (2.8)$$

The second term is the exchange term, which is obtained by forming the pairs $(\vec{x}_e', \vec{x}_h'')$ and $(\vec{x}_e'', \vec{x}_h')$. $\vec{R}_{x_e',x_h''}$ and $\vec{R}_{x_e'',x_h'}$ are the center-of-mass coordinates of these pairs. The pairs $(\vec{x}_h''', \vec{x}_e''')$ and $(\vec{x}_h, \vec{x}_e)$ are kept fixed as a reference system. The exchange contribution will be shown to be essential in producing the repulsive exciton-exciton interaction. All the formulas for the exciton density matrices given in this chapter should only be applied in the low-density limit $na_0^{*3} \ll 1$.

III. EQUATIONS OF MOTION FOR THE REDUCED EXCITON DENSITY MATRICES

In this section we will study the time development of the exciton order parameter $\Psi(\vec{R})$ and of the reduced exciton density matrices $\rho_1(\vec{R}, \vec{R}')$ and $\rho_2(\vec{R}, \vec{R}'; \vec{R}, \vec{R}')$. The equations of motion for these three quantities are needed for the derivation of the four basic conservation laws of a superfluid.[12] The kinetics of the reduced exciton density matrices can be obtained from the equations of motion for the reduced electron-hole density matrices by applying the projection techniques described in Sec. II.

We will limit ourselves to the investigation of a spinless electron-hole system. In a system of particles with spin, the interaction between the excitons has been shown to be attractive, which leads to the formation of exciton molecules,[13] which themselves can undergo a Bose condensation.[14] Furthermore, we will not study the subtle problem of how the exciton interaction is modified due to their interaction with phonons. The assumption is made that the thermal relaxation time of the exciton gas is small as compared to the exciton lifetime. Under these conditions one can take into account the action of an external pump field (e.g., laser excitation) by means of chemical potentials for the electrons and holes. Otherwise, we do not include any explicit interaction of the system with photons. Especially, we do not study the complications for the condensation which are brought about by the polariton effect. Both assumptions, that of a long lifetime and that of the absence of the polariton effect are fulfilled in indirect band-gap materials. In the following sections we will treat the slightly simpler case of direct semiconductors. In Sec. VI B the modifications are given, which are necessary for excitons in indirect semiconductors. If the coupling to the phonons is too weak to guarantee a thermal quasiequilibrium distribution, one could try to establish the condensation by a selective accumulation of excitons in one k state by means of a specific optical excitation.[15]

The total Hamiltonian of the electron-hole system in the effective mass approximation is

$$H = \int d^3x_e\, \psi^\dagger(\vec{x}_e)[(-\hbar^2/2m_e)\nabla_{x_e}^2 - \mu_e + E_g]\psi(\vec{x}_e) + \int d^3x_h\, \phi^\dagger(\vec{x}_h)[(-\hbar^2/2m_h)\nabla_{x_h}^2 - \mu_h]\phi(\vec{x}_h)$$
$$+\frac{1}{2}\int d^3x_e \int d^3x_e'\, V(|\vec{x}_e - \vec{x}_e'|)\psi^\dagger(\vec{x}_e)\psi^\dagger(\vec{x}_e')\psi(\vec{x}_e')\psi(\vec{x}_e) + \frac{1}{2}\int d^3x_h \int d^3x_h'\, V(|\vec{x}_h - \vec{x}_h'|)\phi^\dagger(\vec{x}_h)\phi^\dagger(\vec{x}_h')\phi(\vec{x}_h')\phi(\vec{x}_h)$$
$$-\int d^3x_e \int d^3x_h\, V(|\vec{x}_e - \vec{x}_h|)\psi^\dagger(\vec{x}_e)\phi^\dagger(\vec{x}_h)\phi(\vec{x}_h)\psi(\vec{x}_e)\,, \quad (3.1)$$

where μ_e and μ_h are the chemical potentials of the electrons in the conduction band and of the holes in the valence band, respectively. E_g is the gap energy, and $V(r)$ is the Coulomb potential $e^2/\epsilon_0 r$, where ϵ_0 is the static dielectric constant.

The time derivative of the electron-hole order parameter can be obtained from the Heisenberg equations for the field operators $\psi(\vec{x}_e)$ and $\phi(\vec{x}_h)$, which have the following form

$$i\hbar\dot{\psi}(\vec{x}_e) = [\psi(\vec{x}_e),\ H] = [-(\hbar^2/2m_e)\nabla_{x_e}^2 + E_g - \mu_e]\psi(\vec{x}_e) + \int d^3x_e''\, V(|\vec{x}_e'' - \vec{x}_e|)\psi^\dagger(\vec{x}_e'')\psi(\vec{x}_e'')\psi(\vec{x}_e)$$
$$-\int d^3x_h''\, V(|\vec{x}_h'' - \vec{x}_e|)\phi^\dagger(\vec{x}_h'')\phi(\vec{x}_h'')\psi(\vec{x}_e) \quad (3.2)$$

and

$$i\hbar\dot{\phi}(\vec{x}_h) = [-(\hbar^2/2m_h)\nabla_{x_h}^2 - \mu_h]\phi(\vec{x}_h) + \int d^3x_h''\, V(|\vec{x}_h'' - \vec{x}_h|)\phi^\dagger(\vec{x}_h'')\phi(\vec{x}_h'')\phi(\vec{x}_h)$$

$$- \int d^3x_e'' \, V(|\vec{x}_e'' - \vec{x}_h|)\psi^\dagger(\vec{x}_e'')\psi(\vec{x}_e'')\phi(\vec{x}_h) \,. \tag{3.3}$$

From these equations we obtain

$$\begin{aligned} i\hbar \frac{\partial}{\partial t}\langle \phi(\vec{x}_h)\psi(\vec{x}_e)\rangle = {} & [-(\hbar^2/2m_e)\nabla^2_{x_e} - (\hbar^2/2m_h)\nabla^2_{x_h} - V(|\vec{x}_e - \vec{x}_h|) - (\mu_e + \mu_h) + E_g]\langle \phi(\vec{x}_h)\psi(\vec{x}_e)\rangle \\ & + \int d^3x_e' [V(|\vec{x}_e - \vec{x}_e'|) - V(|\vec{x}_h - \vec{x}_e'|)]\langle \psi^\dagger(\vec{x}_e')\psi(\vec{x}_e')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle \\ & + \int d^3x_h' [V(|\vec{x}_h - \vec{x}_h'|) - V(|\vec{x}_e - \vec{x}_h'|)]\langle \phi^\dagger(\vec{x}_h')\phi(\vec{x}_h')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle \,. \end{aligned} \tag{3.4}$$

Multiplying this equation with $\varphi^*(r)$ and integrating over the relative coordinate $\vec{r}$, we find [according to Eq. (2.7)]

$$\begin{aligned} i\hbar\dot{\Psi}(\vec{R}) = {} & [-(\hbar^2/2M)\nabla^2_R + E_g - E^b_{\text{ex}} - (\mu_e - \mu_h)]\Psi(\vec{R}) + \int d^3r \int d^3x_e' \int d^3x_e'' \, \delta^3(\vec{x}_e' - \vec{x}_e'')[V(|\vec{x}_e - \vec{x}_e'|) - V(|\vec{x}_h - \vec{x}_e'|)] \\ & \times \langle \psi^\dagger(\vec{x}_e')\psi(\vec{x}_e'')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle \varphi^*(r) + \int d^3r \int d^3x_h' \int d^3x_h'' \, \delta^3(\vec{x}_h' - \vec{x}_h'')[V(|\vec{x}_h - \vec{x}_h'|) - V(|\vec{x}_e - \vec{x}_h'|)] \\ & \times \langle \phi^\dagger(\vec{x}_h')\phi(\vec{x}_h'')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle \varphi^*(r) \,, \end{aligned} \tag{3.5}$$

where the exciton binding energy $E^b_{\text{ex}} = e^2/2\epsilon_0 a_0^*$ is the lowest eigenvalue of the Wannier equation

$$[-(\hbar^2/2m)\nabla^2_r - V(r)]\varphi(r) = -E^b_{\text{ex}}\varphi(r) \,. \tag{3.6}$$

The reduced mass m is given by $m_e m_h/M$. The exciton Bohr radius a_0^* is equal to $\epsilon_0\hbar^2/me^2$. In the potential terms of Eq. (3.5) δ functions have been inserted to obtain formally the two unpaired field operators at different locations. As a next step, we transform these density matrices into complete electron-hole-pair density matrices by inserting the missing electron and hole operators and by averaging over the relative motions of these pairs, because we are only interested in situations in which nearly all electrons are bound into the lowest exciton states. The integrations over $\vec{x}_e'$ and $\vec{x}_e''$ (or $\vec{x}_h'$ and $\vec{x}_h''$) in the first (second) potential term are now changed into integrations over the center-of-mass coordinates $\vec{R}'$ and $\vec{R}''$. In the low-density limit, where the excitons are well separated and the overlap is very small, we can approximate the δ functions by

$$\delta^3(\vec{x}_e' - \vec{x}_e'') \approx \delta^3(\vec{x}_h' - \vec{x}_h'') \approx \delta^3(\vec{R}' - \vec{R}'') \,.$$

This approximation holds as long as $na_0^{*3} \ll 1$. Equation (3.5) then takes the form

$$\begin{aligned} i\hbar\dot{\Psi}(\vec{R}) = {} & [-(\hbar^2/2M)\nabla^2_R + E_g - E^b_{\text{ex}} - (\mu_e + \mu_h)]\Psi(\vec{R}) \\ & + \int d^3r \int d^3r' \int d^3r'' \int d^3R' \int d^3R'' \, \delta^3(\vec{R}' - \vec{R}'')[V(|\vec{R} - \vec{R}' + \alpha(\vec{r} - \vec{r}\,')|) \\ & - V(|\vec{R} - \vec{R}' - \beta\vec{r} - \alpha\vec{r}\,'|) + V(|\vec{R} - \vec{R}' - \beta(\vec{r} - \vec{r}\,')|) - V(|\vec{R} - \vec{R}' + \alpha\vec{r} - \beta\vec{r}\,'|)] \\ & \times \langle \psi^\dagger(\vec{x}_e'')\phi^\dagger(\vec{x}_h'')\phi(\vec{x}_h')\psi(\vec{x}_e')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle \varphi^*(r)\varphi^*(r')\varphi(r'') . \end{aligned} \tag{3.7}$$

It is shown in the Appendix that the procedure of supplementing each unpaired field operator by its corresponding partner in the bound state and of subsequent averaging over the internal exciton motion is correct in the low-density limit $na_0^{*3} \ll 1$, where the contribution of the higher-excited exciton bound state and of the ionization continuum states to the reduced density matrices is still negligibly small and where the excitons are spatially well separated.

The electron-hole density matrix is again decomposed into the product of the wave functions of the internal motion of the three pairs and into an exciton three-leg function $\rho_{3/2}$ which describes the center-of-mass motion

$$\begin{aligned} \langle \psi^\dagger(\vec{x}_e'')\phi^\dagger(\vec{x}_h'')\phi(\vec{x}_h')\psi(\vec{x}_e')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle = {} & \varphi^*(r'')\varphi(r')\varphi(r)\rho_{3/2}(\vec{R}, \vec{R}'; \vec{R}'') \\ & - \varphi^*(r'')\varphi(r_{x_e, x_h'})\varphi(r_{x_e', x_h})\rho_{3/2}(\vec{R}_{x_e', x_h}, \vec{R}_{x_e, x_h'}; \vec{R}'') \,, \end{aligned} \tag{3.8}$$

where as in Eq. (2.8) the exchange terms have been taken into account. The exchange contribution simplifies considerably if the masses of electron and hole differ strongly. For a heavy hole mass, e.g., one has $\alpha \approx 1$, $\beta \approx 0$. In this case Eq. (3.7) can be written as

$$i\hbar\dot{\Psi}(\vec{\mathrm{R}})=[-(\hbar^2/2M)\nabla_R^2+E_g-E_{\mathrm{ex}}^b-(\mu_e+\mu_h)]\Psi(\vec{\mathrm{R}})+\int d^3R'\,W(|\vec{\mathrm{R}}-\vec{\mathrm{R}}'|)\rho_{3/2}(\vec{\mathrm{R}},\vec{\mathrm{R}}';\vec{\mathrm{R}}'),\tag{3.9}$$

where the effective scattering potential between two excitons is given by

$$\begin{aligned}W(|\vec{\mathrm{R}}-\vec{\mathrm{R}}'|)=\int d^3r\int d^3r'\,[&V(|\vec{\mathrm{R}}-\vec{\mathrm{R}}'|)+V(|\vec{\mathrm{R}}-\vec{\mathrm{R}}'+\vec{\mathrm{r}}-\vec{\mathrm{r}}'|)\\&-V(|\vec{\mathrm{R}}-\vec{\mathrm{R}}'-\vec{\mathrm{r}}'|)-V(|\vec{\mathrm{R}}-\vec{\mathrm{R}}'+\vec{\mathrm{r}}|)][|\varphi(r)|^2|\varphi(r')|^2\\&-\varphi(|\vec{\mathrm{r}}+\vec{\mathrm{R}}-\vec{\mathrm{R}}'|)\varphi^*(r)\varphi(|\vec{\mathrm{r}}'+\vec{\mathrm{R}}'-\vec{\mathrm{R}}|)\varphi^*(r')].\end{aligned}\tag{3.10}$$

The same effective potential has been found in the framework of the boson treatment for excitons.[6] In Ref. 6 it has been shown that due to the exchange term the potential $W(|\mathrm{R}'-\mathrm{R}|)$ is repulsive. The Pauli principle, which governs the statistics of the constituting electrons and holes causes thus a repulsive interaction between two excitons. For particles with spin the Pauli principle is not so influential. The interaction between two excitons in which both electrons and holes have opposite spins is attractive and leads to the formation of exciton molecules. The interaction between these bi-excitons can in turn be shown to be repulsive[14] so that the condensation of bi-excitons can be treated in analogy to the exciton condensation.

Equation (3.10) has apart from the appearance of the excitation energy E_g-E_{ex} the same form as the order parameter equation of an interacting Bose system.[12] If we factorize the three-leg function

$$\begin{aligned}\rho_{3/2}(\vec{\mathrm{R}},\vec{\mathrm{R}}';\vec{\mathrm{R}}')&=|\Psi(\vec{\mathrm{R}}')|^2\Psi(\vec{\mathrm{R}})+\tilde{\rho}_1(\vec{\mathrm{R}};\vec{\mathrm{R}}')\Psi(\vec{\mathrm{R}}')+\tilde{\rho}_1(\vec{\mathrm{R}}';\vec{\mathrm{R}}')\Psi(\vec{\mathrm{R}})+\tilde{\rho}_a(\vec{\mathrm{R}},\vec{\mathrm{R}}')\Psi^*(\vec{\mathrm{R}}')\\&=\rho_1(\vec{\mathrm{R}}';\vec{\mathrm{R}}')\Psi(\vec{\mathrm{R}})+\tilde{\rho}_1(\vec{\mathrm{R}};\vec{\mathrm{R}}')\Psi(\vec{\mathrm{R}}')+\tilde{\rho}_a(\vec{\mathrm{R}},\vec{\mathrm{R}}')\Psi^*(\vec{\mathrm{R}}').\end{aligned}\tag{3.11}$$

Equation (3.10) takes the form which for He II has been derived by Fröhlich,[16] if one neglects the anomalous function $\tilde{\rho}_a(\vec{\mathrm{R}},\vec{\mathrm{R}}')$. In equilibrium, where the space and time derivatives are zero, we find for the chemical potential [neglecting the noncondensate part in Eq. (3.11)]

$$\mu=\mu_e+\mu_h\approx E_g-E_{\mathrm{ex}}^b+n\int d^3R'\,W(|\vec{\mathrm{R}}'-\vec{\mathrm{R}}|),\tag{3.12}$$

which has been shown in Ref. 6 to be equal to

$$\mu\approx E_g-E_{\mathrm{ex}}^b(1-\tfrac{26}{3}\pi n a_0^{*3}).\tag{3.13}$$

The same result has been derived by Keldysh and Koslov.[5]

Now we turn to the derivation of the equation of motion for the first reduced exciton density matrix. As above, we start with the equation of motion for the corresponding electron-hole density matrix which is obtained directly from Eqs. (3.2) and (3.3). One finds

$$\begin{aligned}i\hbar\frac{\partial}{\partial t}\langle\psi^\dagger(\vec{\mathrm{x}}_e')\phi^\dagger(\vec{\mathrm{x}}_h')\phi(\vec{\mathrm{x}}_h)\psi(\vec{\mathrm{x}}_e)\rangle=&\Big(-\frac{\hbar^2}{2m_e}\nabla_{x_e}^2-\frac{\hbar^2}{2m_h}\nabla_{x_h}^2-V(|\vec{\mathrm{x}}_e-\vec{\mathrm{x}}_h|)\\&+\frac{\hbar^2}{2m_e}\nabla_{x_e'}^2+\frac{\hbar^2}{2m_h}\nabla_{x_h'}^2+V(|\vec{\mathrm{x}}_e'-\vec{\mathrm{x}}_h'|)\Big)\langle\psi^\dagger(\vec{\mathrm{x}}_e')\phi^\dagger(\vec{\mathrm{x}}_h')\phi(\vec{\mathrm{x}}_h)\psi(\vec{\mathrm{x}}_e)\rangle\\&+\int d^3x_e''\,[V(|\vec{\mathrm{x}}_e-\vec{\mathrm{x}}_e''|)-V(|\vec{\mathrm{x}}_e'-\vec{\mathrm{x}}_e''|)-V(|\vec{\mathrm{x}}_h-\vec{\mathrm{x}}_e''|)+V(|\vec{\mathrm{x}}_h'-\vec{\mathrm{x}}_e''|)]\\&\times\langle\psi^\dagger(\vec{\mathrm{x}}_e')\phi^\dagger(\vec{\mathrm{x}}_h')\psi^\dagger(\vec{\mathrm{x}}_e'')\psi(\vec{\mathrm{x}}_e'')\phi(\vec{\mathrm{x}}_h)\psi(\vec{\mathrm{x}}_e)\rangle\\&+\int d^3x_h''\,[V(|\vec{\mathrm{x}}_h-\vec{\mathrm{x}}_h''|)-V(|\vec{\mathrm{x}}_h'-\vec{\mathrm{x}}_h''|)-V(|\vec{\mathrm{x}}_e-\vec{\mathrm{x}}_h''|)+V(|\vec{\mathrm{x}}_e'-\vec{\mathrm{x}}_h''|)]\\&\times\langle\psi^\dagger(\vec{\mathrm{x}}_e')\phi^\dagger(\vec{\mathrm{x}}_h')\phi^\dagger(\vec{\mathrm{x}}_h'')\phi(\vec{\mathrm{x}}_h'')\phi(\vec{\mathrm{x}}_h)\psi(\vec{\mathrm{x}}_e)\rangle.\end{aligned}\tag{3.14}$$

We proceed precisely as above. First, we project Eq. (3.14) into the exciton space by multiplying with $\varphi^*(r)\varphi(r')$ and by integrating over r and r'. In the potential terms we insert the corresponding partners to the unpaired operators and average over the internal motion of these newly generated pairs. Finally, we decompose the fourth-order electron-hole reduced density matrix according to Eq. (2.8) and arrive in the case $m_h\gg m_e$ at

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

$$\times\rho_2(\vec{R}, \vec{R}''; \vec{R}'', \vec{R}'), \tag{3.15}$$

where again the interaction potential W appears, which has been defined in Eq. (3.10). Equation (3.15) has again the same form as the equation of motion for the first reduced density matrix of a system of Bose particles, which interact through the two-particle potential $W(|\vec{R}' - \vec{R}|)$.[12]

In Sec. IV we will also need the time derivative of the diagonal elements of the second reduced exciton density matrix. From the equation of motion for the fourth reduced electron-hole density matrix we deduce

$$i\hbar\frac{\partial}{\partial t}\rho_2(\vec{R}, \vec{R}'; \vec{R}, \vec{R}') = -[\nabla_R\vec{\nu}(\vec{R}, \vec{R}') - \nabla_{R'}\vec{\nu}(\vec{R}', \vec{R})], \tag{3.16}$$

where

$$\vec{\nu}(\vec{R}, \vec{R}') = \frac{\hbar}{2iM}\lim_{\vec{R}''\to\vec{R}}(\nabla_R - \nabla_{R'})\rho_2(\vec{R}, \vec{R}''; \vec{R}', \vec{R}'').$$

This result too is of the same form as that for an interacting Bose system.[12] The obtained equations of motion for the exciton order parameter Ψ and the reduced exciton density matrices ρ_1 and ρ_2 show that the kinetics of a condensed exciton gas is indeed that of a Bose gas with an effective interaction potential W and lent thus further support to the boson method of Ref. 6.

IV. CONSERVATION LAWS

The equations of motion for Ψ, ρ_1, and ρ_2 allow one to derive the conservation laws for the particle density n, the particle current density $\vec{j}$, the energy density E, and for the extra hydrodynamical variable which is characteristic for a condensed system, namely, the superfluid velocity $\vec{v}_s$:

$$\begin{aligned}&\frac{\partial}{\partial t}n + \nabla\vec{j} = 0,\\ &\frac{\partial}{\partial t}\vec{j} + \nabla\overleftrightarrow{\Pi} = 0,\\ &\frac{\partial}{\partial t}E + \nabla\vec{Q} = 0,\\ &\frac{\partial}{\partial t}\vec{v}_s + \nabla(\tfrac{1}{2}\vec{v}_s^{\,2} + \mu/M) = 0.\end{aligned} \tag{4.1}$$

Here, $\overleftrightarrow{\Pi}$ is the stress tensor and $\vec{Q}$ the energy current density. The exciton density is given by the diagonal element of ρ_1:

$$n(\vec{R}) = \rho_1(\vec{R}; \vec{R}). \tag{4.2}$$

From Eq. (3.15) we find at once the first of the conservation laws Eq. (4.1) with an exciton particle current density

$$\vec{j}(\vec{R}) = \frac{\hbar}{2Mi}\lim_{\vec{R}'\to\vec{R}}(\nabla_R - \nabla_{R'})\rho_1(\vec{R}; \vec{R}'). \tag{4.3}$$

Applying the off-diagonal derivative to Eq. (3.15), one finds in the limit $\vec{R}'\to\vec{R}$ the conservation law for the exciton current density. After some calculations (for details of the derivation see Ref. 12) the stress tensor turns out to be

$$\overleftrightarrow{\Pi}(\vec{R}) = -\frac{\hbar^2}{4M}\lim_{\vec{R}'\to\vec{R}}(\nabla\otimes\nabla)\rho_1(\vec{R}; \vec{R}') - \frac{1}{4}\int d^3r\,\frac{\partial W(r)}{\partial r}\,\frac{\vec{r}\otimes\vec{r}}{r}$$
$$\times\int_{-1}^{1}dt\,\rho_2\left(\vec{R} + (t-1)\frac{\vec{r}}{2},\ \vec{R} + (t+1)\frac{\vec{r}}{2};\ \vec{R} + (t-1)\frac{\vec{r}}{2},\ \vec{R} + (t+1)\frac{\vec{r}}{2}\right). \tag{4.4}$$

The symbol $\otimes$ is the sign for a tensor product. The last term simplifies in the hydrodynamic limit [which means the case of negligible spatial variations on a scale given by the range of the effective potential $W(r)$]

$$-\frac{1}{2}\int d^3r\,\frac{\partial W(r)}{\partial r}\,\frac{\vec{r}\otimes\vec{r}}{r}\,\rho_2(\vec{R}, \vec{R}+\vec{r}; \vec{R}, \vec{R}+\vec{r}). \tag{4.5}$$

This form was first derived by Martin and Schwinger.[17] In order to obtain the third conservation law, we have to calculate first the exciton energy density from the electron-hole Hamiltonian [Eq. (3.1)]. Taking the expectation value of Eq. (3.1) and applying the techniques which have been developed in Sec. III, we find

$$\begin{aligned}\langle H\rangle = &\int d^3R\int d^3R'\int d^3r\int d^3r'\,\varphi^*(r)\varphi(r')\,\delta^{(3)}(\vec{R} - \vec{R}')\\ &\times[-(\hbar^2/2M)\nabla_R^2 - (\hbar^2/2m)\nabla_r^2 - V(r) - (\mu_e + \mu_h) + E_g]\langle\psi^\dagger(\vec{x}_e')\phi^\dagger(\vec{x}_h')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle\\ &+\frac{1}{2}\int d^3R\int d^3R'\int d^3R''\int d^3R'''\int d^3r\int d^3r'\int d^3r''\int d^3r'''\,\delta^{(3)}(\vec{R} - \vec{R}'')\delta^{(3)}(\vec{R}' - \vec{R}''')\\ &\times\varphi(r)\varphi(r')\varphi^*(r'')\varphi^*(r''')[V(\vec{R} - \vec{R}' + \alpha(\vec{r} - \vec{r}')) + V(\vec{R} - \vec{R}' - \beta(\vec{r} - \vec{r}')) - V(\vec{R} - \vec{R}' + \alpha\vec{r} - \beta\vec{r}')\\ &- V(\vec{R} - \vec{R}' + \alpha\vec{r}' - \beta\vec{r})]\langle\psi^\dagger(\vec{x}_e)\phi^\dagger(\vec{x}_h)\psi^\dagger(\vec{x}_e')\phi^\dagger(\vec{x}_h')\phi(\vec{x}_h'')\psi(\vec{x}_e'')\phi(\vec{x}_h''')\psi(\vec{x}_e''')\rangle.\end{aligned} \tag{4.6}$$

To obtain this result, one has to give special care to the treatment of the electron-hole interaction term of the Hamiltonian [Eq. (3.1) (last row)]. Firstly, we consider the operators $\psi^\dagger(\vec{x}_e')$ and $\phi^\dagger(\vec{x}_h')$ [and similarly $\phi(\vec{x}_h)$, $\psi(\vec{x}_e)$] as paired and obtain thus the electron-hole interaction $V(r)$ in the Wannier-Hamiltonian of the exciton. Then, we get the next-higher-order contribution of this term by considering the operators as unpaired and by supplementing each of the four operators with the corresponding partner. In this manner we obtain a contribution of the effective interaction between two excitons [the two negative contributions in the last term of Eq. (4.6)].

Introducing ρ_1 and ρ_2 by Eqs. (2.4) and (2.8), we obtain from $\langle H\rangle + \mu N = \int E(\vec{R})d^3R$ the energy density

$$E(\vec{R}) = \lim_{\vec{R}'\to\vec{R}} \left[+(\hbar^2/2M)\nabla_R \nabla_R' + E_g - E_{\text{ex}}^b \right]\rho_1(\vec{R};\vec{R}') + \frac{1}{2}\int d^3R'\, W(|\vec{R}-\vec{R}'|)\rho_2(\vec{R},\vec{R}';\vec{R},\vec{R}'), \tag{4.7}$$

which is apart from the appearance of the excitation energy of the same form as the energy density of an interacting Bose system (see Ref. 12). The time derivative of $E(\vec{R})$ together with the equations of motion for ρ_1 and ρ_2 [Eqs. (3.15) and (3.16)] give the third conservation law with an energy current density

$$\begin{aligned}\vec{Q}(\vec{R}) = {} & \vec{j}(\vec{R})(E_g - E_{\text{ex}}^b) + \frac{i\hbar^3}{16M^2}\lim_{\vec{R}'\to\vec{R}} (\nabla_R - \nabla_{R'})(\nabla_R - \nabla_{R'})^2\rho_1(\vec{R};\vec{R}') \\ & + \frac{1}{2}\int d^3R\, W(\vec{R}-\vec{R}')\vec{\nu}(\vec{R},\vec{R}') - \frac{1}{8}\int d^3r \left(\frac{\partial W(r)}{\partial r}\right)\frac{\vec{r}}{r} \\ & \times\left\{\vec{r}\int_{-1}^{1} dt\left[\vec{\nu}\left(\vec{R}+(t-1)\frac{\vec{r}}{2},\ \vec{R}+(t+1)\frac{\vec{r}}{2}\right)+\vec{\nu}\left(\vec{R}+(t+1)\frac{\vec{r}}{2},\ \vec{R}+(t-1)\frac{\vec{r}}{2}\right)\right]\right\}.\end{aligned} \tag{4.8}$$

$\vec{\nu}$ has been defined in Eq. (3.16). The first term in Eq. (4.8) is specific to the exciton problem. It shows that the exciton energy current density is primarily given by the flow of excitation energy. The remaining terms in Eq. (4.8) are the same as those appearing in $\vec{Q}$ for liquid helium.[12] In the hydrodynamic limit the last expression in Eq. (4.8) reduces to

$$-\frac{1}{4}\int d^3r\,\vec{r}\{[\vec{\nu}(\vec{R},\vec{R}+\vec{r})+\vec{\nu}(\vec{R}+\vec{r},\vec{R})]\cdot\nabla_r W(r)\}. \tag{4.9}$$

Finally, we will derive the last of the hydrodynamic equations given in Eq. (4.1), namely, the equation of motion for the superfluid velocity $\vec{v}_s$. Writing the order parameter in the form

$$\Psi(\vec{R}) = [n_0(\vec{R})]^{1/2} e^{i\theta(\vec{R})},$$

one obtains the contribution of the condensate to the current density from Eq. (4.3) in the form

$$n_0(\hbar/M)\nabla_R\theta(\vec{R}) = n_0\vec{v}_s(\vec{R}). \tag{4.10}$$

Equation (4.10) defines $\vec{v}_s$ as the gradient of the phase of the order parameter. Equations of motion for both n_0 and $\vec{v}_s$ can be obtained from Eq. (3.9) for $\Psi(\vec{R})$. However, the equation for n_0 is no extra hydrodynamic equation, because n_0 approaches always its equilibrium value very fast, i.e., in a nonhydrodynamical way. For the comparison with the theory for superfluid helium in Ref. 12, one has to take into account the explicit appearance of the chemical potentials in the Hamiltonian [Eq. (3.1)]. This gives rise to a phase factor in the order parameter which is equal to $e^{i\mu t/\hbar}$. Using the identity $\theta = (\dot{\Psi}/\Psi - \dot{\Psi}^*/\Psi^*)/2i$ and Eq. (4.10), we obtain the equation of motion for $\vec{v}_s$ from the order parameter equation (3.9) with a chemical potential

$$\mu = E_g - E_{\text{ex}}^b + \text{Re}\left(\int d^3R'\, W(|\vec{R}-\vec{R}'|)\rho_{3/2}(\vec{R},\vec{R}';\vec{R}')/\Psi(\vec{R})\right). \tag{4.11}$$

In equilibrium and under the neglection of the noncondensate, Eq. (4.11) reduces to the result, Eq. (3.12).

We have now completed the derivation of the four basic conservation laws for a superfluid. In the following chapter, we will use explicit approximations for the reduced density matrices in order to get the full hydrodynamic and thermodynamic two-fluid model for an exciton system.

V. TWO-FLUID MODEL

The four conservation laws [Eq. (3.1)], together with the hydrodynamic relations between the various quantities which appear in these equations, describe the hydrodynamics of a superfluid completely.[12] The thermodynamic relations are especially simple in a coordinate system K^0 in which the condensate and thus also the superfluid component is at rest, i.e., $\vec{v}_s^0 = 0$ and $\vec{v}_n^0 = \vec{v}_n - \vec{v}_s$, where $\vec{v}_n$ is the velocity of the normal component. Here one has

$$n = n_s + n_n, \tag{5.1}$$

$$\vec{j}^{\,0} = n_n\vec{v}_n^0, \tag{5.2}$$

$$\Pi^0_{ij} = p\delta_{ij} + Mnv^0_{ni}v^0_{nj}\,, \tag{5.3}$$

$$\vec{Q}^0 = \vec{v}^0_n(E^0 + p - n_s\mu)\,, \tag{5.4}$$

where p is the pressure. The entropy density S is given by

$$nST = E^0 + p - n\mu - Mn_n(v^0_n)^2\,. \tag{5.5}$$

The Galilei transformations for the quantities in (5.1)–(5.4) are given in Ref. 18. Though the phenomenological two-fluid model for superfluid helium and for condensed excitons is the same, one has to keep the physical differences between the two systems in mind. While in the case of helium M is the real mass of a helium atom, it is for excitons just the effective total mass which describes the dynamical behavior of excitons in the crystal. Thus, for excitons the two-fluid model does not describe the superfluid flow of real mass but that of excitation energy.

In the following we will calculate the relevant hydrodynamical quantities of the superfluid exciton system. The derivation of the conservation laws expressed the quantities in terms of the reduced exciton density matrices. Now, we will evaluate these density matrices by assuming local thermal equilibrium. The evaluation is simplified by the fact that the exciton gas is equivalent to a system of boson particles, which interact through the effective potential $W(|R - R'|)$, as has already been shown in Ref. 6 and has been demonstrated again from a different point of view in the preceeding sections. The range of the validity of the given derivation has been shown to be $na_0^{*3} \ll 1$. On the other hand, the Bogolubov approximation for a condensed Bose gas is valid as long as the depletion of the condensate is small, i.e., $(n - n_0)/n \ll 1$. We can approximately replace the effective potential $W(\vec{R} - \vec{R}')$ by $W_0\delta^3(\vec{R} - \vec{R}')$, where W_0 is given by

$$W_0 = \tfrac{26}{3}\pi E^b_{\mathrm{ex}} a_0^{*3} \tag{5.6}$$

[see Eqs. (3.12) and (3.13)]. This procedure is equivalent to expressing the action of the potential in terms of a scattering amplitude.[19] Then, one can show that at $T = 0$ (see, e.g., Ref. 12)

$$\frac{n - n_0}{n} = \frac{n^{1/2}}{3\pi^2}\left(\frac{MW_0}{\hbar^2}\right)^{3/2}\,. \tag{5.7}$$

Inserting W_0, E^b_{ex} and using the definition of the exciton Bohr radius, one finds the following condition for the validity of the Bogolubov approximation

$$(M/m)^3 na_0^{*3} \ll 1\,. \tag{5.8}$$

This condition is more restrictive than $na_0^{*3} \ll 1$, especially in the case of strongly different electron and hole masses. For the following, we will assume that this more restrictive low-density criterion is fulfilled.

It is now straightforward to evaluate the exciton reduced density matrices, which are given by

$$\begin{aligned}
&\Psi(\vec{R}) = \langle\hat{\psi}(\vec{R})\rangle\,,\\
&\rho_1(\vec{R};\,\vec{R}') = \langle\hat{\psi}^\dagger(\vec{R}')\hat{\psi}(\vec{R})\rangle\,,\\
&\rho_{3/2}(\vec{R},\,\vec{R}';\vec{R}'') = \langle\hat{\psi}^\dagger(\vec{R}'')\hat{\psi}(\vec{R}')\hat{\psi}(\vec{R})\rangle\,,\\
&\rho_2(\vec{R},\,\vec{R}';\,\vec{R}'',\,\vec{R}''') = \langle\hat{\psi}^\dagger(\vec{R}''')\hat{\psi}^\dagger(\vec{R}'')\hat{\psi}(\vec{R}')\hat{\psi}(\vec{R})\rangle\,,
\end{aligned} \tag{5.9}$$

where $\hat{\psi}$, $\hat{\psi}^\dagger$ are Bose field operators. In a homogeneous situation the field operator is according to Bogolubov[20] given by

$$\begin{aligned}
\hat{\Psi}(\vec{R}) &= \Psi(\vec{R}) + \hat{\tilde{\psi}}(\vec{R})\\
&= e^{i\theta(\vec{R})}\left(n_0^{1/2} + V^{-1/2}{\sum}' e^{i\vec{K}\cdot\vec{R}}\frac{\hat{\xi}_K + L_K\hat{\xi}^\dagger_K}{(1 - L_K^2)^{1/2}}\right),
\end{aligned} \tag{5.10}$$

where $\hat{\xi}_K$, $\hat{\xi}^\dagger_K$ are the operators in which the Hamiltonian is diagonal

$$H = H_0 + {\sum}' E_K\hat{\xi}^\dagger_K\hat{\xi}_K\,, \tag{5.11}$$

where

$$H_0 = n(E_g - E^b_{\mathrm{ex}}) + nW_0\left(n + {\sum}' L_K\right).$$

The excitation spectrum E_K and the transformation function L_K are given by

$$\begin{aligned}
&E_K^2 = (T_K^2 + 2T_K\Delta)\,, \quad L_K = (E_K - T_K - \Delta)/\Delta\,,\\
&T_K = \hbar^2K^2/2M\,, \qquad \Delta = nW_0\,.
\end{aligned} \tag{5.12}$$

In thermal equilibrium the number of elementary excitations is given by

$$\langle\hat{\xi}^\dagger_K\hat{\xi}_K\rangle = n_K(v^0_n,\,T) = \{\exp[(E_K - \vec{v}^0_n\cdot\hbar\mathrm{K})/k_BT] - 1\}^{-1}\,. \tag{5.13}$$

In the coordinate system K^0 the reduced exciton density matrices are within the Bogolubov approximation given by

$$\begin{aligned}
&\Psi^0(\vec{R}) = \sqrt{n_0}\,,\\
&\rho^0_1(\vec{R};\,\vec{R}') = n_0 + \tilde{\rho}_1(\vec{R};\,\vec{R}')\,,\\
&\tilde{\rho}^0_1(\vec{R};\,\vec{R}') = \langle\hat{\tilde{\psi}}^\dagger(\vec{R}')\hat{\tilde{\psi}}(\vec{R})\rangle e^{i[\theta(\vec{R}') - \theta(\vec{R})]}\,,\\
&\rho^0_{3/2}(\vec{R},\,\vec{R}';\,\vec{R}') = [\,\rho^0_a(\vec{R},\,\vec{R}') + \rho^0_1(\vec{R}',\,\vec{R}') + \tilde{\rho}^0_1(\vec{R};\vec{R}')]\,n_0^{1/2}\,,\\
&\rho^0_a(\vec{R},\,\vec{R}') = \langle\hat{\tilde{\psi}}(\vec{R})\hat{\tilde{\psi}}(\vec{R}')\rangle\, e^{-i[\theta(\vec{R}) + \theta(\vec{R}')]}\,,
\end{aligned} \tag{5.14}$$

$$\rho_2^0(\vec{R}, \vec{R}'; \vec{R}, \vec{R}') = n_0^2 + n_0[\tilde{\rho}_1^0(\vec{R}, \vec{R}') + \tilde{\rho}_1^0(\vec{R}'; \vec{R}) + \tilde{\rho}_1^0(\vec{R}; \vec{R}) + \tilde{\rho}_1^0(\vec{R}'; \vec{R}') + \rho_a^0(\vec{R}, \vec{R}') + \rho_a^0(\vec{R}, \vec{R}')] .$$

The equations (5.10)–(5.14) allow us to calculate n, $\vec{j}^{\,0}$, $\overleftrightarrow{\Pi}^0$, E^0, $\vec{Q}^0$, and μ according to the formulas derived in Sec. IV. Because the calculations of these quantities are, with minor differences, the same as those given in Ref. 12, we will just list the results up to second order in the relative velocity $\vec{v}_n^0$. For the total density [Eq. (4.2)] one finds

$$n = n_0 + \frac{1}{3\pi^2}\left(\frac{M\Delta}{\hbar^2}\right)^{3/2} + \frac{1}{V}\sum{}' \frac{T_K+\Delta}{E_K}\left(n_K(0,\ T) + \frac{M(v_n^0)^2}{3} T_K \frac{\partial^2 n_K(0,\ T)}{\partial E_K^2}\right). \tag{5.15}$$

At $T=0$, the depletion is given by the second term on the right-hand side of Eq. (5.15). This result was used in Eq. (5.7). The exciton current density [Eq. (4.3)] is obtained as

$$\begin{aligned}\vec{j}^{\,0} &= \vec{v}_n^0\left[-\frac{2}{3V}\sum{}' T_K\left(\frac{\partial n_K(0,\ T)}{\partial E_K} + \frac{m(v_n^0)^2}{5} T_K \frac{\partial^3 n_K(0,\ T)}{\partial E_K^3}\right)\right] \\ &= \vec{v}_n^0 n_n ,\end{aligned} \tag{5.16}$$

which defines the normal fluid density n_n. Further, we find for the pressure [Eqs. (4.4) and (5.1)]

$$\begin{aligned}p &= E^0 - (E_g - E_{\text{ex}}^b) - \frac{1}{2V}\sum{}'\Big((E_K - T_K - \Delta)(E_K - T_K) + \tfrac{2}{3} T_K(T_K+\Delta) n_K(0,\ T) \\ &\quad - \frac{Mv_n^{0^2}}{5}\frac{\partial^2}{\partial E_K^2} n_K(0,\ T) T_K^2 (T_K+\Delta)\Big) E_K^{-1} ,\end{aligned} \tag{5.17}$$

for the chemical potential [Eq. (3.13)]

$$\mu = E_g - E_{\text{ex}}^b + W_0\left[n + \frac{1}{V}\sum{}'\left(\frac{1}{2}\frac{T_K - E_K}{E_K} + n_K(0,\ T)\frac{T_K}{E_K} + \frac{M(v_n^0)^2}{3}\frac{T_K}{E_K}\frac{\partial^2 n_K(0,\ T)}{\partial E_K^3}\right)\right], \tag{5.18}$$

for the energy density [Eq. (4.7)]

$$E^0 = n(E_g - E_{\text{ex}}^b) + \tfrac{1}{2} W_0 n^2 + \frac{1}{V}\sum{}'\left(\tfrac{1}{2}(E_K - T_K - \Delta) + n_K(0,\ T)E_K + \frac{M(v_n^0)^2}{3}\frac{\partial^2 n_K(0,\ T)}{\partial E_K^2} T_K E_K\right), \tag{5.19}$$

and for the energy current density [Eq. (4.13)]

$$\vec{Q}^0 = \vec{j}^{\,0}(E_g - E_{\text{ex}}^b) - \vec{v}_n^0\frac{2}{3V}\sum{}' E_K^2\left(\frac{\partial n_K(0,\ T)}{\partial E_K} + \frac{M(v_n^0)^2}{5}\frac{\partial^3 n_K(0,\ T)}{\partial E_K^3} T_K\right). \tag{5.20}$$

The listed results fulfill the two-fluid relation [Eq. (5.4)]. Finally, one gets for the entropy density [Eq. (5.5)]

$$nST = \frac{1}{V}\sum{}'\left[n_K(0,\ T)\left(E_K + \frac{2}{3}\frac{T_K(T_K+\Delta)}{E_K}\right) + \frac{M(v_n^0)^2}{3}\frac{\partial^2 n_K(0,\ T)}{\partial E_K^2}\left(E_K T_K - \tfrac{2}{5} T_K^2 \frac{T_K+\Delta}{E_K}\right)\right]. \tag{5.21}$$

The results are in such a form that it is natural to use n, T, and $\vec{v}_n$ as independent thermodynamic variables. For these variables the following thermodynamic identity holds

$$dF^0 = \mu dn - nSdT - M\vec{j}^{\,0}\cdot d\vec{v}_n^0 , \tag{5.22}$$

where F^0 is the free-energy density

$$F^0(n,\ T,\ \vec{v}_n^0) = E^0 - nTS - M\vec{v}_n^0\cdot\vec{j}^{\,0} = -p + n\mu . \tag{5.23}$$

Finally, we mention that in the low-temperature limit n_n, E^0, n, $\vec{Q}^0$, and nST all vary with the fourth power of the temperature.

VI. EXTENSIONS

A. Static homogeneous electromagnetic fields

An interesting question is, how one can establish a gradient in the chemical potential, which could cause an acceleration of the superfluid component. One obvious way to achieve this is to generate a gradient in the exciton concentration. One might further think to influence the chemical potential via electromagnetic fields. For the time being, we will limit ourselves to static and homogeneous fields $\vec{H}$ and $\vec{E}$ (for interactions of a condensed exciton system with a light field see Ref. 21). As usual, we choose the magnetic vector potential $\vec{A}(\vec{r})$ in the form

$$\vec{A}(\vec{r}) = \tfrac{1}{2}(\vec{H}\times\vec{r}) . \tag{6.1}$$

Then, the electron-hole Hamiltonian takes the form

$$H=\int d^3x_e\psi^\dagger(\vec{x}_e)\left[\left(\frac{\hbar}{i}\nabla_{x_e}-\frac{e}{c}\vec{A}(\vec{x}_e)\right)^2\frac{1}{2m_e}+e\vec{x}_e\cdot\vec{E}+E_g-\mu_e\right]\psi(\vec{x}_e)$$
$$+\int d^3x_h\phi^\dagger(\vec{x}_h)\left[\left(\frac{\hbar}{i}\nabla_{x_h}+\frac{e}{c}\vec{A}(\vec{x}_h)\right)^2\frac{1}{2m_h}-e\vec{x}_h\cdot\vec{E}-\mu_h\right]\phi(\vec{x}_h)+H_{\text{pot}}, \tag{6.2}$$

where H_{pot} is the same potential operator as in Eq. (3.1). From the work on single excitons in static electromagnetic fields, one knows (see, e.g., Ref. 22) that one can treat this problem by making the following *ansatz* for the exciton wave function

$$\psi_{\text{ex}}(\vec{r},\vec{R})=V^{-1/2}\exp\{i[\vec{K}-(e/c\hbar)\vec{A}(\vec{r})]\cdot\vec{R}\}\varphi(r), \tag{6.3}$$

where $\varphi(r)$ obeys the equation

$$-E^b_{\text{ex}}(\vec{H},\vec{E})\varphi(r)=\left[-\frac{\hbar^2\nabla^2}{2m}-\frac{e^2}{\epsilon_0 r}+\frac{ie\hbar}{c}\left(\frac{1}{m_e}-\frac{1}{m_h}\right)\vec{A}(\vec{r})\cdot\nabla\right.$$
$$\left.+\frac{e^2}{2mc^2}\vec{A}^2(\vec{r})-\frac{2e\hbar}{Mc}\vec{A}(\vec{r})\cdot\vec{K}+e\vec{E}\cdot\vec{r}\right]\varphi(r). \tag{6.4}$$

The field-dependent additional terms in the reduced Wannier equation are the Zeeman term, the diamagnetic term, the Lorentz force term, and the Stark term. The last two contributions can be put together to produce an effective electric field term $e\vec{r}\vec{E}_{\text{eff}}=e\vec{r}\cdot[\vec{E}+(\vec{v}\times\vec{H})e/c]$, where $\vec{v}=\hbar\vec{K}/M$. Making use of the ansatz (6.3) we can generalize the derivation of the equations of motion for the reduced exciton density matrices. The assumption of ODLRO now takes the form

$$\langle\psi^\dagger(\vec{x}_e')\phi^\dagger(\vec{x}_h')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle=\exp\left(\frac{i\hbar}{2ec}\left[(\vec{H}\times\vec{R})\vec{r}-(\vec{H}\times\vec{R}')\vec{r}'\right]\right)$$
$$\times\varphi^*(r')\varphi(r)\rho_1(\vec{R};\vec{R}'). \tag{6.5}$$

Similarly, the order parameter is given by

$$\Psi(\vec{R})=\int d^3r\langle\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle\exp\left(-\frac{i\hbar}{2ec}(\vec{H}\times\vec{R})\vec{r}\right)\varphi^*(r). \tag{6.6}$$

With Eqs. (6.5) and (6.6) one gets in the presence of fields formally the same results as in Sec. III, with the only difference that $\varphi(r)$ is now the lowest solution of Eq. (6.4) and that the exciton binding energy E^b_{ex} is replaced by the field-dependent eigenvalue of (6.4), i.e., by $E^b_{\text{ex}}(\vec{H},\vec{E})$. With the same modifications the conservation laws and all the two-fluid expressions remain valid. Specifically, the chemical potential is given by

$$\mu\approx E_g-E^b_{\text{ex}}(\vec{H},\vec{E})(1-\tfrac{26}{3}\pi n a_0^{*3}), \tag{6.7}$$

showing that one could indeed generate gradients in the chemical potential by spatially, slowly varying electromagnetic fields.

B. Indirect semiconductors

In an indirect-band gap material with isotropic effective masses near the extrema at $\vec{p}_C=\hbar\vec{k}_{e0}$ and $\vec{p}_V=-\hbar\vec{k}_{h0}$, the kinetic part of the Hamiltonian is given by

$$H_{\text{kin}}=\int d^3x_e\psi^\dagger(\vec{x}_e)\left(\frac{\hbar^2}{2m_e}(i\nabla_{x_e}-\vec{k}_{e0})^2+E_g-\mu_e\right)\psi(\vec{x}_e)$$
$$+\int d^3x_h\phi^\dagger(\vec{x}_h)\left(\frac{\hbar^2}{2m_h}(i\nabla_{x_h}-\vec{k}_{h0})^2-\mu_h\right)\phi(\vec{x}_h), \tag{6.8}$$

while the potential part is again unchanged. The Wannier equation of an exciton in such an indirect semiconductor can be transformed into a hydrogen-like equation by

$$\psi_{\text{ex}}(\vec{r},\vec{R})=V^{-1/2}e^{i(\vec{K}\cdot\vec{R}+i\vec{k}_0\cdot\vec{r})}\varphi(r), \tag{6.9}$$

where $\vec{k}_0=\alpha\vec{k}_{e0}+\beta\vec{k}_{h0}$ is the relative wave vector at the extrema. $\varphi(r)$ obeys the usual reduced Wannier equation

$$\left(-\frac{\hbar^2}{2m}\nabla_r^2-\frac{e^2}{\epsilon_0 r}\right)\varphi(r)=-E^b_{\text{ex}}\varphi(r). \tag{6.10}$$

The total exciton energy is given by

$$E=E_g-E^b_{\text{ex}}+(\hbar^2/2M)(\vec{K}-\vec{K}_0)^2, \tag{6.11}$$

where $\vec{K}_0=\vec{k}_{e0}-\vec{k}_{h0}$ is the center-of-mass wave vector at the extrema. The assumption of ODLRO therefore takes the form

$$\langle\psi^\dagger(\vec{x}_e')\phi^\dagger(\vec{x}_h')\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle$$
$$=e^{i[\vec{k}_0\cdot(\vec{r}-\vec{r}')]}\varphi^*(r')\varphi(r)\rho_1(\vec{R};\vec{R}') \tag{6.12}$$

and

$$\Psi(\vec{R})=\int d^3r\langle\phi(\vec{x}_h)\psi(\vec{x}_e)\rangle\varphi^*(r)e^{-i\vec{k}_0\cdot\vec{r}}. \tag{6.13}$$

With Eqs. (6.12) and (6.13) one can follow the derivation of motion for Ψ, ρ_1, and ρ_2 of Sec. III. All the results remain valid provided that $i\nabla_R$ is replaced by $(i\nabla_R-K_0)$. The lowest chemical potential [Eq. (3.13)] is no longer obtained for a constant-order parameter but for an order parameter which varies as $\Psi(R)=n_0^{1/2}e^{i\vec{K}_0\cdot\vec{R}}$, i.e., for a condensate which moves with a velocity $\vec{v}_s=\hbar\vec{K}_0/M$. Pulling the extra phase factor out of the order parameter and the reduced density matrices, e.g., $\Psi(\vec{R}) = \Psi^0(\vec{R})e^{i\vec{K}_0\cdot\vec{R}}$ and $\rho_1(\vec{R}; \vec{R}') = \rho_1^0(\vec{R}, \vec{R}')e^{i\vec{K}_0\cdot(\vec{R}-\vec{R}')}$, we go over to a coordinate system moving with $\vec{v}_s = \hbar\vec{K}_0/M$. In this coordinate system the formulas of Sec. IV for the hydrodynamic quantities expressed in terms of the reduced density matrices (with index 0) are valid.

APPENDIX

We will check the validity of supplementing each unpaired field operator with its partner in the bound state and of subsequent averaging over the internal exciton motion. For this purpose we confine ourselves to $T=0$ and to a ground state for a system of N excitons, which is of the form

$$\langle \vec{x}_1, \vec{x}_2, \ldots, \vec{x}_N ; \vec{y}_1, \vec{y}_2, \ldots, \vec{y}_N | 0\rangle = (V^N N!)^{-1} \sum_P (-1)^P \prod_l \varphi(\vec{x}_l, \vec{y}_{Pl}), \tag{A1}$$

where $\varphi(\vec{x}_l, \vec{y}_k)$ is the lowest exciton wave function for an electron at $\vec{x}_l$ bound together with a hole at y_k. P generates all permutations of the N indices. In (A1) no contributions of higher-excited exciton states are taken into account. This is correct if the zero point energy due to the localization of the excitons in the system is much smaller than the exciton binding energy, i.e., $\hbar n^{2/3}/2M \ll e^2/2a_0^*\epsilon_0$, which yields $na_0^{*3}(m/M)^{3/2} < 1$.

A typical expression which we have to evaluate is

$$\langle \psi^\dagger(\vec{x}_e)\psi(\vec{x}_e)\rangle = \int \prod d^3x_i d^3y_i \langle \vec{x}_1, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_N | \rho\psi^\dagger(\vec{x}_e)\psi(\vec{x}_e) | \vec{x}_1, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_N\rangle . \tag{A2}$$

At $T=0$ the density matrix operator is simple $\rho = |0\rangle\langle 0|$, so that (A2) gives

$$\sum_j \int \prod_i d^3x_i\, d^3y_i \langle \vec{x}_1, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_N | 0\rangle\langle 0 | \vec{x}_1, \ldots, \vec{x}_{j-1}, \vec{x}_e, \vec{x}_{j+1}, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_N\rangle \delta^{(3)}(\vec{x}_j - \vec{x}_e)$$

$$= \sum_j \int \prod_i d^3x_i d^3y_i \sum_{P,P'} \frac{(-1)^{P+P'}}{V_N N!} \prod_l \varphi^*(\vec{x}_l, \vec{y}_{Pl})\varphi(\vec{x}_l, \vec{y}_{P'l})\delta^{(3)}(\vec{x}_j - \vec{x}_e) . \tag{A3}$$

Decomposing the permutation $P' = P + Q$, we can sum over the permutations P and find

$$(1/V^N) \sum_j \int \prod_i d^3x_i d^3y_i \sum_Q (-1)^Q \prod_l \varphi^*(\vec{x}_l, \vec{y}_l)\varphi(\vec{x}_l, \vec{y}_l)\delta^{(3)}(\vec{x}_j - \vec{x}_e) . \tag{A4}$$

The contribution of the off-diagonal elements $Ql \neq l$ is by a factor $(a_0^{*3}/V)N = na_0^{*3}$ smaller and can in the low-concentration limit be neglected. The result is

$$(1/V) \sum_j \int d^3y_j \varphi^*(\vec{x}_e, \vec{y}_j)\varphi(\vec{x}_e, \vec{y}_j) = n . \tag{A5}$$

On the other hand, we approximated in Sec. III expressions such as (A2) in the following manner

$$\int d^3x'_e \prod d^3x_i\, d^3y_i \delta^{(3)}(\vec{x}_e - \vec{x}'_e)\langle \vec{x}_1, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_N | \rho\psi^\dagger(\vec{x}'_e)\phi^\dagger(\vec{x}'_h)$$

$$\times \phi(\vec{x}_h)\psi(\vec{x}_e) | \vec{x}_1, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_N\rangle \varphi(r')\varphi^*(r)\, d^3r\, d^3r' . \tag{A6}$$

This expression is equal to

$$\sum_j \int d^3x'_e \prod d^3x_i d^3y_i \delta^{(3)}(\vec{x}_e - \vec{x}'_e) \langle \vec{x}_1, \ldots, \vec{x}_{j-1}, \vec{x}_e, \vec{x}_{j+1}, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_{j-1}, \vec{y}_h, \vec{y}_{j+1}, \ldots, \vec{y}_N | 0\rangle$$

$$\times \langle 0 | \vec{x}_1, \ldots, \vec{x}_{j-1}, \vec{x}'_e, \vec{x}_{j+1}, \ldots, \vec{x}_N ; \vec{y}_1, \ldots, \vec{y}_{j-1}, \vec{y}'_h, \vec{y}_{j+1}, \ldots, \vec{y}_N\rangle \varphi(r')\varphi^*(r)\delta^{(3)}(\vec{x}_j - \vec{x}_e)\delta^{(3)}(\vec{y}_j - \vec{y}_h) d^3r\, d^3r'$$

$$= \frac{1}{V^N N!} \sum_j \int d^3x'_e \prod d^3x_i\, d^3y_i \delta^{(3)}(\vec{x}_e - \vec{x}'_e) \sum_{P,P'} (-1)^{P+P'}$$

$$\times \prod_l \varphi^*_{j'}(\vec{x}_l, \vec{y}_{Pl})\varphi(\vec{x}_l, \vec{y}_{P'l})\delta^{(3)}(\vec{x}_j - \vec{x}_e)\delta^{(3)}(\vec{y}_j - \vec{y}_e)\varphi(r')\varphi^*(r) d^3r\, d^3r' , \tag{A7}$$

where we indicated by the index j' that in $\varphi^*_{j'}$ one has to substitute $\vec{x}_j \to \vec{x}'_e$ and $\vec{y}_j \to \vec{y}'_h$. The main contribution $Q=0$ yields

$$\frac{1}{V}\quad {}_{j} \int d^3x'_e \delta^3(\vec{x}_e - \vec{x}'_e)\varphi^*(\vec{x}'_e, \vec{y}'_h)\varphi(\vec{x}_e, \vec{y}_h)\varphi^*(r)\varphi(r') d^3r\, d^3r' . \tag{A8}$$

Going over to an integration over $\vec{R}'$, we get with $\delta^{(3)}(\vec{x}_e - \vec{x}_e') \approx \delta^{(3)}(\vec{R} - \vec{R}')$ again the result (A5). The last approximation is justified, if the mean exciton separation is much larger than the extension of an exciton, i.e., $\langle |\vec{R} - \vec{R}'| \rangle \approx n^{-1/3} \gg a_0^*$ or $n a_0^{*3} \ll 1$. So we see, that the procedure used in Sec. II is correct in the low-density limit, where higher-excited states do not contribute and where the excitons are still well separated.

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