

ory devices^[10]. The signals corresponding to "ones" and "zeros" are the presence or absence of domains at definite points of the film.

The prospects of the development of domain devices are determined by the high density with which they record information, their low power consumption, their sufficiently high readout rate, and their cheapness.

For these purposes the optimum set of properties is possessed by epitaxial films of the rare-earth ferrites with the garnet structure: these films are ferrimagnetic^[11-13].

At present methods have been developed for: controlling the motion of the cylindrical domains (recording and shifting of information), the production of domains (introduction of information), and registration (detection) of domains^[14-15]. Control of the domain motion implies both a controlled migration of a domain in the plane of the crystal and the immobilization of it at definite locations—magneto-optical traps. For this purpose one uses methods based on the interaction of the domains with the nonuniform magnetic fields produced by current or ferromagnetic appliqué. Domain production is accomplished by the motion of the original domains when the same, but somewhat modified appliqué is used. The readout of information is performed by different methods: inductive, galvanomagnetic and magneto-optical, and magnetic probes. Preference is given to galvanomagnetic and magneto-optical detectors^[16-18].

A number of recorders based on cylindrical domains have already been constructed, e.g., a 1000-bit shift detector for a recorded-information density of 2.5×10^5 bit and a readout rate of 3×10^5 bit/sec^[19].

2. **Thermomagnetic Recording.** This technique combines the optical method of recording and readout with the magnetic method of information storage. A small section of a ferromagnetic film is heated with the aid of a laser in the presence of a magnetization-reversing magnetic field. As a result, the direction of the magnetization of this section of the film is reversed after cooling, while the remaining part retains its original magnetization. Because this method is connected with the local heating of a magnetic film, it is called a thermomagnetic method. Magneto-optical effects are used for the readout. The first thermomagnetic recording was accomplished in 1958 by Mayer by heating a MnBi film with an electron beam^[20]. In view of the construction of lasers and the progress made in the area of magnetic-film production, this method attracts much attention.

In a recording on magnetic films with the aid of a laser beam, the dimension of the spots is limited by light diffraction, and can, in the limit, be comparable with the wavelength of the light. This allows us to achieve a high, information-recording density $\sim 10^8$ bit/cm². It is possible to achieve at the same time a sufficiently high recording rate and an even higher readout rate. A laser of less intensity can be used for the readout and, therefore, a readout is done without destroying the recorded information. Thermomagnetic recording is performed at the Curie point, at the balance point, at the point of rearrangement of the magnetic structures, on films with a strip structure, etc.

Optical memories, including the thermomagnetic

ones, have a great future. These memories guarantee the possibility of a holographic mode of recording. In the limit, the capacity of a holographic memory may reach 10^{10} bit/cm². In perspective, the transition to local systems with holographic introduction and retrieval of information promises the creation of elements for tremendously high-speed electronic computers.

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N. A. Borisevich. Afterglow of Complex Molecules in the Gaseous Phase. Molecular spectroscopy and luminescence were adopted as two of the main research directions of the Physics Institute of the BSSR Academy of Sciences when the Institute was being organized. Investigations in molecular spectroscopy, including laser spectroscopy, constitute at present a significant proportion of the themes of the Institute. In the present paper we consider only specific problems of the spectroscopy of polyatomic molecules in the gaseous phase.

Investigations of rarefied gases and vapors allow us

to directly obtain information about emitting and absorbing centers undistorted by an extraneous medium. From the point of view of molecular spectroscopy, it is important that the distribution of the excited molecules of the rarefied vapors and gases be not the equilibrium distribution not only with respect to the electronic, but also with respect to the vibrational states. On the other hand, by gradually varying the intrinsic vapor pressure, or by adding extraneous gases at different pressures, we get the opportunity to systematically study the vibrational relaxation of the excited molecules, as well as the role of the medium in the formation of the spectral-luminescent characteristics of the molecules. These and other pieces of information are necessary both for the construction of a theory of luminescence, and for the solution of a number of practical problems.

The first investigations of the luminescence of molecules in the gaseous phase were performed by Terenin and his students^[1]. Subsequently, systematic investigations of the spectroscopy of the gaseous state of complex molecules developed in Neporent's laboratory^[2,3] and in our laboratory at the Physics Institute of the BSSR Academy of Sciences^[4]. Initially, the main efforts were directed at the study of fluorescence—the nonequilibrium vapor radiation of duration of the order of 10^{-9} – 10^{-8} sec.

The main results obtained by our laboratory in this direction are as follows. A relation has been found between the fluorescence spectra, the spectra of the quantum yield and the lifetime of molecules in the gaseous phase and the various energy characteristics of an ensemble of excited molecules. In particular, relations have been obtained which establish a connection between the fluorescence quantum yield—its dependence on temperature and on the energy of the exciting quantum—and the differences between the mean energies of the molecules participating in the processes of absorption, emission, and nonradiative transitions. From the experimental data which have been obtained on the dependence of the yield on frequency and temperature and which turned out to be different in different electronic absorption bands, we have been able to determine the indicated mean-energy differences and to consider the problem of the activation barrier of nonradiative transitions.

A consistent allowance for the statistical nature of the emission and absorption processes enabled us to introduce into luminescence such new concepts as effective excitation energy, selective energy, and effective excitation energy for fluorescent molecules, and to find theoretically expressions connecting these quantities with the temperature dependence of the absorption coefficient and the fluorescence intensity. For molecules of diverse degrees of complexity, the spectra of the effective excitation energy, the selective energy, and the effective excitation energy for fluorescent molecules have been experimentally obtained, and they essentially supplement the absorption and fluorescence spectra. Analysis of these distinctive spectra allowed us to obtain information about the dependence of the Einstein coefficients on the energy of the initial and final vibrational levels of the combining electronic states, as well as about the form of the distribution function for the molecules. From the point of view of

the study of the energetics of molecules, the demonstration that the effective excitation energy, i.e., the mean electronic-vibrational energy actually gained by the molecules in the process of excitation, is different from the energy of the exciting quantum is important. These differences are especially large in the anti-Stokes region.

The application of the universal relation between the luminescence and absorption spectra^[5] to the investigation of the excited states of the molecules in rarefied vapors proved to be quite fruitful. Methods for determining the temperatures of the excited molecules, the inversion frequency, and the vibrational heat capacity of vapors have been worked out on the basis of this relation. These characteristics have been obtained experimentally for diverse complex and semicomplex molecules. Analysis of them has led to the conclusion that even in the case of complex molecules, for which continuous electronic spectra are typical, deviations from the complete redistribution of the vibrational energy over the internal degrees of freedom of the excited molecule are possible. Subsequently, the universal relation between the luminescence and absorption spectra was generalized by finding a relation between these characteristics and the luminescence yield and duration^[6].

The investigations in the gaseous phase proved to be important for the solution of the anti-Stokes fluorescence problem, which had been a controversial problem for a number of years.

These problems have been considered in detail in the monograph: "The Excited States of Complex Molecules in the Gaseous Phase"^[4].

We shall discuss below the nonequilibrium afterglow of molecules in the gaseous phase for the case when the decay time of the radiation is of the order of 10^{-6} – 10^{-2} sec. Three types of vapor afterglow are known: α -phosphorescence or thermally activated, delayed fluorescence, β -phosphorescence, and delayed annihilation fluorescence or cooperative luminescence. Interest in the investigation of the persistent types of molecular radiation, which carry information about the triplet states of the molecules, has risen sharply in recent years. I shall discuss in somewhat greater detail only the search for α -phosphorescence and the triplet-triplet transfer of energy in gaseous systems.

The first observation of the afterglow of a vapor of polyatomic molecules was accomplished by Almy^[7] for diacetyl, an observation which has only recently been correctly interpreted as β -phosphorescence. Subsequently, Dikun discovered the afterglow of anthracene and phenanthrene vapors and surmised that it was α -phosphorescence^[8]. Parker and Hatchard^[9] showed that the afterglow observed by Dikun was not α -phosphorescence and called it delayed annihilation fluorescence.

In our search for α -phosphorescence, we proceeded along a somewhat different line. A careful measurement of the absorption spectra of the vapors of anthraquinone and its various derivatives showed that anthraquinone and its β -derivatives possess a weak absorption band (its Kravets integral is equal to 10^6 sec⁻¹), which arises owing to the $n - \pi^*$ transition localized in the carbonyl group $C = O$ ^[10]. We estimated by an indirect method

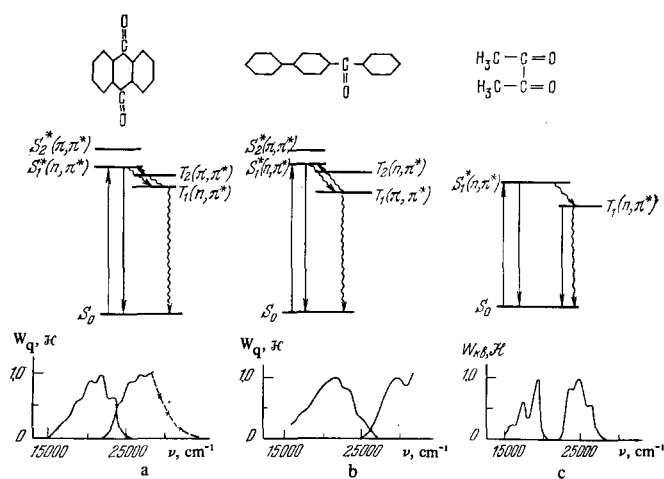


FIG. 1. Electronic-energy-level diagrams and the luminescence (W_q) and absorption (κ) spectra of a) anthraquinone, b) 4-phenylbenzophenone, and c) diacetyl vapors.

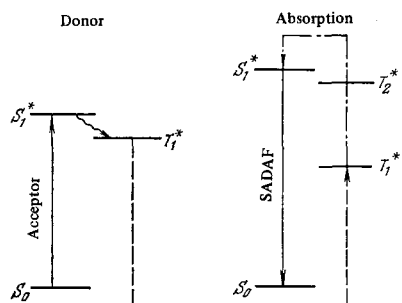


FIG. 2. Electronic-level diagrams for the donor-acceptor pair, diacetyl-anthracene.

and then measured the duration of the luminescence of anthraquinone. It proved to be of the order of 10^{-4} sec. Figures 1 a) and b) show the energy-level diagram for anthraquinone and 4-phenylbenzophenone, their luminescence bands, and the first absorption bands. The energy-level diagram for benzophenone is similar to that of anthraquinone. Their spectral characteristics are also similar. The afterglow of the vapor of these molecules cannot be considered to be delayed annihilation fluorescence, since the dependence of the emission intensity on the intensity of the exciting radiation turned out to be linear and not quadratic. Besides the afterglow, whose τ depends on the vapor pressure, a weak fluorescent radiation was detected^[11]. We interpreted the afterglow to be α -phosphorescence on the following basis. The phosphorescence and long-wave absorption bands overlap. The β -phosphorescence spectrum of the diacetyl vapor and the absorption spectrum do not overlap (Fig. 1c). It follows from the formulas for the quantum yield γ and the emission duration τ that, in contrast to β -phosphorescence, the γ and τ of α -phosphorescence should not vary proportionally as the temperature is varied, and that the kinetics of the quenching of these emissions by oxygen is different. The experimental study of the dependence of γ and τ on temperature and of the quenching of the luminescence of diacetyl and three other molecules confirms the inference that the nonequilibrium radiation is β -phosphorescence in the

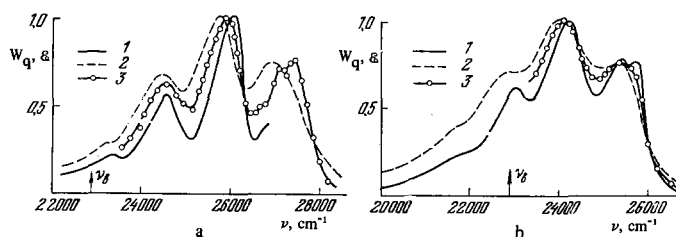


FIG. 3. Fluorescence (1, 2) and diacetyl sensitized anti-Stokes delayed annihilation fluorescence ϵ (3) spectra of a) anthracene and b) 9- and 10-dimethylantracene vapors. (for 1) $\lambda_e = 3650 \text{ \AA}$, 2) $\lambda_e = 3130 \text{ \AA}$, and 3) $\lambda_e = 4360 \text{ \AA}$).

first case and α -phosphorescence in the second^[12,13].

Finally, the proof that the afterglow of anthraquinone, benzophenone, and 4-phenylbenzophenone vapors arises as a result of transitions between an excited singlet level and the ground level is provided by the overlap in the electron transition frequency region of the diagrams of the combining states constructed from the phosphorescence and absorption spectra, as well as by the coincidence of the electron transition frequencies found from these diagrams^[14].

The triplet-triplet transfer of energy from anthraquinone, benzophenone, and 4-phenylbenzophenone to diacetyl was studied. We used the triplet-triplet energy transfer from carbonylic compounds to polynuclear aromatic molecules to produce a sensitized anti-Stokes delayed annihilation fluorescence (SADAF) of the latter molecules^[15]. The level diagram for the donor-acceptor pair for which the SADAF should be observed is shown in Fig. 2. Diacetyl was chosen as the energy donor; anthracene, 9- and 10-dimethylantracene, and 9- and 10-diphenylantracene served as the energy acceptors. As is well-known, in the gaseous phase anthracene emits delayed annihilation fluorescence (DAF). The anthracene derivatives also emit DAF.

If the cuvette contains only the vapor of anthracene or any of its derivatives, then no emission is induced when it is illuminated with light of wavelength $\lambda_e = 4360 \text{ \AA}$, since the vapor does not absorb such photons. If, however, diacetyl is introduced into the cuvette, then emission by the anthracene is observed. The duration of this radiation was measured. It turned out to be of the order of 10^{-3} sec and equal to the duration of the DAF of these compounds. Its intensity increased quadratically with increase of the intensity of the exciting radiation and with increase of the vapor pressure of the donor, i.e., the observed sensitized afterglow of the anthracenes does not differ in any way from their DAF. As an example, we present in Fig. 3 the spectra of the SADAF and the ordinary fluorescence of anthracene and 9- and 10-dimethylantracene vapors. The measurement of the SADAF spectra is considerably simpler than the measurement of the DAF ones, since a phosphoroscope is not required, as the excitation of the SADAF is not at all accompanied by short-lived fluorescence which spectrally coincides with the SADAF.

Since the spectra of the ordinary fluorescence of rarefied vapors differ for different quanta $h\nu_e$ of the exciting radiation, we can find E_T by comparing them with the SADAF spectrum and taking into account the fact that $h\nu_e = 2E_T$. For anthracene the SADAF spec-

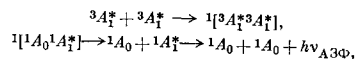
trum coincides with the fluorescence spectrum excited by the radiation $\lambda_e = 3340 \text{ \AA}$ ($\nu_e = 30000 \text{ cm}^{-1}$). Consequently, the energy E_T of the lower triplet level of anthracene in the gaseous phase is equal to $15000 \pm 500 \text{ cm}^{-1}$, which is in good agreement with the data obtained by other methods.

In conclusion, let us discuss the kinetics of the triplet-triplet energy transfer in gaseous systems and the mechanism underlying the appearance of the SADAF. For many donor-acceptor pairs in liquid solutions, the quenching of the triplet states of the donor takes place at a constant rate close to the diffusion rate. This, apparently, is the result of the prolongation of the encounter between a triplet donor molecule and an acceptor molecule (the donor and acceptor molecules undergo multiple collisions in one encounter).

In the gaseous phase, such a prolongation of an encounter, owing to the influence of the medium molecules, does not occur, and this allows us to determine the efficiency per gas-kinetic collision of the energy transfer from a triplet donor molecule to an acceptor molecule. As investigations^[16] have shown, the quenching of the diacetyl phosphorescence by anthracene in the gaseous phase is described by the Stern-Folmer equation, and the efficiency of the quenching by anthracene of the phosphorescence of diacetyl vapor is found to be 0.22.

The main problem in the study of the energy-transfer processes is the establishment of the mechanism of the bimolecular interactions that occur in the course of a transfer. Numerous investigations in recent years, chiefly those carried out by Terenin and Ermolaev^[17-19], have shown that the triplet-triplet electronic-energy transfer process takes place through the exchange mechanism.

The question as to the nature of the intermolecular interactions responsible for the appearance of the DAF cannot be considered as solved. Several hypotheses about the triplet-triplet annihilation mechanism have previously been put forward^[20]. In particular, it was assumed that the production of short-lived eximers is the intermediate stage of the triplet-triplet annihilation that leads to the emission of delayed fluorescence. Since the formation of 9- and 10-diphenylanthracene eximers is, on account of steric hindrances, not possible^[21], the very existence of delayed annihilation radiation in the vapor of this substance indicates the possibility of such a triplet-triplet annihilation mechanism in which the formation of the eximer as an intermediate product is not absolutely necessary. Also responsible for the appearance of DAF and SADAF are, apparently, the exchange-resonance interactions:



i.e., the formation of the exchange complexes ${}^1[{}^3A_1^*{}^3A_1^*]$ and ${}^1[{}^1A_0^1A_1^*]$ constitutes the intermediate stage.

There are up till now no data on the annihilation fluorescence of the aromatic ketones in both gaseous and condensed media. An explanation to this is provided by analysis of the spectra of the quantum yield of these compounds. Although the duration of the luminescence of anthraquinone and benzophenone vapors remains constant in the region ν_e under study, its quantum yield

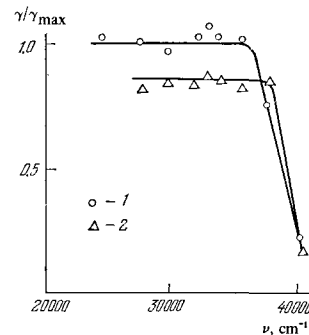


FIG. 4. Spectra of the quantum yield of the luminescence of anthraquinone (1) and benzophenone (2) vapors.

(Fig. 4), as well as the quantum yield of the internal conversion in the short-wave excitation region, decreases rapidly. This is connected with the very effective photochemical conversions of molecules in highly excited singlet states^[22]. The DAF mechanism presupposes the cumulation in one molecule of the energy of two triplet molecules, which is more than 54000 cm^{-1} for anthraquinone and about 50000 cm^{-1} for benzophenone. Therefore, even for a fairly efficient triplet-triplet annihilation, the quantum yield of their DAF will be small, since the excited molecule produced as a result of the annihilation falls into the range of energies corresponding to quite intense nonradiative degradation.

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B. I. Stepanov. Lasers Based on Complex Organic Compounds. The prospects of the use of organic compounds of the dye type to generate radiation was reliably validated in 1964 by the staff of the BSSR Academy of Sciences Physics Institute. The principal distinctive features of the anticipated generation were predicted at the same time. The generation was experimentally realized in 1966^[1]. As it turned out, the capacity for generation is a widespread property of complex molecules. At the BSSR Academy of Sciences Physics Institute alone about 350 generating compounds have been studied. The principal sources of energy loss and a number of relationships connecting the generation properties with the structure of the molecules are already known. This allows us to conduct a directed search for prospective molecules for the construction of lasers for different purposes. Knowing the optical characteristics of a solution, we can compute a priori the generation parameters (the efficiency, power, energy, frequency, and duration). Where they are comparable, the theoretical and experimental results practically coincide. By measuring the generation parameters, we can in principle obtain new information about the properties of the excited states of the most active substances.

Lasers based on organic compounds possess a number of specific distinctive features which make them widely applicable for the solution of the most diverse scientific and technological problems. Their main property is the possibility of producing radiation of any wavelength in the range 2400–11180 Å^[2-4]. At the BSSR Academy of Sciences Physics Institute a device, called "Raduga" (Rainbow), has been developed which allows the realization of a smooth tuning of the laser radiation in the entire range of from 3600 to 10600 Å without having to replace the pumping source and the resonator elements^[3]. Also of extreme importance is the possibility of controlling the spectral characteristics of the generation. The width of the spectrum generated by a given solution can be changed at the experimenter's will within the limits of six orders of magnitude—from 10³ to 10⁻²–10⁻³ Å^[1,2,5]. To narrow the band one introduces into the resonator different strobing elements. The smooth tuning of the generation frequency is realized within the limits of the luminescence band (up to 1000 Å) by a simple rotation or shift of a strobing element. An operational frequency tuning with electronic control has also been accomplished, and this allows us to vary the frequency according to a prescribed law, even during the period of one generation pulse, and thereby insert

necessary information into the radiation.

Also possible is the production of generation with diverse time characteristics. There are in many laboratories operational, ultranarrow ($\sim 10^{-11}$ sec) light-pulse generators based on dyes^[2]. When pumped with a nitrogen laser, generation with a high pulse-repetition rate (50–100 Hz^[6]) is easily realized. At the BSSR Academy of Sciences Physics Institute a tunable laser with a repetition rate of up to 50 Hz and with ordinary pumping by a pulse tube has been tested^[7]. In the USA a steady generation has been accomplished with pumping by an argon laser^[8].

Theoretical and experimental investigations have shown that when generators based on the best dyes are excited by pulse tubes, their energy characteristics are close to the characteristics of solid-state lasers. The highest energy (>100 J) has for the present been obtained in the investigations^[9].

The preparation of microgenerators in the form of films and fibers has been reported lately, and this is important for integral optics. Owing to the high amplification factor and the wide band, organic compounds are used as light amplifiers.

Generators based on organic compounds are already being quite extensively used: in laser spectroscopy, nonlinear optics, photochemistry and luminescence, in high-resolution spectroscopy in the probing of plasmas and the atmosphere, in holography, medicine, and biology, i.e., wherever we need to tune the frequency of the laser radiation and the action on a substance under the conditions of a one-photon or a many-photon resonance. The practical utilization of generators of this type will expand rapidly from now on.

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