# BRILLIANT START AND WONDERFUL CONTINUATION (COLLEAGUE'S MEMOIRS)

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Time cannot be stopped. Here is Mykhailo Semenovych joining the club of 80-year-old persons working at our native Institute of Physics of the National Academy of Sciences of Ukraine. This event stimulated me to write down these short memoirs, because we started our scientific ways at approximately the same time. I remember the time after my entering the postgraduate study, when our research supervisor, Academician Antonina Fedorivna Prikhotko, suggested me to research the absorption and dispersion spectra of naphthalene and 1,2-benzanthracene crystals in a wide temperature interval of 20–300 K and to analyze the relationship between them. I lodged in the same room, where M.S. Brodyn successfully studied exciton absorption and dispersion in anthracene crystals, as well as other crystals of the same family.

M.S. Soskin demonstrates the unique low-temperature interferometer for studying the crystal dispersion at low temperatures to the Nobel Prize winner Academician P.L. Kapitsa (Kyiv, 1971)

Metallic cryostats developed for the first time at the Department of Crystal Physics by V.P. Babenko, a jack of all trades, and V.S. Medvedev, who arrived from Moscow, were the best cryostats in the world at that time. They were supplied with special quartz windows to carry out measurements within the whole optical spectral range. Dispersion was measured on a miniature low-temperature Jamin interferometer, which had been developed by I.V. Obreimov, Academician of AS of the USSR, the first Director of the Ukrainian Physico-Technical Institute at Kharkiv. The interferometer could be completely disposed in a wide-neck cryostat. In Fig. 1, the legendary Academician P.L. Kapitsa, who visited our institute, when participating in the Kiev Low-Temperature Conference, examines this cryostat with a great interest (before him stands his son S.P. Kapitsa, the creator and the permanent presenter of the TV program "Ochevidnoe – neveroyatnoe" (Evident, but incredible), which he has been running for 40 years).

In the course of researches, M.S. Brodyn and I proposed some improvements for the optical channel of a cryostat, which were necessary for the quantitative measurements of the crystal dispersion to be more convenient. Those propositions were implemented by V.P. Babenko. This design was described in our common publication in the foreign journal (V.P. Babenko, M.S. Brodyn, and M.S. Soskin, A cryostat for dispersion measurements at low temperatures, Cryogenics **2**, 365 (1962)). In those years, it was a very rare event of publishing a work in a foreign journal (below, I shall tell more about the accompanying problems). For this reason, the results obtained by Soviet scientists were almost unknown to the world scientific community; in particular, it was true for a cycle of works on molecular crystals.

Conditions for carrying out researches in a separate room were remarkable, and the clothing style was informal, the latter being especially important, because the summer was hot that year.

It is known that, in classical electrodynamics, there are integral Kramers–Kronig relations between the real, $\varepsilon'(\omega)$, and imaginary, $\varepsilon''(\omega)$, parts of the dielectric permittivity function $\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)$, which determine the refractive and absorption indices (the optical constants) of the given medium. The relations look like

$$\varepsilon'(\omega) - 1 = \frac{1}{\pi}\text{v.p.}\int\limits_{-\infty}^{+\infty} \frac{\varepsilon''(x)}{x-\omega}dx, \tag{1a}$$

$$\varepsilon''(\omega) = \frac{1}{\pi}\text{v.p.}\int\limits_{-\infty}^{+\infty} \frac{\varepsilon'(x)-1}{x-\omega}dx, \tag{1b}$$

where $\omega$ is the frequency, which spans the whole frequency interval, and the notation v.p. means the principal value of the integral. Hence, those indices are not independent of each another. The validity of the Kramers–Kronig relations has been verified repeatedly for various media, in various aggregate states (crystals, liquids, solutions), and at various temperatures. A conclusion has even been drawn that the measurement of either the absorption or dispersion curve only would be enough, because the other curve could be easily *calculated* taking advantage of the Kramers–Kronig relations.

However, it turned out not the case for excitons in crystals. M.S. Brodyn and I managed to demonstrate it for the first time using molecular crystals as an example. For this purpose, the Kramers–Kronig relations were transformed into the form, which explicitly included the refractive index $\mu$ and the integral absorption coefficient $\chi$ of the medium, namely,

$$\mu^2(\omega) - \chi^2(\omega) - 1 = \frac{4}{\pi}\int\limits_{0}^{\infty} \frac{x\,\mu(x)\,\chi(x)}{x^2-\omega^2}dx. \tag{2a}$$

The integral equation (2a) does not allow the dispersion curves to be express explicitly in terms of the absorption spectrum for the corresponding component of the polarized spectrum. Therefore, while making calculations, it is convenient to change from the integral to a sum of Gaussian curves, which include the peak values $A_i$ and the half-widths $\delta_i$ of the absorption bands. Then, as was shown by Prof. E.Y. Rashba (the winner of the "excitonic" Lenin Prize, of which I shall tell more down here), formula (2a) looks like

$$\mu^2(\omega) = 1 + \chi^2(\omega) - \frac{4}{\sqrt{\pi}}\sum_i A_i w\left(\frac{v-v_i}{\delta_i}\right), \tag{2b}$$

where the function

$$w(z) = e^{-z^2}\int\limits_0^z e^{x^2}\,dx \tag{2c}$$

was tabulated, which cardinally facilitated calculations.

First, we tested a crystal of 1,2-benzanthracene. The spread of the dispersion curve at 20 K turned out 4.4±0.9 times larger than that calculated following the Kramers–Kronig relations! As the temperature grew, the discrepancy with the theory decreased and, at last, disappeared at room temperature.

We always shared information with each other concerning the results obtained and discussed them together. M.S. Brodyn addressed the results of measurements presented in his Ph.D. thesis. For our common pleasure, it turned out that, for anthracene, the most challenging molecular crystal, the discrepancy with the Kramers–Kronig relations attained even larger value of $6 \pm 1$! Moreover, even at room temperature, the discrepancy for anthracene amounted to $2 \pm 0.4$. The physics of the revealed violation of the Kramers–Kronig relations is associated with the spatial dispersion of excitons, which has certainly not been taken into account, when deriving those relations in 1926–1927 on the basis on the causality and *locality* principles applied to the dielectric/optic constants of the medium. We showed our results to Academician A.F. Prikhotko, who approved and supported them. As a result, the paper by M.S. Brodyn, M.S. Soskin, and A.F. Prikhotko was publuished (Violation of Kramers–Kronig relations for molecular crystals at various temperatures, Optika i Spektroskopiya **6**, 1 (1959)). They have every reason to consider it as one of landmark works for the physics of excitons in molecular crystals.

At this point, it is worth telling about the publication rules at that time. I know them very well, because 30 years ago I was the chairman of the expert commission at our institute. First, the absence of the subject matter of invention was obligatory. Since the techniques applied in many of our works, especially those dealing with nonlinear optics and quantum electronics, were really original, certificates of invention have to be executed (for instance, I have 49 such certificates). For a work to be published in a foreign journal, it was necessary to prove that it did not contain essentially new results.

Therefore, our work, which, in essence, became a new direct proof of the spatial dispersion of excitons, was first published in the Optika i Spektroskopiya journal. Fortunately, good results find their way, although it may happen slowly. When I checked my citation index last year (including my *h-index,* which equals 27 now), I saw that the paper concerned started to be cited ***half a century*** after its publication (!): the relevant reference data referred to the pages of *domestic* (untranslated) issue of this journal.

The "excitonic epoch" was formally ended by awarding a team of 10 physicists the Lenin Prize (the highest scientific award in the USSR) for the discovery and the research of exciton states in various classes of solids. The Kyiv physicists, Academicians O.S. Davydov and A.F. Prikhotko, composed the core of the team. Some other employees of the Institute of physics, namely, M.S. Brodyn, V.L. Broude, and A.F. Lubchenko, were also awarded. Unfortunately, no room in the list of participants was found for M.T. Shpak, who had discovered and studied excitonic luminescence in molecular crystals.

Certainly, the studies of exciton states in molecular crystals continued for several more years. However, the main contribution had already been made. But here is quantum electronics coming! Last year, the physicists of the whole world widely celebrated the 50-th anniversary since T. Maiman observed the generation in a ruby laser, the event having become a starting point of a laser era. It became clear at once that the epoch of optical quantum electronics came. The leading employees of the Department of crystal physics M.S. Brodyn, V.L. Broude, and M.T. Shpak responded to the challenge of time by engaging themselves with enthusiasm into the researches in this domain. Academician A.F. Prikhotko, who was always sensitive to such challenges, organized a small seminar, where new works devoted to lasers, as well as probable directions of our researches, were discussed; she encouraged such works as much as possible. It was decided that, at the first stage, the implementation of the lasing frequency tuning would be the main task for the Department of crystal physics. A.F. Prikhotko never restrained anybody of employees, if she saw that they started new researches in challenging branches of modern physics. Therefore, in 1965, three new departments were separated out from the Department of crystal physics. These were the Department of Nonlinear Optics (DNO), the Department of Optical Quantum Electronics (DOQE), and the Department of Photoactivity (DPA), under the direction of M.S. Brodyn, V.L. Broude, and M.T. Shpak, respectively. I am sure that the DNO (the Russian abbreviation, ONO, means IT) obtained the most promising name – it was suggested by M.S. Brodyn, – because nonlinear optics will always be challenging, and the life confirms it every day.

The formed groups headed by M.S. Brodyn, V.L. Broude, and M.T. Shpak were transformed in due time into departments, with each of them having selected its specific way. In particular, M.S. Brodyn demonstrated that the generation in homogeneous semiconductors occurs with the participation of the same Wannier–Mott excitons, owing to their radiative annihilation simultaneously with the emission of longitudinal optical phonons, localizations of excitons at impurities, exciton-to-exciton interaction. M.S. Brodyn suggested to use a series of mixed semiconductors belonging to the $A_2B_6$ group, which have a variable energy gap width and, hence, different lasing frequencies, for the jump-like tuning of the optical generation frequency. A collection of such crystals characterized by a high optical quality, which was necessary to obtain lasing, were grown up by the employee of the Institute of Semiconductor Physics of the NASU Mykola Vitrikhovskyi, and the employee of the DNO Violetta Reznichenko carried out necessary experiments, which completely confirmed the proposed idea.

In the Department of Optical Quantum Electronics, dispersion resonators were developed, which enabled a smooth tuning of the lasing frequency within the amplification band of a laser active medium to be carried out. Taking advantage of such a prism dispersion resonator, the frequency tuning of a solid-state laser was executed for the first time. Laser frequency tuning by 500 $\mathrm{cm}^{-1}$ was attained for a neodymium-glass laser. At last, in the Department of Photoactivity, the lasing frequency tuning in lasers based on dyes of new types was studied and implemented.

In 1974, all those works executed at the Institute of Physics of the NASU were awarded the State Prize of Ukraine with the motivation "For the development of physical bases to control the stimulated radiation frequency and the creation of tunable lasers".

It is the common knowledge that science does not mark time. In the 1970s, there emerged the dynamic holography (holography on dynamic gratings recorded in nonlinear media) at the DOQE. Today, this direction is successfully developed by S.G. Odoulov, Corresponding member of the NASU. Lasers on dynamic gratings, which allow the generation of laser beams with the inversion of a wave front, have been proposed and created. Simultaneously, M.S. Brodyn studies processes of laser beam self-action in nonlinear media, such as self-focusing, self-defocusing, and selfdeflection. The methods for measuring the corresponding nonlinearities have been elaborated, and the mechanisms of the discovered self-action have been established.

Simultaneously, Russian (at S.I Vavilov State Optical Institute, St.-Petersburg) and Belarusian (at B.I. Stepanov Institute of Physics of the Academy of Sciences of Belarus, Minsk) scientists were developing new methods aimed at the transformation of a spatial structure of light beams. The cycle of all those works was awarded in 1982 the State Prize of the USSR. Of Ukrainian scientists, M.S.Brodyn, S.G. Odoulov and I were marked. For today, M.S. Brodyn is the laureate of *all* the highest State Prizes of the USSR and Ukraine in science!

M.S. Brodyn successfully continues to develop new directions in nonlinear optics. For instance, the researches of nonlinear effects in island films have been started. Within those years, M.S. Brodyn has trained a number of talented disciples; among them, there are Corresponding member of the NASU I.V. Blonsky, Dr. Sci. in physics and mathematics A.A. Borshch, and others.

At the end of my memoirs, I wish Academician M.S. Brodyn many happy returns of the day and new creative victories!