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Invited Paper

Progress in exciton spectroscopy: personal perspective

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Abstract

In this Lecture, I display my personal perspective on the progress in physics of excitons during the last 50 years. Preference is given to the areas of this extensive field which I participated. They include molecular excitons, giant oscillator strength inherent in bound excitons, self-trapping problem, strong magnetic fields including spectroscopy of the quantum Hall effect, etc. Critical importance of international scientific collaboration is also discussed.

Keywords: Excitons; Molecular excitons; Self-trapping; Magnetoexcitons

1. Introduction

Receiving the ICL Prize is a high honor for me. In my perception, I share it with my colleagues, theorists and experimenters, and my teachers. I wish to use this remarkable opportunity to mention their names and specify some of their contributions.

This Prize is an International award. This notion signifies the fact that science has no borders. In the Soviet Union, we lived and worked in isolation from the International community. Nevertheless, invisible connections existed even then and were of great importance for research. I will illustrate this remarkable fact by several examples.

In what follows, I will concentrate on the progress in physics of excitons, a specific part of my scientific activity most closely connected with the scope of this Conference. By necessity, what I will display is my personal perspective on this field: I will speak about some of the problems which I participated.

2. Excitons 50 years ago

It is easy for me to make that landmark – exactly 50 years ago I graduated from the Kiev University. It was the childhood of excitons. The notions of Frenkel and Wannier–Mott excitons were known from textbooks. Polarized narrow exciton bands of aligned polymers known as J-aggregates (Scheibe polymers) were investigated. Some assignments for exciton bands were proposed for alkali-halides. In Kiev, A.F. Prikhot'ko and her group discovered absorption bands of molecular crystals sharply polarized along crystal axes. This polarization signified a collective phenomenon. The bands were ascribed to excitons forming exciton multiplets separated by Davydov splitting.

Devil was in details. When it came to assignment of the specific absorption bands, there were no criteria for distinguishing exciton bands from impurity bands. It was accepted that in anisotropic crystals exciton bands are polarized along the crystal axes, while impurity bands – along the axes of these molecules. However, many of the bands supposedly related to defects were surprisingly strong and had puzzling polarization.

Reliable criteria for assigning exciton and impurity bands were needed.

3. Bound excitons: Anomalous polarization and giant oscillator strength

Solomon I. Pekar, the leading theorist of the Physical Institute in Kiev, proposed a competition for explaining anomalies in the impurity spectra. I developed a theory

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(1956) based on the quasi-resonance between impurity and free exciton states in molecular crystals and won the competition. Because of the attractive potential of an impurity, bound exciton can be created in the vicinity of it. If this potential is weak, bound states are shallow and have a large radius. Under these conditions, *the "impurity band" borrows intensity from the exciton absorption band or gives its intensity to the exciton continuum*. The closer the impurity band is to the edge of the exciton energy band, the stronger the effect is. It results in *a giant change in the intensities of impurity bands and their polarization ratios*. This phenomenon is called Rashba effect.

The excitement was followed by disappointment. *JETP* rejected my paper as being of no general interest. It was published in a new Russian journal *Optika i Spektroskopiya* [1]. This made the difference: *JETP* was already being translated into English, while *Opt. Spektrosk.* was not. There was little chance of the paper to survive.

Fortunately, Vladimir L. Broude found model systems for checking the theory. These were isotopic solutions of chemically identical molecules. By changing the isotopic content of host and guest molecules, he could move an impurity band with respect to the exciton multiplet and observe changes in the band intensity and polarization. Broude and Elena F. Sheka confirmed the theory. This success permitted us to explain strong impurity features near exciton bands and begin investigating energy bands of molecular excitons.

The mechanism investigated for molecular excitons is universal and is applicable also to bound excitons in semiconductors. All that was needed was a soluble theoretical model. With Givi Gurgenishvili, a student from Georgia, I showed (1962) that the effect in semiconductors is much stronger than in molecular crystals because of the small exciton mass which results in *giant oscillator strength* [2].

Radiative transformation of excitons into biexcitons also has giant crossection (A.A. Gogolin and Rashba, [3]). At the same time, E. Hanamura [4] predicted giant two-photon production of biexcitons.

4. Landau's criticism

In the winter 1960/61 I came to Landau to discuss with him my recent prediction of the electric-dipole spin-resonance (EDSR) and my theory of bound molecular excitons. It was easy to reach him. I called him at home from the Institute of Physical Problems where he worked. Nobody introduced me. Landau asked me only two questions (where I came from and the subject of my paper) and proposed to meet in one hour. He lived in the Institute.

Actually, I wished to get Landau's opinion, first of all, about EDSR. Indeed, EDSR which is now a quite common notion [5], was not confirmed experimentally yet. I was confident about bound excitons because the theory has already been confirmed by experiment. Therefore, I decided to start with spin–orbit phenomena and EDSR. I was lucky! It was known that the first Landau's impression was very important.

For one hour we walked along the corridor, and Landau performed and kept the calculations in his memory. It was only a couple of times that we came to the blackboard in one of the offices to write some equations. He checked arguments, asked questions, and had no objections. Finally, he proposed me to give a talk at his seminar. I was highly delighted! Landau's ability to accumulate science was incredible. At the seminar it was he who answered most of the questions. His answers were brief and exact.

Surprisingly, when I started to tell Landau about bound excitons, his reaction was momentary: "You will be wrong". I used the credit which I earned by my work on EDSR, and spoke to him briefly three times during those three days. He was friendly but there could be no compromise about scientific problems. Gradually Landau started changing his mind: "I see. You have a good problem". Finally, he made his opinion and avoided discussing details by disappearing with a joke: "I am not so smart".

The following is my reconstruction of Landau's vision of the problem. Quantum mechanics is not applicable to excitons directly because electrodynamics is essentially involved. [In fact, long-range interaction results in the LT (longitudinal–transverse) splitting in the exciton energy spectrum and, therefore, violates the three-fold degeneracy typical for quantum p-states]. Landau advised me to consider the resonant light scattering by bound excitons rather than light absorption. It is remarkable that Landau was able to outline the framework of a rigorous theory for a field where he never worked by himself. I argued that the role of electrodynamics is a minor for weak exciton transitions, and neglecting it simplifies the final results and makes the essence of the phenomenon clear.

Later on, electrodynamics was taken into account by me [6] and different authors (V.I. Sugakov [7]; J.J. Hopfield [8]). About 40 years passed since then, and giant oscillator strength had been identified in a huge amount of papers. For bound excitons, electrodynamics corrections have neither been sought nor observed yet. It was only recently that A.L. Ivanov et al. reported that these corrections have been detected in some biexciton spectra where similar problems are involved [9].

I spoke to Landau only then. Next winter the tragic car accident occurred.

5. Exciton energy bands of molecular crystals

Energy spectrum of elementary excitations is one of the basic problems of solid state physics. Unfortunately, the powerful methods developed for charge carriers are not applicable to neutral particles – molecular excitons. Different experimental techniques were needed. *Imperfections*, such as lattice defects and intramolecular phonons, *turned out to be effective probes for investigating exciton energy bands of perfect molecular crystals*.

The isotopic technique permits one to change, in a well-controlled way, the spectral separation between the bound exciton and free exciton bands. It allowed us to establish an effective criterion for distinguishing exciton and impurity bands in the absorption spectra of molecular crystals. *Only those absorption bands which give their intensity to the isotopic guest bands coming nearer to them are exciton bands*. With this technique, Broude et al. [10] assigned the exciton doublet in naphthalene. The problem was controversial because of a strong anomaly in the polarization ratio of the doublet.

Finding the exciton dispersion law $\varepsilon(\boldsymbol{k})$ and the density of states $\rho(\varepsilon)$ in the exciton band is another important problem. I have shown that $\rho(\varepsilon)$ *can be found from the intensity distribution in the spectra of optical conversion of electronic excitons into vibrational excitons and vice versa*. Such experiments could not be done in Kiev then, but the American group of S.D. Colson et al. measured these spectra for two crystals and for the first time reconstructed $\rho(\varepsilon)$ from experimental data [11]. They coined the term "hot-band spectroscopy" for this technique. We had the pleasure of meeting all this group in Tallinn at the European Congress on Molecular Spectroscopy (1973).

Vibronic spectra correspond to the simultaneous excitation of an exciton and an intramolecular phonon. For crystals with narrow exciton bands, intramolecular phonon can be considered as a stable quasiparticle. As a result, vibronic states can be treated similarly to the problem of the isotopic doping though the theory is actually more involved.

This research allowed us to develop a *unified approach to exciton and vibronic spectra of molecular crystals* and reconstruct the energy spectra for some of them [12].

All work on molecular excitons is closely linked for me with Broude, a brilliant experimenter and a charming person. He designed equipment and performed experiments. Broude was the first to confirm Davydov's theory by establishing the polarization of the exciton triplet in benzene. Broude passed away in 1978 after battling cancer for two years. Half a year before his death the impossible happened. He was given permission to go abroad for the first time in his life, and to the US. He was already very exhausted, but such a chance could not be lost! He crossed the US from coast to coast and visited a number of labs where he knew people from their work and where people knew of him. American colleagues took touching care of him. I remember him, excited and enthusiastic, giving his seminar about that trip. It was his last seminar appearance.

6. Self-trapping of excitons

This Conference is an appropriate place to pay tribute to S.I. Pekar. His seminal paper [13] on the adiabatic polaron strongly influenced the following development of the theory of self-trapping phenomena. *It was the very first paper on polarons*, and the term *polaron* was proposed in it. Pekar's work on multi-phonon optical transitions [14] and his paper [15] on exciton polaritons and additional light waves which originated the field are also closely related to the scope of this Conference. He had been my teacher whom I admired.

My first research was on self-trapping of excitons. It was focused on the problem which sounded as heretic then: *can free and self-trapped excitons coexist*? My theory [16] showed that they can coexist in three dimensions (3D), i.e., in crystals, but not in an isolated polymer chain. Free and self-trapped states are separated by a barrier. These statements are non-trivial. When coupling to polar phonons is strong, self-trapping of electrons proceeds by monotonic lowering of their energy, and I believe that under these conditions the free electron states even do not exist in a true mathematical sense.

It took about 20 years before the coexistence of free and self-trapped excitons was demonstrated experimentally. It was a matter of growing crystals perfect enough to suppress trapping of metastable free excitons by defects. The success was achieved in mid 70s for rare gas solids by J.M. Debever et al. [17] in France, I. Fugol' and her group [18] in Kharkov, and G. Zimmerer and his collaborators [19] in Hamburg, and for alkali halides by Ch.B. Lushchik and his group [20] in Estonia. For molecular crystals the coexistence was discovered by A. Matsui and H. Nishimura [21] in Japan.

Observation of coexistence posed exciting theoretical problems. The large self-trapping energy and the atomic size of self-trapped states in rare gas solids and alkali halides implied that the typical height of the barrier is small compared to the atomic scale and, therefore, its spatial extent is large compared to the atomic spacing. Hence, the continuum approximation could be applied and *escapes from free states* (the "false vacuum") *can be described by some field theories*. I worked on this problem with S.V. Iordanskii and A.S. Ioselevich from the Landau Institute. We investigated collective tunneling of the exciton–phonon system through the barrier, the role of the Arrhenius activation process, the magnitude of the pre-exponential factor which turned out to be large, and its temperature dependence (reviewed in Ref. [22]). In particular, self-trapping of hot excitons produced by light was a challenging problem. The initial energy of the hot exciton increases the self-trapping rate only if it is coherently transferred to the lattice, but the probability of this transfer is small because of low lattice frequencies. Nevertheless, this process can produce *a bypass flow of hot excitons into the self-trapped states prior to the exciton* *thermalization*. It was observed in pyrene by A. Matsui and his group [23].

Existence of two types of self-trapped excitons in rare gas solids, quasi-atomic and quasi-molecular with a broken symmetry [24], posed another theoretical problem: *does the relaxation path to the molecular states pass through higher-energy atomic states*? With my student F.V. Kusmartsev we showed by symmetry arguments [25] that, because of the degeneracy of the valence band, the barrier for self-trapping into broken-symmetry states is lower than for symmetric ones. So, the bypass of atomic states is rather probable.

Self-trapping is accompanied by large energy transfer from an exciton to the lattice. We considered possible mechanisms of the defect production [26] and sputtering [27] resulting from transferring this energy to only a few lattice degrees of freedom.

During this work we collaborated closely with experimenters in the USSR, and after Perestroika has begun I also enjoyed meeting a number of active workers in this field in Germany, France, Japan, Israel, and around the world.

Self-trapping in lower dimensions is rather different than in 3D. E.g., large self-trapped stated are possible in 1D for non-polar coupling (Rashba [16]; T. Holstein [28]), while in 3D they always have atomic scale. However, I will not comment on this subject in more detail here.

7. L.D. Landau Institute for Theoretical Physics

I worked in the Landau Institute since 1966 when it was established. The Institute is internationally known. Many people asked me about the Institute and the origin of its success. I feel it is appropriate to discuss this subject here briefly.

After the car accident in early 1962 when Landau had to stop working, four of his former students decided to organize a new Institute. It was a way to transfer the Landau tradition to the next generation. The Academy of Sciences supported the project since it required no investment of money. Originally the Institute had about 12 scientists. Most of them were about 35 years old at that time. Later on some more people joined us.

The Institute consisted of two branches, in Moscow and in Chernogolovka, a campus near Moscow. In Chernogolovka we used a small building of 9 rooms which was granted to us by Nikolai N. Semenov, Director of the Institute of Chemical Physics and Nobel Laureate. It was designed for some offices of his Institute like the Party Committee. Semenov made a brave and right decision! Usually we worked at home and came to the Institute for discussions and meet visitors and our students. For our regular seminars we used the conference hall of the Institute for Solid State Physics. Unfortunately, Chernogolovka was closed to foreigners. First of the foreigners came here only in 1988. In order to interact with foreigners we used a single room which we had at our disposal, with their kind permission, in the Kapitsa Institute of Physical Problems in Moscow.

To be in touch with the external world, we subscribed *Physical Review* and *Physical Review Letters* for our library using our own money. To maintain contact with theorists from around the USSR, small informal conferences were called every two or three years, in the outskirts of Odessa on the Black Sea. They were called "Odessa"s. There was no preliminary Program and Proceedings, but there were free and heated discussions. People came with their new papers and presented them on the Sessions which usually were held in the afternoon. In the morning one could see along the beach picturesque small and large groups of people arguing, discussing science, and performing calculations.

Were there any special secrets of the success of the Landau Institute? All of us were convinced that *the professional competence was the critical issue for the success of the new Institute.* The problem was how to achieve this goal living inside the System which was corrupt and professionally incompetent. To reduce the damaging effect of the System on our work, we established an internal procedure of unofficial secret vote which was binding for one year. This vote of the "Directorate" reduced the pressure on the individual members of the Institute by sharing responsibility in important and sensible issues. The vote was taken after a careful scientific hearing, but preceded any official process. The two most important issues were the admission of new scientists to the Institute and awarding the Doctor of Science degrees. It was the highest scientific degree in the USSR and we felt it a high responsibility on the part of the Institute in awarding it.

The same criterion was applied to our students from the Moscow Institute for Physics and Technology. We had our student group there, and many of us worked there part-time. The new generations of the Landau Institute were our main success. Many of these people are now internationally known for their scientific achievements.

8. Strong magnetic fields and the Quantum Hall Effect

Strong magnetic field enhances the effect of the Coulomb interaction on optical spectra. Excitons turn into magnetoexcitons. Their binding energy and oscillator strength increase (R.J. Elliott and R. Loudon [29]). In 3D, for appropriate directions of the field, hyperbolic excitons arise near the saddle points of the energy spectrum [30]. In 2D the effect is most fundamental since new electronic phases appear.

Many problems of the spectroscopy of interacting 2D electrons in a strong magnetic field can be treated in terms of excitons, especially under the conditions of the Quantum Hall Effect, i.e., for integer values of the filling factor ν. Filling factor measures the concentration of electrons in units of the multiplicity of Landau levels.

For integer values of ν, optical properties of strongly pumped quantum wells in a strong magnetic field can be described in terms of *magnetic excitons and deexcitons, elementary excitations of a neutral plasma with population inversion*. Deexcitons are bound states of a "hole" at a filled Landau level in the conduction band and an electron at an empty Landau level in the valence band. These excitations are stable at the Fermi edge but decay away from it. The theory (Yu.A. Bychkov and Rashba [31]) takes consistently into account the Coulomb interaction and establishes connection between the absorption and emission spectra. The restriction on ν can be relaxed [32] because of the hidden symmetry inherent in 2D electron–hole systems in a strong field. Our theory has been initiated by the data of J.C. Maan and his collaborators (Grenoble) and compared with the data of L.V. Butov and V.D. Kulakovskii from Chernogolovka [33]. It was then that I felt that we were gradually becoming a part of the international community.

The Fractional Quantum Hall Effect (FQHE) in semiconductors is one of the most fundamental problems of the modern solid state physics. In a strong magnetic field the system of interacting 2D electrons acquires unique properties. At some values of ν, electrons form incompressible quantum liquids. In an incompressible state, a gap Δ opens in the spectrum and new elementary excitations appear. These excitations, quasielectrons and quasiholes, carry fractional charges. For example, for $\nu = \frac{1}{3}$, quasiparticles carry charges equal to $\pm\frac{1}{3}$ of the elementary charge. They obey fractional statistics and therefore are called *anyons*; they are neither bosons nor fermions. Under these conditions, excitons are *anyon excitons* – they consist of *a hole and three quasielectrons*. Optical investigation of this state is challenging.

I.V. Kukushkin and V.B. Timofeev from Chernogolovka discovered that the position, $\omega = \omega(\nu)$, of the emission band originating from the radiative recombination of quasielectrons on neutral acceptors, shows some kind of singularities when ν passes through quantized values, say $\nu = \frac{1}{3}$. The research was continued in Stuttgart with K. von Klitzing and his group. This success raised a theoretical problem: *can the gap Δ be found from optical data*? My student Vadim Apal'kov and I showed [34] *that the singularities are cusps of the square root type*, *and that the cusp strength depends on Δ* and some parameters of the heterojunction. The theory suggested a way to find the gaps from the experimental data. The proposed method was checked and used in later research [35].

Another puzzle came from intrinsic emission spectra investigated by D. Heiman et al. [36]. They found doublets and some different features in these spectra. These data stimulated research on anyon excitons. Since 1992, I found my new home in the friendly and professional environment of the Physics Department of the University of Utah in Salt Lake City. Here, in collaboration with Vadim Apal'kov, Fedor Pikus, and Michael Portnoi, young researchers from Russia, I developed a theory of anyon excitons [37,38]. To my best knowledge, the puzzle of the doublets still remains unsolved and our theory has no experimental confirmation. The intrinsic spectroscopy of the FQHE is still not understood. However, I really hope that anyon excitons exist and will be discovered.

9. Which problems I missed?

The list of the problems which I discussed above is rather long. Nevertheless, it makes only a small part of the contemporary physics of excitons. I realized how fast it grew when we worked with Michael Sturge about 20 years ago on editing the Volume "Excitons" [39], a collection of review papers. My recollections on this collaboration should appear in Sturge's *Festschrift* [40].

So, what I missed? First of all, collective properties of excitons of which most prominent are exciton (electron–hole) liquids and Bose condensation. Nonlinear spectroscopy of excitons and fast transients should also be mentioned. I feel, that the central theoretical problem in fast transients is the interference of bound and free states and considered it recently [41] as applied to some simple models. There are also polaritons in different dimensions and other problems.

However, I am not much upset that I missed these problems. I have never considered the spectroscopy of excitons to be my sole occupation. I tried many times to run away from it to learn about different problems and contribute to them. I am happy that I had a chance to work on semiconductor devices, spin–orbit coupling, organic metals, size effects, etc. It provided me with a wider outlook on our science. I came back to the physics of excitons because of irresistible attraction of new experimental findings, and the work with experimenters was always rewarding for me. My theoretical work often included difficult analytical calculations. Nevertheless, I tried to complete it with simple final formulae and models uncovering the underlying physics.

If this paper reflects my excitement by experiment, I have fulfilled my task.

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