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IN MEMORY OF LEONID VENIAMINOVICH KELDYSH

Coherent states of excitons

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Abstract. The concept of a coherent exciton state is formulated. It is shown that for this state, a macroscopic wave function can be introduced such that it satisfies a nonlinear equation of the type familiar in the phenomenological theory of a superfluid liquid. The corresponding nondissipative flux is the flux of energy. For excitons interacting with an electromagnetic field, a coupled system of Maxwell equations and Ginzburg–Pitaevskii-type equations (phenomenological theory of Bose liquid) is obtained.

Keywords: excitons, coherent state of excitons, macroscopic wave function, phenomenological theory of superfluid liquid, Bose-liquid equations, Ginzburg–Pitaevskii equations

There has been a large number of recent theoretical papers on exciton condensation in crystals [1–14]. There are, in fact, three different problems at issue here. One is the thermodynamically equilibrium rearrangement of the electronic spectrum due to the instability of the original spectrum under the electron–electron interaction [6–8]. Another is the Bose condensation of nonequilibrium excitons (for example, those excited by light) [1–5]. The third is the coalescence of excitons into a dense phase, i.e., condensation in the same sense of the word in which any gas condenses into a liquid [14, 15].

Although the problem of Bose condensation of nonequilibrium excitons was the first to appear in the literature, it remains the subject of the most fundamental difference of opinion among researchers. Early studies [1–3] assumed that excitons, which consist of two Fermi particles (an electron and a hole), are bosons and that theoretical results for Bose gases and Bose liquids (which consist of structureless particles) directly apply to a system of excitons. Subsequent analyses [4, 5] showed that the deviation of the exciton statistics from the Bose statistics must be taken into account simultaneously with introducing the exciton–exciton interaction, and that sufficiently large exciton densities make the very concept of the exciton meaningless. But at low densities, a system of excitons does indeed behave like a weakly nonideal Bose gas and, notably, can exhibit superfluid motion in a crystal. However, it was argued recently in [13] that unlike a system of true bosons, a system of excitons cannot be superfluid in principle. In this paper, we focus on the analysis of this situation and on resolving the problem of what the Bose condensation and excitonic superfluidity mean from the physical (observational) standpoint. We show, in particular, that the conclusions in Ref. [13] are based on a misconception.

By an exciton, as usual, we mean an itinerant electronic excitation in a crystal not associated with charge and mass transfer. In the simplest molecular crystal or semiconductor models, the exciton is, respectively, an excited single-molecule state transferred resonantly between elementary cells of the crystal (Frenkel exciton) or a hydrogen-like bound electron–hole state (Wannier–Mott exciton). Thus, simply by definition, the motion of an exciton cannot involve a flow of matter or electric charge. Excitons transfer their excitation energy and, possibly, properties such as the angular momentum and the electric and magnetic moments whenever appropriate. Therefore, the superfluidity of nonequilibrium excitons can also well imply the existence of undamped energy flows (with a reservation to be made below) or, for example, the existence of polarization, but it does not imply a superfluid mass or charge transfer, whereas the proof of the impossibility of exciton superfluidity in Ref. [13] totally relies on the analysis of mass transfer.

We also note in passing that the formal proof in Ref. [13] has no relation to nonequilibrium excitons because it assumes that all electrons have the same chemical potential and hence the system is in full thermodynamic equilibrium. In actual fact, however, the condensation of nonequilibrium excitons implies that the electron–hole system is not fully in equilibrium in the sense that although the electrons, holes, and excitons are in equilibrium among themselves and with the lattice, the total number of excitons and electron–hole pairs is determined not by thermodynamic equilibrium but by a certain external excitation source. Such a situation readily occurs in real conditions, because recombination is in most cases much slower than the thermalization of electrons and holes and their binding into excitons. For example, in germanium at liquid helium temperatures, the thermalization time $\lesssim 10^{-9}$ s and the exciton formation time from electrons and holes are of the same order of magnitude for the electron and hole concentrations $n_{e,h} \gtrsim 10^{12}$ cm^{-3} and the exciton lifetime $\gtrsim 10^{-5}$ s. The exciton lifetime can be much longer if the exciton recombination is spin-forbidden.

We can now be more precise about the concept of the superfluid flow of excitons. Clearly, in contrast to liquid helium and superconductors, the superfluid flow of excitons

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exists not arbitrarily long but only during the exciton lifetime, and the transition of the system of excitons into a superfluid state means that the flow damping time is determined not by the exciton scattering time but by the exciton lifetime, which is longer by several orders of magnitude.

Excitons are most commonly viewed as certain quasiparticles in a crystal, and from this standpoint their Bose condensation is the accumulation of a macroscopic number of such particles in a single state. The same situation, however, can also be described in other terms: as is known [16, 17], excitons are in fact the quanta of normal vibrations of the electron density in a crystal, similar in many respects to plasmons. Their Bose-condensed state is then a coherent definite-phase electron density wave with a finite amplitude (rather than with an amplitude of the order of V^{-1}, where V is the system volume). As regards the statement on superfluidity, this means that introducing effects that are nonlinear in the amplitude results in the complete suppression of scattering processes for such a wave.

We now turn to a more formal analysis of the problem posed. The secondary-quantized electron Hamiltonian of the crystal has the usual form

$$H = -\frac{\hbar^2}{2m_0}\int \psi_\alpha^+(\mathbf{x})\,\nabla^2\psi_\alpha(\mathbf{x})\,\mathrm{d}^3x - \sum_{\mathbf{n},k} Z_k e^2 \int \frac{\psi_\alpha^+(\mathbf{x})\,\psi_\alpha(\mathbf{x})}{|\mathbf{x}-\mathbf{R}_{\mathbf{n},k}|}\,\mathrm{d}^3x + \frac{e^2}{2}\int \frac{\psi_\alpha^+(\mathbf{x})\,\psi_\beta^+(\mathbf{x}')\,\psi_\beta(\mathbf{x}')\,\psi_\alpha(\mathbf{x})}{|\mathbf{x}-\mathbf{x}'|}\,\mathrm{d}^3x\,\mathrm{d}^3x', \tag{1}$$

where $\psi_\alpha^+(\mathbf{x})$ and $\psi_\alpha(\mathbf{x})$ are fermion operators satisfying the commutation relations $[\psi_\alpha(\mathbf{x}),\psi_\beta^+(\mathbf{x}')]_+ = \delta_{\alpha\beta}\delta(\mathbf{x}-\mathbf{x}')$, the symbol $[...]_+$ denotes the anticommutator, $\hbar$, m_0, and e are the Planck constant, the electron mass, and the electron charge, and Z_k and $\mathbf{R}_{\mathbf{n},k}$ are the atomic number and the radius vector of the nucleus at the kth position in the $\mathbf{n}$th unit cell. For simplicity, we consider the nuclei to be rigidly fixed, and the subsequent treatment is carried out first in the mean-field approximation. Then the operator $\psi_\alpha(\mathbf{x})$ can be decomposed into electron (positive-frequency) and hole (negative-frequency) parts:

$$\psi_\alpha(\mathbf{x}) = \psi_\alpha^{(\mathrm{e})}(\mathbf{x}) + \psi_\alpha^{(\mathrm{h})+}(\mathbf{x}),$$
$$\psi_\alpha^{(\mathrm{e})}(\mathbf{x}) = \sum_{j>j_0} a_j\chi_{j\alpha}(\mathbf{x}), \qquad \psi_\alpha^{(\mathrm{h})+}(\mathbf{x}) = \sum_{j\leqslant j_0} a_j\chi_{j\alpha}(\mathbf{x}), \tag{2}$$
$$[a_j,\,a_{j'}^+]_+ = \delta_{jj'}, \qquad [a_j,\,a_{j'}]_+ = 0.$$

Here, $\chi_{j\alpha}(\mathbf{x})$ is the set of Hartree–Fock basis functions, where the indices $j\leqslant j_0$ ($j>j_0$) label the states with filled (empty) electronic bands. These functions must evidently have the Bloch form

$$\chi_{j\alpha}(\mathbf{x}) = \exp\left(\frac{\mathrm{i}}{\hbar}\,\mathbf{p}\mathbf{x}\right)u_{\mathbf{p}l\alpha}(\mathbf{x}),$$

where $\mathbf{p}$ is the quasimomentum and l labels bands. Thus, in Eqns (2), $j=\{\mathbf{p},l\}$, and because our discussion concerns nonmetal crystals, the summation over $j\leqslant j_0$ means a summation over all $\mathbf{p}$ within the first Brillouin zone and over $l\leqslant l_0$.

The function $\chi_{j\alpha}(\mathbf{x})$ satisfies the Hartree–Fock equations

$$\int h_{\alpha\beta}(\mathbf{x},\mathbf{x}')\,\chi_{j\beta}(\mathbf{x}')\,\mathrm{d}^3x' = \varepsilon_j\chi_{j\alpha}(\mathbf{x}), \tag{3}$$

$$h_{\alpha\beta}(\mathbf{x},\mathbf{x}') = \delta_{\alpha\beta}\delta(\mathbf{x}-\mathbf{x}')\left\{-\frac{\hbar^2}{2m_0}\nabla^2 - \sum_{\mathbf{n},k}\frac{Z_k e^2}{|\mathbf{R}_{\mathbf{n},k}-\mathbf{x}|} + \frac{e^2}{2}\int\frac{g_{\beta\beta}(\mathbf{y},\mathbf{y})}{|\mathbf{x}-\mathbf{y}|}\,\mathrm{d}^3y\right\} - e^2\,\frac{g_{\alpha\beta}(\mathbf{x},\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}, \tag{4}$$

where

$$g_{\alpha\beta}(\mathbf{x},\mathbf{x}') = \sum_{j\leqslant j_0}\chi_{j\alpha}(\mathbf{x})\,\chi_{j\beta}^*(\mathbf{x}').$$

The function $g_{\alpha\beta}$ is the operator of projection onto the subspace of filled electron states and coincides with the limit value of the electron Green's function

$$G_{\alpha\beta}^{(0)}(\mathbf{x}t;\mathbf{x}'t') = -\frac{\mathrm{i}}{\hbar}\left\langle\left(T\psi_\alpha^{(0)}(\mathbf{x}t)\,\psi_\beta^{+(0)}(\mathbf{x}'t')\right)\right\rangle_0$$

as $t'\to t-0$. Here, as usual, $\langle T...\rangle_0$ denotes the ground-state average of the chronologically ordered operator product and $\psi_\alpha^{(0)}(\mathbf{x}t)$ is the electron Fermi field operator in the interaction representation.

The exciton states of interest to us are described by the two-particle two-time Green's function

$$G_{\alpha\beta,\gamma\delta}^{(2)}(\mathbf{x},\mathbf{y},t;\mathbf{x}',\mathbf{y}',t') = -\frac{\mathrm{i}}{\hbar}\left\langle T\psi_\alpha^+(\mathbf{x}t)\,\psi_\beta(\mathbf{y}t)\,\psi_\gamma^+(\mathbf{x}'t')\,\psi_\delta(\mathbf{y}'t')\right\rangle_0,$$

where $\psi_\alpha(\mathbf{x}t)$ are Heisenberg operators.

Introducing the full set of excited states $|J\mathbf{P}\rangle$ (where $\mathbf{P}$ is the total quasimomentum and J is the set of all other quantum numbers) and their corresponding energy levels $E_{J\mathbf{P}}$, we can write the function $G^{(2)}$ in the form

$$\mathrm{i}\hbar G_{\alpha\beta,\gamma\delta}^{(2)}(\mathbf{x},\mathbf{y},t;\mathbf{x}',\mathbf{y}',t') = \sum_{\mathbf{P},J}\left\{\varphi_{\alpha\beta}^{\mathbf{P}J}(\mathbf{x},\mathbf{y})\varphi_{\gamma\delta}^{\mathbf{P}J*}(\mathbf{x}',\mathbf{y}')\times\exp\left[\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\,\frac{\mathbf{x}+\mathbf{y}-\mathbf{x}'-\mathbf{y}'}{2}-E_{J\mathbf{P}}(t-t')\right)\right]\right\}\ \text{at}\ t>t',$$

$$\mathrm{i}\hbar G_{\alpha\beta,\gamma\delta}^{(2)}(\mathbf{x},\mathbf{y},t;\mathbf{x}',\mathbf{y}',t') = \sum_{\mathbf{P},J}\left\{\varphi_{\alpha\beta}^{\mathbf{P}J*}(\mathbf{x},\mathbf{y})\varphi_{\gamma\delta}^{\mathbf{P}J}(\mathbf{x}',\mathbf{y}')\times\exp\left[-\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\,\frac{\mathbf{x}+\mathbf{y}-\mathbf{x}'-\mathbf{y}'}{2}-E_{J\mathbf{P}}(t-t')\right)\right]\right\}\ \text{at}\ t<t',$$

where

$$\exp\left(\frac{\mathrm{i}}{2\hbar}\mathbf{P}(\mathbf{x}+\mathbf{y})\right)\varphi_{\alpha\beta}^{\mathbf{P}J}(\mathbf{x},\mathbf{y}) = \left\langle 0\middle|\psi_\alpha^+(\mathbf{x})\,\psi_\beta(\mathbf{y})\middle|J\mathbf{P}\right\rangle. \tag{5}$$

Because of the translation symmetry of the problem,

$$\varphi_{\alpha\beta}^{\mathbf{P}J}(\mathbf{x}+\mathbf{R}_\mathbf{n},\mathbf{y}+\mathbf{R}_\mathbf{n}) = \varphi_{\alpha\beta}^{\mathbf{P}J}(\mathbf{x},\mathbf{y}), \tag{6}$$

where $\mathbf{R}_\mathbf{n}$ is an arbitrary lattice vector. Passing to the Fourier time representation of the quasimomentum, we obtain

$$G_{\alpha\beta,\gamma\delta}^{(2)}(\mathbf{x},\mathbf{y};\mathbf{x}',\mathbf{y}';\mathbf{P}E) = \sum_J\frac{2E_{J\mathbf{P}}}{E^2-(E_{J\mathbf{P}}-\mathrm{i}\delta)^2}\,\varphi_{\alpha\beta}^{J\mathbf{P}}(\mathbf{x},\mathbf{y})\varphi_{\gamma\delta}^{J\mathbf{P}*}(\mathbf{x}',\mathbf{y}'), \quad \delta\to+0. \tag{7}$$

The discrete values of $E_{J\mathbf{P}}$ (at fixed $\mathbf{P}$), i.e., the poles of Eqn (7), correspond to excitons, and the corresponding function $\varphi_{\alpha\beta}^{\mathbf{P}J}(\mathbf{x},\mathbf{y})\exp[(\mathrm{i}/2\hbar)\mathbf{P}(\mathbf{x}+\mathbf{y})]$ defined by Eqn (5) can be viewed as the exciton wave function, with the respective variables $(\mathbf{x},\alpha)$ and $(\mathbf{y},\beta)$ referring to the electron and the hole. Hence, the only possible definition for the exciton creation and annihilation operators $J\mathbf{P}$ is apparently

$$B_{J\mathbf{P}}^{+}=\frac{1}{\sqrt{V}}\int\exp\left(\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}(\mathbf{x}+\mathbf{y})\right)\times\psi_{\alpha}^{+}(\mathbf{x})\,\varphi_{\alpha\beta}^{J\mathbf{P}}(\mathbf{x},\mathbf{y})\,\psi_{\beta}(\mathbf{y})\,\mathrm{d}^3x\,\mathrm{d}^3y\,, \tag{8}$$

$$B_{J\mathbf{P}}=\frac{1}{\sqrt{V}}\int\exp\left(-\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}(\mathbf{x}+\mathbf{y})\right)\times\psi_{\alpha}^{+}(\mathbf{x})\,\varphi_{\alpha\beta}^{J\mathbf{P}+}(\mathbf{x},\mathbf{y})\,\psi_{\beta}(\mathbf{y})\,\mathrm{d}^3x\,\mathrm{d}^3y\,, \tag{9}$$

where $\varphi_{\alpha\beta}^{J\mathbf{P}+}(\mathbf{x},\mathbf{y})=[\varphi_{\beta\alpha}^{J\mathbf{P}}(\mathbf{y},\mathbf{x})]^*$ and V is the normalization volume. By writing the commutator of these operators,

$$\begin{aligned}[B_{J\mathbf{P}},B_{J'\mathbf{P}'}^{+}]=\frac{1}{V}\int\psi_{\alpha}^{+}(\mathbf{x})\Bigg\{&\exp\left(-\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}'\mathbf{x}\right)\varphi_{\alpha\gamma}^{J'\mathbf{P}'+}(\mathbf{x},\mathbf{z})\\&\times\exp\left(-\frac{\mathrm{i}}{2\hbar}(\mathbf{P}-\mathbf{P}')\mathbf{z}\right)\varphi_{\gamma\beta}^{J\mathbf{P}}(\mathbf{z},\mathbf{y})\exp\left(\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}\mathbf{y}\right)\\&-\exp\left(\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}\mathbf{x}\right)\varphi_{\alpha\gamma}^{J\mathbf{P}}(\mathbf{x},\mathbf{z})\exp\left(\frac{\mathrm{i}}{2\hbar}(\mathbf{P}-\mathbf{P}')\mathbf{z}\right)\\&\times\varphi_{\gamma\beta}^{J'\mathbf{P}'+}(\mathbf{z},\mathbf{y})\exp\left(-\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}'\mathbf{y}\right)\Bigg\}\psi_{\beta}(\mathbf{y})\,\mathrm{d}^3x\,\mathrm{d}^3y\,\mathrm{d}^3z\,,\end{aligned} \tag{10}$$

it is easy to see that they are not at all of the Bose type in general. The situation is simplified in the mean-field approximation, however, where a complete orthogonal set of one-electron states exists. In this case, the exciton wave function can be presented as a superposition of the products of electron and hole states,

$$\varphi_{\alpha\beta}^{J\mathbf{P}}(\mathbf{x},\mathbf{y})=\sum_{l>l_0,\,l'\leqslant l_0,\,\mathbf{p}}u_{\mathbf{p}l\gamma}(\mathbf{x})(\varphi_{\alpha\beta}^{J\mathbf{P}})_{ll'}^{\gamma\delta}u_{\mathbf{p}-\mathbf{P}l'\delta}^{*}(\mathbf{y})\,. \tag{11}$$

The products $\varphi\varphi^{+}$ and $\varphi^{+}\varphi$ are then the projection operators onto the mutually orthogonal subspaces of electron and hole states, and therefore, decomposing the operators ψ in Eqn (10) into the electron and hole parts and using the orthonormalization condition for the $\varphi^{J\mathbf{P}}$,

$$\frac{1}{V}\int\varphi_{\alpha\beta}^{J\mathbf{P}+}(\mathbf{x},\mathbf{y})\,\varphi_{\beta\alpha}^{J'\mathbf{P}}(\mathbf{y},\mathbf{x})\,\mathrm{d}^3x\,\mathrm{d}^3y=\delta_{JJ'}\,, \tag{12}$$

we can transform Eqn (10) to the form

$$\begin{aligned}[B_{J\mathbf{P}},B_{J'\mathbf{P}'}^{+}]=\delta_{JJ'}\delta_{\mathbf{P}\mathbf{P}'}-\frac{1}{V}\int\Bigg\{&\psi_{\alpha}^{(\mathrm{e})+}(\mathbf{x})\exp\left(\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}\mathbf{x}\right)\\&\times\varphi_{\alpha\gamma}^{J\mathbf{P}}(\mathbf{x},\mathbf{z})\exp\left(\frac{\mathrm{i}}{2\hbar}(\mathbf{P}-\mathbf{P}')\mathbf{z}\right)\varphi_{\gamma\beta}^{J'\mathbf{P}'+}(\mathbf{z},\mathbf{y})\\&\times\exp\left(-\frac{\mathrm{i}}{2}\,\mathbf{P}'\mathbf{y}\right)\psi_{\beta}^{(\mathrm{e})}(\mathbf{y})+\psi_{\alpha}^{(\mathrm{h})+}(\mathbf{x})\exp\left(-\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}'\mathbf{y}\right)\\&\times\varphi_{\beta\gamma}^{J'\mathbf{P}'+}(\mathbf{y},\mathbf{z})\exp\left(\frac{\mathrm{i}}{2\hbar}(\mathbf{P}-\mathbf{P}')\mathbf{z}\right)\varphi_{\gamma\alpha}^{J\mathbf{P}}(\mathbf{z},\mathbf{x})\\&\times\exp\left(\frac{\mathrm{i}}{2\hbar}\,\mathbf{P}\mathbf{x}\right)\psi_{\beta}^{(\mathrm{h})}(\mathbf{y})\Bigg\}\,\mathrm{d}^3x\,\mathrm{d}^3y\,\mathrm{d}^3z\,.\end{aligned} \tag{13}$$

It follows from this equation that the operators B are close to the Bose type for weakly excited states of the system, their commutation relations deviating from those of Bose operators by a quantity of the order of $n_{\mathrm{e}}a^3$, where n_{e} is the density of electron excitations and a is the effective exciton radius, which is determined by the way $\varphi(\mathbf{x},\mathbf{y})$ decays at large $|\mathbf{x}-\mathbf{y}|$: $\varphi(\mathbf{x},\mathbf{y})\lesssim\mathrm{const}\times\exp[-|\mathbf{x}-\mathbf{y}|/a]$ at $|\mathbf{x}-\mathbf{y}|\gg a$.

If excitons were pure bosons and their interaction could be ignored, the coherent exciton states could be defined in the usual way,

$$|\beta,J\mathbf{P}\rangle=\exp\left[\beta B_{J\mathbf{P}}^{+}\exp\left(\frac{\mathrm{i}}{\hbar}\,E_{J\mathbf{P}}t\right)-\beta^{*}B_{J\mathbf{P}}\exp\left(-\frac{\mathrm{i}}{\hbar}\,E_{J\mathbf{P}}t\right)\right]|0\rangle\,. \tag{14}$$

However, including deviations of the exciton statistics from the Bose statistics together with the interaction between excitons leads to the fact that both the form of the operator $B_{J\mathbf{P}}$ and the energy value $E_{J\mathbf{P}}$ change as the exciton wave amplitude β is varied. The coherent exciton states must therefore be defined in a more general way:

$$|\varphi\rangle=\exp\left\{\int\left[\psi_{\alpha}^{+}(\mathbf{x})\varphi_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_{\beta}(\mathbf{y})\exp\left(\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\,\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right)-\psi_{\alpha}^{+}(\mathbf{x})\varphi_{\alpha\beta}^{+}(\mathbf{x},\mathbf{y})\psi_{\beta}(\mathbf{y})\exp\left(-\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right)\right]\mathrm{d}^3x\,\mathrm{d}^3y\right\}|0\rangle. \tag{15}$$

The function $\varphi_{\alpha\beta}$ entering this definition does not coincide with any of the $\varphi_{\alpha\beta}^{J\mathbf{P}}$ but tends to one of them in the low-density limit, and its exact form (as well as the value of μ) must be determined from the Schrödinger equation

$$\left(\mathrm{i}\hbar\,\frac{\partial}{\partial t}-H\right)|\varphi\rangle=0\,.$$

Letting D_{φ} denote the operator in the right-hand side of Eqn (15), we transform it to the form

$$D_{\varphi}D_{\varphi}^{+}\left(\mathrm{i}\hbar\,\frac{\partial}{\partial t}-H\right)D_{\varphi}|0\rangle=0\,,$$

which is equivalent to

$$\left(\mathrm{i}\hbar D_{\varphi}^{+}\,\frac{\partial D_{\varphi}}{\partial t}-\tilde{H}\right)|0\rangle=0\,, \tag{16}$$

where $\tilde{H}=D_{\varphi}^{+}HD_{\varphi}$.

Equation (16) cannot be satisfied rigorously by any choice of the function $\varphi_{\alpha\beta}(\mathbf{x},\mathbf{y})$ because the form of function (15) does not take multiparticle correlation effects into account and corresponds in its meaning to describing the state of the system in the mean-field approximation. This situation is not specific to excitons. For any system of interacting bosons, it is only in the mean-field approximation that Eqn (14) determines the coherent states. All the correlation corrections can be calculated by a diagram technique for strongly non-equilibrium states, as discussed in Ref. [18]. In this paper, we confine ourselves, as already mentioned, to the lowest (mean-field) approximation. The function $\varphi_{\alpha\beta}(\mathbf{x},\mathbf{y})$ then has a structure similar to that in Eqn (11), i.e., its expansion in the set of functions $\chi_{j\alpha}(\mathbf{x})$ contains only electron states for the variable $\mathbf{x}$ and only hole states for $\mathbf{y}$. Therefore, the operator

D_φ can be rewritten in the form

$$D_\varphi = \exp\left\{\int\left[\psi_\alpha^{(e)+}(\mathbf{x})\varphi_{\alpha\beta}(\mathbf{x},\mathbf{y}) \times \exp\left(\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right)\psi_\beta^{(h)+}(\mathbf{y}) - \psi_\beta^{(h)}(\mathbf{y})\varphi^*_{\alpha\beta}(\mathbf{x},\mathbf{y}) \times \exp\left(-\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right)\psi_\alpha^{(e)}(\mathbf{x})\right]\mathrm{d}^3x\,\mathrm{d}^3y\right\}. \quad (17)$$

As is known, this unitary operator performs a linear transformation of the operators $\psi^{(e)}$ and $\psi^{(h)}$. Physically, this means that the partial redistribution of electrons due to the creation of a large number of excitons results in a change in the concept of a hole (the subspace of filled states) in the system, and state (15) is the vacuum state for the operators redefined in this way. The formulas for passing to the new operators are

$$\psi_\alpha^{(e)}(\mathbf{x}) \to D_\varphi^+\psi_\alpha^{(e)}(\mathbf{x})D_\varphi = \int\left\{C_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\alpha^{(e)}(\mathbf{y}) + \exp\left(\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right)S_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\beta^{(h)+}(\mathbf{y})\right\}\mathrm{d}^3y,$$

$$\psi_\alpha^{(h)}(\mathbf{x}) \to D_\varphi^+\psi_\alpha^{(h)}(\mathbf{x})D_\varphi = \int\left\{\tilde{C}_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\alpha^{(h)}(\mathbf{y}) - \exp\left(\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right)\psi_\beta^{(e)+}(\mathbf{y})S_{\beta\alpha}(\mathbf{y},\mathbf{x})\right\}\mathrm{d}^3y, \quad (18)$$

where

$$C_{\alpha\beta}(\mathbf{x},\mathbf{y}) = \delta_{\alpha\beta}\delta(\mathbf{x}-\mathbf{y}) + \sum_{n=1}^{\infty}\frac{(-1)^n}{(2n)!}(\varphi\varphi^+)^n,$$
$$S_{\alpha\beta}(\mathbf{x},\mathbf{y}) = \varphi_{\alpha\beta}(\mathbf{x},\mathbf{y}) + \sum_{n=1}^{\infty}\frac{(-1)^n}{(2n+1)!}\varphi(\varphi^+\varphi)^n. \quad (19)$$

The product of the operators φ and φ^+ in Eqns (19) is understood in the sense of integral convolution. With Eqns (18) and (19), Eqn (16) becomes

$$0 = \int\left\{\psi_\alpha^{(e)+}(\mathbf{x})\tilde{h}^{(e)}_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\beta^{(e)}(\mathbf{y}) + \psi_\alpha^{(h)+}(\mathbf{x})\tilde{h}^{(h)}_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\beta^{(h)}(\mathbf{y}) + \psi_\alpha^{(e)+}(\mathbf{x})Q_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\beta^{(h)+}(\mathbf{y})\exp\left(-\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right) + \psi_\beta^{(h)}(\mathbf{y})Q^*_{\alpha\beta}(\mathbf{x},\mathbf{y})\psi_\alpha^{(e)}(\mathbf{x})\exp\left(\frac{\mathrm{i}}{\hbar}\left(\mathbf{P}\frac{\mathbf{x}+\mathbf{y}}{2}-\mu t\right)\right) + \frac{e^2}{2}\frac{N\left[\psi_\alpha^+(\mathbf{x})\psi_\beta^+(\mathbf{y})\psi_\beta(\mathbf{y})\psi_\alpha(\mathbf{x})\right]}{|\mathbf{x}-\mathbf{y}|}\right\}\mathrm{d}^3x\,\mathrm{d}^3y\,|0\rangle, \quad (20)$$

where $N[...]$ is the normally ordered product of the operators $\psi = \psi^{(e)} + \psi^{(h)+}$. Although the electron–electron interaction term looks formally the same as before the transformation, it is different because the operators $\psi^{(e)}$ and $\psi^{(h)}$ and the function $G^{(0)}_{\alpha\beta}$ are different.

In the mean-field approximation, Eqn (20) takes the form

$$\left\{\psi^{(e)+}\tilde{h}^{(e)}\psi^{(e)} + \psi^{(h)+}\tilde{h}^{(h)}\psi^{(h)} + \psi^{(e)+}Q\psi^{(h)+} + \psi^{(h)}Q^+\psi^{(e)}\right\}|0\rangle = 0 \quad (21)$$

(we again use a symbolic expression where products are understood as integral convolutions). Here, as in Eqn (20), the following notation is used:

$$\tilde{h}^{(e)} = C\left(h^{(e)}-v\right)C - S\left(h^{(h)}-v^+\right)S^+ + CVS^+ + SV^+C - \mu S^+S, \quad (22)$$

$$Q = C\left(h^{(e)}-v\right)S + S\left(h^{(h)}+v^+\right)\tilde{C} - CV\bar{C} + SV^+S - \mu CS. \quad (23)$$

The matrix $\bar{C}$ differs from C by the permutation $\varphi \rightleftarrows \varphi^+$, and the quantities V and v are defined as

$$V_{\alpha\beta}(\mathbf{x},\mathbf{y}) = \frac{e^2}{|\mathbf{x}-\mathbf{y}|}\int S_{\alpha\gamma}(\mathbf{x},\mathbf{z})\,\tilde{C}_{\gamma\beta}(\mathbf{z},\mathbf{y})\,\mathrm{d}^3z, \quad (24)$$

$$v_{\alpha\beta}(\mathbf{x},\mathbf{y}) = \frac{e^2}{|\mathbf{x}-\mathbf{y}|}\int S_{\alpha\gamma}(\mathbf{x},\mathbf{z})S^+_{\gamma\beta}(\mathbf{z},\mathbf{y})\,\mathrm{d}^3z. \quad (25)$$

We also note the relations $CC + SS^+ = 1$ and $CS = S\tilde{C}$ and the fact that the quantities

$$n_e(\mathbf{x}) = \int S_{\alpha\beta}(\mathbf{x},\mathbf{y})\,S^+_{\beta\alpha}(\mathbf{y},\mathbf{x})\,\mathrm{d}^3y,$$
$$n_h(\mathbf{x}) = \int S^+_{\alpha\beta}(\mathbf{x},\mathbf{y})\,S_{\beta\alpha}(\mathbf{y},\mathbf{x})\,\mathrm{d}^3y \quad (26)$$

determine the densities of excited electrons and holes, which are periodic with the crystal lattice period for the class of states considered so far.

The necessary and sufficient condition for Eqn (21) to hold is clearly $Q \equiv 0$, because the first two terms in the left-hand side give zero when acting on the vacuum. Thus, Eqn (21) reduces to

$$Q_{\alpha\beta}(\mathbf{x},\mathbf{y}) = 0, \quad (27)$$

which, by Eqns (19) and (23)–(25), is a nonlinear integro-differential equation for $\varphi_{\alpha\beta}(\mathbf{x},\mathbf{y})$, and the exciton chemical potential μ is defined as an eigenvalue of Eqn (27).

The analysis [2] of solutions of Eqn (27) in the general case is hardly possible, and we therefore confine ourselves to relatively low excitation densities $n_{e,h}a^3 \ll 1$, for which Eqn (27) can be expanded in powers of φ using the relations

$$C \simeq 1 - \frac{1}{2}\varphi\varphi^+, \qquad \tilde{C} \simeq 1 - \frac{1}{2}\tilde{\varphi}^+\varphi, \qquad S \simeq \varphi - \frac{1}{6}\varphi\varphi^+\varphi.$$

The lowest approximation, linear in φ, yields

$$\int\left\{h^{(e)}_{\alpha\gamma}(\mathbf{x},\mathbf{z})\,\varphi_{\gamma\beta}(\mathbf{z},\mathbf{y}) + h^{(h)}_{\gamma\beta}(\mathbf{z},\mathbf{y})\,\varphi_{\alpha\gamma}(\mathbf{x},\mathbf{z})\right\}\mathrm{d}^3z - \left(\frac{e^2}{|\mathbf{x}-\mathbf{y}|}+\mu\right)\varphi_{\alpha\beta}(\mathbf{x},\mathbf{y}) = 0, \quad (28)$$

which is the Schrödinger equation for Coulomb-interacting electrons and holes. The way they actually interact in a crystal is much more complicated, but it is easy to show that after the summation of all correlation corrections that are linear in φ, Eqn (28) becomes exactly an equation for the exciton wave functions $\varphi^{J\mathbf{P}}$. In what follows, we replace the index J by 0, having the lowest excitonic branch of the spectrum in mind.

Thus, in the lowest approximation,

$$\varphi = \sqrt{n}\,\varphi^{0\mathbf{P}}\,, \qquad \mu^{(0)} = E_{0\mathbf{P}}\,,$$

where the normalization factor $\sqrt{n}$ is determined by the mean exciton concentration. The solvability condition for the next-approximation equation ($\sim \varphi^3$) yields a level shift $\mu^{(1)}$ proportional to n:

$$\mu^{(1)} = \varkappa n\,,$$

$$\varkappa = e^2 \int \mathrm{d}^3x\,\mathrm{d}^3y \left\{ \int \left(\frac{1}{|\mathbf{x}-\mathbf{y}|} + \frac{1}{|\mathbf{x}-\mathbf{z}|} \right) \times \varphi_{\alpha\beta}(\mathbf{x},\mathbf{y}) \varphi^{+}_{\beta\gamma}(\mathbf{y},\mathbf{z}')\, \varphi_{\gamma\delta}(\mathbf{z}',\mathbf{z})\, \varphi^{+}_{\delta\alpha}(\mathbf{z},\mathbf{x})\, \mathrm{d}^3z\,\mathrm{d}^3z' - \sum_{\alpha,\beta} \frac{1}{|\mathbf{x}-\mathbf{y}|} \left(\left| \int \varphi_{\alpha\gamma}(\mathbf{x},\mathbf{z})\, \varphi^{+}_{\gamma\beta}(\mathbf{z},\mathbf{y})\, \mathrm{d}^3z \right|^2 + \left| \int \varphi^{+}_{\alpha\gamma}(\mathbf{x},\mathbf{z})\, \varphi_{\gamma\beta}(\mathbf{z},\mathbf{y})\, \mathrm{d}^3z \right|^2 \right) \right\}. \tag{29}$$

The constant $\varkappa$ can also be calculated for the pure Coulomb case. Summing all the corrections for the constant is a more difficult problem [4], and it is more convenient to simply regard it as a phenomenological parameter.

Until now, we have been considering stationary coherent exciton states with a fixed total quasimomentum $\mathbf{P}$, i.e., with the same mean excitation density n for all elementary cells in the crystal. In a more general case, wave packets are composed of states of this type. To introduce such packets, we specify a transformation D_Φ of a more general type,

$$D_\Phi = \exp \left\{ \int \left[\psi^{(\mathrm{e})+}_{\alpha}(\mathbf{x})\, \Phi_{\alpha\beta}(\mathbf{x},\mathbf{y},t) \exp\left(\frac{\mathrm{i}}{\hbar}\,\mu t \right) \psi^{(\mathrm{h})+}_{\beta}(\mathbf{y}) - \psi^{(\mathrm{h})}_{\beta}(\mathbf{y}) \exp\left(-\frac{\mathrm{i}}{\hbar}\,\mu t \right) \Phi^{+}_{\alpha\beta}(\mathbf{x},\mathbf{y},t)\, \psi^{(\mathrm{e})}_{\alpha}(\mathbf{x}) \right] \mathrm{d}^3x\,\mathrm{d}^3y \right\},$$

where we assume that the dependence of $\Phi_{\alpha\beta}(\mathbf{x},\mathbf{y},t)$ on t is slower than $\exp\left((\mathrm{i}/\hbar)\mu t\right)$. Similarly to the discussion above, the equation for Φ then takes the form

$$\mathrm{i}\hbar\,\frac{\partial \Phi}{\partial t} - Q\{\Phi\} = 0\,.$$

For this equation, we seek a solution of the form $\Phi(\mathbf{x},\mathbf{y},t) = \Phi(\mathbf{x}+\mathbf{y},t)\varphi^{0\mathbf{P}}(\mathbf{x},\mathbf{y})$ assuming that $|a\nabla\Phi(\mathbf{X},t)/\Phi| \ll 1$, i.e., considering $\Phi(\mathbf{X},t)$ as a slowly varying amplitude. We set $\mathbf{P}$ equal to $\mathbf{P}_\mathrm{m}$, a value for which $E_{0\mathbf{P}}$ reaches a minimum; in its neighborhood, $E_{0\mathbf{P}} \approx E_0 + (\mathbf{P}-\mathbf{P}_\mathrm{m})^2/2m$. Then, in the lowest approximation, disregarding nonlinear terms in the derivatives of $\Phi(\mathbf{X},t)$, we obtain $\mu = E_0$, and the next approximation yields an equation for $\Phi(\mathbf{X},t)$,

$$\mathrm{i}\hbar\,\frac{\partial \Phi}{\partial t} + \frac{\hbar^2}{2m}\nabla^2\Phi - \varkappa|\Phi|^2\Phi = 0\,, \tag{30}$$

equivalent, as is known, to the phenomenological hydrodynamics equations for a superfluid liquid (for $\varkappa > 0$).

The discussion above has been concerned with excitons that have virtually no interaction with light. The condensation of dipole-active excitons should automatically create an electromagnetic field accompanying them, also in a coherent state. Writing the full Hamiltonian for the particle–field system, we readily obtain the following system of equations for the coherent-state amplitudes:

$$\mathrm{i}\hbar\,\frac{\partial \Phi}{\partial t} + \frac{\hbar^2}{2m}\nabla^2\Phi + (\hbar\omega - E_0)\Phi - \varkappa|\Phi|^2\Phi = \mathbf{E}\mathbf{d}\,,$$

$$\mathrm{rot}\,(\mathrm{rot}\,\mathbf{E}) + \frac{\tilde{\varepsilon}}{c^2}\left(\frac{\partial}{\partial t} - \mathrm{i}\omega \right)^2 \mathbf{E} = -\frac{4\pi}{c^2}\,\mathbf{d}\left(\frac{\partial}{\partial t} - \mathrm{i}\omega \right)^2 \Phi\,. \tag{31}$$

Here, ω is the mean field frequency, $\mathbf{d}$ is the dipole moment matrix element for an exciton transition, $\tilde{\varepsilon}$ is the crystal dielectric constant with the contribution from the exciton state under consideration subtracted, and $\mathbf{E}(x,t)$ is the complex field amplitude in terms of which the real field is expressed as

$$\frac{1}{2}\left(\mathbf{E}(x,t)\exp\left(-\mathrm{i}\omega t\right) + \mathbf{E}^*(x,t)\exp\left(\mathrm{i}\omega t\right) \right).$$

System of equations (31) incorporates the effects of frequency and spatial dispersion and those of nonlinear polarizability.

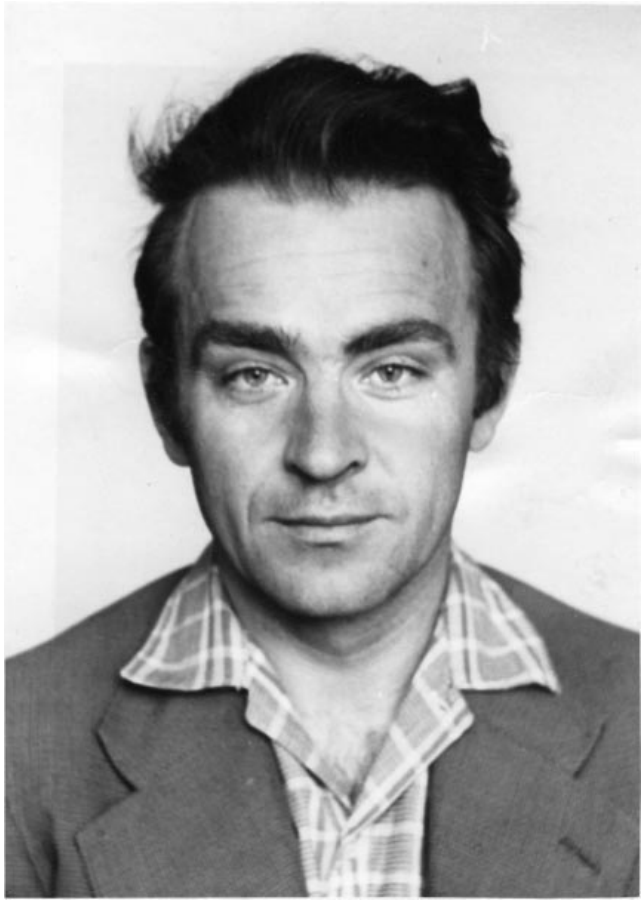
In the case of high-symmetry crystals, exciton states can be degenerate, requiring several functions Φ to be introduced for their description. While this complicates system (31), the qualitative results remain unchanged.

Note from the Editors

L V Keldysh's paper "Coherent states of excitons" presented in this memorial issue of *Physics–Uspekhi* was first published in 1972 in the Tamm memorial collection [19] and went relatively unnoticed by the physics community. The paper is of quite fundamental importance, however.

In the latter half of the 1960s and the early 1970s, there was an interesting discussion in the literature as to whether Bose condensation and superfluidity are possible in a system of excitons in a semiconductor. Within quite a short time, the discussion grew quite confused, and it was the objective of Keldysh's work to clarify a number of fundamental issues that arose. The paper emphasizes that there are three fundamental problems that should be recognized in the area. One of these is directly related to the rearrangement of the electron spectrum of a semimetal due to the Bose condensation of electron–hole pairs in the ground (equilibrium) state in the Keldysh–Kopaev model of an exciton dielectric [6]. The second problem is related to the possibility of Bose condensation and superfluidity in a nonequilibrium exciton gas produced by the optical pumping of a semiconductor [1, 4]. Finally, the third problem is about the formation (condensation) of electron–hole droplets in highly excited semiconductors as excitons break up to form a sufficiently dense electron–hole Fermi-liquid phase [20]. Experimentally, most real (multi-valley) semiconductors exhibit precisely this last scenario (it is this scenario which Keldysh predicted in his concluding speech at the Ninth International Conference on the Physics of Semiconductors held in Moscow in 1968 [20]).

At the same time, the direct analogy existing between the excitonic insulator model and BCS superconductivity led to contradictory opinions among researchers, some claiming that this model allows a superfliud electron–hole pair condensate (which manifests itself in superconductivity-type phenomena) [8], and others fully ruling out that superfluidity can occur in a system of excitons [11]. It is to explain some of these contradictions that Ref. [19] is now reproduced in *Physics–Uspekhi*.



Leonid Veniaminovich Keldysh in the late 1960s
(which was exactly the period of his active excitonic studies).
(Courtesy of Galina Nikolaevna Mikhailova, co-author of Ref. [23].)

This study is mainly concerned with whether superfluidity can be exhibited by *nonequilibrium* excitons that are produced in a semiconductor exposed to an external excitation source. It is emphasized that moving excitons cannot produce either a flow of matter or an electric current but can imply the existence of undamped (up to the exciton lifetime) energy or polarization flows, but not a superfluid mass or charge transfer, which immediately invalidates the 'general' proof of Kohn and Sherringtone, who additionally, at the very outset, assumed a thermodynamic equilibrium (i.e., a system of the excitonic insulator type).

Most of the paper focuses on the explicit construction of coherent excitonic states taking their generally nonbosonic nature into account. The result, Eqn (30), is directly analogous to the Gross–Pitaevskii equation for a superfluid liquid, with the dipole interaction with an external electromagnetic field (31) taken into account.

As regards the possibility of superfluidity in an excitonic insulator, this debate was completely resolved in a later paper by Keldysh and Guseinov [21], in which it was shown that allowing interband transitions in this model turns a second-order transition into a first-order one, and therefore the excitonic insulator state has no properties distinguishing it from the usual dielectrics.

The author of these lines was at the time Keldysh's postgraduate student at the FIAN (Lebedev Physical Institute) Theory department, named after I E Tamm shortly before. The publication of the memorial collection [19] was a major event in the life of the department. An impressive feature of the collection was the list of referenced authors, which included prominent Soviet and foreign scientists. Some of the department staff used to come to the seminars with a copy of this volume to ask its authors for an autograph. False modesty prevented me from doing this — and quite regrettably, because the autographs made those copies uniquely valuable.

Incidentally, my postgraduate research (electrons in disordered systems) had no relation to the condensation of excitons, which was then Keldysh's primary concern, and therefore my role here is simply that of an unprejudiced witness. Keldysh's prediction of electron–hole droplets in Ref. [20] was followed by a rather long break in his publications in this area. The first sufficiently detailed account of the theoretical foundations of this concept also appeared in a relatively hard-to-access paper collection [22] in 1971. Over a number of years, Keldysh's interests were centered on the experimental confirmation of this phenomenon, sometimes to the extent of coauthoring experimental studies [23]. As is known, the general picture of the formation of electron–hole droplets he gave in Ref. [20] received striking experimental confirmation, and experimental and theoretical research in this field has intensified worldwide [24].

Paper [19] stands alone in this sense, and I can offer some conjectures as to its origin. All of Keldysh's students knew about his large notebooks into which, when at home, he wrote down his calculations on a wide range of solid-state physics problems and where he described his results in detail, often without later publishing them as journal papers.

For example, E G Maksimov told me that in those years he was actively involved in attempts at constructing a consistent theory of electron–phonon interaction in metals, to extend and improve the traditional Fröhlich Hamiltonian approach by correctly using the adiabatic approximation and introducing multiparticle effects. In Maksimov's words, Keldysh also devoted much attention to these problems and occasionally showed his results to Maksimov, but did not publish anything at all.

In my view, Ref. [19] appeared as a reply to Kohn and Sherrington's paper [13], which made some points Keldysh did not agree with. This led him to perform a number of 'private' calculations, as it were, that remained hidden for a number of years in his notebooks until the opportunity came to publish them in the Tamm memorial collection. I may be wrong, but all of us, his students, recall those notebooks quite often. It would be interesting to find them and examine them for interesting results, which they are almost certain to contain and which Keldysh did not manage to publish.

M V Sadovskii

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