

PERSONALIA

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PROFESSOR Sveshnikov, doctor of physical-mathematical sciences and outstanding specialist in the field of luminescence, passed away on September 10, 1962.

Sveshnikov began his scientific career at Moscow University where, after graduating from the physics department, he became a staff member of the Scientific Research Institute of Physics at the University and started working under the guidance of S. I. Vavilov.

At this time Vavilov and his school were particularly concerned with the energetics of luminescence and were investigating the causes of decreases in the luminescence efficiency of solutions. Sveshnikov naturally took part in this work. After an initial study devoted to the luminescence yield and absorption spectra of dye solutions in the range of minimal concentrations, Sveshnikov turned to investigation of quenching of the luminescence of solutions by extraneous impurities that do not absorb in the visible region. At first this work was carried out at Moscow State University, and then in 1933 it was transferred to the State Optical Institute in Leningrad, to which organization Sveshnikov moved at Vavilov's invitation. He continued to be active at this Institute until the end of his life.

Quenching by non-colored impurities occurs only in a case of close approach of the quencher molecule to an excited molecule of the luminescent substance. Consequently, diffusion of the interaction molecules during the lifetime of the excited state plays an important part in this process, for it results in the mutual approach of these molecules; diffusion is particularly significant in low-viscosity media.

From the start of his scientific career, Sveshnikov manifested an ability to grasp the essence of the investigated phenomenon and describe it in mathematical terms. Hence, in addition to rigorous experimentation, Sveshnikov always considered the theoretical aspect of the problem. This valuable combination of skilled experimentation and profound theoretical analysis continued to be characteristic of Sveshnikov in his subsequent investigations of luminescence.

Questions of quenching of luminescence and in particular of development of the diffusion theory of quenching by extraneous impurities became one of Sveshnikov's main interests and continued to hold his attention for nearly thirty years. His candidate of sciences dissertation, defended at the State Optical Institute in 1936, was devoted to this subject.



During these years, Sveshnikov also performed organizational functions, acting as Vavilov's deputy in the Luminescence Laboratory of the State Optical Institute.

Another field in which Sveshnikov was interested was chemiluminescence. In his studies, he elucidated the complex nature of chemiluminescence processes, which differ in different substances but generally involve the appearance of excited molecules at one of the stages of the complicated chemical transformations that occur in chemiluminescing media.

Sveshnikov's third area of interest was investigation of the persistent luminescence of organic compounds. Many of the significant results of his studies were incorporated in his doctoral dissertation, defended at the Physical Institute of the Academy of Sciences in 1951.

After the death of Vavilov, Sveshnikov, his student and close associate, took over the direction of the

Luminescence Laboratory. He devoted much time to guiding the work of young scientists at the Laboratory, many of whom developed into independent researchers.

During this period, Sveshnikov and his collaborators carried out a great many investigations. The theory of luminescence quenching by extraneous impurities was developed further and subjected to exhaustive experimental verification. The role of triplet states in luminescence processes and in energy transfer in solutions was thoroughly studied.

Sveshnikov is the author of 95 scientific papers and reviews, published in leading Soviet journals.

Throughout his working life, Sveshnikov endeavored to utilize the results of his luminescence research for the solution of various practical problems and to promote the application of his research in industry.

Sveshnikov was an excellent colleague, always ready to share his voluminous knowledge and experience with his co-workers.

During the last years of his life Sveshnikov was seriously ill. First he suffered a heart attack, and then it was discovered that he had cancer of the stomach. An operation in December 1960 did not effect a cure but merely delayed his demise. Despite his very poor state of health, he continued to work intensively to the very last, directing the Laboratory and carrying out theoretical investigations, thereby giving proof both of great courage and of his love for science.

INVESTIGATION OF QUENCHING OF THE LUMINESCENCE OF SOLUTIONS BY EXTRANEOUS NON-ABSORBING SUBSTANCES

His series of investigations of quenching of the luminescence of solutions by extraneous non-absorbing substances constitutes one of Sveshnikov's principal contributions to the science of luminescence. Sveshnikov paid particular attention to this problem during his entire scientific life; in fact, the results of his last studies in this field were published only after his death. Sveshnikov's great interest in quenching of luminescence is explained by the fact that this effect is essential to understanding of the basic laws of luminescence; moreover, investigation of quenching is very important for establishing the kinetics of bimolecular chemical reactions in solutions.

The ground work for development of the theory of quenching of the luminescence of solutions by extraneous non-absorbing substances was laid back in the 1920's by S. I. Vavilov. However, the diffusion theory developed by Vavilov was incapable of explaining the experimentally observed nonlinear dependence of quenching on the quencher concentration and on the viscosity of the solvent.

Sveshnikov's candidate dissertation was devoted to elucidating the reasons for this discrepancy. In the course of this work Sveshnikov formulated a variant of the diffusion theory of quenching that differed radi-

cally from the earlier theories in the method of calculating the number of effective encounters (encounters leading to quenching) between the quencher molecules and the excited molecules. Sveshnikov showed that in calculating the diffusion during the lifetime of the excited molecules ($\sim 10^{-8}$ sec) one cannot neglect the nonstationary term in the solution of the corresponding diffusion equations. Taking this term into account made it possible to describe correctly the concentration dependence of quenching. At this stage of its development, however, Sveshnikov's theory still did not supply in rigorous mathematical form an adequate explanation for the often observed sharply nonlinear dependence of the quenching on the fluidity of the solvent; it was, however, supplemented by him by a convincing qualitative characterization of the phenomenon.

Comparing the experimental data on the variation of quenching and the rotational depolarization of the luminescence of solutions, obtained using different methods for varying the viscosity, Sveshnikov showed that the slowing down of the rate of increase of quenching with increasing fluidity of the solvent is connected with decrease of the theoretical parameter characterizing the probability for quenching in the encounter of an excited molecule with a quencher molecule, which probability, in turn, is determined by decrease of the duration of the encounter.

Subsequently, these qualitative inferences were incorporated into the theory by Sveshnikov in rigorous mathematical form; in one of his last investigations, Sveshnikov, while retaining the original approach to the problem, developed a new variant of the diffusion theory that quantitatively explained all the known experimental data regarding the quenching of the luminescence of solutions by extraneous non-absorbing substances, including the viscosity dependence of quenching.

It must be noted, however, that even the first version of Sveshnikov's theory was a significant step forward; even in this version of the theory Sveshnikov not only correctly described the experimental facts but also predicted new effects. Thus, it follows from the theory that the decay law for quenched solutions must be nonexponential even when the decay of the luminescence of the original unquenched solution is exponential.

Experimental proof of this important inference would be not only a decisive substantiation of the validity of the basic premises of the diffusion theory but also of the fundamental formulas of Smoluchowski, characterizing nonstationary Brownian motion, upon which the diffusion theory was based. In the 1930's—during the period of development of the theory—experimental techniques were not adequate for detecting this very minor deviation from exponential decay, amounting to only a few nanoseconds. Some twenty years later there were constructed in Sveshnikov's laboratory, under the supervision of A. M. Bonch-Bruevich, spe-

cial large aperture precision fluorometers. Recently, at the suggestion of Sveshnikov, some of his students developed a fluorometer with an ultrahigh modulation frequency for the specific purpose of investigating the decay laws of nanosecond luminescence.

By virtue of these developments fluorometric methods have been accepted in laboratory practice for investigating the luminescence of organic luminophors. By direct fluorometric measurements at different frequencies of modulation of the exciting light Sveshnikov and his co-workers proved that the decay law for quenched solutions is actually nonexponential and that the character of the deviation is not only in qualitative but also in good quantitative agreement with the diffusion theory.

Many years after creation of the diffusion theory, Schmüller, and subsequently Sveshnikov himself, observed nonproportionality between the decrease in efficiency and persistence of luminescence incident to quenching of the luminescence of viscous solutions by extraneous non-absorbing substances. Sveshnikov showed that this result follows directly from his theory—with no further assumptions—and, like the nonexponential decay law, is due to the dependence of the quenching probability on the time elapsed from the instant of excitation.

In conjunction with detailed analysis of the effect, Sveshnikov called attention to the necessity in the case of nonexponential decay to distinguish clearly between the mean persistence of luminescence and the mean lifetime of the excited state, a distinction that many earlier investigators failed to take into account.

The method developed by Sveshnikov for calculating the number of effective encounters is also of great interest from the standpoint of the theory of bimolecular chemical reactions in solutions. In the middle 1930's, when the theory of diffusion quenching was being developed, chemists were generally of the opinion that the number of effective (leading to reaction) collisions is proportional to the total number of molecular encounters between the molecules of the dissolved substances and in the first approximation does not depend on the viscosity of the solvent. Sveshnikov showed, however, that the independence of the reaction rate of the viscosity of the solvent, following from this assumption, is observed only in the extreme case when the probability for reaction incident to one encounter is small. In the other extreme case, when the probability for reaction in an encounter is 100 per cent, the number of effective collisions is simply equal to the number of encounters and the reaction rate is proportional to the fluidity of the solvent.

Sveshnikov's method for calculating the number of effective encounters between reacting molecules, based on application of diffusion laws, since the end of the 1930's, has gained general acceptance and is commonly used in investigating the kinetics of chemical reactions in solutions.

The development of quenching due to increase in the concentration of the luminescing substance is much more complicated than the development of quenching under the influence of extraneous impurities. In the 1930's Sveshnikov, in collaboration with P. P. Feofilov carried out an investigation of concentration quenching and concentration induced depolarization of fluorescence; this investigation is still one of the most comprehensive experimental studies of this effect. On the basis of the experimental data Sveshnikov and Feofilov proposed a formula that established the dependence of concentration depolarization on the persistence of luminescence of dyes (in the experiments the persistence was varied by adding extraneous non-absorbing substances). The results of this study are still used as an experimental criterion for checking theoretical constructs. Subsequently, S. I. Vavilov used these experimental results in developing his theory of concentration-induced depolarization.

INVESTIGATION OF THE PHOSPHORESCENCE OF ORGANIC COMPOUNDS AND THE KINETICS OF INTRAMOLECULAR TRANSITIONS IN SOLID SOLUTIONS OF ORGANIC LUMINOPHORS

A second important group of studies carried out by Sveshnikov is constituted by his investigations of the phosphorescence of organic compounds. In 1945, when Sveshnikov initiated his studies of this interesting and important field of luminescence, it was impossible to have a clear physical picture of the effect owing to the lack of adequate experimental data.

Sveshnikov carried out a large number of significant experimental investigations of this process, and as the basis for creating a mathematical theory of the effect he used the diagram of three molecular levels proposed by A. Jablonski and the then recent interpretation by A. N. Terenin of the phosphorescence metastable level as a triplet state. At the time, experimental data for verifying this hypothesis were lacking; there were also extant other hypotheses, for example, the Franck-Livingston hypothesis that the molecule in the metastable state is a tautomer of the unexcited molecule. It was a widely held opinion that the fluorescence and phosphorescence of organic solutions can be emitted by different centers corresponding to different types of interaction of the luminescent substance with the rigid solvent. Consequently, Sveshnikov's first studies in this field were devoted to proving the identity of fluorescence and phosphorescence centers by investigating the dependences of the relative yield of both forms of emission on the intensity of the exciting light. The results of Sveshnikov's studies substantiated the hypothesis regarding the triplet nature of the metastable state.

Later, Sveshnikov in collaboration with P. P. Dikun carried out an analysis of the vibrational structure of the phosphorescence spectra of benzene and its deriva-

tives, and showed that the phosphorescent state is one of the electronic states of the molecule. In further studies, Sveshnikov investigated the influence of the medium on the persistence of phosphorescence. These investigations also showed that the phosphorescence processes occur primarily in the activator molecule and that the medium plays an auxiliary if sometimes significant role. Final substantiation of these concepts follows from the experiments of P. P. Dikun, Sveshnikov's student, in which he detected and investigated the phosphorescence of some aromatic compounds in the vapor state.

The next important stage in Sveshnikov's series of investigations of phosphorescence was study of the modes of population of the metastable triplet state. Investigations of the variation of the ratio of the phosphorescence and fluorescence yields as a function of the wavelength of the exciting light and comparison of concentration induced depolarization of fluorescence and phosphorescence showed that the population of the triplet state is realized via the fluorescent state, i.e., the lower excited singlet state into which the molecule enters from higher electronic levels after elimination of its excess energy. In transition from this singlet level to the ground state fluorescence is emitted, but part of the energy goes not to the ground level but to the triplet level. Prior to Sveshnikov's experiments, some authors assumed that the transition to the phosphorescent triplet state is realized directly from high-lying electronic-vibrational levels to which the molecule is raised as a result of absorption of a photon.

These studies enabled Sveshnikov to analyze the kinetics of phosphorescence in detail. He carried out calculations of the transitions between three molecular levels, characterizing them by definite probabilities. In constructing the differential equations for the population of each of the levels all the assumption were convincingly supported by arguments based either on Sveshnikov's own results or extensive data from the literature. As a result of solution of the differential equations there was deduced a mathematical relationship between the transition probabilities and the experimentally determined luminescence characteristics of the substance (quantum efficiency, persistences of fluorescence and phosphorescence, etc.).

Among the assumptions made by Sveshnikov in deriving the equations characterizing the kinetics of intramolecular transitions was the assumption that the luminescence decay law is exponential, as was observed for a large number of organic luminophors during the first stages of decay. However, development of experimental techniques made it possible to establish the existence of some deviation from exponential decay at the later stages of the process. This served as the basis for the hypothesis that there simultaneously occur two processes: monomolecular and bimolecular, the kinetics of the latter being determined by the recombination probability. Nevertheless,

in solid solutions of organic luminophors the monomolecular process is the predominant one. In later papers published by Sveshnikov in collaboration with A. V. Aristov there were adduced additional arguments in favor of the transition scheme used for calculating intramolecular transitions and, in particular, it was shown that the transitions between the fluorescent and phosphorescent states of a molecule occur directly between isoenergetic levels of these states.

Use of the equations describing the kinetics of intramolecular transitions enabled Sveshnikov to give a correct interpretation of some of the earlier observed experimental facts, for example, the increase in the probability for transitions from the fluorescent to the metastable state with increasing temperature and concentration. However, measurement of the usual luminescent characteristics does not yield sufficient information for determining all the transition probabilities. A complete set of equations can be constructed only by taking into account the dependence of the luminescence intensity or the absorption of the specimen on the density of the exciting radiation. Sveshnikov and Aristov proposed a relatively simple method for quantitative evaluation of this dependence from the "saturation" of the luminescence of optically thin layers at high excitation densities. The advantage of this method as compared with absorption methods, is that in using it there is no necessity for taking into account the overlapping of the absorption spectra of the molecule in the metastable and ground states and also of the absorptive power of short-lived products of reversible photochemical reactions.

Use of these new experimental data in conjunction with the earlier known characteristics makes it possible to calculate the probabilities for all transitions, i.e., the probabilities necessary for solution of the fundamental question of the ways of expenditure of excitation energy by the excited molecule, for investigation of the kinetics of some chemical reactions, and for choice of the optimum conditions for "accumulation" of molecules in the metastable state. The last is particularly important in connection with the recent use of some organic media for realizing masers and lasers; the theory developed by Sveshnikov can be used for the practical end of determining the efficiency of pumping.

Sveshnikov was keenly interested in investigating almost all the basic characteristics of phosphorescence. Thus, in collaboration with P. P. Dikun and A. A. Petrov, Sveshnikov in 1951 published a comprehensive study of the mean decay rate of the phosphorescence of about 50 different benzene derivatives. In this paper it was emphasized that the persistence of phosphorescence must be determined not only by intercombination forbiddenness but also in a number of cases by symmetry considerations.

In recent years, V. A. Pilipovich, under the supervision of Sveshnikov, carried out a detailed investiga-

tion of deviations of a number of compounds from the exponential decay law.

Sveshnikov devoted much attention to investigation of the polarization of phosphorescence. Under his guidance, V. L. Ermolaev measured the dependence of the degree of polarization on the wavelength of the exciting light and showed that for a number of dyes the direction of the moment of the electronic transition corresponding to phosphorescence is perpendicular to the moment of the transition associated with fluorescence.

In other studies under the supervision of Sveshnikov there were also investigated the influence on the phosphorescence of the viscosity of the solvent, the quenching of phosphorescence by extraneous quenchers, excitation of phosphorescence in the anti-Stokes region, and molecular association in the range of high concentrations.

Sveshnikov was undoubtedly one of the outstanding specialists in the field of phosphorescence of organic compounds. Formulas derived by Sveshnikov as far back as 1948 are still extensively used by investigators concerned with the problems of the kinetics of organic luminophors.

WORK ON CONCENTRATION QUENCHING OF LUMINESCENCE

In the course of the past decade, in addition to other problems involved in the luminescence of organic compounds, Sveshnikov considered different concentration induced effects in luminescent solutions. Sveshnikov was of the opinion that investigation of the luminescence properties of solutions with different concentrations is an effective approach to elucidating the nature and the laws governing the interaction of organic molecules in the excited state. These investigations were also of practical value, inasmuch as they involved the study of the emission of luminophors that are significant in the field of biology; at the same time they were a good method for investigating the liquid state of matter. Emphasizing the complexity of concentration effects, Sveshnikov noted that the interaction of molecules in the liquid state may change with changes in the experimental conditions. Thus, in investigating alcohol solutions of acriflavine and acridine orange it was found that reduction of the temperature from room temperature to liquid air temperature is accompanied by transition from migration quenching to associative quenching.

The migration theory of concentration effects, taking into account energy transfers between monomers, was developed by S. I. Vavilov, T. Förster, D. L. Dekster, and M. D. Galanin. According to this theory, transfer of excitation energy from monomer to monomer should lead to differences between the rates of decay of the components of the emission polarized parallel and perpendicular to the electric vector of

the linearly polarized exciting light; this leads to depolarization of the emission, increasing in degree at the later stages of decay. Precise measurements carried out by means of the State Optical Institute's fluorometer not only substantiated the existence of this effect, which was detected earlier by M. D. Galanin, but also showed that the magnitude of this effect is in good agreement with the predictions of the Galanin-Förster theory.

The significant role played by migration of energy in the process of luminescence in concentrated solutions has now been firmly established. However, there still exist different views regarding the nature of the quenching mechanism involved in the transfer.

Investigations of the shapes of concentration versus persistence curves for glycerol solutions of fluorescein, rhodamine B and acriflavine, carried out by Sveshnikov and his co-workers, showed that excitation energy transfer to nonluminescing dimers plays a significant role in the quenching of the luminescence of these solutions.

Sveshnikov and his co-workers also carried out comprehensive research on the quenching of luminescence in solutions of aromatic hydrocarbons, in which diffusion of the luminescent molecules plays an important part. The observed viscosity dependence of the efficiency and persistence of fluorescence for solutions of anthracene and its derivatives substantiated the diffusion nature of the quenching effect. It was also found that concentration quenching of the diffusion type is satisfactorily described by the theory developed by Sveshnikov for quenching of fluorescence by extraneous non-absorbing quenchers.

In the course of investigation of the fluorescence of solutions of benzene and its derivatives there were carried out for the first time fluorometric measurements in the ultraviolet region and the persistences of the fluorescence of benzene, many of its derivatives and naphthalene were determined. In the solutions of some aromatic hydrocarbons there were detected luminescent excited dimer-eximers. In experiments initiated by Sveshnikov the properties of eximers were investigated for the first time by fluorometric techniques in the cases of solutions of benzene, pyrene and some meso-derivatives of anthracene. Simultaneous phase and de-modulation measurements of the persistence of fluorescence of eximers together with measurements of different modulation frequencies made it possible to prove that the eximers are characterized by a specific nonexponential decay law. This is connected with the fact that eximers appear during the lifetime of the monomers in the excited state, as a result of collisions of monomers in the process of diffusion.

Sveshnikov also considered the influence of high activator concentrations on the phosphorescence of organic compounds. It was proved by careful measurements, carried out under his supervision, that con-

centration-induced changes of phosphorescence are not connected with transfer of energy from the phosphorescent level, for depolarization is not evinced in the decay process.

Concurrent investigation of the concentration affected behavior of the fluorescence and phosphorescence of the same compounds led to the conclusion that at high concentrations there occurs associative interaction between the activator molecules, which leads to changes in the probability for transitions between the levels: the probability for nonradiative transitions from the fluorescent to the phosphorescent and from the phosphorescent to the ground states increases.

WORK ON CHEMILUMINESCENCE

At an early stage of his scientific career Sveshnikov became interested in chemiluminescence—luminescence associated with certain chemical reactions. Although this phenomenon (in the form of bioluminescence) has been known for a long time, actually chemiluminescence is still one of the least studied forms of luminescence. At the end of the 1930's, when Sveshnikov began his investigations in this field, there existed a number of different hypotheses for explaining the mechanism of chemiluminescence, but the scanty data available at that time were inadequate for deciding between the different and often conflicting hypotheses proposed by different investigators. After analyzing the different hypotheses, Sveshnikov noted that they could all be divided into two distinct groups: in one it is assumed that the excited molecule appears as a result of chemical interaction between two molecules of the initial substance; in the other it is assumed that it appears as a result of successive transformations of one molecule. These two hypothetical chemiluminescent mechanisms imply entirely different dependences of the intensity (proportional to the reaction rate) on the concentration of the chemiluminescing substance: a quadratic dependence in the former case and a linear in the latter. Sveshnikov's investigations of the chemiluminescence of phthalic acid, hydrazines and lucigenin in the process of oxidation by hydrogen peroxide in an alkaline medium unambiguously showed that the concentration dependence of the initial chemiluminescence intensity is nearly linear. This at once made it possible to discard all hypotheses implying quadratic dependence.

As a result of Sveshnikov's investigation of the kinetics of chemiluminescence interesting data were also obtained on the influence of different factors (concentration of the reacting substances, viscosity of the solvent, etc.) on the intensity of the emission and on the character of its variation with time. These data showed that the emission of light is the result of complex interweaving of different chemical processes, characterized by different rates highly sensitive to the experimental conditions.

Sveshnikov's work on chemiluminescence, carried out with the care and thoroughness characteristic of all his studies, has made a significant contribution to our understanding of the mechanism of this phenomenon.

In speaking of Sveshnikov's work on chemiluminescence one should not omit mention of his popular-scientific publications on this topic. In his interesting and lively articles he not only called attention to this interesting effect but gave his readers a full account of the scientific results obtained in investigating this phenomenon.

Sveshnikov was by no means a narrow specialist. By way of illustration of the width and diversity of his interests one can cite his reviews on chromatography, a topic that at first glance might seem to be far from the field of his scientific activity. The publication of these reviews was stimulated by his desire to call attention of investigators to the refined and potentially very useful chromatographic procedure for separating complicated mixtures and to show the role played in the development of this procedure by its inventor, the Russian scientist M. S. Tsvet.

Throughout his life Sveshnikov paid a great deal of attention to the problems of practical utilization of luminescence. Sveshnikov and his students developed a number of original instruments and devices. Thus, in collaboration with M. G. Kachurin and P. P. Dikun, Sveshnikov developed a luminescence method for quality control of dyeing of cloth, which is now extensively used in Soviet textile mills.

The slow neutron detectors developed by B. Ya. Sveshnikov and T. V. Timofeeva are now extensively used in different types of nuclear equipment. Sveshnikov also contributed to research aimed at developing luminescent flaw detection.

In all the diverse fields of science and technology with which Boris Yakovlevich Sveshnikov came into contact he showed himself to be an inventive, erudite specialist and at the same time a modest, deeply humane person, always willing to help with his advice and to encourage every useful initiative. He will remain such forever in the memory of all his acquaintances, friends and comrades.

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