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On the Seventy Fifth Anniversary of the Birth of S. I. Vavilov

*THE CONTRIBUTION OF ACADEMICIAN S. I. VAVILOV TO THE STUDY OF LIGHT**

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THE greater the distance from which we contemplate the creative path of great scientists, the more clearly do we see the influence which they exerted on the course of scientific development. In our age of rapid development of the natural sciences a fifteen-year period is sufficient to trace the extension and development not only of already established trends and concepts, but of remarks and thoughts, made as if in passing, not yet formulated in the form of clear assertions, which being intuitively fruitful, actually develop, become concrete, and finally grow to be entire scientific fields.

A great scientist always looks into the future, and attempts to see the outlines and ways of development of future science. Vavilov was such a scientist. Being for many years the scientific director of the Optics Institute and being engaged ex officio in the study of all problems of optical science and technology on a country-wide scale, he was in his private scientific interests always faithful to that basic problem to which he devoted his entire scientific life and which he defined by the title of his last monograph—"The Microstructure of Light."

Micro-optics, as understood by S. I. Vavilov, is the study of elementary interaction processes of light and matter. And whereas, as he used to say, classical "macro-optics" can be compared with thermodynamics, the analog of micro-optics is the molecular theory of matter. Micro-optics comprises an analysis of elementary processes of absorption and radiation of light; approaching here spectroscopy in its classical and quantum aspects, it encompasses a circle of phenomena specific for limitingly small powers of the light flux, and analyzes the microscopic mechanism of interference phenomena; finally, it investigates phenomena unknown to macrooptics which attest to the impossibility of considering the elementary light sources to be isolated from the medium in which they are located. The investigation of precisely these phenomena determined by the interaction of the quantized magnetic field of the light wave and the

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matter, treated either as an aggregate of classical operators, or as a quantum mechanical system, constituted the main avenues of development of optics during the last decades.

The fifteen years which have passed since the publication of "The Microstructure of Light," were marked by important events in optics. New trends and fields appeared and old fields, apparently exhausted, and in any case of little promise, took on new life. Turning the pages of Vavilov's "Microstructure," one can see that many of the rapidly developing new trends in optics were anticipated by the perspicacity of the author of these "Investigations and Sketches."

The entire aggregate of micro-optical processes of the interaction of radiation and matter appears with exhausting completeness and diversity in luminescence phenomena, with the investigation of which Vavilov's creative life was most closely connected. Vavilov's interest in luminescence which enables one to penetrate by comparatively simple means into the intimate mechanism of the processes of light absorption and emission, was excited in him still when, as a student working in P. P. Lazarev's laboratory, he carried out an investigation into the photochemistry of dyes. The complex molecules of organic dyes with which he started his investigations of luminescence, turned out to be an extraordinarily suitable object for the study of elementary optical processes. It was precisely because of the complexity of these systems that it turned out to be possible to produce simple classical models so close in their spirit to the style of Vavilov's scientific thought, always exceedingly clear and specific.

Well known is the immense role Vavilov fulfilled in establishing and developing the study of luminescence in this country, and the influence of his work and the work of his school on the development of the corresponding field in world science. Disregarding the purely spectroscopic aspects of the problem, it is sufficient to say that the formulation of basic laws specific to luminescence as a process of transformation of light energy by matter, laws which determine the energetics and kinetics of this process, is connected with his name.

Both during Vavilov's lifetime and during the subsequent years, the extent of the work in the field of luminescence has grown at an ever increasing rate. Whereas, as Vavilov used to say, until the October revolution the physicists, chemists, and engineers who specialized in luminescence in Russia could be counted on the fingers of one hand, and in 1948 there were hundreds of such specialists, at present this army counts thousands of people.

Luminescence became popular not only because it entered into the mode of life of millions of people, together with fluorescent illumination and the television screens, but also because one of the greatest scientific achievements of our time is based on luminescence phenomena. Einstein's idea of stimulated emission found its practical expression in the production of radiation generating devices: masers and lasers. Although, because of the vagaries of the history of science, the practical realization of optical generators of stimulated emission, lasers, came over the radio band, bringing along with it an unknown terminology which is sometimes strange to optics, all the premises for its realization and adequate optical description of the entire range of laser phenomena were available in the study of luminescence and in classical wave optics. It is quite obvious that solid-state lasers could not have been produced if, as a result of spectroscopic and luminescence investigations, the energy schemes of the corresponding systems and the probabilities of the transitions of the systems from one electronic state to another had not been established.

The production of lasers was a powerful stimulus for the development of investigations in luminescence, in particular the luminescence of activated crystals. Luminescence attracted the attention of additional thousands of specialists and hundreds of radiophysicists, and representatives of other specialties far removed from optics began speaking its language.

Vavilov did not live to see the construction of lasers. One can, however, imagine very clearly the sincere enthusiasm with which he would have greeted this closest to him in spirit discovery.

How close Vavilov came to the problem of obtaining coherent radiation from a macroscopic ensemble of elementary particles, one can judge after reading in "The Microstructure of Light":

"Is it impossible to obtain coherent light in the course of a sufficiently long time from two different particles of matter separated by a distance of several particle diameters? Apparently not. If two (or more) such particles are simultaneously in an excited state which lasts long compared with the period of the light oscillations, then there will inevitably appear between them resonance interaction or (in the quantum interpretation) exchange forces. As a result of this, the emission of both particles must become coherent, phase-related. Extremely strong excitation and a luminescent medium providing long-lasting molecular "spontaneous" emission (for example, the uranyl salts. . .) are required for this experimentally."

How close this situation is to that realized in contemporary lasers. However, this was written ten years before the construction of a laser.

Vavilov's name is also indissolubly connected with the birth of such a new field as the optics of "superluminal" velocities. This work of Vavilov's is well known; therefore it is sufficient to say about it a few words. Analyzing the totality of the peculiar properties of the universal emission of pure liquids under the action of the gamma rays of radium, observed by

P. A. Cerenkov in 1933, Vavilov concluded that this emission cannot be treated like common radioluminescence. Decisive for this conclusion was the duration of the emission which, as Vavilov showed, can serve as the only unambiguous criterion for separating luminescence from other forms of nonequilibrium radiation. As is well known, the criterion of duration was subsequently introduced by him into his definition of luminescence. Separating the Cerenkov radiation from luninescence, Vavilov came to the conclusion that a new type of radiation occurs here and he asserted that the emission is not due directly to the gamma rays, but to the fast electrons which appear in the scattering of the gamma rays. Further investigations and, above all, theoretical work of I. E. Tamm and I. M. Frank showed that Cerenkov's radiation accompanies the propagation of electrons in a medium whose velocity exceeds the phase velocity of light in that medium. An elementary theory of Cerenkov radiation which explains above all the peculiar spatial distribution of the radiation field can be constructed by means of a micro-optical analysis of the structure of the interference field of light waves which appear when a superfast electron passes through the medium. On the other hand, as V. L. Ginzburg showed, the peculiarity of Cerenkov radiation can also be explained from considerations about light quanta and from the principle of conservation of momentum. Noting this, Vavilov emphasized that we are not dealing here with two explanations, but that "both interpretations combine into a single one in accordance with the dual corpuscular-wave nature of light."

As early as in the Twenties, Vavilov raised the question concerning the limits of the fulfilment of the superposition principle of light waves. The linearity of the equations of classical optics which follows from this principle appeared to be a firm basis of optics. Violation of the superposition principle should lead to a disturbance of the independent passage of the light flux through space in which other light pulses propagate and should give rise to the "self-scattering" phenomenon of light. In the absence of such "self-scattering," which surprised already Huygens, Lomonosov saw a convincing argument against the corpuscular theory of light, according to which there should have occurred, when intense light beam intersected, a "mixing up of the rays." Lomonosov's opponents—the defenders of the corpuscular theory—sought a way out by assuming the light corpuscles to be extraordinarily small.

The discovery of the quantum nature of light phenomena forced anew the raising of the question concerning the principle of superposition and the linearity of the equations of optics. Although attempts to observe the self-scattering of light have invariably given negative results, the limits of the empirical accuracy of the principle of superposition remained rather undetermined. After convincing himself by his own experiments of the futility of attempts to determine these limits under terrestrial experimental conditions with the most intense sources in existence at that time, Vavilov estimated them from the brightness of the solar corona observed in space, where light beams of enormous intensity intersect. Estimates yielded as an upper limit of the effective cross section of quanta an extraordinarily small value (10^{-40} cm^2), which turned out in subsequent theoretical calculations to be by many orders of magnitude smaller. Now we know that these cross sections are so small that even today's experimental resources do not make it possible to observe the self-scattering of light in space devoid of matter.

A different case is the propagation of light in matter where appreciable violation of the linearity can be observed.

As early as during the development of quantum theory in 1920, Vavilov published the results of experiments checking the independence of the absorption coefficients of matter of the intensity of the transmitted light. These experiments were carried out in an extremely broad range—the light intensity varied by a factor of 10^{20}. At that time, the most interesting region appeared to be the region of limitingly low intensities where according to the hypothesis of M. Planck, the creator of quantum theory, concerning the continuous nature of the absorption and the quantum nature of the emission, one could expect a strong increase of the absorption. The constancy of the absorption coefficient established in Vavilov's experiments in a broad intensity range refuted Planck's "hybrid" hypothesis and attested to the fact that absorption is also of a quantum nature.

Later Vavilov again turned to the problem of the linearity of the absorption, but now in the other limiting case—for very large intensities of the transmitted light. In experiments with a very high intensity of light (a condensed spark) which were carried out in conjunction with V. L. Levshyn in 1925 a 1.5% decrease of the absorption coefficient of uranium glass was successfully observed with a mean error of ± 0.3%. The phenomenon was interpreted as a result of the relatively long duration of the luminescence of uranium glass ($\sim 10^{-4}$ sec) and the possibilities of accumulation of an appreciable fraction of active centers in an excited state connected with it. In the case of the so-called phosphors which have excited states lasting on the order of seconds and longer, appreciable violations of linearity can be readily observed even at comparatively small intensities, these violations being in a number of cases so great that they can serve as the basis of special "absolute" photometers. Such a "nonlinear" photometer constructed at the time in accordance with Vavilov's idea can be considered to be a first realization of a new and original principle of photometry.

The violation of the linearity of the equations of optics in the interaction of radiation with matter made it possible for Vavilov to pose the problem and outline the directions of development of the new trend of "nonlinear" optics which considers critically the constancy of such characteristics of matter as the absorption, dispersion, double refraction, dichroism, etc. As Vavilov wrote in 1944, special "problems of this kind are solved not in a rigorous analytic manner, but with the aid of very crude and primitive simplifications. Sometimes, and even often, this is sufficient, but nevertheless one should already long ago have been able to solve these problems lege artis. One should not forget that the real optics of matter with which we are dealing is in general cases nonlinear and its treatment requires a 'nonlinear' mathematical apparatus."

Thus, fifteen years before the realization of the first lasers, when a broad staging of experimental investigations in the field of nonlinear optics became possible, Vavilov formulated perfectly clearly the necessity of setting up the theoretical premises of such investigations, bearing in mind above all the problems of astrophysics. It was impossible to foresee that the theory of nonlinear optics phenomena would be required in the so near future for terrestrial experiments.

As is well known, in recent years nonlinear optics developed most intensively both in its experimental and theoretical aspects. This development occured under a strong influence of the ideas and methods of the nonlinear theory of oscillations which came into existence somewhat earlier in radio physics. A large contribution to the development of nonlinear optics was due to our compatriots R. V. Khokhlov and S. A. Akhmanov. At present, in addition to these simplest nonlinear phenomena in luminescent systems, of which Vavilov spoke, a large number of phenomena have been observed and predicted. Some of these are of first-class conceptual and applied significance. These are above all the generation of the second and higher harmonics, sum and difference frequencies in the interaction of waves in crystals, optical detection—the change of the polarization constant of a dielectric in the field of a powerful light wave, stimulated Raman and Brillouin scattering, summation of the electron excitation energy of activated crystals, etc. Allowance for nonlinear phenomena is quite essential in analyzing the operation of solid-state lasers, in the study of luminescence phenomena in strongly excited luminors, etc. The scope of investigations in nonlinear optics can be judged even from the fact that its problems are discussed at special symposia at which hundreds of participants gather. Thus there appeared and is developing rapidly before our eyes a new, most promising, trend in physics which originated from Vavilov.

At times the deviations from linearity can be very strong. Thus we succeeded recently in observing instances in which the intensity of the visible luminescence excited as a result of the summation in activated crystals of several infrared quanta, turned out to be proportional to the third power of the intensity of the exciting light.

The last years of Vavilov's scientific activity were marked by his ever increasing interest in the problem of interaction of elementary light sources and the medium. In the usual macro-optical approach it is assumed that to solve problems on the propagation of light in a medium, it is sufficient to characterize the light source by means of its spectrum, intensity, and state of polarization, and specify the constants characterizing the medium—its absorption coefficient, index of refraction, and its optical activity. However, such a description which ignores the mutual coupling of the source and the medium, while suitable for considering processes which take place in the "far field" at distances from the source which exceed the wavelength of the emitted light, turns out in a series of cases to be insufficient. Thus, in considering processes in media which absorb and emit light simultaneously, for example in macroscopic light sources or in luminescent media, in which the emitting molecules are surrounded by other molecules (identical or of a different kind) which are capable of absorbing the emitted light, one must give up the conception that the light source and the medium are separate and go over to a more general conception of the light source and the medium as an integral, organically coupled, entity. Here one must consider not only the "passive" role of far removed absorbing particles which weaken the already formed radiation, but also the "active" participation of absorbing particles, located close to the radiating particle, in the formation of the radiation. This active role of the closely located absorbing particles manifests itself in an inductive resonance coupling which they form with the radiating particles.

The first consequence of setting up of inductive coupling in luminescent systems should be a change in the radiating ability of luminescent molecules and of the absorbing ability of the unexcited ones located near the radiating ones. Unfortunately, the experimental study of these phenomena undertaken by Vavilov during the very last years of his life, which formed the subject of one of his last scientific publications, has not been further developed.

The presence of inductive resonance coupling between molecules in solutions leads also to the fact that when the molecules come sufficiently close to one another, i.e., with a sufficient concentration of dissolved matter, the energy of the electron excitation from the absorbed light quantum becomes transferable from excited to unexcited molecules. This explains a series of peculiar phenomena observed with increasing concentration of the luminescent matter in solutions.

Having developed a theory of such concentration phenomena on the basis of data obtained in studying luminescent solutions, where the inductive resonance occurs between chemically identical molecules, Vavilov showed experimentally that this identity of the molecules is by no means essential for the inductive resonance. The excitation energy can also migrate between molecules of different kinds. The only thing that is essential is the presence of common frequencies of the molecules—a necessary condition for the possibility of resonance interaction.

As numerous investigations of recent years have shown, the introduction of the concept of migration of the electron excitation energy in luminescent media (both amorphous and crystalline) broadens considerably the conceptions of the processes possible in them. Wandering in such a system which can, generally speaking, be a multicomponent system and can contain an aggregate of centers that are both donors and acceptors of energy, the excitation energy can be realized in this or that form depending on the parameters which govern the migration and on the relative probability of the various ways of realization of the migrating energy: spontaneous emission from a donor or acceptor, radiationless conversion into the ground or lower excited states, photochemical processes, etc. During the last few years our knowledge about these processes has broadened; in addition to the simplest phenomena known to Vavilov—quenching and sensitized luminescence—we now have such phenomena as were observed and investigated by N. A. Tolstoĭ and his co-workers, the phenomenon of "nonlinear" quenching connected with the interaction of two excited centers. In our most recent work we observed the phenomenon of summation—accumulation of electron excitation energy in rare-earth activated ionic crystals which is also connected with the energy migration and leads, in particular, to conversion of infrared into visible radiation with a sharp nonlinearity of the coupling between the emitted and exciting light (up to the third power) which we mentioned above. Very close to the ideas developed by Vavilov are the phenomena observed several years ago in the USA, in which two elementary interacting systems were brought into two different excited states by means of a single absorbed quantum. All those and many other related phenomena attest to the birth of another new trend in optics—the optics of elementary interacting systems. Strictly speaking this is no longer optics, more correctly, not only optics, but both molecular physics and physical chemistry; in other words, this is a point of contact of many disciplines, at which, as is well known, discoveries are most probable.

We could touch only on a few basic trends of modern optics, attempting to show that their roots are to some extent connected with the circle of Vavilov's main scientific interests. In certain cases these trends are the direct result of the development of his ideas, in others they were developed under a very strong influence of his ideas. In establishing the generic relationship of these trends with Vavilov's work, we should not forget that in his scientific legacy there are still many insufficiently evaluated statements and remarks. Vavilov's scientific and popular works, his brilliant appearances and speeches, which by rights are a part of the valuable fund of our native and world culture, will undoubtedly also in the future turn out to be a source of fruitful ideas.

Translated by Z. Barnea