CONFERENCES AND SYMPOSIA

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A scientific session of the Physical Sciences Division of the Russian Academy of Sciences (RAS) dedicated to the 120th anniversary of the birth of Sergei Ivanovich Vavilov was held in the Conference Hall of the P N Lebedev Physical Institute, RAS, on 30 March 2011.

The following reports were put on the session's agenda posted on the web site www.gpad.ac.ru of the Physical Sciences Division, RAS:

(1) **Masalov A V** (P N Lebedev Physical Institute, RAS, Moscow) "S I Vavilov and nonlinear optics";

(2) **Basiev T T** (Laser Materials and Technology Research Center, A M Prokhorov General Physics Institute, RAS, Moscow) "Luminescent nanophotonics and high-power lasers";

(3) **Vitukhnovsky A G** (P N Lebedev Physical Institute, RAS, Moscow) "Advances in luminescent light sources and displays";

(4) Aleksandrov E B (Ioffe Physical Technical Institute, RAS, St. Petersburg) "Sergei Ivanovich Vavilov and the special theory of relativity";

(5) **Bolotovsky B M** (P N Lebedev Physical Institute, RAS, Moscow) "Vavilov–Cherenkov effect";

(6) **Vizgin V P** (S I Vavilov Institute of the History of Natural Sciences and Technology, RAS, Moscow) "Sergei Ivanovich Vavilov as a historian of science";

(7) **Ginzburg A S** (Knowledge Society) "Academician S I Vavilov—a devotee of the enlightenment and the first president of the Knowledge Society of the USSR".

The papers written on the basis of reports 1–4 and 6 are given below. The main contents of report 5 is reflected in the paper "Vavilov–Cherenkov radiation: its discovery and application" [*Usp. Fiz. Nauk* **179** 1161 (2009); *Phys. Usp.* **52** 1099 (2009)] published earlier by B M Bolotovsky.

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S I Vavilov and nonlinear optics

A V Masalov, Z A Chizhikova

Sergei Ivanovich Vavilov was a distinguished Russian physicist, outstanding organizer, eminent teacher, and enlightener. It was precisely his activity that promoted the revival and progress of physics research in our country after the devastation of the 1920s. Owing to his efforts as a scientist and organizer, our country became a world power with regard to scientific investigations.

While on the subject of S I Vavilov's scientific heritage, he greatly advanced the science of luminescence in various media, to begin with. In I M Frank's apt remark [1], Vavilov transformed the knowledge about luminescence from a description of a collection of facts to a rigorous science. In particular, he gave a more exact definition of the phenomenon of substance luminescence, introduced the notions of the energy and quantum yields of luminescence of substances, ascertained that the quantum yield is independent of the wavelength of the exciting light (Vavilov's law), elaborated the techniques for measuring the luminescence yield, and studied polarization characteristics of luminescent radiation and its relation to the density of luminescent particles. Proceeding from this knowledge, jointly with his colleagues he developed the luminescence method of substance analysis. This method, which received ample recognition even in his lifetime, is also topical today, especially so in the study of the properties of nanoparticles.

Much has been well written about S I Vavilov's role in the discovery of the Vavilov–Cherenkov effect. However, as regards the history of the invention of fluorescent lamps — daylight lamps — it has not been adequately covered. This history dates back to the time when S I Vavilov introduced the notion of luminescence yield and first revealed by direct measurements that the quantum yield of luminescence may approach 100% in a number of media. It is precisely the high quantum yield of luminescence that underlies the several-fold energy superiority of fluorescent lamps over ordinary incandescent lamps. In modern times, when we attempt to consider scientific investigations from the standpoint of

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S I Vavilov in 1925–1926, when the first experimental investigation in nonlinear optics was carried out.

financial advantage, it would be instructive to give an estimate of the savings which fluorescent lighting (i.e., research on the fluorescence yield) has given our country over the past decades.

The fact that media with a high quantum yield of luminescence exist favored the advent of lasers. T Maiman, the inventor of the first ruby laser, reminisced [2] that he was dissatisfied with the available data about the low quantum yield of optical luminescence in ruby, because a high quantum yield would be natural and only special reasons could be responsible for lowering it. In the USSR program of creating lasers [3], luminescent crystals were highest on the list of the most promising laser media. It also comes as no surprise that the first lasers in our country were made from ruby crystals in institutions which were S I Vavilov's 'offsprings'—at the P N Lebedev Physical Institute of the USSR Academy of Sciences (FIAN) and the State Optical Institute (GOI) [3].

The story of the first experimental investigation performed by S I Vavilov (in collaboration with V I Levshin) in nonlinear optics is well known. This investigation was carried out in 1925, and its results were presented in Ref. [4]. (S I Vavilov's portrait displayed on this page dates back to precisely that time.) However, the essence and details of the experiment are little known. Here, we report on several features and details of Vavilov's first experiment mentioned above and give more information about nonlinear optical studies conducted by M D Galanin—S I Vavilov's pupil and disciple, who supervised for many years the Laboratory of Luminescence established by S I Vavilov at FIAN.

S I Vavilov's and V I Levshin's paper [4] is often cited as the first observation of a nonlinear optical effect. This is

precisely the paper which tells us about the lowering of absorption in a medium with increasing light intensity. Such a statement is undoubtedly true, with the reservation that S I Vavilov pondered and wrote notes about the limits of the validity range of the Bouguer-Lambert-Beer law of 'linear' light absorption back in 1919. One can read about it in the materials collected in the RAS Archive. The paper "O sootnoshenii mezhdu flyuorestsentsiei i fosforestsentsiei v tverdykh i zhidkikh sredakh" ("On the relation between fluorescence and phosphorescence in solid and liquid media") [4] was submitted for publication in December 1925. Different experiments were described in that paper. Its Section 4, entitled "On the feasibility of absorption lowering in fluorescent or phosphorescent media irradiated by the light of a spark", was concerned with the violation of the 'linear' light absorption law. Premising the description of their experiment, Vavilov and Levshin distinctly formulate the mechanism responsible for the expected lowering of substance absorptivity with increasing incident radiation intensity. This lowering must take place because of the reduction in the number of absorbing molecules in the ground state due to the light absorption. The authors of Ref. [4] devised the formula for the intensity of radiation transmitted through the medium, which should quantitatively describe the effect: $J = J_0 \exp \left[-N(1-x) \alpha \right]$. Here, x describes the fraction of molecules that are 'out of the game' due to the absorption of light, and α and N are the absorption cross section of the molecules and their number per unit cross section of the sample (the product of molecular concentration and the sample thickness). Should we describe the effect in presentday terms, we would write out precisely the same formula (to within the notation). Also given was an estimate of the fraction x under continuous irradiation in the limiting case of the weak effect: $x = J_{abs}\tau/(Nh\nu)$, where J_{abs} is the absorbed light flux, τ is the lifetime of excited molecules of the absorbing substance, and hv is the photon energy. Simultaneously, the authors of Ref. [4] consider, apart from the case of continuous irradiation, the special case wherein the light source emits pulses of a duration shorter than the molecular lifetime. As a result, they draw a conclusion that maximizing the effect requires selecting a medium with the longest possible lifetime. They select uranium glass (in modern sets of color optical glass samples, uranium glass is designated as JS19). Uranium glass possesses a very long lifetime of the excited state ($\sim 10^{-5}$ s). For a light source they employed a spark with a glow duration of $< 10^{-6}$ s—supposedly the highest-power source available at that time. To verify the role of lifetime, the authors prepared a second sample-a cell with a solution of fluorescein. The lifetime of fluorescein molecules in a solution is shorter than the duration of a spark flash, and under these conditions one would expect a linear character of light absorption, i.e., the absence of the soughtafter effect. Proceeding from the data about the spark light energy, the authors estimated the sought-after effect of the lowering of uranium glass absorptivity at 2%. The setup for observing light absorption in uranium glass and fluorescein employed a noteworthy layout. The latter was not given in the paper, but it was described in sufficient detail.

Figure 1 represents a schematic of the setup reconstructed from its description in Ref. [4]. The light of a spark was focused on a sample in such a way that one half of the image of the spark passed through the medium under investigation, and the other half passed by the medium. The transmitted light of the spark was focused on the slit of a spectro-



Figure 1. Schematic representation of the setup for observing light absorption in uranium glass: 1—spark, 2—lenses, 3—uranium glass or a cell with fluorescein, 4—sample-free region, 5—attenuation filter, 6—spectrophotometer, 7—polarizer, and 8—observer's eye.

photometer so that the light passing through the sample was focused onto one half of the slit height, and the other half was illuminated by the light that passed by the sample. The spectrally decomposed radiation at the output of the spectrophotometer was analyzed by the human eye. In the spectrophotometer, a polarizer was mounted in the path of the light transmitted through the sample, which made it possible to attenuate this light to the intensity level of the light that passed by the sample. As is well known, in the visual comparison of two illuminated fields observed in one field of view, it is possible to discern a very small difference in visual field illuminances and thereby equalize the illuminances to at least within several tenths of a percent.

Therefore, the observer's task during repetitive spark flashes was to equalize, by way of polarizer rotation, the fields of the light passed through the sample and the light passed by it. This procedure was repeated for each sample in two series of 50 measurements. In one series, an attenuation filter was introduced into the light beam in front of the sample to lower the intensity of sample irradiation; in this case, the nonlinear effect according to estimates would not be expected to occur. In the second series, the same attenuation filter was placed after the sample, and the light of the highest possible intensity passed through the sample. In this case, one would expect a manifestation of absorption nonlinearity for uranium glass, i.e., a disturbance of the balance in illuminance of the light fields under observation. The observer restored the balance of illuminances by slewing the polarizer and reading its new position. The difference between the readings of the angular polarizer positions in the two series was converted to a change in absorption. The authors estimated the reproducibility of these absorption measurements at $\pm 0.3\%$ (this was done from the results of measurements with fluorescein; most likely the accuracy was limited by the instability of the spark discharge). Transposition of the attenuation filter resulted in a lowering of the absorptivity of the uranium glass by 1.5% with an increase in light intensity. The sign of the effect, like its magnitude compared to the estimate (2%, see above), testifies to the validity of the result. Also, it is amazing how reasonably and optimally the measurements were made: a comparison object was utilized, intensity measurements relied on equalization of illuminances, and the trick of transposing the attenuation filter was taken advantage of. As a consequence, the experimental observations proved to be a success, despite the absence of photoelectric recorders. In modern nonlinear optics, the trick of attenuation filter transposition has come to be generally accepted.

S I Vavilov described in full measure the significance of this investigation in his book *Mikrostruktura Sveta* (*The Microstructure of Light*) [5], which was published late in 1950. He saw an advance copy of the book not long before

his death. The book [5] gave a very vivid and complete picture of how the properties of separate radiators-atoms and molecules of a substance-form the characteristics of the emitted light. In his book, S I Vavilov once again addressed the mechanism of absorption lowering in a medium with increasing light intensity; nowadays we call this mechanism the population saturation of absorbing molecules and its corresponding manifestation is termed the bleaching of the medium. S I Vavilov also introduces the term nonlinear optics. He writes: "The greater the number of molecules in an excited state in the propagation of light through the medium, i.e., the higher the light power, the greater must be the lowering of the absorbed energy fraction, because the excited molecules cease to absorb the light in the previous manner prior to returning to their normal state. Absorption must therefore depend on the power of the light flux.... 'Nonlinearity' in an absorbing medium should be observable not only in relation to light absorption. The latter is related to dispersion, and therefore the velocity of light propagation through the medium, generally speaking, should also depend on the light power. In the general case, the dependence on the light power, i.e. the violation of superposition principle, should be observable for the same reason in other optical properties of the medium as well-in birefringence, dichroism, optical rotation, etc." Here, S I Vavilov generalizes the manifestation of nonlinearity and predicts other nonlinear phenomena. Much more recently, with the advent of lasers, appropriate effects were discovered and studied in different media in the course of their radiation-induced bleaching (for the accompanying nonlinearities in dye solutions, see, for instance, Ref. [6]).

It is noteworthy that, in his reasoning about nonlinear optical effects, S I Vavilov unequivocally associates 'nonlinearity' with the violation of the superposition principle: the action of several light waves on a medium in the mode of nonlinear interaction is not reduced to the sum of the individual actions of these waves. By this reasoning, S I Vavilov foresees the foundation for the future mathematical description of nonlinear optical phenomena. Indeed, when diverse nonlinear optical phenomena were discovered in the 1960s, owing to the advent of lasers, and the construction of an adequate mathematical apparatus for their description became a necessity, the first and supposedly the most important step of theoretical nonlinear optics involved the formulation of the nonlinear relationship between the medium polarization and the light wave field inducing the latter. It is precisely this relationship that S I Vavilov's reasoning about the violation of the superposition principle is fully applicable to.

It should be noted that S I Vavilov also analyzed the opposite limiting case of weak light fluxes when discussing the applicability problem for the law of linear light absorption—the Bouguer–Lambert–Beer law—at high radiation intensities. In doing so, he took advantage of a quantum treatment of radiation and considered light as a flux of rare single photons [7].

The birthday of a new field of physics—nonlinear optics—may be dated to either 1925, when the first experimental work [4] was carried out, or 1950, when the book *Mikrostruktura Sveta* was printed, in which the term 'nonlinear optics' was introduced and important generalizations were made. We emphasize that physics in 1950 and, in the first place, optics were on the eve of revolutionary changes: four years remained before the emergence of

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quantum electronics, and ten years before the advent of lasers. It so happened that the breakthrough in the development of coherent radiation sources in the optical range, i.e. lasers, was made using luminescent crystals. By that time, information about the luminescence of substances made up a mature field of knowledge with a wealth of experimental data; at FIAN and other research institutions there were teams of experienced experts in luminescence — the pupils and disciples of Vavilov's scientific school. It is safe to say that the way to success in the implementation of the first lasers in our country was paved not only by the insight of the founders of quantum electronics, N G Basov and A M Prokhorov, and their organizational activity, but also by the achievements of S I Vavilov's school in the area of luminescence.

With the advent of lasers, research in the field of nonlinear optics substantially broadened in scope. A great variety of new nonlinear effects were discovered, their nonlinearity mechanisms being different from that in S I Vavilov's first experiment. In modern nonlinear optics, it is possible to single out the two biggest classes of nonlinearity mechanisms: the mechanisms with a nonlinear electronic response, and those with the response of atomic and molecular nuclei. Electronic nonlinearities are characterized by an anharmonic response of electrons in substances to the action of a harmonic light field. The class of electronic nonlinearities may be subdivided into the groups of resonance and nonresonance nonlinearities (depending on the ratio between the frequency of the light field and atomic transition frequencies). The nonresonance electronic nonlinearity is responsible for second harmonic generation, the generation of sum and difference frequencies, multiphoton absorption, and several other nonlinear phenomena. The resonance electronic nonlinearity is primarily responsible for the bleaching of a medium, as well as for other nonlinear effects. S I Vavilov's and V L Levshin's experiment [4] demonstrates the manifestation of precisely the resonance nonlinearity of substance molecules. In the nonlinear mechanisms involving responses of atomic and molecular nuclei, the field-induced motion of electrons remains harmonic, but this motion is the reason for the displacement of atomic cores, which shows up as a change in optical properties of substances. Among these mechanisms, mention should be made of electrostriction, orientation of optically anisotropic molecules, excitation of molecular vibrations, and some others. These mechanisms are responsible for the nonlinearity of the refractive index of a medium, induced birefringence, stimulated light scattering, and other effects. It is valid to say that S I Vavilov's first experiment turned out to be merely a window on the world of diverse nonlinear optical phenomena.

After the creation of lasers, experimental work on nonlinear optics poured forth as from a horn of plenty. It was evident that nonlinear mechanisms are highly diverse and go beyond the scope of the effect of light on energy level populations in a substance. Many nonlinear effects occurred with no light absorption at all (second harmonic generation, nonlinear refractive index, etc.). A breakthrough in the theoretical description of nonlinear optical effects was made in our country by R V Khokhlov, S A Akhmanov, and their collaborators: a nonlinear material equation was introduced to relate the response of a medium in the form of polarization to the magnitude of the electric field strength in a light wave: P = P(E). In this case, of fundamental importance for the theory was going over from the radiation intensity (as with S I Vavilov) to the electric field strength of the wave, and from energy level populations to the polarization of the medium. This made it possible to construct a consistent theoretical description of a wealth of diverse nonlinear optical effects, in which the differences between nonlinearity mechanisms were 'concealed' in the form of the material equation P = P(E). Methods for the solution of Maxwell equations with one type of the material equation or another were also elaborated. Furthermore, the material equation permitted reformulating the superposition principle making it possible to differentiate linear and nonlinear optical effects. Among the nonlinear effects are those wherein the polarization of a medium exposed to the sum of different fields is not equal to the sum of polarizations induced by each of the fields separately.

S I Vavilov's merit in the formation of nonlinear optics as a new avenue in light–matter interaction science is marked by the fact that the regular International Conference on Nonlinear Optics in Novosibirsk bears his name.

At FIAN, S I Vavilov set up the Laboratory of Luminescence, which he supervised until the last days of his life. After 1963, this laboratory was headed by his pupil and disciple M D Galanin (S I Vavilov's last postgraduate student). In mid-1961, Moscow's first laser was put into operation in Galanin's team — a ruby laser [8]. After the creation of lasers, M D Galanin, along with Z A Chizhikova and other members of the Laboratory of Luminescence, undertook in the period from 1963 to 1973 the 'development of Vavilov's nonlinear optics' and performed many pioneering studies with the employment of laser radiation. They published more than ten papers concerned with the observation of new nonlinear effects. They discovered two-photon absorption and dichroism in liquids, luminescence quenching by intense light fluxes, and anti-Stokes Raman light scattering by the electronic levels of dye molecules; they investigated superluminescence in molecular crystals under laser irradiation and the luminescence of dyes from the second excited level, and they studied the features of luminescence under excitation by picosecond light pulses. These papers were published in scientific journals of the highest prestige at that time, including Pis'ma Zh. Eksp. Teor. Fiz. (JETP Lett.) [9]; many of them are appropriate for citation in textbooks.

M D Galanin's closest colleague, who worked with him on the making of the ruby laser—A M Leontovich—also carried out several important nonlinear optical studies, in particular, on the resonance interaction of short light pulses with ruby and neodymium ions in crystal matrices [10]. Together with his colleagues, he succeeded in realizing the mode of coherent light pulse amplification, wherein the pulse duration is shorter than the period of medium phase memory; this mode is no longer described in terms of the populations of the ground and excited ion states.

In the organization of research at the Laboratory of Luminescence of FIAN, S I Vavilov attached special significance to the work of scientific seminars. Regular seminars in the Laboratory of Luminescence, or colloquia, as they were called at that time, were conducted jointly with the meetings of the Commission on Luminescence of the USSR Academy of Sciences beginning from 1945. Reports about these seminars have been retained since January 1947. The records were made by seminar secretaries for almost 65 years, and for the last 24 years the records were made by Z A Chizhikova. The seminars are described in greater details in Ref. [11]. Here are examples of scientific reports borrowed from these records. At the seminar of 5 October 1949, S I Vavilov and M D Galanin gave a report entitled

"Izluchenie i pogloshchenie sveta induktivno svyazannykh molekul" ("Emission and absorption of light by inductively coupled molecules"). This work was among the first in a long series of investigations into the migration of excitation energy in substances. Held in June of 1949 was a seminar with five reports on the subject of luminescent light sources; among the speakers was V A Fabrikant with a report "Raboty VEI po lyuminestsentnym lampam"¹ ("Work on luminescent lamps at VEI"). The seminars were held regularly on Wednesdays at 10 a.m. A regular seminar was conducted by S I Vavilov on Wednesday, 24 January 1951—on the day before his death. Since S I Vavilov's death, the seminars of the Laboratory of Luminescence have been held on Wednesdays as before. The 1900th seminar was held in October 2010. Beginning from 1976, Vavilov Readings are held annually in honor of S I Vavilov's birthday late in March, at which reports on topical subjects of modern optics are given by leading scientists from FIAN and other institutes. Nobel Prize Laureates I M Frank, N G Basov, A M Prokhorov, and V L Ginzburg, as well as other famous scientists from Moscow research institutes and from institutes in other cities of our country, have participated in the work of the seminars and Vavilov Readings. This year saw the 35th Vavilov Readings. The seminar of the Laboratory of Luminescence is a special monument to its founder—S I Vavilov—and is undoubtedly among the unique phenomena of FIAN.

In a brief report, it is impossible to overview the numerous achievements made in nonlinear optics over the past years. We shall merely cite several examples where nonlinear optics 'work'.

(1) First of all, this is the development of laser technology. Each time a new laser medium emerges or a laser is designed to provide specific radiation parameters, there is a need to calculate its operating conditions. This may only be done by using balance equations, which is nothing but a description of the resonance nonlinearity of the active medium. In this case, the nonlinear optical description acts as an engineering science.

(2) Special nonlinear media ensure laser operation in unique lasing modes. For instance, in lasers which generate pulses of picosecond and femtosecond duration, use is made of bleachable media and media with a nonlinear refractive index. To amplify short pulses, advantage is taken of parametric crystals whose operation is underlain by the mechanism of nonresonance electronic nonlinearity. The duration of picosecond and femtosecond laser pulses is measured exclusively with instruments which rely on nonlinear optical phenomena, because the methods of direct photodetection do not offer the requisite time resolution.

(3) Nonlinear media make it possible to substantially broaden the wavelength range in which coherent radiation may be obtained. In this case, wide use is made of crystals which generate the second optical harmonic, and of converting media utilizing stimulated Raman light scattering. To continuously tune the wavelength of laser radiation, advantage is taken of parametric crystals with a nonresonance electronic nonlinearity.

(4) The spectroscopic technique of ultranarrow atomic resonances, which is applied in the development of optical frequency standards and precise clocks, is inseparably linked with resonance nonlinear phenomena. Owing to the narrowness of the atomic resonances, the nonlinearity of the

interaction with light manifests itself even for milliwatt radiation power.

(5) Lasers and the nonlinear media capable of ensuring optical rectification and differential frequency generation find application in the modern technology of generating and detecting terahertz radiation.

(6) Nonlinear interaction effects that emerge in the data transmission by light pulses in optical fiber communication lines restrict the technical communication capabilities. In this case, even a weak nonlinearity of the refractive index of the light guide material has an adverse influence owing to accumulation of the disturbing effect over a long propagation path. However, 'useful' nonlinearity—optical pulse amplification due to stimulated Raman scattering in the light-guide material—also finds use in light guides.

(7) Optical memory cells developed for quantum computers rely exclusively on the resonance nonlinearity effects in different media.

(8) To generate optical radiation in nonclassical quantum states (sub-Poissonian, squeezed, etc.), only nonlinear processes that ensure the multiphoton nature of an elementary act of interaction with light are suitable. This light is requisite for unique instruments intended for ultrasensitive optical measurements, permitting one to overcome the standard quantum sensitivity limit.

(9) Among the spectroscopic methods for studying a substance, supposedly only one nonlinear optical method has gained acceptance—the technique of coherent Raman spectroscopy. Its realization necessitates two laser sources, one of which is continuously tuneable in wavelength.

(10) To record weak infrared (IR) radiation, use is sometimes made of the mixing of IR waves with visible radiation in a nonlinear medium. In this case, the IR frequency is transferred to the visible range in the course of sum (or difference) frequency generation, where the means of high-sensitive photodetection is available.

The above examples do not exhaust the subject of the role and place of nonlinear optics in modern science and technology.

During the years of the 'laser boom' in the 1960s and 1970s, FIAN and several academic institutes expanded to take on the graduating students of the Moscow institutes of higher education, who improved laser technologies and discovered new nonlinear optical effects. A considerable portion of them was made up of the graduating students of the Moscow Institute of Physics and Technology (MIPT). At this point, there is no escape from recalling a little-known aspect of S I Vavilov's activity — his participation in the establishment of MIPT. As President of the USSR Academy of Sciences, he advocated the idea of organizing in our country a highest-level educational institution aimed at preparing research physicists, which was expressed by P L Kapitza and like-minded scientists. In this connection, in 1946 S I Vavilov become president of the board of the Higher Physicotechnical School of the USSR (later MIPT). Academician G S Landsberg-S I Vavilov's fellow scientist—was assigned to organize the teaching of optical disciplines there. During the period of the laser boom, hundreds of graduating students from MIPT joined FIAN, the Institute of Spectroscopy, and other research centers of the USSR Academy of Sciences and ensured a world level of achievements in the laser area.

To summarize, it is valid to say that the advancement of the science of luminescence in media, which is due to a great extent to S I Vavilov and his successors, was conductive to the

¹ All-Union Electrotechnical Institute (VEI in Russ. abbr.), Moscow.

successful development of laser research in our country. The pioneering experiments performed by S I Vavilov and V L Levshin opened the window onto the world of diverse nonlinear phenomena in optics. Thanks to the application of laser light sources, S I Vavilov's pupils and successors made a major contribution to the discovery and investigation of new nonlinear optical effects. The spirit of devotion to science displayed by S I Vavilov and his personal example of selfless labor under incredibly difficult conditions are still helpful in retaining the high scientific level of optical research in our country.

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Luminescent nanophotonics, fluoride laser ceramics, and crystals

T T Basiev, I T Basieva, M E Doroshenko

When taking up some of Sergei Ivanovich Vavilov's scientific publications and his fundamental work — the book *Mikros-truktura Sveta* (*The Microstructure of Light*) [1]— one arrives at the conclusion that they laid the foundation for the modern nanophotonics of laser and luminescent materials.

Sergei Ivanovich stated that any light source may be characterized by three attributes: radiation energy, spectrum, and the state of polarization. In this regard, he emphasized that they are nothing more than average macroscopic characteristics. Concealed behind them is an extremely complicated microoptics world, due to which these average characteristics are formed. To investigate the nature of light and expose the relation between its properties and the properties of the elementary emitters generating light field, it is necessary to penetrate into this world of microoptics (or nanophotonics, as it is customarily called nowadays).

S I Vavilov assigned to microoptics (nanophotonics) the properties of very small emitters, the manifestations of the lifetimes of excited molecular states and, lastly, the interactions of luminous molecules with the surrounding medium. He placed special emphasis on the fact that the neighboring molecules determine the initial, principal, chain of optical excitation energy transfer (migration) in the medium [1].

Being aware that an increase in the particle concentration results in a shortening of the distance between optically active molecules and, accordingly, in a strengthening of the interaction between them, S I Vavilov and his collaborators studied this phenomenon in detail and discovered characteristic 'nontrivial' concentration dependences of the excited state lifetime, the polarization, and the yield of luminescence. As far back as the 1930s, S I Vavilov and his colleagues discovered that the concentration dependences of the excited state lifetime and the quantum yield were different; this was a direct indication that the kinetic curves of luminescence decay measured in their work were nonexponential.

Unfortunately, in those distant years there was technically no way of instantaneously exciting phosphor and measuring with high precision the kinetics of luminescence decay; nor was there a theory providing a quantitative description of the decay kinetics of a particle ensemble with the inclusion of microinteractions.

The first expressions for the decay kinetics of a statistical ensemble of luminous particles (donors) were due to Förster [2] (1948) and Galanin [3] (1955) in the form of a square-root dependence for the two-particle dipole–dipole quenching

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Uspekhi Fizicheskikh Nauk **181** (12) 1334–1340 (2011) DOI: 10.3367/UFNr.0181.201112j.1334 Translated by E N Ragozin; edited by A Radzig interaction and the donor-acceptor energy transfer:

$$I(t) \approx \exp\left(-\sqrt{Wt}\right),\tag{1}$$

where I(t) is the normalized luminescence intensity, and W is the quenching rate.

More recently, this law was generalized for higher-order multipolar interactions. The general formula has the form [4–6]

$$I(t) \approx \exp\left[-\left(Wt\right)^{d/S}\right],\tag{2}$$

where S is the multipolarity order: S = 6, 8, and 10 for dipole-dipole, dipole–quadrupole, and quadrupole–quadrupole interactions, respectively, and d is the geometric dimensionality of space: d = 1, 2, 3.

It should be emphasized that so complicated form of luminescence decay kinetics is an important instrument for revealing the microstructure of light, which Vavilov spoke of, i.e., for determining the microefficiency (W_0) and multipolarity (S) of the dominant ion-ion interaction, the concentration dependence W(c), the degree of particle ordering, and the dimension d of confined geometries in complex molecular systems of nanometer and atomic scales.

At present, the kinetics of energy transfer permits revealing not only the microoptics of interactions in a phosphor and a laser medium, but also the features of the spatial distribution of the NdO₈, PrO₈, and SmO₈ rare-earth ion molecular nanocomplexes in laser glasses.

An example is provided by the Nd–Nd concentration quenching in laser matrices. Not only may neodymium radiate energy, but two closely spaced ions may quench each other due to dipole–dipole interaction which transfers excitation to quenched levels. Furthermore, optical excitation may migrate from ion to ion and stay on the upper laser level.

The special feature consists in the fact that donor-donor interaction C_{DD} usually is strictly resonance, and its efficiency is therefore always higher than the efficiency of quenching donor-acceptor interaction C_{DA} . When $C_{DD} \ge C_{DA}$, the quenching kinetics for neodymium and many other ions is described by the simple expression

$$I(t) = \exp\left(-Wt\right),\tag{3}$$

with one parameter for the average quenching rate. On the face of it this is a facilitation for researchers, but in reality four characteristics enter into this parameter: the multipolarity of the interaction, the elementary transfer rate, the distance of closest approach, and the dimension of space. It was not clear how to determine these four unknown microparameters from one rate macroparameter, i.e., how to penetrate into microoptics from macrooptics. One way involves the use of low temperatures to 'freeze' migration [7, 8], while the other involves the utilization of samarium ions, whose migration interactions are strongly suppressed, $C_{\text{DD}} \rightarrow 0$ [9–11], to fulfill the function of probing (model) ions.

The general formula for the three-dimensional case of statical quenching ($C_{DD} = 0$) has the form (see, for instance, Refs [7, 9–11])

$$I(t) = \begin{cases} \exp\left[-W_{\rm in}t\right], & W_{\rm in} = \frac{7c_{\rm A}C_{\rm DA}}{R_{\rm min}^{S}}, & t < t_{\rm b}, \end{cases}$$
(4a)

$$\left(\exp \left[- (W_{\rm F} t)^{3/S} \right], \quad W_{\rm F} = k c_{\rm A}^{S/3} C_{\rm DA}, \quad t > t_{\rm b}, \ (4b) \right.$$



Figure 1. Sm–Sm quenching kinetics in lithium–lanthanum–phosphate glass for different Sm concentrations: Sm concentration of 2.5×10^{20} cm⁻³ (1), 9.2×10^{20} cm⁻³ (2), and 23×10^{20} cm⁻³ (3).

where W_{in} is the quenching rate at the initial stage, W_F is the quenching rate at the disordered stage described by the Förster law, t_b is boundary time of the change of these stages, R_{min} is the distance of the closest possible approach of active particles, k is a constant, and c_A is the acceptor concentration.

As discussed above, this more complicated, power-law form of kinetics is highly important for revealing the microstructure of intermolecular interactions, which Vavilov wrote of, i.e., for determining the multipolarity order, the microefficiency, and other transfer microparameters.

From the slope of the decay kinetics curve [7–11] rectified in a log-log plot of intensity vs time at a late stage of decay, $t \ge t_b$ (Fig. 1), we find the degree of nonexponentiality d/S(for Sm–Sm transfer, d/S = 0.375); hence, for the known dimension of space (d = 3), the multipolarity order S(Sm-Sm)=8.

From the slope of the same kinetics curve, although plotted in the coordinates $t^{d/S} - \ln(I)$, we determine the average rate of nonexponential decay, $W_{\rm F} \sim c_{\rm A}^{S/3} C_{\rm DA}$, whence we find the transfer microefficiency $C_{\rm DA}$ from the known *S* and the acceptor concentration $c_{\rm A}$.

Interestingly, the initial decay stage, $t < t_b$, is indicative of ordered exponential kinetics, which reflects distance ordering in the location of luminescent particles or the volume around one particle, which is forbidden to approach. By substituting the already known microparameters S, C_{DA} , and the concentration c_A into the formula $W_{in} = 7c_A C_{DA}/R_{min}^S$ for the quenching rate at the initial stage, we find the last microcharacteristic, R_{min} , which characterizes the degree of short-range order in the ensemble. The energy transfer microparameters determined for several rare-earth ions in glass are collected in Table 1.

It is significant that R_{\min} enters the expression for W_{in} (which also describes the quenching rate in a massive crystal in the presence of ultrafast migration) with a large exponent,

Ions	S	C_{DA}	<i>t</i> _b , ms	$W(R_{\min}), s^{-1}$	R_{\min} , Å
Nd-Nd	6	$0.03 \text{ nm}^6 \text{ ms}^{-1}$	0.59	1.7×10^3	5.1-5.4
Sm-Sm	8	$0.23 \text{ nm}^8 \text{ ms}^{-1}$	0.04	2.5×10^4	5.6
Eu-Eu	10	$4 \times 10^{-3} \text{ nm}^{10} \text{ ms}^{-1}$	0.12	$8 imes 10^3$	4.7

 Table 1. Microparameters of Sm–Sm and Nd–Nd quenching in lithium– lanthanum–phosphate glass and of Eu–Eu migration in sodium–borosilicate glass [7–11].

which permits controlling the quenching rate and the quantum yield by selecting media with different R_{\min} .

Cooperative nanodimensional energy transfer is another interesting example in which the kinetics are highly important.

Until 1999, developers of laser crystals believed that the Ce³⁺ ion, like La and Lu ions, is optically inactive (since it has no f-f transitions in the visible and near-infrared (IR) spectral ranges) and that it may be the main cation forming the laser matrix and not leading to the quenching of activators. However, it turned out that this is not so, and that the quenching by Ce ions does exist. We found that, for instance, for Nd³⁺ ions [12] (and more recently for the Ho³⁺ and Tm^{3+} ions as well [13]), the quenching rises sharply (quadratically) with increasing Ce^{3+} ion concentration. In particular, the lifetime of Tm^{3+} ions in CeF₃ crystals turned out to be 10 times shorter than in LaF₃. We undertook a comprehensive study of this new effect to find that it was due to the energy transfer from one donor ion, for instance, Tm^{3+} , simultaneously to two Ce³⁺ ions, which act as a common twoparticle acceptor. In this case, the single-particle donor luminescence energy must be at resonance with the doubled Ce-ion absorption energy.

We also discovered the phenomenon of cooperative energy transfer in the case of $\text{Er} \rightarrow 3\text{Ce}$ quenching in lanthanum-cerium trifluoride [12] and in the Tb $\rightarrow 2\text{Yb}$ transfer in ytterbium garnet [14]; in the former case, three Ce ions acted as a single cooperative acceptor, and the concentration dependence of the quenching rate was cubic.

It should be emphasized that prior to our studies [12–16], experiments [17, 18] were carried out only on the cooperative transfer from a two-particle donor to a single-particle acceptor (2Yb \rightarrow Tb up-conversion). The transfer probability was very low: the rate was equal to about 2 s⁻¹, which is well below the radiative transition rates for rare-earth ions, $10^2 - 10^5$ s⁻¹, and therefore it could not be measured directly.

We revealed that the radical difference of 'down-process' (i.e., of down-conversion—the cooperative energy transfer from a single-particle donor to two-particle acceptors) from up-conversion is the inclusion of summation over the set of pair acceptor versions throughout the cation (cerium) crystal sublattice, with the result that the total probability of transfer (of down-conversion) rises steeply (by several orders of magnitude).

In this case, the energy transfer rates are comparable to the radiative transition probabilities for donor ions, or even exceed them by an order of magnitude. These features should be taken into account in the development and investigation of laser materials, which has previously been overlooked. Cooperative sensibilization with down-conversion is of special interest, because it may permit advancing lasing to the mid-IR wavelength range $(3-6 \ \mu m)$ and, furthermore, raising the quantum yield (200-300%) due to splitting one 'heavy' photon into several 'light' ones. The problem of space-averaged cooperative energy transfer was theoretically considered assuming the continuous medium and a zero particle dimension [19–21]. In the case of quenching by two-particle acceptors, the kinetic dependence at long times has the following form

$$I_{2}(t) = \exp\left[-(W_{2}t)^{d/(2S-d)}\right],$$

$$W_{2} = \frac{d}{S-d}\left[\Gamma\left(\frac{2S-2d}{2S-d}\right)\right]^{(2S-d)/d}\frac{c_{A}^{2S/d}}{t_{0}},$$
(5a)

and in the case of quenching by three-particle acceptors, one finds

$$I_{3}(t) = \exp\left[-(W_{3}t)^{d/(3S-2d)}\right],$$

$$W_{3} = \left(\frac{d}{S-d}\right)^{2} \left[\Gamma\left(\frac{3S-3d}{3S-2d}\right)\right]^{(3S-2d)/d} \frac{c_{A}^{3S/d}}{2t_{0}}, \quad (5b)$$

where W_2 and W_3 are the average cooperative quenching rates, Γ is the gamma function, and t_0 is the time of an elementary act of cooperative energy transfer to two or three acceptor particles located at the closest distance from the donor.

We performed Monte Carlo simulations and arrived at a good agreement between numerical experiment and the theoretical prediction by the example of a simple cubic lattice. Figure 2 shows cooperative energy transfer to twoparticle acceptors. One can see that the slopes of simulated and theoretical intensity curves plotted on a logarithmic scale against time taken on the d/(2S - d)-power scale are in good agreement at a late stage, which testifies to the coincidence of the functional laws of decay in the theory and in the computer simulations. A characteristic feature of the thus found kinetic dependences is that the fractional exponent d/(2S-d) of time t depends heavily on the interaction multipolarity S and the space dimension d. For instance, for a one-dimensional problem and a quadrupole-quadrupole interaction, the exponent of t is very small (0.053), while for a threedimensional problem and a dipole-dipole transfer the exponent is much greater (0.333). So that the slope of the nonexponential kinetic dependence changes sharply with variations of multipolarity order S and space dimension d, and may serve as a 'probe' both for S and for d.



Figure 2. Cooperative luminescence quenching: simulations by the Monte Carlo method (solid lines), and analytical expression (5a) (dashed lines).

When we consider the dependence of the transfer rate on the number (concentration) of acceptors [see expression (5a)], we see that it also strongly depends on the dimension and multipolarity of interaction $(c_A^{2S/d})$. The exponent of the concentration dependence therewith is very large, from 4 to 20, unlike that for conventional concentration quenching.

In recent years, *nanostructured media* have occupied a special place in the physics of phosphors and laser media. Nanodispersed materials, photonic crystals, and nanopowders doped with fluorescent ions are important from the standpoint of using them in medicine, biology, and optics.

This brings up the question: how do the optical properties of luminescent media change in going over from massive bodies to nanodimensional ones? The objective was to determine the peculiarities and find analytical expressions for the relaxation kinetics of optically excited impurity ions in nanoparticles.

We applied the Monte Carlo method [22] to simulate the kinetics of energy transfer from donor ions to acceptor ions randomly located in spherical nanoparticles 5–50 nm in size, which is much smaller than the radiation wavelength. It was found that for nanoparticles the decay proceeded slower and the quantum yield was higher, and that this effect proved to be stronger for the nanoparticles of smaller size.

The analytical expression for the static quenching in nanoparticles was derived in the form [23, 24]

$$I_{\text{st_nano}}(t) = \left(1 - \frac{(W_{\text{F}}t)^{1/S}}{2k_1 N_{\text{A}}^{1/3}}\right)^3 \exp\left[-(W_{\text{F}}t)^{3/S}\right] + \frac{3}{k_1} \frac{N_{\text{A}}^{1/3}}{(W_{\text{F}}t)^{2/S}} \exp\left[-\frac{(W_{\text{F}}t)^{3/S}}{2}\left\{1 - 2k_0 \frac{(W_{\text{F}}t)^{1/S}}{N_{\text{A}}^{1/3}}\right\}\right], \quad (6)$$

where k_0 , k_1 are constants, and N_A is the average number of acceptors in one nanoparticle. For not-too-small nanoparticles, the second term in the curly brackets on the right-hand side of expression (6) may be discarded.

Expression (6), both with constant and with timedependent amplitude coefficients of the summands, provides an adequate description of the simulated kinetics (Fig. 3). The initial stage of quenching kinetics in nanoparticles shows a power-law time dependence [the first term on the right-hand side of expression (6)], which is similar to the dependence for quenching kinetics in a bulk sample [see expression (4b)]. However, even when the luminescence intensity decays by only one-two orders of magnitude $(I(t) \sim 0.1 - 0.01)$, the second term on the right-hand side of expression (6) comes into play, and a smooth transition to slower kinetics occurs. These kinetics are also linearized in $t^{3/S} - \ln(I)$ coordinates, but they have another macroscopic transfer rate. According to our analysis, the microphysics of quenching in nanoparticles manifests itself in the fact that those donors which are located in the volume of nanoparticles and have acceptors in the total sphere of quenching (in a 4π solid angle) initially decay. Then, as the bulk donors decay, the quenching of the donors that are located in the surface layer and have acceptors only in a quenching hemisphere (in a 2π solid angle) makes itself evident. To put this another way, a twofold lower number of acceptors come into play compared to the volume case, and the quenching proceeds with a $2^{S/d}$ lower macroscopic rate (4-to-1000 times lower).

The next example is *supermigration in nanoparticles*. As discussed above, in the energy transfer and luminescence quenching in the supermigration mode, when $C_{\text{DD}} \gg C_{\text{DA}}$



Figure 3. Two-stage kinetics of luminescence quenching in nanoparticles: Monte Carlo simulations (circles), and analytical expression (solid curve with triangles) consisting of two terms.

and the excitation migrates along donors quite fast, averaging of the quenching rates of different donors occurs, and then we normally observe the so-called kinetic limit — a monoexponential decay with a single average rate. And again this brings up the question: how is it possible to go over to microparameters, to microoptics, when we have only one rate macroparameter W (the average quantity) and many unknown microparameters?

It was found [25] that the kinetics in an ensemble of similar nanoparticles at long times cease to be monoexponential and pass into a nonexponential, power-law stage similar to statical quenching. And from this complex law it is possible to find, as before, all the sought-after microparameters: S, d, C_{DA} , and R_{\min} . Roughly speaking, when we consider a bulk crystal with monoexponential decay kinetics, then on dispersing it to nanoparticles we arrive at nonexponential kinetics and next can determine all sought-after interaction microparameters in the system.

At long times, the exact analytical solution [25] has the form

$$I(t) = \exp\left[-N_{\rm D}\left(\frac{W_{\rm F}t}{N_{\rm D}}\right)^{3/S} \left\{1 - \frac{9\Gamma(1 - 4/S)}{16\Gamma(1 - 3/S)} \times \frac{1}{R}\left(\frac{C_{\rm DA}t}{N_{\rm D}}\right)^{1/S}\right\}\right] \approx \exp\left[-N_{\rm D}\left(\frac{W_{\rm F}t}{N_{\rm D}}\right)^{3/S} \times \left\{1 - \frac{0.6}{N_{\rm A}^{1/3}}\left(\frac{W_{\rm F}t}{N_{\rm D}}\right)^{1/S}\right\}\right],\tag{7}$$

and this dependence agrees nicely with the data of computer simulations.

Our analysis gives evidence that solution (7) is similar to the static-case solution, with the exception of one important feature: the decay rate now depends on N_D —the number of donors in a nanoparticle, or the donor concentration. Previously, in a bulk specimen and in statics, this dependence was absent.

Figure 4 displays kinetic dependences for nanoparticles in this supermigrative mode, when $C_{DD} \gg C_{DA}$. For specimens of nanoparticles with different diameters D, as well as different donor and acceptor concentrations, one can readily observe the two-stage character of the kinetics, when the luminescence intensity is plotted on a doubly logarithmic



Figure 4. Two-stage kinetics of supermigrative luminescence quenching in a bulk crystal and nanoparticles with different donor concentrations; α is the slope, relative to the *x*-axis, of the kinetic curve plotted on a doubly logarithmic scale.

scale and time is plotted on a logarithmic scale: an exponential law for $t < t_b$ with a decay rate close to the kinetic limit in a bulk specimen is replaced with a power-law, nonexponential dependence for $t > t_b$ (Fig. 4a). A rectification of the kinetic curve in $t^{3/S} - \ln(I)$ coordinates is evident at long times (Fig. 4b). As before, from the slope of the curve plotted in $\ln(t) - \ln(\ln(I))$ coordinates for $t \ge t_b$, we find the exponent d/S of the power law and, hence, either the space dimension or the interaction multipolarity. Then, from the slope of the curve in $t^{3/S} - \ln(I)$ coordinates, we find the average quenching rate $W_{nF} = W_F N_D^{S/3-1}$; on substituting c_A , N_D , D, and Sinto it, we determine the microparameter C_{DA} . From the known t_b or from the decay rate at the initial stage, one can find the elementary donor-acceptor transfer rate at the distance of closest approach (in a donor-acceptor pair) or R_{\min} .

Below, we enlarge on several achievements in the development of *modern laser materials* and on the interrelation between their properties and interaction microprocesses at a nanoscopic level in a laser medium.

Figure 5 illustrates a specimen of fluoride laser nanoceramics with no analogue in the world of laser materials, which was developed in our work. The fluoride base was selected, because alkali-earth fluorides possess a broad forbidden band and a narrow phonon spectrum, providing a unique possibility of making laser materials with a record broad wavelength spectrum, from the ultraviolet region to the mid-IR. Furthermore, these materials are sufficiently strong, are moistureproof, tolerate a high level of doping, and possess a high thermal conductivity. Low refractive indices, both linear and nonlinear, permit obviating undesirable losses, spurious oscillations, stimulated Raman scattering, and self-focusing.

Rare-earth (RE) ions in fluoride crystals (for instance, Yb and Er ions) have a small lasing transition cross section and long metastable level lifetimes, which lower superluminescence losses and make the pump source cheaper. This is significant in the making of high-power laser amplifiers and oscillators.

Furthermore, a special feature of alkali-earth fluoride doping by RE ions consists in the fact that the RE ions enter them primarily in the form of clusters — two-, four-, and eight-ion clusters. This must manifest itself in luminescence and laser characteristics. Specifically, while the clusterization of neodymium ions is an adverse factor owing to the Nd–Nd cross-relaxation, which we considered above, for other ions, for instance, Yb, Er, Ho, and Tm, clusterization may well play a positive role. In particular, the theory of migration-



Figure 5. A girl holding in her hand a ceramics specimen obtained by the hot pressing technique.

controlled quenching suggests that the reciprocal of the quantum yield is proportional to the rate of optical excitation migration along laser-active ions and nanoclusters to quenchers. The migration rate, in turn, is proportional to the number of quenchers; uncontrollable impurity ions quite often act as quenchers, which usually imposes the requirement of high purity of initial materials and consequently leads to their high price. However, the quenching rate depends even more strongly on the laser ion concentration N_{las} , the clusterization degree $n_{\rm cl}$, and the interaction multipolarity S [26]: $W_{\rm mig} \sim c_{\rm A} (N_{\rm don})^{(S-2)/3} = c_{\rm A} (N_{\rm las}/n_{\rm cl})^{(S-2)/3}$. Here, $N_{\rm don} = N_{\rm las}/n_{\rm cl}$ is the concentration of clusters along which optical excitation migration takes place. Therefore, by raising the degree of clusterization $(n_{\rm cl} = 2-8)$, we can lower the migration rate $W_{\rm mig} \sim (1/n_{\rm cl})^{(S-2)/3}$ by a power-law factor [the exponent (S-2)/3 varies from 4/3 to 8/3] and thereby weaken the quenching of excitations by a factor of 6–100 (!), depending on the multipolarity order S. That is, by going over

to cluster activation, it is possible to moderate quenching and ensure a higher efficiency and a higher quantum yield in laser and luminescent materials, even for a higher number density of laser particles and quenchers, which makes it possible to relax the requirements on the purity of the starting raw material and to lower its price.

Ytterbium ions in different matrices have aroused considerable recent interest as the working ions under selective optical pumping by laser diodes due to their extremely simple diagram of electron energy levels and the consequential absence of different unfavorable processes, like cross-relaxation or absorption from the excited state, which significantly lower the efficiency of lasing, especially so for high dopant concentrations [27, 28]. The use of ytterbium ions with high enough concentration (2-9%) in fluoride crystals provides a way of obtaining broad absorption spectra, which are convenient to pump by diode lasers, and broad luminescence spectra, which make it possible to realize femtosecond lasing and output wavelength tuning over a rather broad range with a high efficiency [29].

Figure 6a demonstrates the best example of the lasing properties of Yb³⁺-ion doped nanostructured crystals and ceramics under laser diode pumping. The differential efficiency amounts to 50% for crystals, and to 45% for ceramics [30]. A broad spectral lasing range, from 1.01 to 1.09 μ m, permits making either tunable or femtosecond lasers with compact and inexpensive diode pumping [31, 32].

Another important example of laser fluoride ceramics is Nd-doped SrF_2 ceramics. For a low doping level (0.5%), this



Figure 6. Comparison of lasing characteristics of (a) laser fluoride ceramics and a similar CaF_2-SrF_2 : Yb^{3+} single crystal, and of (b) SrF_2 : Nd^{3+} single crystal and SrF_2 : Nd^{3+} ceramics, and SrF_2-LaF_3 : Nd^{3+} ceramics under selective diode pumping and similar experimental conditions.

ceramics is characterized by a high thermal conductivity and a high quantum yield.

The highest-efficiency Nd-ion lasing is realized with Nd–F complexes (L-centers of tetragonal symmetry) possessing one of the shortest laser wavelengths in the ${}^{4}F_{3/2} - {}^{4}I_{11/2}$ optical transition (1.037 µm) and a lifetime of 1.1 ms of the metastable level, which is long for Nd ions [33]. The latter is of considerable importance in simplifying and making cheaper laser diode pumping, as well as in storing high inversion in the laser medium. Should the need arise to raise the concentration of active Nd ions, optically inactive La ions would be used as a co-activator to suppress the aggregation of Nd ions with each other and thereby suppress their self-quenching [34].

Figure 6b represents the best results in the generation of coherent radiation by neodymium ions in laser crystals and the nanoceramics of two types: $SrF_2(Nd)$, and $SrF_2(LaNd)$. A rather high differential efficiency was obtained in a quasicontinuous mode: 24% for the crystal, 19% for $SrF_2(Nd)$ ceramics, and 18% for $SrF_2(LaNd)$ ceramics in measurements under similar conditions [35]. The best result for the crystal in the absence of reflection loss was 37%, which gives hope that new ceramic materials will be competitive with a widely accepted neodymium laser glasses.

Another fresh example of newly developed laser ceramics is provided by $SrF_2: Pr^{3+}$ — the first ceramics operating in the visible spectral region (639 nm), where the requirements imposed on scattering loss are more stringent than in the near-IR region. Furthermore, the short-wavelength blue (445 nm) source required for the laser diode pumping of Pr ions complicates the attainment of lasing still more because of possible radiation scattering, induced loss, and photoionization. We managed to make an $SrF_2: Pr^{3+}$ ceramics which provides a low threshold of red (639 nm) lasing under continuous diode pumping and a sufficiently high efficiency (9%) [36–38]. This opens up the way for producing compact ceramic multicolor [red, green, blue (RGB)] coherent light sources for laser displays and projectors.

Let us consider the example of fluoride crystals, as well as CaF₂: Er and SrF₂: Er ceramics with Er₆F₃₆ nanoclusters intended for mid-IR lasers. In this case, clusterization plays a positive role of fundamental importance: Er³⁺ ions possess a self-terminating laser transition ${}^{4}I_{11/2} - {}^{4}I_{13/2}$ (2.7 µm) with a long lifetime (9 ms) of the lower laser level, and lasing by single Er³⁺ ions is rapidly quenched owing to equalization of the populations of the upper and lower laser levels. Owing to short Er-Er separations in Er₆F₃₆ nanoclusters, a strong upconversion energy transfer $Er({}^{4}I_{13/2}) + Er({}^{4}I_{13/2}) \rightarrow$ $Er({}^{4}I_{11/2})$ with depopulation of the lower laser level ${}^{4}I_{13/2}$ and population of the upper energy level ${}^{4}I_{11/2}$ emerges under high-intensity pumping. This Er³⁺-ion mode, which was first discovered in heavily doped YAG-Er³⁺ laser crystals [39, 40], subsequently enabled realizing the mode of cw lasing. For CaF_2 : Er and SrF_2 : Er crystals exposed to transverse laser diode pumping, it was possible to realize in this mode a threemicrometer cw lasing with a high efficiency (5-7%) and an output power of up to 2 W [41].

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Advances in light sources and displays

A G Vitukhnovsky

Commemorating in 2011 the 120th anniversary of the birth of our outstanding compatriot Sergei Ivanovich Vavilov, an optical scientist, it is pertinent to note that his teacher Petr Petrovich Lazarev was the founder of the journal *Uspekhi Fizicheskikh Nauk*. This relationship imposes certain requirements on the report about modern light sources and alphanumeric displays, given below.

S I Vavilov laid the foundations of the science of luminescence in our country. Apart from Sergei Ivanovich's substantial contribution to the development of basic notions about the nature of luminescence, it was due to his organizational talent that our country obtained new light sources the fluorescent lamps so well known to everyone. Under S I Vavilov's supervision, his associates and students set up an entire branch of power engineering and made a significant contribution to saving electric energy. The high-efficiency phosphors made with the direct participation of S I Vavilov enabled setting up domestic production of TV sets with the shortest possible delay.

A team of scientists supervised by S I Vavilov were awarded the 1951 Stalin (State) Prize for their achievements in the "Development of fluorescent lamps". All recipients of this major award need to be mentioned: S I Vavilov (awarded posthumously), V L Levshin, V A Fabrikant, M A Konstantinova-Shlezinger, F A Butaeva, and V I Dolgopolov. At present, the application of fluorescent lamps, primarily based on thoroughly modernized compact fluorescent lamps, is the solution of choice for illumination.

A few words about the history of light lamps. The year 1872 saw the advent of the first incandescent lamp, which completed the millennial search and revolutionized illumination technology. This happened in Russia, and the first to conjecture the air evacuation from a glass bulb and placing there a carbon rod incandesced by electric current was the brilliant Russian scientist Aleksandr Nikolaevich Lodygin. On May 20, 1873 lamps of his design went on in St. Petersburg. These were eight lanterns with Lodygin lamps. Unfortunately, the pioneer's laurels went not to A N Lodygin but to the outstanding American inventor Thomas Alva Edison, who received the corresponding patent [1]. Edison merely connected with wires a Lodygin

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Uspekhi Fizicheskikh Nauk **181** (12) 1341–1344 (2011) DOI: 10.3367/UFNr.0181.201112k.1341 Translated by E N Ragozin; edited by A Radzig lamp, an electric generator, a socket, and a plug to make a circuit!

Subsequently, carbon rods were replaced with tungsten spirals. The development of such light sources as mercury, halogen, sodium, and xenon lamps continued. These endeavors were undertaken due to the imperfections of incandescent lamps. Being the best in their time (for 70–80 years), incandescent lamps nevertheless possessed several obvious disadvantages, above all a low luminous efficiency. In particular, the first incandescent lamps exhibited a luminous efficiency of only 1.5 Im W^{-1} . Nowadays, it is ten times higher and amounts to $10-15 \text{ Im W}^{-1}$.

About 10 years ago, a new achievement in electronics entered the realm of lighting engineering: third type of light sources (after thermal and gas-discharge) appeared—lightemitting diodes (LEDs). Today, LEDs are no longer exotic and are competent partners of incandescent and gas-discharge lamps. The efficiency of light sources based on inorganic (semiconductor) light-emitting diodes ranges up to 90 lm W⁻¹ [2] over a relatively long lifetime.

The *virtues of light-emitting diodes* are worthy of mention. The electric current in a light-emitting diode, unlike that in an incandescent lamp or a fluorescent lamp, transforms directly into optical radiation rather than into heat, and theoretically this may proceed almost without losses. Indeed, a lightemitting diode barely heats up at all (with due heat removal), which makes it irreplaceable for certain applications. Furthermore, a light-emitting diode emits in a narrow part of the spectrum, its color is pure, which is particularly appreciated by designers, and ultraviolet and infrared radiations are absent, as a rule. A light-emitting diode is mechanically durable and extremely reliable, its service life amounting to almost 100 thousand hours, which is almost 100 times that of an incandescent lamp, and 5-10 times that of a fluorescent lamp. Lastly, light-emitting diodes are lowvoltage electrical appliances and are therefore safe.

The invention of the *organic* light-emitting diode (OLED) [3, 4] in 1987 should be regarded as the next stage of development. Having a low energy consumption, an OLED affords a remarkable color rendering for a low cost and a luminous efficiency of up to 100 lm W^{-1} with the use of phosphorescent organic materials. The characteristics of light sources are collated in Table 1.

At the present time, laboratory specimens of OLED structures exhibit characteristics comparable to those of the best light-emitting diodes from leading world manufacturers. However, it is pertinent to note that the program for the development of the light-emitting diode industry, elaborated by the US Department of Energy (US DOE Solid State Lighting Roadmap, July 2011), follows a strategy whereby the OLED and LED technologies are regarded as mutually complementary technologies rather than competing ones. Among the main disadvantages of the LED are its low overall brightness and a rather poor flexibility. It is precisely this circumstance which gives OLEDs an advantage over LEDs in general illumination systems, for instance, in office lighting.

The world level of OLED technology development has entered the stage of commercialization. This technology accounts for a steadily growing share in the market, which is exemplified by display applications. Considering the scientific and technological achievements, the huge total amount of financing in the world, and the development programs adopted by the leading States and biggest corporations,

Category	Туре	Luminous efficiency, lm W ⁻¹	Radiant efficiency, %
	Candle	0.3	0.04
and the faith and	100-W incandescent lamp (220 V)	13.8	2.0
	Linear fluorescent lamp	60	9.0
	White light-emitting diode	10-90	1.5-13
	White OLED	102	15.0

Table 1. Characteristics of different light sources.

OLED technology in the area of lighting will undoubtedly meet with success.

We now turn to advancements in the area of alphanumeric displays (Fig. 1). Quite evident is the progress in connection with changing from classical displays based on electron-beam tubes (recall bulky 'Rubin' TV sets and the dreams of Sony TV sets) to fine plasma panels, and subsequently to modern liquid crystal (LC) monitors which not only are the screens of modern TV sets and notebooks, but also are used in a countless number of so-called gadgets (cell phones, navigators, etc.). However, progress is unstoppable, and different versions of organic light-emitting devices come up to take the place of LC displays: a polymer lightemitting diode (PLED) based on conducting polymers; an OLED based on 'small' molecules of organometallic complexes, and a QD-OLED [an organic matrix with quantum dots (QDs) implanted into it] which makes use of flexible substrates and hybrid materials.

Analysts at Research and Markets (Dublin, Ireland), the leading source for international market research and data, are certain that displays which rely on organic light-emitting diode (OLED) technology will become the main 'engine' of the industrial sector in the next decade. In any case, this





Figure 2. Energy level structure of the most popular colloidal cadmium chalcogenide quantum dots.

conclusion was reached in their report "Energy Efficient Displays Technologies to 2020—Organic Light Emitting Diodes (OLED) Displays Set to Propel Growth of the Industry."

OLED displays have a huge market potential: according to experts' estimates, their sales volume will reach 10.6 billion dollars by 2020.

For several years, OLED technology has been believed to hold the greatest promise for displays development. In the period from 2005 to 2009, the corresponding market grew on average by 33.9% per year to expand from 256 to 822 million dollars. In the next ten years, an annual growth of 25.5% will persist, according to the analysts at Research and Markets. To date, mobile devices are the main application area of OLED displays. The displays employed in cell phones (this segment now accounts for 65% of the total volume in money terms), digital cameras, players, and other devices of this sort are small in size. The demand will begin to grow when OLED panels of large area come to TV sets, monitors, and personal computers.

Organic displays will supposedly take the place of liquid crystal ones. Therefore, the main factors which moderate the spread of OLEDs are the constant improvement and cost reduction of LC displays. However, analysts believe that technological innovations and a change to mass production will allow reducing the cost of OLED displays.

There is good reason to enlarge on the latest achievement in the area of modern displays — QD-OLED technology. The key element of devices of this kind is a colloidal quantum dot (a nanocrystal) 2–7 nm in size. As a rule, use is made of socalled core–shell quantum dots. Quantum dots prepared by the colloidal chemistry methods [5] are exemplified in Fig. 2, which also depicts the behavior of their energy levels.

Also shown in Fig. 2a are a shell-free CdS quantum dot and the spatial distance distribution of electrons and holes. A CdSe/CdS quantum dot (Fig. 2b) corresponds to the so-called type I nanoheterostructure, where electrons and holes are located in the core. A CdTe/CdSe quantum dot (Fig. 2c) exhibits a different distribution of electrons and holes. This nanoheterostructure belongs to type II: its electron resides primarily in the shell, while its hole is located in the nanoparticle core. (For the classification of heterostructures, see, for instance, Ref. [6].)

By placing quantum dots between two n- and p-type organic conductor layers and applying voltage to the outer electrodes, it is possible to excite the quantum dots and



Figure 3. Operating diagram of the simplest organic light-emitting diode (display pixel) with semiconductor quantum dots (QD-OLED).

eventually obtain electron-hole radiative recombination [7]. The simplest schematic diagram of a QD-OLED is depicted in Fig. 3.

An obvious advantage of using this scheme is the possibility of tuning the radiation wavelength, which is determined only by the nanoparticle size, as well as its stability (durability, which is achieved by using an inorganic material as the emitter) and the low cost of colloidal nanoparticle synthesis. The high quantum yield of quantum dot electroluminescence is not the least of the factors as well.

Figure 3 demonstrates how the energy structure changes when moving from a bulk material (Fig. 4a, E_g is the energy gap width) to a nanodimensional (Fig. 4b, E_g is the lowest transition energy) one. This quantity is defined by a simple formula: $\Delta E = = h/(2md^2)$, which relates the diameter d of a nanoparticle (quantum dot) to the transition energy.

Clearly, by changing the nanoparticle (quantum dot) size it is possible to obtain radiation in different parts of the visible spectrum, which is required for obtaining a full-color display.

At the present time, the excitation mechanism of quantum dots in QD-OLEDs is still unclear. Let us consider the possible quantum-dot excitation mechanisms illustrated in Fig. 5.

Direct electron-hole recombination may occur at a quantum dot, resulting in its excitation and the consequential emission of a photon with the appropriate energy.



Figure 4. Energy structure of two types of objects: bulk (a), and nanodimensional (b).



Figure 5. Mechanisms of excitation of quantum dots at the interface between n-type (electron transport) and p-type (hole transport) organic layers.

Another scenario is also possible: electron-hole recombination may occur either in the n-type organic layer or in the ptype layer. In these cases, electron excitation energy transfer proceeds from the excited molecule in the organic layer to the quantum dot. Such an energy transfer follows the Förster mechanism [8]. In Ref. [9], for instance, an investigation was made of electron excitation energy transfer from a blue-lightemitting organic conjugated poly[(9,9-dihexylfluorenyl-2,7diyl)-alt-co(9,ethyl-3,6-carbazole)] polymer to a colloidal CdSe/ZnS core-shell quantum dot. It was shown that the energy transfer proceeded by the Förster mechanism and the polymer acted as the donor, while the quantum dot as the acceptor. The Förster radius was determined equal to (80 ± 15) Å. Investigations into the processes at the organic layer-quantum dot interface are required to make an efficient display pixel. There are also several physical problems which need to be solved to optimize the operation of QD-OLEDs.

It is well known that a quantum dot exposed to continuous excitation emits light discretely [10]. This effect is termed blinking luminescence. Figure 6 exhibits the luminescence intensity of a single quantum dot under continuous excitation. One can clearly see the intervals of light emission and the intervals without light.

The time-varying luminescence intensity of a quantum dot under continuous excitation is characterized by the states



Figure 6. Blinking luminescence of a single quantum dot.



Figure 7. Association of the blinking luminescence of a quantum dot with the capture and release of a charge carrier by a trap.



Figure 8. Illustration of the role of the thickness of a core-shell quantum dot shell.

'ON' (light is emitted) and 'OFF' (light is not emitted). One explanation of this effect reduces to variation of the charge of the quantum dot. A dot which 'loses' charge does not exhibit luminescence, but after a lapse of time the charge may 'return' (this is in fact the capture and release of charges by traps at the interface between the quantum dot and the surrounding medium). This process is schematically presented in Fig. 7. Such a phenomenon plays an adverse role in the making of an efficient display pixel. In this connection, it is necessary to investigate the cause of blinking luminescence and find ways to suppress it. There are only primary indications of what role the core–shell quantum dot thickness plays in this phenomenon (see, for instance, Ref. [11]). Figure 8 depicts the blinking luminescence of a quantum dot with different shell thicknesses.

In a brief report it is impossible to cover all aspects of the progress in the area of modern light sources and displays, but even the individual case of employing a QD-OLED as a pixel makes evident the vast possibilities of using hybrid nanomaterials. Combining colloidal semiconductor nanoparticles (quantum dots) and organic interfaces does lead to a qualitatively new display type, which possesses a long operating lifetime, a high luminous efficiency, and the possibility of tuning the output radiation wavelength throughout the visible spectral range. It is evident that optoelectronic devices of this kind would be in demand by the industry. On the other hand, the complexity of the organic interface–quantum dot system is of considerable interest for fundamental physics.

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Direct experimental demonstration of the second special relativity postulate: the speed of light is independent of the speed of the source

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In memory of S I Vavilov and his Postdoc A M Bonch-Bruevich

1. Introduction

Special relativity is undoubtedly the most famous physical theory. The popularity of the special theory of relativity (STR) is related to the simplicity of its main principles, the

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Uspekhi Fizicheskikh Nauk **181** (12) 1345–1351 (2011) 10.3367/UFNr.0181.2011121.1345 Translated by V S Zapasskii; edited by A Radzig imagination-staggering paradoxicality of the conclusions, and its key position in 20th-century physics. Special relativity has brought unprecedented fame to Albert Einstein, and it is this fame that became one of the reasons for incessant attempts to revise the theory. Among professional physicists, the debates around STR ended more than 50 years ago. A quotation from Wikipedia: "All the experimental data of the high-energy physics, nuclear physics, spectroscopy, astrophysics, electrodynamics, and other fields of physics, within the experimental errors, perfectly agree with the STR. In particular, in quantum electrodynamics (unification of the STR, quantum theory and Maxwell equations), the value of the anomalous magnetic moment of electron coincides with theoretical calculations to within 10^{-9} ."

Still, editorial boards of physical journals continue to be bombarded by amateurish proposals to revise the STR [1] (see also paper [2]). In spite of an infinite amount of evidence of the validity of the STR available nowadays, efforts to refute or to essentially revise it do not cease, being motivated by the insufficient reliability of experimental confirmations of its basic principles, including, in particular, its second postulate, which states the constancy of the speed of light for all inertial reference systems regardless of the light source velocity. It is noteworthy that, most frequently, the criticism is directed at earlier experiments aimed at searching for the 'ether wind' [3], which were traditionally considered as almost the only experimental proof of the validity of the STR. While not penetrating into the pages of serious scientific literature, the attempts to revise the STR overwhelm the mass media and Internet, which cannot help disorienting unprofessional readers, including schoolchildren and students. The situation was additionally aggravated in the years of celebration of the centenary of the relativity theory, counted from the date of publication of the historical article by Einstein [4], considered as the birthday of the STR.¹ At the same time, distrust of the STR (from the side of social community unencumbered by knowledge) also existed 60 years ago, when S I Vavilov charged his PhD student A M Bonch-Bruevich with the experiment on direct verification of the second postulate of the special relativity [10].

The incessant attacks on the STR are motivated by discrepancies in evaluation and interpretation of the first relativistic experiments by Fizeau, Michelson, and others. Specifically, one of Michelson's successors — Miller [11] — insisted, until his last years, that, in those experiments, a certain seasonal systematic effect was observed, which he interpreted as a partial drag of the 'luminiferous ether' by Earth upon its orbital motion around the Sun. After definitive establishment of the validity of the STR these experiments have practically ceased to be reproduced, with the accuracy of such measurements still remaining rather low.

There are few who know that the first famous negation of the existence of the 'ether wind' was made by Michelson [12] in 1881 on the basis of rather unconvincing observations. The achieved accuracy of the measurements only slightly exceeded the magnitude of the effect proper expected based on the

¹ Of the innumerable critical publications, we will restrict ourselves by mentioning only two: the review article of N Noskov, divesting 'centennial relativistic fraud' [5], and the recent publication by Sokolovs [6] reviving the old 'ballistic' hypothesis of Ritz [7]. The jubilee of the STR was celebrated in a peculiar way by St. Petersburg Polytechnical University, which published again, in 2009, the pretentious monograph *Myths of the Relativity Theory* by A A Denisov [8], whose extravagant constructions had been refuted by lecturers of the same university 20 years ago [9].

hypothesis of a 'fixed luminiferous ether'. (It is not a surprise that Einstein did not want to acknowledge this experiment as the one that inspired him to create the STR). In subsequent experiments, much more definite results have been obtained. However, usually some systematic component of the velocity of ether wind (about 10% of the velocity V of the orbital motion of Earth) was observed. Only in the late 1920s was a sufficiently definite negative result achieved: the upper bound of the ether wind velocity was reduced to $\sim 3\%$ of V [3]. Further refinement of these results soon lost its topicality in view of much indisputable evidence of the STR validity accumulated in the process of development of nuclear physics and accelerators, which themselves could not be constructed without using the relativity theory. This knowledge, however, remained the domain of professionals, while popular presentations of the STR traditionally appealed to the Michelson experiments as the only justification of the STR. It is exactly this gap between understanding the measure of validity of the STR by professionals and the wider public that stimulated President of the Academy of Sciences of the USSR S I Vavilov to demonstrate in the middle of the last century the independence of the speed of light from the source velocity in a 'first-order' experiment. Vavilov planned direct measurements of the speed of light c emitted by a source moving with a high speed v, in contrast to indirect measurements in Michelson's experiment, where the expected effect was to be proportional to the square of the ratio v/c.

At that time, the postulate of the independence of the speed of light was explicitly supported only by astronomical observations of double stars. According to de Sitter's idea [13], if the speed of light is dependent on the source velocity, the trajectories of motion of binaries should crucially differ from the observed ones (consistent with celestial mechanics). His argument, however, encountered objection (reproduced in Ref. [6]) related to the role of interstellar gas which, as a refracting medium, should have been regarded as a secondary source of light. From this point of view, the light emitted by a moving source loses the memory of its initial velocity, while propagating in the interstellar medium. Since the characteristics of this medium are known with poor accuracy (as are the absolute distances to the stars), such a position allows one to cast doubts upon most astrophysical proofs of the constancy of the speed of light. (In particular, this kind of criticism was directed at a far more later publication [14], in which the question of existence of variable stars was used as radical proof of the validity of the second postulate of special relativity: for the linear dependence of the speed of light on the source velocity, the light of star of a variable brightness should lose the intensity modulation with increasing distance due to thermal spread of the ray velocities of elementary emitters. So, such stars, in this case, would have been unknown.)

S I Vavilov proposed that his Postdoc working for the degree of Doctor of Sciences, A M Bonch-Bruevich, design a setup with a beam of fast excited atoms as the light source. In the process of detailed elaboration of the would-be experiment, it was found that there was no chance of getting a reliable result because, for the experimental technique of that time, it was impossible to produce beams with the needed velocity and density: the increment of the speed of light in the framework of ballistic theory was expected to be about a few percent, while the light beam intensity was estimated to be too low. The experiment was not realized. After the premature death of S I Vavilov in January 1951, the plan of the

experiment was revised on G S Landsberg's initiative, who proposed comparing the speed of the light emitted by two equatorial edges of the rotating Sun. A M Bonch-Bruevich wrote 50 years later [10]: "This proposal deprived the experiment of its original elegance, but was perhaps the only opportunity to lead it to the end even in a strongly deformed shape." The result of this experiment, however, could not be considered proof of the independence of the speed of light of the source velocity, because the light from the Sun was transmitted through a glass objective of a telescope which, in conformity with the concept of reemission of light by the refracting medium, should have equalized the velocities of the two light beams (to say nothing of the effect of Earth's atmosphere).

Since then, attempts to experimentally prove the second postulate of the STR have been repeatedly undertaken (see, e.g., monographs [15, 16] and recent comprehensive reviews by G B Malykin [17, 18]). All the authors of these papers arrived at the conclusion of the validity of the postulate. But this could not cease the flow of critical publications, in which objections against the ideas of the experiments were put forward or their accuracy was questioned. The latter was related, as a rule, to the smallness of the light source velocity compared to the speed of light. The revival of interest in the ballistic hypothesis happened in 1962, when the experimental work of W Kantor, who allegedly discovered changes in the speed of the light passed through a moving glass plate was published [19]. Kantor's work raised a wide discussion, but soon after its results were refuted on the basis of test experiments. Nevertheless, the ballistic hypothesis still remains popular among critics of the STR. In 1980, the Presidium of the Academy of Sciences of Ukraine supported setting up large-scale experiments with M I Duplishchev to check the Ritz hypothesis. The experimentalist, following Kantor's concept, measured the speed of the light passing through a fastly moving refracting medium, which was considered a secondary light source. The author came to the conclusion of the validity of the idea of summation of the speed of light and light source velocity in agreement with the 'corpuscular (ballistic) Newton-Ritz theory,' but did not manage to publish his results in respectable journals. In 2008, an account of these experiments [20] was published by his daughter on a commercial basis.

It seems to us that it is high time to return to S I Vavilov's proposal. Now, that idea can be realized in its 'original elegance', because at present physics has at its disposal an extremely bright ultrarelativistic source. This is a synchrotron emitter with the light emitted by a bunch of electrons moving along a curved trajectory with velocity very close to the speed of light. Under these conditions, the speed of light in a perfect laboratory vacuum can be easily measured. Following the logic of the ballistic hypothesis, this speed should be equal to double the speed of light from a fixed source! This is a very rough effect whose discovery (if it exists) would not require resorting to any special tricks. Indeed, it suffices, for this purpose, to measure the time of flight of a measured distance by the light pulse in a vacuum.

Leaving aside for the moment the details and concrete versions of the experiment, it makes sense to summarize arguments in favor of the expediency of its arranging. Of course, for professional physicists, there are no doubts about the results of such an experiment. In this sense, the experiment seems useless. However, the direct demonstration of the constancy of the speed of light has great tutorial value,



Figure 1. Schematic of the experiment: M1-M4—corner magnets, IOS—glass plate input/output system, L—collecting lens, D—photodetector, CU—control unit for the IOS, L1 and L2—induction coils.

restricting the room for further speculations about the insufficient reliability of the foundations of the theory of relativity. Physics, in its evolution, returned repeatedly to the reproduction or refinement of its basic experiments by implementing them with new technical tools. In this case, we do not mean to measure the speed of light with a higher accuracy; we are talking about filling the gap in the experimental validation of the STR basic principles, which should simplify the perception of this fairly paradoxical theory. We may say that we are dealing with a demonstrative experiment for future textbooks on physics.

2. Experimental

As a pulsed light source, we utilized in these experiments a source of synchrotron radiation (SR)—the Siberia-1 electron storage ring at the Kurchatov Center of Synchrotron Radiation and Nanotechnologies of the National Research Centre 'Kurchatov Institute' (NRC KI) [21]. A general view of the Siberia-1 storage ring with a schematics of the SR output and experimental setup is presented in Fig. 1.

The magnetic system of the Siberia-1 electron storage ring forming the closed electron orbit comprises four corner magnets (M1–M4) separated by four 60-cm-long rectilinear segments. The magnetic field induction in the stationary electron orbit reaches 1.5 T. The radius R of the stationary electron orbit in the corner magnets is R = 1 m. The nominal electron energy in the storage ring amounts to 450 MeV. Synchrotron radiation created by relativistic electrons in the corner magnets extends over a wide spectral range — from IR and visible to X-ray, with the characteristic wavelength of 61.3 Å. The SR leads to an energy loss of 3.69 keV per round trip for each electron in the beam.

To compensate for the radiative loss of the electron beam in each turn, a radio-frequency (RF) resonator is placed in segment l of the storage ring. The power provided by the RF oscillator creates, at the accelerating gap of the resonator, a voltage with an amplitude of 15 kV and a frequency of 34.53 MHz, equal to that of the electron bunch orbital



Figure 2. Vacuum unit of the glass plate input/output system.

rotation. Under these conditions, the longitudinal distribution of electron density in the bunch is Gaussian with a standard halfwidth of 0.3 m.

The angle between the axis of the SR output channel, which is tangent to the stationary orbit in magnet M3, and the axis of the fourth rectilinear segment, succeeding magnet M3, comprises 30°. This means that the emission point (the beginning of the path of the SR along the channel axis) is at a distance of $\pi R/3$ from the input end of magnet M3. The length of the channel from the point of emission to the output sapphire window measures 7.2 m.

A glass plate input/output system was placed in the SR channel, at a distance of 1.8 m from the point of emission. It was designed as a vacuum unit (Fig. 2) with a movable frame inside it with two apertures. One of these apertures is supplied with a 1 mm-thick glass window. The frame is mounted on small wheels so that it can move across the axis of the SR channel and be fixed in two extreme positions: in one position, the SR beam passes through the glass plate, while in the other, it passes through the open aperture. The butt end

of the movable frame is supplied with a small permanent magnet, while on the outer tube, surrounding the frame, two magnetic coils are mounted, connected oppositely. Upon feeding a DC voltage from the control unit to the coils with one polarity or the other (Fig. 3), due to the interaction of the permanent magnet with the field of the coils, the frame moves in one direction or the other. In this way, the glass window is moved into the zone of the SR beam or is removed from this zone.

The experiment was run using two setups. In the *first*, the time of flight of the light pulse through a fixed distance in a vacuum was measured for two cases: (i) the SR pulse entered the test segment passing through the open aperture of the frame, and (ii) the SR pulse entered the test segment passing through a hole with a thin glass window transparent in the visible spectral range. In the *second* setup, the speed of the light pulse in the vacuum was measured in a straightforward manner: the distance passed by the light was divided by the time of flight.

At the butt end of the vacuum tube of the channel was mounted the output flange with a sapphire window 2.4 cm thick (transmission range $0.17-5.5 \mu m$). At a distance of 3 cm from the output window was placed a collecting lens 1.4 cm thick which focused the SR beam on the sensitive area of the photodetector mounted at a distance of 7 cm from the lens. As the photodetector, we used an Si pin-photodiode Hamamatsu S5972 (spectral range $0.32-1 \mu m$, bandwidth 500 MHz, effective area of sensitivity 0.5 mm²). A schematic of the pindiode circuit is shown in Fig. 4.

When the light pulse hits the pin-diode, the voltage created by the photocurrent on the load resistor (50 Ω) is fed, through an RF cable, to one of the inputs (50 Ω) of a Tektronix TDS3052C two-channel oscilloscope (bandwidth 500 MHz). On the other input (50 Ω) of the oscilloscope, a synchronizing sinusoidal RF signal is fed from the pick-up loop of the resonator. To exclude errors related to phase shifts of the signals, we used cables of the same type (RC-50) and the same length (8 m) for transporting the useful signal from the load resistor of the pin-diode and the signal of synchronization.



Figure 3. Schematic of the control unit.



Figure 4. Schematic electric circuit of the photodetector with pin-diode.

Thus, the observer should see in the ideal case a comb of successive near-Gaussian periodic SR pulses with a standard width of 1 ns following each other with a frequency of 34.53 MHz, which are overlapped onto a sinusoidal signal with a frequency of 34.53 MHz.

3. Experimental results

Setup 1. Figures 5 and 6 display experimental oscillograms of pulses of the pin-diode and synchronization signals from the resonator loop. The oscillograms were recorded for the same electron current in the storage ring. As one can see, they are identical from the viewpoint of phase relations between the signals. In other words, the position of the pulses from the pin-diode remained the same with respect to the synchronization signals, regardless of whether the light passed through the glass plate or not.

This means that the speed of the light emitted by the relativistic electrons in a vacuum is equal to the speed of the light that passed through the glass plate in a vacuum, i.e., it does



Figure 5. The light signal (channel 1) for the case of light traveling in a vacuum through the test segment with an open input aperture and the synchronization signal (channel 2).



Figure 6. The light signal (channel 1) for the case of the light traveling in a vacuum through the test segment with the input aperture closed by the glass plate and the synchronization signal (channel 2).

not depend on the light source velocity. This result refutes the Ritz ballistic hypothesis.

Indeed, if the speed of the light emitted by the relativistic electrons in a vacuum were equal to 2c, the light would move along a path 7.2 m long from the point of emission to the output sapphire window in 12 ns.

On the other hand, if the glass plate is placed in the path of the SR beam at a distance of 1.8 m from the point of emission, this plate becomes a source of secondary emission propagating in the forward direction with the velocity c. In this case, the time taken by the light to pass the distance of 7.2 m should be the sum of the times needed to pass the distance from the point of emission to the glass plate (1.8 m) and the distance from the glass plate to the output sapphire window (5.4 m). This would give, in total, 3 ns + 18 ns = 21 ns.

Thus (neglecting the short time delay of light in the glass plate related to its own refractive index n > 1), we should detect, on the oscillograms presented in Figs 5 and 6, a phase shift between the optical signals, corresponding to the different instants of appearance of the two input signals:

21 ns - 12 ns = 9 ns.

U

 $U_{\rm m}$

 $U_{\rm RF}$

0

 φ

It should be noted that the amplitude of the light signal on the oscillogram of Fig. 5 (the aperture with no glass plate) is slightly smaller than that on the oscillogram of Fig. 6 (the aperture with the glass plate), which is explained by insignificant vignetting of the light beam in the horizontal direction by the side edges of the glassless aperture.

Setup 2. Now, using the oscillograms obtained in the first experimental setup, we can estimate the speed of the light emitted by relativistic electrons in vacuum.

The path length *l* passed by the light pulse from the point of emission to the output sapphire window equals 7.2 m. To calculate the speed of light, we have to measure the time taken by the light to traverse this distance. To do this, we have to know the instant when the electron bunch passes the point of emission in the stationary orbit of the storage ring inside the magnet M3 (see Fig. 1). As a synchronizing signal, we can use the voltage from the pick-up loop of the RF resonator. This loop is oriented in the resonator in such a way that the phase of its output voltage is shifted by 180° with respect to the voltage across the accelerating gap of the resonator. The synchronized electrons pass through the gap at a certain phase φ_s of the accelerating voltage. With a knowledge of

 $\Delta \phi$



this phase and taking into account the configuration of the stationary orbit of the Siberia-1 storage ring, we can calculate the instant of time (phase) when the electron bunch passes the point of emission.

Figure 7 shows phase dependence of the voltage across the accelerating gap of the resonator. The condition of stability of motion of relativistic electrons in the storage rings requires the electron bunch to pass through the gap of the RF resonator when its voltage decreases. In other words, the equilibrium phase φ_s corresponds to the falling portion of the RF-voltage curve.

The quantity φ_s can be calculated based on the following reasons. Each time that the synchronized particle passes through the gap of the RF resonator, it acquires the energy increment $\Delta W = 3.69$ keV compensating for the SR-related loss per round trip. In turn, one has

$$\Delta W = q U_{\rm RF} \,, \tag{1}$$

where q = 1 is the electron charge, and U_{RF} is the voltage across the gap of the resonator at the moment of passage of the synchronized particle. It is known that

$$U_{\rm RF} = T U_{\rm m} \cos \varphi_{\rm s} \,. \tag{2}$$

Here, φ_s is the phase of the synchronized particle counted from the peak of the voltage, T = 0.99 is the time-of-flight coefficient defined as a result of averaging of the accelerating RF voltage in the resonator gap, both over time and over longitudinal coordinate. Notice that in the case of a uniform strength of the RF field in the accelerating gap of the resonator of length *d*, we have

$$U_{\rm RF}(t) = U_{\rm m} \cos \left(\omega t + \varphi\right), \quad T = \frac{\sin \psi}{\psi},$$

where $\psi = \pi d/\lambda_{\rm RF}$, $\lambda_{\rm RF}$ is the wavelength of the accelerating voltage, and $U_{\rm m} = 15$ kV is the RF voltage in the resonator. From Eqn (2), with allowance for Eqn (1), we find

$$\varphi_{\rm s} = \arccos\left(\frac{\Delta W}{qTU_{\rm m}}\right).\tag{3}$$

Substituting all the known values into formula (3), we obtain

$$\varphi_{\rm s} = \arccos\left(\frac{3.69}{0.99 \times 15}\right) = 75.61^{\circ}.$$

From here, in accordance with Fig. 7, one finds

$$\Delta \varphi_{\rm s} = 90 - 75.61 = 14.39^{\circ} \,,$$

or, in time units, with allowance made for the fact that the RF oscillation period (with a frequency of 34.53 MHz) is equal to 28.96 ns, it gives

$$\Delta t_{\rm s} = \frac{14.39^{\circ} \times 28.96 \text{ ns}}{360^{\circ}} \approx 1.16 \text{ ns}.$$

Let us calculate the electron-bunch time of flight, t_e , of the path *L* from the accelerating gap in the resonator to the point of emission in the magnet M3. First, based on the geometry of the Siberia-1 storage ring (see Fig. 1), we find the path length *L*:

$$L = 0.22 \text{ m} + \frac{\pi R}{2} + 0.6 \text{ m} + \frac{\pi R}{2} + 0.6 \text{ m} + \frac{\pi R}{3}$$
$$= 0.22 + 1.57 + 0.6 + 1.57 + 0.6 + 1.05 = 5.61 \text{ m},$$

where R = 1 m.

 φ_{s}



Figure 8. Measuring the light pulse delay in the photodetector with respect to the phase of the synchronized particle in the accelerating gap.

Now, taking into account that the velocity of the ultrarelativistic electrons is virtually equal to the speed of light ($c = 2.997924 \times 10^8 \text{ m s}^{-1}$), for the time of flight t_e of the path length L by the electron bunch, we have

$$t_{\rm e} = \frac{L}{c} = \frac{5.61 \text{ m}}{2.998 \times 10^8 \text{ m s}^{-1}} \approx 18.71 \times 10^{-9} \text{ s} = 18.7 \text{ ns.}$$

Consider now the oscillogram depicted in Fig. 8, which reproduces the oscillogram of Fig. 5. In our case, the phase of the voltage (synchronization signal taken from the pick-up loop in the resonator) is shifted by 180° with respect to that of the voltage across the accelerating gap (which is schematically shown in Fig. 8 by the dashed line).

Summing up the above time interval $\Delta t_s = 1.16$ ns and the time interval $\Delta t \approx 42.20$ ns measured by the oscilloscope between the fixed cursors (solid and dashed vertical lines on the oscillograms), we find, from the oscillogram of Fig. 8, the time delay t_{sum} in the light signal appearance in the detector with respect to the moment of passing through the accelerating gap by the corresponding electron bunch:

 $t_{\rm sum} \approx 43.36 \text{ ns}$.

We consider here the light pulse B emitted by the electron bunch entering the accelerating gap of the resonator at the phase φ_s , whereas the light pulse A is emitted by the electron bunch that had entered the gap one round trip earlier. By subtracting, from this delay, the time of flight t_e of the path length L by the electron bunch, we find the time t_d for the light to traverse the path from the point of emission to the detector:

 $t_{\rm d} = t_{\rm sum} - t_{\rm e} \approx 43.36 \text{ ns} - 18.71 \text{ ns} = 24.65 \text{ ns}.$

The total path length of the light pulse counted from the entrance into the sapphire window to the detector equals 13.8 cm: 2.4 cm (sapphire window, refractive index of sapphire n = 1.765) + 10 cm (air, 3 cm before the lens and 7 cm after the lens) + 1.4 cm (glass of the lens with the refractive index n = 1.52). Light characterized by a speed in a vacuum of 30 cm ns⁻¹, with allowance made for the refractive indices in sapphire and glass, traverses this path in 0.55 ns. The time

delay of the electric signal formation in the detector is neglected here.

Bearing in mind the last remark, we find t_{SR} —the time taken by the light to pass the distance from the point of emission to the output sapphire window (l = 7.2 m):

$$t_{\rm SR} = 24.65 \text{ ns} - 0.55 \text{ ns} = 24.10 \text{ ns}$$
.

And, finally, we evaluate the speed of the light emitted by relativistic electrons in a vacuum to be

$$c_{\rm SR} = \frac{l}{t_{\rm SR}} = \frac{7.20 \text{ m}}{24.10 \times 10^{-9} \text{ s}} \approx 2.99 \times 10^8 \text{ m s}^{-1}.$$

The result thus obtained differs from the CODATA recommended value of the speed of light in vacuum by no more than 0.5%.

4. Remarks

In the course of the experiments, considerable efforts were made to eliminate stray pick-up of the accelerating RF voltage to the optical signal detection channel. This synchronized pick-up contained several harmonics of the fundamental frequency and thus strongly distorted initially the useful signal. We managed to get rid of it practically completely using double-screened cables, both in the signal and in the synchronizing channels. The degree of suppression of the interference is demonstrated in Fig. 9, which reproduces Fig. 6 with the photodetector covered with black paper.

The width of the observed optical signal well correlated with the expected value, while its shape revealed a spurious 'ringing' at the trailing edge of the pulse, related to oscillatory processes in the photodetector electric circuits. This distortion of the signal, however, did not affect the accuracy of the measurements. Some idea about achieved measurement accuracy is given by Fig. 10, demonstrating oscillograms of the optical and synchronization signals after digital averaging. The time scale is here extended by a factor of 2.5 compared to the previous oscillograms.

A brief synopsis of this paper is given in Ref. [22].



Figure 9. Residual signal of the RF stray pick-up (channel 1) and the synchronization signal (channel 2).

4 ns

Figure 10. The noise-suppressed signal extended in time.

5. Conclusions

In this work, we directly measured for the first time (to the best of our knowledge) the speed of the light emitted by an ultrarelativistic source. The results obtained here are incompatible with the Ritz ballistic hypothesis which implies adding the speed of light to the light source velocity. It is shown that inserting a glass plate into the light beam does not affect the speed of its propagation to within fractions of a percent, whereas, according to Ritz's hypothesis, the speed of light after its passing through a fixed window should decrease by a factor of 2. The measurements of the speed of light pulse in a vacuum yielded a value differing from its table value by less than 0.5%. The results of the measurements can be considered as the most straightforward evidence of the validity of the STR second postulate.

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Sergei Ivanovich Vavilov as a historian of science

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1. Introduction

"One can hope that the history of science will sometime itself become science. A warrant of this is the obvious growth of natural science and technology and hundreds of thousands of people creating the history of science on the globe in our sight. It is impossible to ignore this powerful natural phenomenon

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which is capable of changing the Earth no less drastically than earthquakes and floods do" (S I Vavilov [1], pp. 3, 4).

In the Soviet period, along with the rapid growth in physical studies, the literature on science history, which was first created by physicists themselves (P P Lazarev, V K Frederiks, A N Krylov, S I Vavilov, et al.), also grew quite rapidly. The work of A N Krylov and S I Vavilov devoted to Isaac Newton, including translations of his Principia and Opticks, corresponded to the highest professional level of historic scientific studies. This work formed the basis for the professional history of physics (at the Institute of the History of Science and Technology in Leningrad and at the Department of Physics of M V Lomonosov Moscow State University). However, the institute in Leningrad headed by N I Bukharin was closed after a while, and the second wave of the institualization of the history of science and technology took place in the postwar years. The Institute of the History of Natural Science and Technology of the Academy of Sciences appeared in Moscow during this wave.

Notice that S I Vavilov played a key role (organizational and conceptual) in the formation of the professional history of science, in particular, physics, both in the prewar and in the postwar periods. Some of his studies are still archetypes of historic scientific studies (Vavilov's book about Newton and the series of his works on the history of optics in the 17th–18th centuries). They contain certain general concepts about the structure of physical knowledge and its development; the relationship between an experiment, the physical foundations of a theory, and its mathematical apparatus; mechanisms of the appearance of a new scientific knowledge and its interaction with social institutions, etc. It is such a scope of concepts that we will call the historiographic concept.

We will consider in our report the historiographic concept of S I Vavilov, whose name was given to the Institute of the History of Natural Science and Technology, RAS not by accident. Beginning from the 1920s and especially from the early 1930s and to his last days, Vavilov combined in an extraordinary way huge organizational and research activities in the field of optics with investigations into the history of physics. A brief chronological and bibliographic reference characterizing S I Vavilov as a historian of science is presented in the Appendix.¹

2. Historiographic concept of S I Vavilov

"...The history of science can and must be the true and only 'theory of knowledge' instead of many artificial epistemological constructions..." (S I Vavilov [1], p. 7).²

The key points of the historiographic concept of S I Vavilov were presented in a 1933 article "Old and new physics" [1] in the collection *To the Memory of Karl Marx* reproduced in the collection *The History and Methodology of Natural Sciences* in 1965, and also in an article "Physics" in 1936 in the *Great Soviet Encyclopedia*, Vol. 57 [3].³

S I Vavilov's contemplations about the driving forces and basic processes governing the history of science often appeared in his diaries as well, to which he made entries each free minute.

Vavilov's historiographic concept can be approximated by the following points.

(i) The history of science must tend to become science, not in the sense of its reduction to 'naive' logical schemes withdrawn from the live human and social and cultural context, but rather in the sense of some synthesis of sciences of natural and humanitarian cycles taking these contexts into account.

(ii) Among the numerous factors determining the subjectmatters and specific features of the development of science, there always exist a small number of dominating factors. Their determination is one of the key issues of a historical scientific investigation.

(iii) "The continuous line of the development of science contains some 'singularities', which look like turning points in history," 'scientific upheavals' of a certain type or scientific revolutions, although the latters (unlike the approach of T Kuhn) follow the concept of the continuity of scientific development.

(iv) The consistent historiographic concept should comprise the description of the process of reception of a new scientific knowledge; only this process can explain the succession and continuity of the development of scientific ideas and theories.

(v) The historical experience acquired from physics teaches that theories are the fundamental systematic units of scientific knowledge and that there are three basic methods of their construction (at least in the field of exact natural science): the method of hypotheses-models, the method of principles, and the mathematical hypothesis method.

(vi) The true theory of knowledge and philosophy of science should be based on the history of science (or even more bluntly, they are themselves the conceptualized history of science or the theoretical historiography of science).

3. The history of science must tend to become science

"...The history of science is a necessary and maybe even sufficient prerequisite for planning science. Therefore, sooner or later the history of science should become science" (S I Vavilov [1], p. 4).

The history of science has grown and become a large special field; however, the problems indicated by Vavilov are still urgent. We will describe them briefly, giving the floor to S I Vavilov himself. Below, we present a mosaic of citations with brief comments.

On the scientific nature of the history of science and planning science based on it: "To understand this process (the growth of natural science and technology — *Authors*), as always, means to master it in many ways and to learn to direct it in the required direction. The history of science is a necessary and maybe even sufficient prerequisite for planning science." [1, p. 4]. Unfortunately, despite progress in the history of science, Vavilov's rather cheerless evaluation of the state of this field of knowledge still remains valid to some degree: "To the present time, it (the history of science in the early 1930s — *Authors*) rests, however, in the cradle of personal characteristics and biographies, chronological dates and, in many cases, quite imperfect documentation.

¹ A brief essay on the historiographic concept of S I Vavilov was published earlier in Ref. [2].

² By the way, remarkable article [1], from which this phrase is taken and which is one of the most conceptual works on the history of science, was not included at that time in the *Collected Works* of Vavilov, maybe because of this phrase.

³ References to pages in paper [3] and other papers included in the *Collected Works* of Vavilov are given according to this collection. References to pages in article [1] are indicated for its reprint in an MSU collection published in 1965.

'The scientific nature' of this history is reduced to naive schemes, in which science is withdrawn from a live changing medium and is treated as an autonomous organism following almost a logical harmony in its development'' [1, p. 4].

Such was Vavilov's vision of the 'scientific visage' of the history of science. The transformation of the history of science to science itself would allow us to predict and plan the development of society, which was considered in the early 1930s as the most urgent issue in our country, which had to be solved based on a scientific foundation. And therefore, science itself should be planned. The latter, according to Vavilov, is possible only based on the study of the historic scientific experience, which should be brought to the scientific level.

But Vavilov warned against a hasty solution to this problem by 'withdrawing' science from the social and cultural context and constructing 'naive' logicized schemes. It seems that Vavilov saw the 'historic scientific nature' of the history of science as a synthesis of the natural science and humanitarian scientific approaches. Therefore, a certain union should exist between scientists working in the field of natural sciences and humanitarians (historians, philosophers, sociologists) to elaborate a certain form of the historic scientific professionalism for solving the problem of the scientific nature of the history of science.

However, instead of this union (which was in fact unreal at that time in the USSR because of the catastrophic degradation of humanities), a vast gap existed between natural and humanitarian sciences, the history of science vexatiously being somewhere in between ("the subject itself was unclear and alien" for historians, while natural scientists "had no time to look back" and "in many cases... they did not have the necessary general historic and philosophic knowledge"). Then, Vavilov specifies a number of fundamental questions on the relation between science and prescientific and nonscientific knowledge, which have been considered but nevertheless have not been solved to date.

"Science as a historical factor" is another important issue in the history of science put forward by S I Vavilov. This question is closely related to another basic issue of historic scientific study-the determination of the driving stimuli of science development. Vavilov points out that "the inner logic of science itself was considered consciously or unconsciously almost the only such stimulus" [1, p. 6]. However, as he shows by the example of optics in the 17th-18th centuries, "it is reasonable to seek such stimuli in the technical challenges of the time, in the social and economic conditions of people and times, etc." [1, p. 6]. At the same time, "it would be erroneous to try to find a detailed parallelism between the history of science and the history of society" (ibid.). "Social and economic factors are the main catalyst of the development of science, but these processes begin from the level that science has already achieved" [1, p. 6]. Beginning from the 16th century, this level is maintained at "a certain height" owing to the "international scientific relations." The questions about the history of science formulated by S I Vavilov in the early 1930s still remain urgent.

4. Definitive "factors of the kinetics of science development." Galilean and Newtonian telescopes

S I Vavilov believed that, despite the progressive and cumulative nature of the development of scientific knowledge, "the course of science is not one-dimensional, possessing a 'width', bifurcations, zigzags, and loops'' [4, p. 235]. He protested against the reduction of the live, multidimensional, and multifactor kinetics to the one-dimensional mode repeating "the time-swept inner logic of today's scientific dogma," but "rarely coinciding with intricate zigzags actually happening'' [4, p. 235].

However, Sergei Ivanovich could distinguish some dominating factors in this intricate multifactor process. For example, one such dominant factor in the development of optics, mechanics, and physics as a whole in the heroic epoch of Kepler, Galileo, and Newton in the 17th century was the telescope. "Siderius Nuncius" (Star Herald by Galileo, in which he describes his applications of a telescope — Authors) wrote S I Vavilov, "compelled the scientific world in the early 17th century to engage in research on dioptric devices, the grinding and polishing of glasses. The history sees Descartes, Spinoza, Newton, kings and princes, abbots and monks, physicists, philosophers and physicians engaged in this activity. This resulted in a very rapid development of the geometrical optics of refracting media, glass machining technologies, the art of construction of optical devices and optical manufacturing in a broad sense" [4, pp. 236, 237]. In another paper, Vavilov distinguishes the same factor which became the key stimulus of the entire creativity of Newton. "The source of the scientific activity of Newton, in which the three main channels-optics, celestial mechanics, and mathematical studies-are intersected, is a reflecting telescope." And then he explains this 'formula': "The search for the perfect shape of optical glasses... is a probable practical motive for the first geometrical works of Newton. The discovery of light dispersion is a direct consequence of work on the improvement of telescopic glasses. The objects of telescopic observations-planets and their satellitesattracted the attention of Newton to celestial mechanics. Finally, the initial aim of prolonged chemical investigations... was to find alloys suitable for manufacturing metal mirrors for reflectors.... Thus, it is reasonable to assume that the external motive (for the development of Newton's investigations - Authors) was a technological problem of the refinement of telescopes" [5, p. 109]. After one and a half decades, S I Vavilov transferred this key thought to his book on Newton, comparing the Newtonian telescope to an overture: "Just as in an overture preceding a large musical piece the main motives of this piece are interlaced, so in the Newtonian telescope we can see the sources of almost all the main avenues of Newton's scientific thinking and work" [6, p. 321]. In other cases, S I Vavilov could also distinguish similar key factors governing the choice of the topical scientific problem and the creation and further development of scientific ideas and constructions.

5. "Singularities" in the "continuous line of the development of science"

Accepting the unidirectional progress in the development of science, Vavilov assumed that this growth is not quite continuous and "the continuous line of the development of science contains some 'singularities', which look like turning points in history" [1, p. 7]. This approach anticipates the concept of scientific revolutions by T Kuhn, which became popular in the 1960s–1970s. By the way, Sergei Ivanovich used a similar term — 'scientific upheaval'.

Attentive reading of his works devoted to Newton and Galileo allows us to understand the peculiarities of the

scientific revolution in the 17th century and some specific features of 'scientific upheavals'. Vavilov believed that one of them was the quantum-relativistic revolution beginning at the end of the 19th century and spreading over the first third of the 20th century. According to Vavilov, the most important among the scientific revolutions belongs to the radical transformation of the fundamental theoretical system of concepts, allowing a more detailed and accurate description of the accumulated experimental material. He emphasized that the roots of revolutionary transformations 'extend far back' and that new theoretical approaches are "the necessary result of the preceding development, spectacular in its ripeness and fruitfulness, but in fact not containing anything qualitatively new" [1, p. 7]. However, it is not easy to agree entirely with the latter statement, which, of course, differs from Kuhn's concept of the scientific revolution [7]. Vavilov believes that the discontinuity aspect obeys the continuum of scientific development, as 'singularities' are the solutions to differential equations, which are continual by their nature. S I Vavilov, giving Newton's predecessors their due, showed the novelty and depth of Newton's breakthrough. This also concerns the quantum-relativistic revolution, although he considered it necessary to reveal classical sources of quantum and relativistic ideas.

6. "Inculcation of the scientific truth"

For a scientific discovery and a new scientific theory to be established in science, it is insufficient to make or construct them, because they should also be accepted by the scientific community. The historians of science only comparatively recently understood the importance of the problem of reception of new scientific knowledge. Thus, a comprehensive study of the reception of the theory of relativity by scientific communities in different countries [8] first appeared only in 1987.

S I Vavilov well understood the importance of this aspect of a historic and scientific study. However, instead of the reception he talked about the 'inculcation', which resembles the assimilation of the results of fundamental studies in practice and technologies. Thus, he said that Galileo possessed the amazing gift of what is now called "the inculcation of the scientific truth." "The truth," he continued, "became the public domain owing to its application, new arguments clear to everybody, due to the active struggle for it..." [4, p. 236]. In his paper on the optical works of Lomonosov, S I Vavilov talked about his tragedy because the rich scientific legacy of Lomonosov "is buried in unread books, unprinted manuscripts, and abandoned and ruined laboratories on Vasil'evskii Island and on the Moika" [9, p. 168].

Sergei Ivanovich noted that the remarkable optical discoveries of Leonardo da Vinci also had the same destiny [4, p. 250].

7. Three main methods for constructing physical theories

The study of Newton's works, the history of optics, and the history of the theories of relativity and quanta being created before his eyes led S I Vavilov to the following classification of the main methods for constructing physical theories: the method of hypotheses-models, the Newtonian method of principles, and the Maxwell mathematical hypothesis (or mathematical extrapolation) method. Vavilov wrote already about the Newtonian method of principles in 1927, showing that his *Opticks* and *Principia* are based on this method opposing the hypothesis-model method, which was popular at that time [5]. However, the latter should not be understated: "Based on the model hypothesis method, the classical theory of heat, light, sound, etc. has grown" [3, p. 156]. The advantages of this method are its clearness and 'intelligibility'. Its limitation lies in the unsubstantiated extrapolation of the macroscopic (human) scale to the microworld.

Vavilov considered principles used in the 'method of principles' as "the ascertaining of an experiment in the adequate mathematical form" [3, p. 156]. "Such principles, mathematically expressed and generalized, play further the role of axioms in geometry, from which logical conclusions are made concerning specific physical problems" [3, p. 157]. The examples of theories constructed by this method are not only the classical mechanics of Newton or his optics, but also classical thermodynamics and the special theory of relativity. The reliability and viability of theories constructed by this method is confirmed by the entire experience in the development of physics. Compared to the model hypothesis method, this method is considerably more abstract and less clear. The common feature of both methods is that "mathematics plays... mainly a service, technical role" in them [3, p. 157].

In the mathematical hypothesis method, which is "the most abstract and detached from experience," but is "very important in modern physics" [3, p. 157], mathematics plays a completely different, creative, heuristic, structure-forming role. Vavilov himself never used this method, but well understood and highly appreciated it. According to Vavilov, "this method was first used by Maxwell with remarkable success in the field of electrodynamics" [1, p. 11], and after that "mathematics acquired incomparably deeper significance for physics." "Mathematics was transformed from an auxiliary tool for quantitative calculations and formulations to a heuristic method allowing a theorist to anticipate experiments..." [1, p. 11]. The creations of the general theory of relativity and quantum mechanics are, in the opinion of Vavilov, "amazing examples of the power of the method of mathematical extrapolation" [1, p. 11].⁴ Vavilov highly appreciated the mathematical hypothesis method and believed that, apart from experiments, it should be supplemented or corrected by methodological regulators, such as the correspondence and simplicity principles [1, p. 12]. Admiring the latest amazing achievements of the general theory of relativity, quantum mechanics, and quantum electrodynamics, Vavilov wrote at the end of his remarkable article in 1933: "The theoretical method applied by Maxwell is infinite, like mathematics, and any scales arbitrarily distant from common human things pose no threat to it. Based on this method, physics can develop infinitely, relying alternatively on experiments and mathematical thought" [1, p. 12].

Vavilov believed that there is no insurmountable boundary between these theoretical methods. In the real work of a theoretical physicist, they are interlaced, passing

⁴ According to Vavilov, "its essence consists... in finding such mathematical forms which, including all particular cases directly found in experiments, would provide simultaneously a considerably broader content. Certainly, the only justification of the correctness of the chosen mathematical form can be its subsequent confirmation in experiments. Deprived of concrete images and models in the new-scale world, a physicist has found in mathematics an infinitely capacious method for the development of a new theory" [1, pp. 11, 12].

to each other (see, for example, paper [5, p. 108]). In this way, the history of science gives rise to the live cognition theory and philosophy of natural science based on the experience of several generations of natural-science researchers and devoid of factitious epistemological schemes.

8. Appendix. S I Vavilov as a historian of science (chronological reference)

After 1922: Vavilov translated foreign books on the theory of relativity (A Einstein, F Auerbach). Vavilov made and edited many translations until 1951.

After 1926: Biographical articles and articles on physics in encyclopedias (78 articles).

1927: "Principles and hypotheses of Newton's optics" review in *Uspekhi Fizicheskikh Nauk* [5]. Later, Vavilov returned many times to the creativity of Newton.

1927: Translation of Newton's Opticks [10].

1928: Monograph *Experimental Foundations of the Theory of Relativity* with chapters inscribed with epigraphs from Newton's *Principia* and *Opticks* [11].

1933: Article "Old and new physics" [1].

After 1934: Chief of the section of the history of physics and mathematics at the Institute of the History of Science and Technology, the USSR Academy of Sciences (Leningrad).

1937: Article "Optical views and works of M V Lomonosov" [9].

After October 1938: Article "Science and technology in the French revolution period" [12].

1943: Biographic book *Isaac Newton* (First edition [6], repeatedly reprinted; see Ref. [13]).

1943: Article "Galileo in the history of optics" [4].

1945: "Essay on the development of physics at the USSR Academy of Sciences for 220 years" [14].

After 1945: The chair of the Commission on the History of Physical and Mathematical Sciences at the Division of Physics and Mathematics (after the death of A N Krylov).

After 1945: A member of the Scientific Council of the Institute of the History of Natural Science (IHNS), the USSR Academy of Sciences, a member of the editorial boards of the *IHNS Proceedings* and the *Scientific Legacy* Series issued at the IHNS.

1946: Report "Physics of Lucretius" [15] (in General Assembly of the USSR Academy of Sciences, 15–19 January 1946).

1946: Translation of Newton's *Lectures on Optics* from Latin.

1946: Report "I Newton's atomism" in England (presented by H Dale) (see Ref. [16]).

1948: Article "Petr Nikolaevich Lebedev" [17] published in the book *People of the Russian Science* and other articles.

1949, January: Address at the session of the USSR Academy of Sciences devoted to the history of the Russian science.

1949: Article "Lenin and philosophical problems of modern physics" issued in *Usp. Fiz. Nauk* [18].

The works of S I Vavilov on the history of physics and his role in the development of the native science are also characterized in biographical book [19] by L V Levshin, essays [20] by T P Kravets, collection of papers [21], and works [2, 22]. The complete list of S I Vavilov's works on the history of science is presented in *IHNS Proceedings* [23].

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