

IN MEMORY OF LEONID KELDYSH, ACADEMICIAN OF THE RUSSIAN ACADEMY OF SCIENCES¹

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One year passed rapidly after the unexpected and regrettable decease of the outstanding theoretical physicist and brilliant personality, academician of the Russian Academy of Sciences Leonid Veniaminovich Keldysh. In his memory we would like to remind some of his fundamental results concerning Bose-Einstein condensation of excitons in semiconductors, reflected in the monograph [1] written by professor D. W. Snoke together with one of us (S.A.M.), as well as about the contribution of academician L. V. Keldysh to the development of physics in the Republic Moldova, taking into account his membership to the Academy of Sciences of Moldova.

In a comprehensive and instructive review [2] L.V. Keldysh pointed out the peculiar properties distinguishing the e - h system from any other such as greatly reduced Coulomb interaction due to dielectric screening in the host crystal, the electron and hole masses of the same order of magnitude or different at most by an order. These cause a dominant role of quantum effects at all temperatures $K_B T < R_{yex}$, where R_{yex} is the exciton binding energy, and lead to very large zero-energy vibrations of exciton in the excitonic molecules. For the same reason, when the electron and hole masses are comparable, nothing like an "excitonic crystal" analogous to solid hydrogen, can exist. The weak van der Waals attraction between the excitons is not able to confine light particles such as excitons or biexcitons. A condensed phase of biexciton molecules i.e. a molecular liquid similar to liquid hydrogen, also cannot exist because the s -wave-scattering length of two biexcitons is large and positive. But an electron-hole liquid (EHL), similar to metallic hydrogen or alkali metals, does exist [3-6]. The collective pairing of electrons and holes in the vicinity of the Fermi surfaces, being quite similar to Bardeen-Cooper-Schrieffer (BCS) pairing in superconductors, is manifested in the appearance of energy gaps near



În imagine, de la stânga la dreapta: acad. L. Keldysh, acad. R. Hohlov, acad. S. Moscalenco.

Moscova, 1975

the Fermi surfaces. These gaps may be considered as a remnant of the binding energy of single exciton and decrease quickly with increasing particle density. The excitonic-insulator state in the nonequilibrium e - h system is a coherent Bose-Einstein condensation (BEC) state of high-density excitons with $n_{ex} a^3 \gg 1$. Keldysh originated the idea of the electron-hole liquid (EHL) and electron-hole droplet (EHD). He pointed out for the first time that the excitons and the excitonic molecules of the types similar to alkali metal atoms and molecules, with comparatively small binding energies, will become EHL_s and electron-hole droplets (EHD) in the case of high intensity excitation. These suggestions were justified completely by the subsequent theoretical and experimental investigations of high density excitons in such crystals as *Ge* and *Si* [4, 5, 7-13]. Brinkman and Rice [9] and Combescot and Nozières [10] showed that the many-valley structures of the conduction bands in *Ge* and *Si*, the fourfold degenerate structure of their valence bands, and the aniso-

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tropy of the corresponding masses not only facilitate but play a decisive role in the formation of the EHL_s and EHD_s. The EHL happens to be much more stable than the exciton and biexciton gases in these crystals.

Another important difference of the *e-h* system from ordinary matter is its essentially nonequilibrium nature, produced by some external action, usually illumination. Then the total number *N* of *e-h* pairs becomes an independent variable with a value controlled by an external source. In certain semiconductors the lifetime of the nonequilibrium *e-h* system may be much longer than the thermalization time, so that the conservation of *N* is not broken by the recombination processes on short time scales. The system appears to be in quasi-equilibrium, the only nonequilibrium parameter being the number of particles *N*.

Furthermore, Keldysh noted that in the description of the nonequilibrium excitons it is necessary to take into account the incomplete equilibrium in the *e-h* system. It is complete in the sense that the electron, holes and excitons are in equilibrium with each other and with the crystal lattice in all parameters except one, namely the total number of the excitons and of the *e-h* pairs is determined not by the thermodynamic equilibrium conditions, but by the external excitation source [14]. The exciton thermalization time is assumed to be much less than its lifetime. In this case the establishment of a quasi-equilibrium distribution function in the exciton band can be assumed.

As Keldysh noted in [2], it is also possible to create an equilibrium excitonic-insulator state, i.e. a population of permanent excitons. This can occur if the binding energy per pair in the excitonic molecule or the EHL is larger than the initial energy band gap for free *e-h* pairs creation. In this case, spontaneous reconstruction of the electronic structure of the crystal occurs. Nothing like superfluidity can arise, however, because the Hamiltonian does not conserve the total number of excitons or *e-h* pairs [15, 16]. In this case the interband scattering matrix elements of the Coulomb interaction are the sources of the creation of *e-h* pairs, lifting the gauge invariance of the Hamiltonian [17].

Keldysh and Kozlov [18] were the first to consider the problem of exciton BEC at $T = 0$ from the many electron-hole point of view. They noted that the excitons are formed from coupled electrons and holes and for this reason do not exactly obey Bose-Einstein statistics.

In the papers of all previous authors the excitons were treated as simple bosons, without taking into account the underlying Fermi constituents. Keldysh-Kozlov-Kopaev (KKK) treated the full many-electron

Hamiltonian and answered positively to the question: whether can we still talk about the Bose condensation?

The deviation of the exciton operators $\psi_{ex,k}^+$ and $\psi_{ex,k}$ from Bose statistics increases as the electron-hole density increases. For this reason, as mentioned by Keldysh and Kozlov, the possibility of considering the exciton system as a weakly nonideal Bose gas a priori is not evident. For exciton densities of the order of $10^{17} - 10^{18} \text{ cm}^{-3}$, the average distance between the excitons starts to become comparable with their radius. In this case the internal structure begins to play an important role. For example, two electrons belonging to two different excitons cannot approach each other closely if they have the same spin projections. Two holes exhibit the same behavior. The Pauli exclusion principle leads to a kinematic exciton-exciton interaction, even in the absence of a dynamic interaction. Keldysh and Kozlov [11, 15] pointed out that the effects connected with the difermion exciton structure appear at the same order of the exciton density as that of the effects tied to the Bose gas nonideality. As a result the systematic investigations of the exciton Bose condensate, even at low densities, cannot be posted as a problem about a weakly nonideal Bose gas. Nevertheless, it was shown that the exciton system with a definite structure possesses many similar properties. In particular, at low temperature a condensation can take place into a single composite boson state with $\vec{k} = 0$.

Coherent macroscopic states slowly varying in space and time have been used by London [19] in his attempts to explain the superfluidity, by Ginzburg and Landau [20] in the theory of superconductibility, by Pitaevskii [21] and Gross [22] in the theory of quantum vortices, and by Keldysh [14] in the theory of coherent excitons and photons.

The use of the Glauber-type unitary transformation permits to introduce into the Hamiltonian the spontaneous symmetry breaking. This allows one to study BEC of excitons not only in the case in which the excitons and the biexcitons are described by the true Bose operators, but also when their underlying electron-hole structure is taken into account [18]. For that case, instead of the true Bose operator the creation operator of the bound electron-hole (*e-h*) pair with the center-of-mass wave vector equal to zero was introduced.

The new operator of the displacement, which breaks the symmetry of the Hamiltonian in the *e-h* representation, was used. This procedure was followed by Keldysh and Kozlov [18], who studied the collective properties of excitons in the *e-h* description.

Bose-Einstein condensation of the coupled *e-h* pairs is well known now as the Keldysh-Kozlov-Kopa-

ev formulation of Bose condensation. It was interpolated by Comte and Nozieres between dense and dilute systems [23].

Following Ref. [14], excitonic BEC means the existence of a coherent wave of electron density, with definite phase and finite amplitude. Superfluidity of the coherent wave could signify that its scattering processes are completely suppressed by the nonlinear effects. Unlike liquid H_e , the superfluid exciton flux cannot exist an arbitrary long time, but only during the exciton lifetime. The flux relaxation time is determined by the exciton lifetime, which is some orders of magnitude greater than the single exciton relaxation time.

The electron-hole description of the exciton BEC in the Keldysh-Kozlov paper [18] as well as the Keldysh-Kopaev [15] description of the excitonic insulator state in semimetals are based on the use of the unitary transformation introducing the coherent macroscopic state formed by the Bose-condensed compound quasiparticles such as the Wannier-Mott excitons in semiconductors or as the electron-hole Cooper-type pairs in semimetals. In the case of semiconductors such unitary transformation introducing the Bose-condensate of the electron Cooper pairs was proposed by Bogoliubov [24] in his refined version of the BCS theory of superconductivity.

Keldysh and Kozlov [18] argued that along with the individual one-fermion excitations, there are also collective two-particle excitations corresponding in the limit $n_{ex} \rightarrow 0$ to the motion of a single exciton as a whole entity, without bond breaking. This excitation changes only the total momentum and the kinetic energy of the translational motion of an exciton.

The Keldysh-Kozlov equations of motion for the two-particle Green's function are to some extent similar to the Beliaev equations for the nonideal Bose gas [25, 26]. Unlike the pure algebraic Beliaev equations, however, Keldysh-Kozlov equations are integral equations over the momentum of the relative motion. For this reason they describe both the excitation and the internal structure.

One naturally expects that the BEC of excitons will lead to superfluidity, which would be detected as anomalous fast exciton diffusion. This possibility, suggested and discussed in many papers [27, 28], was subjected to criticism by Kohn and Sherrington [29]. They affirmed that the exciton system, while it is a collective of compound e - h quasiparticles, can undergo the phenomenon of BEC. But unlike true bosons, in their opinion, the composite e - h quasibosons cannot exhibit the phenomenon of the superfluidity. Keldysh [14] analyzed the latter argument and pointed that it is based on a misunderstanding. It is necessary

from the beginning to determine exactly what exciton superfluidity is and to take into account the exciton features properly. As noted by Keldysh, unlike the flux of atoms, the exciton flux is not accompanied by matter transfer. On this point there is complete accordance with the Kohn and Sherrington paper [29]. But the exciton can transfer their creation energy, their kinetic energy of translational motion, and such things as angular electric and magnetic moments, if they exist. Therefore, the superfluidity of the exciton gas, which exists only as a nonequilibrium excitation state of the crystal, means the energy or the polarization transfer, but not of the mass or of the charge. The proof of the impossibility of the exciton superfluidity in Ref. [29] was completely based on an investigation of the mass transfer. The experimental discovery of the cavity polaritons superfluidity even at room temperature completely confirmed these statements [30].

Considerable efforts for about 50 years of many generations of experimental and theoretical physicists investigating the optical properties of semiconductors were undertaken to understand and to realize experimentally the Bose-Einstein condensation of excitons. The multiple attempts of many groups of investigators were crowned by the discovery of the BEC of exciton polaritons in microcavities [31, 32]. The essential progress over last 30 years was achieved in the study of the BEC of excitons in Cu_2O crystals [33].

All these impressive achievements of the contemporary physics were stimulated in a great manner by the fundamental theoretical investigations of academician Leonid Keldysh, whose great authority between the specialists ensured success and inspired enthusiasm.

We remember with gratitude about multiple visits to Chișinău of academician L.V. Keldysh in the frame of gala sessions of the Department of General Physics and Astronomy of the USSR Academy of Sciences, of the conferences organized by the Scientific Councils on the Problems of Coherent and Nonlinear Optics and Solid State Physics as well as of the exciton seminars. All of them taken together influenced essentially and beneficially on the development of physics in Moldova. Many of our present and former doctors in physico-mathematical sciences discussed and promoted their theses in the frame of the scientific seminars and departments headed by L.V. Keldysh.

Academician L. V. Keldysh left an indelible impression in the consciousness of many generations of physicists, who learnt from him and tried to align to him. His presentations at scientific conferences and seminars were fascinating for both experienced researchers and students since he possessed the art of de-

scribing the most complicated phenomena in a simple but realistic fashion. We are proud that academician L.V. Keldysh was honorable member of the Academy of Sciences of Moldova. There is no doubt that we were privileged by fate to meet in our scientific ways a most outstanding, noble and generous personality.

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