IN MEMORY OF ACADEMICIAN A. N. TERENIN (1896-1967)

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In 1970, it will have been three years since the death of the outstanding Soviet scientist, physical chemist, hero of socialist labor, Academician Aleksandr Nikolaevich Terenin.

The scientific interests of Terenin were extremely broad and diversified. As a student of the most prominent Russian and Soviet physical optician D. S. Rozhdestvenskii, Terenin began his scientific activity with works in the field of atomic spectroscopy. His first investigations [1-3] were devoted to the explanation of the structure of the energy levels of a number of metal atoms by resonance fluorescence methods. Even these first works, carried out with great experimental skill at the start of the twenties during a period of vigorous scientific development with respect to the structure of the atom, brough Terenin widespread fame. A huge contribution of Terenin to the knowledge of atomic spectra was his discovery in 1928 together with Dobretsov [4] of the ultrafine structure of spectral lines caused by the presence of nuclear spin.

However, beginning with the middle twenties, Terenin's circle of scientific interest shifted more and more to the field of spectroscopy and photochemistry of molecules. In 1925, using a method of optical excitation previously discovered by him for the vapors of halide salts, Terenin discovered the phenomenon of photodissociation of molecules to form excited atoms and radicals and the new method based on this for determing the dissociation energy of molecules [5, 6], meanwhile proposing the start of a large cycle of works devoted to the study of the nature of primary photochemical processes in gaseous compounds [7].

In the thirties, Terenin, while heading the already well-known collective of scientists and co-workers in the laboratories he organized in the State Optical Institute and in Leningrad University, labored over a broad front using various methods of investigation. Continuing his works on the photodissociation of molecules, Terenin together with Neuimin [8] extended these investigations to the vacuum UV region of the spectra, which permitted a study of the photochemical reactions of oxygen, water, ammonia, alcohols, and other compounds. Together with Popov [9], he was one of the first to begin to use mass spectrometric techniques for the analysis of the products of photoionization of molecules. Terenin simultaneously concerned himself with the problems of exchange and transformation of electronic and vibrational energy of excitation of molecules. With this goal in mind, he studied the effect of contaminating gases on the intensity and vibrational structure of the fluorescent spectra of the products of photodissociation of molecules (N. P. Prilezhaeva, Neuimin, Tibilov) [7], and propounded experiments for explaining the effect of contaminating gases on the fluorescence of complex organic compounds (Vartanyan, Neporent) [10], and studied the processes of deactivation of vibrational energy with respect to the IR discharge emission in molecular gases (Neuimin) [11]. In this period also we find the first works of Terenin and his scientists with respect to the luminescence of crystallophosphors (Klement) [12], the IR spectroscopy of organic compounds dissolved in gases under high pressure [13], as well as investigation of fluorescence and phosphoresence of layers of organic compounds at low temperatures [14] which form the basis of the study of photochemical reactions of molecules in this state.

At the end of the thirties and start of the forties, Terenin transferred his interest to the study of photo processes in organic dye molecules. In 1941, together with Vartanyan [15], he detected the semiconductor properties of layers of organic dyes. In 1943, his studies of the phosphorescence of complex organic molecules with conjugated bonds, including dye molecules, led him to the hypothesis concerning the biradical (triplet) nature of the excited metastable state of molecules [16, 17], which for the first time permitted one to arrive at a correct explanation of the mechanism of phosphorescence and photochemical reactions of complex organic compounds and layed the foundation for modern photochemistry.

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In the mid forties, Terenin introduced the problem of spectral emission of photochemical reaction of chlorophyll and its analogs, having in view an explanation of the mechanism of the first stage of photosynthesis in plants and the reproduction of its separate stages within the living cell [18]. In 1948, as a result of works carried out under the direction of Terenin in the photobiochemistry laboratory he organized, Krasanovskii discovered the reversible reaction of photoreduction of chlorophyll, which is extremely important for photosynthesis in plants [19]. Somewhat earlier, Terenin and Karyakin [20] observed for the first time the reversible phototransfer of a photon between organic molecules.

In the ensuing years, Terenin became extremely interested in the problems of the transfer and migration of energy, especially in biochemical processes [21]. In 1952, he and Ermolaev [22] detected and subsequently studied in detail the phenomenon of sensitization of phosphorescence of organic compounds caused by nonemissive transfer of the energy of electronic excitation of molecules with respect to the triplet levels. Simultaneously with the problems of energy transfer, Terenin evolved investigations of the photoelectrical properties of dyes and other organic compounds including chlorophyll (Vartanyan, Putseiko, Evstigneev) [19]. He studied in detail the mechanism of spectral sensitization by dyes of photoelectric processes in semiconductors (Putseiko, Akimov) [23, 24]. Using the methods of high-speed and pulse spectroscopy he studied the intermolecular transfer of an electron [25]. He also carried out a study of the dark and photochemical reactions of dyes with respect to the absorption spectra in the visible and IR regions and via the EPR method (Siorov, Kholmogorov) [19].

Having developed the investigation in the direction of more and more complex objects, including biochemical systems, Terenin at the same time did not curtail his study of photo processes in simple gaseous compounds. Included in his postwar works of this cycle are the study of the sensitized fluorescence in vapors of organic compounds (Karyakin) [26], the investigation of the IR emission of vibrational-excited molecules in electrical discharge (Dodonova), and the photosynthesis of organic compounds from simple gases under the action of vacuum UV radiation (Dodonova, Sidorova) [27]. The works of Terenin and Vilesov [28] with respect to the energetics and mechanism of photoionization processes in the vacuum UV spectral region, which encompassed a broad circle of objectives from simple gases to complex dye molecules and biologically active compounds, were the subject of broad development in the fifties and sixties. In these very same years, under Terenin's direction were carried out investigations of the photoelectric properties of organic polymers which culminated in the discovery of a new class of organic semiconductors [29], two-quantum photo reactions were studied [30], works with respect to the IR spectra of complexes with catalytically active compounds [31] were accomplished via spectroscopy at high pressures [32], and many others.

Not limiting himself to a study of the photophysical and photochemical processes in gaseous and condensed phases, Terenin in the thirties entered into a study of the effects of the interaction of optical radiation with the surface boundary of a gas–solid and, associated with this, the more general problems of physicochemical surface phenomena. Here, as well as in other fields, Terenin addressed himself chiefly to spectral and other optical methods using them to solve such problems as the degree of deformation and structural changes of molecules during adsorption, energy exchange between molecules and solids, and the mechanism of elementary acts of chemical reactions proceeding on the surface. We should like to here discuss in more detail works of this type which occupy such a central position in the scientific activity of Terenin.

Having organized a laboratory of the optics of surface phenomena in Leningrad University in 1935, Terenin worked out for it a broad program of investigations which provided for both the obtaining of data on the state of adsorbent–adsorbate systems as well as direct observance of the effects and transformations caused by light absorption. One of the problems which the laboratory encountered was the study of the possibility with the help of light to actively affect the course of heterogeneous, catalytic reactions. A spectral-manometric method, which has subsequently assumed widespread extension in the works of Soviet and foreign scientists, played an important role in these investigations. The application of this method even in the first year of the laboratory's work permitted Terenin together with Kastarov, Val'nev, and Tagantsev to enter into a planned study of the photodesorption and photodecomposition of adsorbed molecules as well as photochemical reactions between molecules in the surface layer [33-36]. As a result of these investigations it was found that light quanta which are not capable of causing the same processes in molecules in the free gaseous state can actively affect the adsorbed molecule. In particular, it was shown that the reaction with a solid can lead to a significant lowering in the boundary value of the quantum energy necessary for disruption of the valence bonds. Terenin together with Kurbatov then detected and studied the effect of increasing the adsorption capabilities of a solid during illumination with visible and UV light, i.e., the photosorption of molecules [33-36].

These works, which were the first systematic investigation in the field of photocatalysis, were developed further in the postwar period when Terenin and Solonitsyn [37] began a study of photosorption and photodesorption of simple gases which occur during absorption of light by an absorbent. During this period, along with manometric measurements for investigating photosorption and photocatalytic processes, Terenin and Solonitsyn, Vilesov, Lisachenko, Kotel'nikov, and Basov began the widespread application of mass spectrometric methods which made possible reliable quantitative analysis of reaction products and permitted the formulation of a number of new problems. For example, owing to the use of oxygen molecules of different isotopic composition, the dynamics of photosorption processes in the oxygen–zinc oxide system were discovered and the simplest photocatalytic reaction of isotopic oxygen exchange was investigated and made it possible to link photosorption processes in the oxygen–oxide system with the photocatalytic activity of oxides in oxidation reactions (Vilesov, Lisachenko) [38].

Of great significance for explaining the mechanism of photosorption and photocatalytic processes in addition to direct manometric and mass-spectrometric experiments were investigations of electronic transitions in the surface layer with respect to a change in the conductivity of absorbents as initiated by Terenin and Putseiko [23] and subsequently developed by Korsunovskii, Solonitsyn, Kotel'nikov, Rapoport, and Basov. These experiments showed that photosorption processes are not caused only by a change in the electronic state of the absorbent during illumination but are associated with the action of light on the surface hydroxyl groups of the adsorbent or with the change in the degree of charging of the surface (structural or previously chemisorbed) oxygen occurring during illumination. A parallel study of the conductivity, photoconductivity, and photocatalytic properties was used to explain the mechanism of photooxidation of water on the surface of semiconductor oxide adsorbents (Korsunovskii).

It should be noted that Terenin, even in his early works, used the concept of electron exchange in the gas–solid system [36, 39] to explain the photosorption of molecules and the quenching of photoluminescence of adsorbents during contact with certain gases (as observed by him and Gachkovskii). In the postwar years, Terenin subjected processes associated with the transfer of an electron in the adsorbent–adsorbate system to comprehensive investigation using such methods as the measurement of the photoconductivity and photo emf of powdered oxides (Putseiko), the quenching of the luminescence of an adsorbent by adsorbed molecules (Tagantsev), the change in the photoelectric work of discharge of semiconductors (Vilesov), and the absorption of semiconductors in the IR (Filimonov) and EPR (Kholmogorov, Rapoport) spectral regions. In the process, Terenin did not limit himself to the investigation of the role of the electronic factor in the physical chemistry of surface phenomena but also set as his goal a study of the effects of adsorption on the photoelectric properties of semiconductors [40]. Of particular importance in these investigations were works devoted to the spectral sensitization of the internal photo effect in semiconductors by adsorbed molecules and the associated sensitization of photographic layers. An extended series of investigations carried out by Terenin and Putseiko, Akimov, and other co-workers, as well as direct measurements of the ionization potentials of adsorbed molecules (Vilesov, Akimov, Bentsa) permitted a solution of the principal problem of the mechanism of the transfer of the energy of excitation from an adsorbed molecule to a solid [24, 41].

Another scientific direction established by Terenin in the middle thirties, viz., the spectroscopy of adsorbed molecules, played an important role in the development of modern concepts of the surface structure of oxide adsorbents and the nature of the forces and adsorption centers. The direct study of the adsorbed state and chemical transformations of molecules on a surface of a solid by optical and radio spectroscopic methods entered into the tasks of works in this direction which formed a basis for the broad application of spectral methods in investigations of the chemistry of a solid surface, adsorption, and heterogeneous catalysis.

The first works of Terenin in the field of the spectroscopy of adsorbed molecules were accomplished in the laboratory of the optics of surface phenomena in 1934-1940 and were devoted to a study of the changes which the absorption spectra of simple molecules undergo in the visible and UV region during adsorption on halide salts and silicone dioxide aerogel [33, 35]. These works particularly provided for determination of the shift of the electronic spectrum of molecules during transition from gaseous to the adsorbed state and its correlation with the energy of adsorption of molecules (Barakan, Kurbatov,). In subsequent years, investigation of the electronic spectra of adsorbed molecules was extended by Terenin to complex aromatic compounds which have a characteristic absorption and undergo more pronounced changes on contact with a solid surface which are caused by the development of chemisorbed states [42]. The use of such indicator molecules permitted spectral observation and study of the active centers of catalyst surfaces. This method

was especially widely used to explain the nature of the active centers of an alumosilicate catalyst. Even in the first works of this series, Terenin and Sidorova [43] found that the adsorption of aromatic amines on active clays of the bentonite type is accompanied by ionization of the adsorbed molecules, which attests to the presence of strongly acidic centers on the surface. Subsequent investigations carried out by Terenin and Kotov [44], particularly the application of "poisoned" catalysts permitted obtaining spectroscopic evidence of the presence of acidic centers of two types on the surface of alumosilicate catalyst proton donors which give up a proton to molecules of the organic bases, and electron acceptors which lead to the formation of cation-radicals. The next step in these investigations was the use of polynuclear aromatic hydrocarbons as indicator molecules, which made possible the observation of the appearance of carbonium ions (Kotov, Barachevskii) on the catalyst surface as well as the application of the EPR method for detecting and identifying various radical forms of adsorbed hydrocarbons (Kholmogorov) [45]. Additional data on the nature of the chemisorbed states and the degree of perturbation of the electronic structure of molecules during adsorption were obtained from a study of the effect of contaminating gases on the adsorption complex and the competing reaction of molecules with different electron-donating properties.

Later, Terenin and Pimenov applied the method of electronic absorption spectra to study the donor-acceptor reactions of organic molecules with the surface of typical semiconductors. As a result of these investigations, the fundamental products of the reaction of such type were detected: positive and negative molecular ions and a charge-transfer complex. In addition, it was found that the fundamental photophysical and photochemical processes occurring in the layer near the surface of the lattice, which are manifested particularly in a change in the optical properties of the adsorbent, change markedly during a donor–acceptor reaction which results in charge separation.

Terenin devoted special attention to the development of the IR spectroscopy of adsorbed molecules. This method, which is currently one of the most direct and important sources of information regarding the state of adsorbed molecules and the structure of surface compounds, was first used by Terenin for the investigation of surface phenomena in 1940. The first works in this field, which he carried out together with Kasparov, involved a desire to study the chemisorption state of ammonia and ethylene molecules on metal surfaces [46, 47]. Although these experiments had a somewhat exploratory nature and were interrupted by the incipient war, the setting up itself of such investigations at that time attests to the experimental skill and profound scientific perspicacity of Terenin who even then foresaw the great potentials of this method of investigation. It is sufficient to say that abroad the first investigations with respect to the IR spectroscopy of adsorbed molecules were established only in 1953. In 1948, the study of the vibrational spectra of adsorbed molecules and surface chemical compounds was again resumed by Terenin in the near IR spectral region, and somewhat later also in the region of fundamental molecular vibrational frequencies. Initially these investigations were focused on a study of the hydroxyl coating of silicone dioxide adsorbents, the existence of which on oxide surfaces Terenin, Yaroslavskii, Kurbatov, and Neuimin proved for the first time by direct spectroscopic experiments [48]. As a result of the entire series of investigations begun by these authors and subsequently continued under the direction of Terenin by Sidorov, Karyakin, and Filimonov, it was shown that the hydroxyl groups of the silicone dioxide surface, due to their acidic nature and capacity for formation of stable hydrogen bonds as well as a result of the presence in them of a dipole moment, are the centers of physical adsorption of a broad spectrum of molecules differing in electronic structure, proceeding from diatomic molecules of simple gases and terminating in molecules of aromatic hydrocarbons [49]. It was found, in addition, that the surface silanol groups can also project as centers of chemisorption of molecules leading to the formation of chemically modified surfaces [50]. These works subsequently permitted the development of a comprehensive study of the role of surface hydroxyl groups in the physical and chemical adsorption of molecules on oxide adsorbents.

After explaining the fundamental principles of the interaction of molecules with surface hydroxyls, Terenin turned to a study of the IR spectra of chemisorbed molecules using them in particular to study the nature of the active centers of catalyst surfaces. The studies of this period encompassed a large number of adsorbents and catalysts including synthetic zeolites, salts, metals, and metal oxides. IR spectroscopy was used, e.g., together with electronic spectra and EPR to study the acidic centers of alumosilicate catalysts and permitted observing electron-accepting centers of the Lewis acid type on its surface and the observation of the appearance of proton acid centers during the adsorption of water (Filimonov, Roev) [51]. Later, these investigations were extended to other adsorbents, during which it was found that the electron-accepting acid centers of different strength which were capable of forming a coordinate bond with adsorbed molecules are present on the surface of a large number of metal oxides as well as several silicone dioxide

adsorbents (Sidorov, Filimonov). In another series of works devoted to the study of a catalyst surface, Terenin, Roev, and Alekseev conducted a comparative study of the adsorption centers of a number of transition metals and their compounds using the nitric oxide molecule as an indicator of the electronic properties of the surface [52]. As a result of these investigations, a definite dependence between the structure of the electronic cloud of the metal atom and the nature of adsorbent-adsorbate bond was revealed. A coincidence of the forms of adsorption of nitric oxide on metal and the corresponding cation was established for several of the metals, which speaks for the discrete nature of the adsorption centers on the metal surface and the participation of its valence electrons in the formation of local bonds with the adsorbed molecules.

In connection with his study of the mechanism of heterogeneous catalytic reactions, Terenin made wide ranging application of IR spectroscopy for elucidating the structure of surface compounds formed due to chemisorption and subsequent chemical transformations of molecules on the catalyst surface. In this category of special importance are investigations of the chemisorption of hydrogen and oxygen on metal oxides and investigations with respect to the adsorption and catalytic decomposition of alcohol. Recently, the study of the vibrational spectra of adsorbed molecules was also carried out in the Terenin laboratory by the method of combined light dispersion (Raskin, Pershina).

The investigation of the luminescence of a catalyst in the oxidation of organic compounds in the gas phase serves as an example of the optical study of the phenomenon of catalysis itself. This form of luminescence, which is associated with the evolution of energy in the first stage of oxidation on the surface and its transmission to the center of luminescence, was first detected in the Terenin laboratory by Kurbatov and subsequently studied by Terenin and Kachur [53].

At the same time that he elaborated the mechanism of activation of molecules during contact with a solid surface, Terenin for the first time applied spectral methods for studying the photoactivation of adsorbed molecules and their photochemical reactions. In particular, the photoionization of benzene molecules in the adsorbed state under the influence of quanta with energies of values one half that required for ionization of benzene in the gaseous state was detected by the method of optical electronic spectroscopy (Barachevskii) [54]. A lowering of the quantum energy causing ionization of the molecules was also observed during the adsorption of benzene derivatives and other aromatic hydrocarbons, during which it was shown that the ionization process is not associated with the absorption of light by the adsorbent lattice, and proceeds as a result of excitation of the adsorbed molecule with subsequent removal of an electron by the electron-accepting center of the surface (Kotov, Pankratov). In addition, application of the spectral methods permitted detection and investigation of an entire series of other photochemical reactions of adsorbed molecules, including those caused by scission of the valence bonds. For example, the photodecomposition of adsorbed alcohol molecules to form unsaturated radicals under the action of light quanta which do not evoke direct photolysis of alcohols under homogeneous conditions (Kholmogorov) was studied by the EPR method. The ability of a number of metal oxides to sensitize photooxidation of alcohols and hydrocarbons in the near UV was detected by IR spectroscopy, and the mechanism of these reactions was investigated (Filimonov). The decomposition of water and ammonium molecules adsorbed on aluminum oxide was studied by manometric and mass-spectrometric methods (Kotel'nikov) [55]. These investigations together with the much earlier investigations of Terenin and Fialkovskaya [56] devoted to the photoactivation of adsorbed molecules of pyridine were an important step in the development of the photochemistry of the adsorbed state of molecules.

It is difficult in a short review to present the complete picture of the diversified scientific activity of Terenin for whom was characteristic an extremely broad range of problems, innovations, diversified methods of investigation, and constant seeking for new paths for the solution of complex scientific problems. While being an eminent specialist in the field of spectroscopy and photochemistry of complex organic compounds, photosynthesis, photoelectronics of organic semiconductors, and while working out problems of the physical chemistry of surface phenomena, Terenin at the same time was deeply involved in organizational, scientific-social, and pedagogical pursuits. For more than ten years, he was the scientific director of one of the largest scientific departments of our country, viz., the State Optical Institute. Terenin made a vast contribution to the development of the optical industry, especially in regard to the development of the construction of indigenous spectral devices. In his latter years, he directed the scientific Soviet with respect to photosynthesis in the Academy of Sciences of the USSR and was a member of the editorial staff of Soviet and foreign journals and represented the Soviet Union in the commission on the structure and spectroscopy of molecules of the International Union of Pure and Applied Chemistry (IUPAC). Terenin was especially interested in the training of scientific specialists. All his scientific and pedagogical activity was unceasingly associated with Leningrad University of which he was selected a professor in 1932. In 1964, Terenin

organized and headed the Department of Molecular Biophysics in the physical faculty of Leningrad State University. Terenin created the Soviet School of Spectroscopists and Photochemists.

The scientific services of Terenin were greatly esteemed and honored both in the Soviet Union and abroad. In 1932, he was selected as a corresponding member and in 1939 as an active member of the Academy of Sciences of the USSR. In 1945, after his work in the field of photochemistry, he was awarded the first degree state prize. His studies with respect to luminescence, molecular spectroscopy, and intermolecular transfer of energy were awarded the Vavilov (1954), Chiamichian (Bolonsk University, 1959), and Finzen (England 1964) gold medals. Terenin was selected as an esteemed member of the French Society of Physical Chemistry and the English Chemical Society. He was awarded the Fourth Order of Lenin, the Order of Labor Red Premium, the Order of the Red Star, and other medals. In 1966, he was awarded the title Hero of Socialist Labor.

Terenin's labors will always be a part of the golden fount of Soviet and world science.

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