



Molecular Crystals and Liquid Crystals

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Vladimir L'vovich Broude (1924–1978)

Indeed it is extremely difficult for us to write about Vladimir L'vovich in the past tense, for us who worked with him and knew him well. We sensed his kind influence, as a man, and as a scientist devoted to science, and his inexhaustible vitality and enthusiasm infected us.

“Vladimir L'vovich Broude's way to science,” relates Professor A. F. Prihot'ko, “started in one of the days in 1947, when two young engineers, Vladimir L'vovich Broude and Vladimir Sergeevich Medvedev, having graduated from the Moscow Institute of Chemical Engineering, where they specialized in cryogenics, came to the Kiev Institute of Physics. The task set before them was to design and organize the cryogenics laboratory. They became members of our then rather diminutive team immediately and not only established the cryogenics laboratory, but also designed a new system for low temperature spectral studies that enormously contributed to the success of the research on crystal spectroscopy. Later, they got thoroughly involved in different fields: V. S. Medvedev in cryogenic engineering, his cryostats are well known by everyone, and Vladimir L'vovich in the spectroscopy experiments. Without specific education in advanced physics and mathematics, he fast became a genuine physicist, well educated, and in command of the theory, while making the best use of his engineering knowledge. His disposition, concern, his candid interest in everything around, always drew him to the focus of any event. He enlivened our department by heated argumentation, humor, fascination, intolerance of any injustice, and infectious enthusiasm. Regarding his experimental results extremely rigorously and being notable for his keenness of observation, Vladimir L'vovich was able to recognize something new where someone else might miss it. Avoiding conventional standards of thinking and conservatism in experiment, he was able to approach results from new and original points of view. Such was the case regarding the deformation effects in crystal spectra, the elucidation of the importance of defects and imperfections in crystals, as well as with excitons in mixed crystals. Discovering a new phenomenon, he always demonstrated it joyfully, as was the case, for instance, with the alter-

nating generation of light on the components of the ruby doublet at the very outset of the laser spectroscopy.”

Vladimir L’vovich Broude started his scientific activities in 1947 at the Institute of Physics of the Ukraine Academy of Sciences by redesigning the experimental techniques. The outcome of the activities counted sufficiently fast with the first paper in 1950 on the spectra of organic crystals at low temperatures. Inconvenient glass and quartz Dewar vessels were replaced by the metallic cryostats with glass or quartz windows. In the new cryostats, metallic and glue conductors were eliminated and the sample was placed in the cold vapor contiguous with the liquid. The microprojector was invented, which was placed with the crystal into the cryostat. (V. L. Broude, V. S. Medvedev, 1951). This solved the problem of making samples, for it enabled one to isolate a single crystal from a polycrystalline sample. Later on the microprojector made possible polarization measurements on substances which are liquid at room temperature.

While working, as an engineer, at inventing the cryostat, Vladimir L’vovich shifted his interest more and more towards the low temperature spectroscopy itself. However, he participated in the design of various cryostats for spectral studies at 20.4 K and 4.3 K (1956, 1958). During his final years of life, while suffering from a serious illness, he built up a large organization for designing and making thermostats and cryostats of large scale use in the Institute of Solid State Physics of the Academy of Sciences of the U.S.S.R.

His first scientific paper in 1950 on nature of coloring of solutions of metals in liquid ammonia marked the transition from the technician-engineer to the physicist-experimenter. In this work ammonia coloring was obtained by injection of electrons from a point electrode. In this way polaron coloring in metal-ammonia solutions was proven experimentally by an independent and convincing method, revealing Vladimir L’vovich’s unique insight in scientific investigation.

His first paper on the spectroscopy of molecular crystals appeared a year later (1951) and soon became a classic work. The success, undoubtedly, was due to Vladimir L’vovich’s starting his activities, as a physicist, in one of the world best experimental schools on spectroscopy of solids founded by I. V. Obreimov and A. F. Prikhod’ko. I. V. Obreimov and his colleagues were pioneers in the systematic investigation of splitting broaden spectra of absorption and luminescence of complex organic compounds at low temperatures. Ya. I. Frenkel appealed to the first papers of that series, performed in the late 1920’s, in his classical paper on excitons (1931).

Improving the experimental technique, A. F. Prikhod’ko obtained the classic absorption spectrum of a super thin naphthalene crystal in polarized light and discovered “crystal” bands having sharp polarization along principal crystallographic axes (1944). These achievements gave rise to a new

approach to the theory of light absorption in molecular crystals: Davydov's theory of exciton multiplets (Davydov splitting, 1948).

Vladimir L'vovich joined molecular crystal spectroscopy on the eve of the next phase, when the interdependence between fundamental experimental results and the theoretical ideas was to be determined, and excitons were to be applied as a working model in the interpretation of spectra. It was in his first spectroscopy paper on electron levels of a molecule and a benzene crystal that the identity of "crystal" bands and exciton multiplets were proven experimentally for the first time (with V. S. Medvedev and A. F. Prikhot'ko, 1951). This paper paved the way for the further extensive experimental investigation of exciton spectra of molecular crystals.

Vladimir L'vovich returned to the benzene crystal spectrum several times during his scientific activities, making its assignment more and more accurate. These results can be found in his review (1961) and his monograph (1965).

Intensive investigations of the subsequent years (1950–1960) allowed Vladimir L'vovich and his collaborators (G. V. Klimusheva and M. I. Onoprienko) to accumulate and analyze a vast amount of experimental material on crystal spectra of benzene homologues, their change at phase transitions, deformations, and so on. Systematization of the material resulted in a two-volume monograph (1965, 1967). These achievements, with the early investigations of I. V. Obreimov and A. F. Prikhot'ko, form the background of our notions on the general structure of exciton spectra.

In the course of these investigations a new range of problems was gradually formulated: a clear-cut and unambiguous assignment of an exciton band in a spectrum, and determination of the genesis and structure of the exciton bands. Solving these problems involved working out a new experimental approach. In this way the spectroscopy of isotopic crystals and of isotopic mixed crystals was developed on the basis of deuterated molecules of benzene and naphthalene. The basic facts underlying them have been found experimentally in investigations of V. L. Broude and his pupils (M. I. Onoprienko, S. M. Kochubey, E. F. Sheka).

Vladimir L'vovich gave attention to the spectra of deuterocompounds for the first time in 1958 while studying electron spectra of some aromatic hydrocarbons (1958). Since 1961 he has started a systematic study of isotopic mixtures which is going on in the molecular crystal spectroscopy till now. One of the principal landmarks in the way of development of this field has become the discovery of impurity exciton bands in spectra of concentrated isotopic solutions of deuterobenzenes (1961). In that investigation, a few exciton multiplets equal to the number of the mixture components was observed for the first time. Studying the spectra of mixed crystals as a function of component concentration allowed V. L. Broude and E. I. Rashba to advance a simple and clear-cut physical model of the observed phenomenon and enabled

them to trace immediately the development of the exciton multiplet from the impurity band. This very fact helped to ascertain the genesis of the components of the exciton multiplet from a particular band in the spectrum of the molecule (1961, 1964).

Later a large intensity change in the impurity band was observed, now known as the Rashba effect (1961), which permitted one to identify the exciton bands unambiguously, to determine boundaries of the exciton bands, and so on. It is due to these investigations that the exciton bands in molecular crystals have become the same "experimental reality" as the electron bands in metals and semiconductors.

During the same period (1957–1965), the interests of Vladimir L'vovich Broude were also focused on the low temperature crystal luminescence. He with his pupil V. V. Feremenko investigated exciton effects in CdS. This was followed by a series of investigations showing, in particular, that a majority of narrow bands near the edge absorption and luminescence of CdS are due to excitons localized on impurities (1957). The generalization of his own experience in studying the luminescence of CdS crystals and the analysis of the data available in literature by that time, resulted in the review written in cooperation with A. F. Prikhot'ko and E. I. Rashba (1959) on principal properties of the exciton luminescence of crystals. It has become a manual for the experiments on crystal luminescence and their analysis.

In further investigations V. L. Broude with his collaborators, M. T. Shpak and E. F. Sheka, for the first time paid attention to the fact that the temperature dependence of the shape of the band-to-band transitions in the spectrum of the exciton luminescence can be used to determine the location of the exciton bands edges, their widths, and gaps between them (1963). A paper in 1965 suggests that such an important parameter as the degree of exciton localization on the impurity site should be determined quantitatively by studying the doublet structure of the vibronic spectra of luminescence. The methods of studying the exciton band structure have been given a wide-scale development which was reviewed by V. L. Broude and E. I. Rashba (1974).

The investigations, performed by Vladimir L'vovich Broude during 1951–1965, made up a considerable portion of the research on exciton spectroscopy, for which, in 1966, a team of Soviet physicists was awarded the highest reward of this country—the Lenin Prize.

Soon after the invention of first optical quantum generators Vladimir L'vovich ardently started to study physical processes in them. Having united some enthusiasts of the new field, he founded the quantum electronics laboratory in the Institute of Physics of the Ukraine Academy of Sciences in 1963. His ideas for using molecular crystals, displaying a rich spectrum of transitions, for tunable lasers date from that time (1962), and also for designing lasers with a dispersive cavity (1965, 1967).

In 1966 Vladimir L'vovich was offered a job at Chernogolovka in the new Institute of Solid State Physics of the Academy of Sciences of the U.S.S.R. He left Kiev for Chernogolovka almost at the same time as his friend E. I. Rashba, who had been offered a job a little bit earlier at the established L. D. Landau Institute for Theoretical Physics. Their friendship sprang up in the middle of the 1950's, due to their common scientific interests in the spectroscopy of molecular crystals. An experimenter and a theorist, they complemented each other, which favored the development of new scientific ideas that were both realistic and original, simultaneously. Their friendship and collaboration were of great importance and of essential significance for the success of the whole program dealing with the techniques for studying exciton band structure.

In Chernogolovka Vladimir L'vovich established the laboratory of optics and spectroscopy for research in some fields of crystal spectroscopy. At that time his interests were extraordinarily diversified. He faced the analysis of experimental results on the basis of the dynamic approach to the vibronic spectra of crystals. This enabled one to describe both the exciton and vibronic spectra of pure and doped crystals (1966). He was interested in holography and in the mechanism of photopolymerization of crystal monomers. He once again returned to the absorption spectra of mixed crystals on deuterio-naphthalenes, obtaining the first quantitative data on absorption coefficients, and also to the analysis of the cluster structure (1971). His interests then shifted to exciton-phonon (1974) and phonon-phonon interactions (1976). He was involved in electron and phonon spectroscopy of liquid crystals and raised the problem of studying the exciton bands of molecular crystals by inelastic neutron scattering.

However, Vladimir L'vovich's main personal scientific interest during the final decade of his life was associated with the spectroscopy of molecular excitons at high levels of optical pumping. The object of the investigation was the anthracene crystal. The first experiments on studying the quantum yield of luminescence of the anthracene crystal were performed with V. S. Medvedev in as early as 1959–1960, then the investigations followed on stimulated luminescence of anthracene with intense pumping performed in Kiev (1964, 1966).

After moving to Chernogolovka Vladimir L'vovich devoted a lot of time to building up an experimental installation for time-resolved spectroscopy. The installation allowed for either a time scan at a fixed wave length or a spectral scan at a given time point in conjunction with a powerful nanosecond pumping pulse. The effects were crowned with success. The pioneering results have been recently obtained by studying the luminescence kinetics of the anthracene crystal at high pumping. Vladimir L'vovich and his collaborators (V. V. Korshunov, I. I. Tartakovskii) as well as his colleagues from the

Institute of Chemical Physics of the Academy of Sciences of the U.S.S.R. (V. A. Benderskii, V. Kh. Brikenshtein, and others) were involved in the experiment. The light generation by excitons in pure and doped anthracene crystals (1973–1974), a local biexciton in the vicinity of defect molecules (1977), and stimulated Raman scattering under resonance conditions (1977) have been observed.

When investigating optical properties of highly pumped anthracene crystals, it was found that the non-equilibrium of the phonon system is of great importance. A number of effects have been observed due to phonon pulse propagation, in particular, the oscillation of the “effective” temperature (1974, 1977). The analysis of these experiments gave rise to a new viewpoint on the propagation of phonon nonequilibrium (1978). The theory of this phenomenon has been suggested by I. B. Levinson.

All the investigations were focused on revealing the exciton-exciton interaction at high concentration. Vladimir L’vovich and his colleagues succeeded in discovering a new long-wave band in the luminescence spectrum of the highly pumped anthracene crystal which may indicate the formation of the dense phase of the molecular excitons.

Death brought to an end the ardent activities of Vladimir L’vovich in the prime of his creative strength.

The contribution made by Vladimir L’vovich to science is not only his personal achievements. Being a man of broad scientific interests, and of exceptional benevolence and charm, he easily got in touch with absolutely different people. He readily imparted his ideas and shared his experience and advice both with his colleagues and young scientists. He brought up many disciples, for whom his ardent enthusiasm for science, and inexhaustible interest for everything new, remains. Vladimir L’vovich continued to work actively with a surprising fortitude up to the end of his life while suffering from a serious illness, and while being aware of his incurable disease.

I would like to finish this sketch with the excellent words said by Antonina Fedorovna Prikhot’ko, who was always a sincere friend, preceptress and adviseress of Vladimir L’vovich Broude. She said, “Surrendering himself wholly to his cause, Vladimir L’vovich took delight from a beautifully performed experiment and a correctly comprehended phenomenon. And it made his life capital, in spite of all the worldly hardships. His scientific generosity increased the number of his ideas, and therefore a trace of him will never become lost. Vladimir L’vovich will always be alive for us—in his ideas, papers, thoughts, aspirations.”

Elena Sheka

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