

# Scientific Session of the Division of General Physics and Astronomy, USSR Academy of Sciences (19-20 December 1973)

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A scientific session of the Division of General Physics and Astronomy of the USSR Academy of Sciences was held on December 19 and 20, 1973 at the conference hall of the P. N. Lebedev Physics Institute. The following papers were delivered:

1. Z. R. Mustel', Supernova Outbursts and Transformations of Elements.

2. Yu. N. Efremov, S. B. Novikov, and P. V. Shcheglov, Development of Ground-Based Optical Astronomy and its Prospects.\*

3. V. S. Letokhov, Selective Laser Irradiation of Matter.

4. D. N. Mirlin, Optical Studies of Surface Vibrations in Ionic Dielectrics and Semiconductors.

5. G. A. Askar'yan, V. A. Namiot, and M. S. Rabinovich, Use of Ultracompression of Matter by Light Reaction Pressure to Obtain Microcritical Masses of Fissile Elements, Ultrastrong Magnetic Fields, and Particle Acceleration.

We publish below brief contents of three of the papers.

V. S. Letokhov. Selective Laser Irradiation of Matter. Recent progress in broadening the range of laser wavelengths and in the development of methods for tuning and frequency control of lasing have enabled us to enter upon systematic study of the possibility of controlling selective chemical reactions with laser radiation. This problem is of fundamental importance for the use of coherent light in chemistry, especially in photochemistry and nuclear chemistry and possibly in biology, medicine, and other fields. The basic idea is to use the difference between the absorption spectra of substances not for analysis of their composition and structure, but for selective treatment of the substances with the object of changing their composition and properties. The general statement of the problem will be found in<sup>[1]</sup>.

A mixture of molecules (atoms) A and B with very closely similar chemical properties is considered, and the problem is to obtain preferential participation of, let us say, molecule A in a chemical reaction. As a coefficient of selectivity we might take the ratio of the number of reacted molecules A to the number of molecules B at equal initial concentrations:  $S = ([N_A])/([N_B]) - 1$ .  $S = 0$  in the absence of selectivity, and  $S \gg 1$

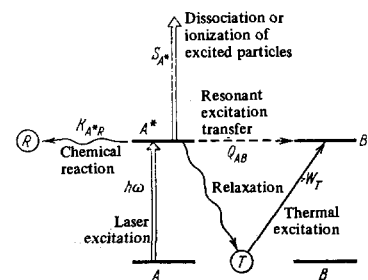
corresponds to high selectivity. If the absorption spectra of molecules A and B have at least one noncoincident absorption line belonging to molecule A, it can be used for selective excitation of molecules A only by laser radiation of the appropriate wavelength (see Figure). The selectively excited molecules A\* can, first of all, enter into a chemical reaction with a certain acceptor R at a higher rate than the unexcited molecules A and B. Secondly, the excited molecules have a dissociation or ionization energy that is smaller than that of unexcited molecules A and B, and this can also be used for selective laser treatment of the substance.

There are two mechanisms of excitation selectivity loss. First, a mixture of gases may be heated as a result of relaxation of excitation, with subsequent thermal (nonselective) excitation of both species of molecules. This mechanism appears, for example, on excitation of vibrational levels of the molecules. Secondly, there is a rather fast resonant transfer of excitation between molecules (atoms) A\* and B. To act selectively on molecules A, it is necessary that the rate of the selectivity-preserving processes (fast chemical reaction, dissociation of excited molecules) greatly exceed the rate of the stability-losing processes. The idea that selective reactions might be made possible by excitation of particles of a given species with monochromatic radiation was advanced 50 years ago<sup>[2]</sup>. A few successful experiments were made during the 1930's<sup>[3]</sup>. Naturally, the idea was resurrected with the advent of the laser<sup>[4]</sup>. But the high intensity of laser radiation makes possible selective photoionization of excited atoms<sup>[5,6]</sup> or photodissociation of excited molecules<sup>[5,6]</sup> at a rate much higher than that at which selectivity is lost due to collision processes. In many cases, this is of fundamental importance for preservation of selectivity, since, like the rates of the various selectivity-loss processes, the rate of chemical reaction of the excited particles is proportional to the number of collisions.

The paper reports results of an experimental study of the thermal mechanism of selectivity loss on excitation of vibrational levels of the NH<sub>3</sub> molecule by CO<sub>2</sub>-laser radiation in the continuous and pulsed modes<sup>[7]</sup> and an investigation of the rate of vibrational-vibrational excitation exchange in a mixture of <sup>14</sup>NH<sub>3</sub> and <sup>15</sup>NH<sub>3</sub> molecules and a buffer gas<sup>[8]</sup>. The measurement method is based on direct observation of the NH<sub>3</sub> vibrational level population kinetics as reflected in the strength of the absorption lines in the 2200 Å range<sup>[9]</sup>. A "bottleneck" effect due to rotational-level relaxation of the molecules<sup>[10]</sup> was observed experimentally in<sup>[8]</sup> on excitation of molecular vibrational levels with a short laser pulse, and population-factor measurements are reported for one sublevel of the C<sub>2</sub>F<sub>3</sub>Cl molecule<sup>[11]</sup>.

Study of the kinetics of selective vibrational excitation of <sup>15</sup>NH<sub>3</sub> molecules mixed with <sup>14</sup>NH<sub>3</sub> made it possible to select conditions for experimental observation of two-stage selective photodissociation of the molecules under the action of a CO<sub>2</sub> laser pulse and a pulse from an ultraviolet radiation source<sup>[12]</sup>. The selectivity of the dissociation was measured after separation of the <sup>14</sup>N and <sup>15</sup>N isotopes. The final products (H<sub>2</sub> and N<sub>2</sub>) of selective photodissociation contained about 80% of <sup>15</sup>N and 20% of <sup>14</sup>N when the initial concentrations were equal<sup>[13]</sup>, giving a selectivity S = 4. Maximum enrichment with a coefficient S ≈ 10–50 was obtained in experiments carried out under special conditions<sup>[13]</sup>.

Classification of processes causing retention of selectivity (fast chemical reaction, photodissociation or photoionization of excited particles) and loss of selectivity (thermal excitation and resonant excitation transfer).



The process of selective two-stage photodissociation is a practically universal tool for laser separation of isotopes. However, selective dissociation can be attained by excitation with radiation at a single frequency when predissociation absorption lines are used<sup>[13,14]</sup>. The paper describes the results of an experiment in selective predissociation of orthoiodine molecules by radiation from an argon laser at  $\lambda = 5145.3 \text{ \AA}$ <sup>[15]</sup>. Use of this method brought about a strong shift in the ortho-I<sub>2</sub>-para-I<sub>2</sub> equilibrium in the direction of para-I<sub>2</sub>, and the predissociation probabilities and relaxation rates were measured for the ortho and para components of the iodine. This method can be used to separate the isotopes <sup>127</sup>I and <sup>129</sup>I.

The conclusion examines the possibilities for attaining selectivity on overlapping absorption lines with single-quantum<sup>[6]</sup> and two-quantum<sup>[16]</sup> excitations of a gas at low pressure in the field of a standing wave, and also the possibility of preventing chemical binding of the dissociation products in the gaseous medium due to escape of molecule fragments from the irradiated gas jet.

<sup>1)</sup>The editors plan to publish, in one of the upcoming issues of this journal, an article by these authors that reflects the text of the paper delivered.

<sup>1</sup> V. S. Letokhov, *Science* **180** (4085), 451 (1973).

<sup>2</sup> E. Hartley, A. O. Pender, E. J. Bowen, and T. R. Merton, *Phil. Mag.* **43**, 430 (1922).

<sup>3</sup> W. Kuhn and H. Martin, *Zs. phys. Chem.* **B21**, 93 (1933); W. Kuhn, H. Martin, and K. H. Eldau, *ibid.* **B50**, 213 (1941).

<sup>4</sup> W. B. Tuffany, H. W. Moos, and A. L. Schawlow, *Science* **157**, 40 (1967).

<sup>5</sup> R. V. Ambartsumyan, V. P. Kalinin, and V. S. Letokhov, *ZhETF Pis. Red.* **13**, 305 (1971) [*JETP Lett.* **13**, 217 (1971)].

<sup>6</sup> R. V. Ambartsumyan and V. S. Letokhov, *IEEE J. Quantum Electron.* **QE-7**, 305 (1971); *Appl. Optics* **11**, 354 (1972).

<sup>7</sup> R. V. Ambartsumyan, V. S. Letokhov, G. N. Makarov, A. G. Platova, A. A. Puzetskii, and O. A. Tumanov, *Zh. Eksp. Teor. Fiz.* **64**, 771 (1973) [*Sov. Phys.-JETP* **37**, 392 (1973)].

<sup>8</sup> R. V. Ambartsumyan, V. S. Letokhov, G. N. Makarov, and A. A. Puzetskii, *Proc. of the Laser Spectroscopy Conference*, Vail, Colorado, USA, 1973.

<sup>9</sup> R. V. Ambartsumyan, V. S. Letokhov, G. N. Makarov, and A. A. Puzetskii, *Chem. Phys. Lett.* **16**, 252 (1972).

<sup>10</sup> V. S. Letokhov and A. A. Makarov, *Zh. Eksp. Teor. Fiz.* **63**, 2064 (1972) [*Sov. Phys.-JETP* **36**, 1091 (1973)].

<sup>11</sup> V. S. Letokhov, A. A. Makarov, and E. A. Ryabov, *Dokl. Akad. Nauk SSSR* **212**, 75 (1973) [*Sov. Phys.-Dokl.* **18**, 603 (1974)].

- <sup>12</sup>R. V. Ambartsumyan, V. S. Letokhov, G. N. Makarov, and A. A. Pureskiĭ, *ZhETF Pis. Red.* **17**, 91 (1973) [*JETP Lett.* **17**, 63 (1973)]; *Dokl. Akad. Nauk SSSR* **211**, 365 (1973).
- <sup>13</sup>V. S. Letokhov, *Chem. Phys. Lett.* **15**, 221 (1972).
- <sup>14</sup>E. S. Yeung and C. B. Moore, *Appl. Phys. Lett.* **21**, 109 (1972).
- <sup>15</sup>V. S. Letokhov, A. A. Makarov, and V. A. Semchishen, *ZhETF Pis. Red.* **18**, 515 (1973) [*JETP Lett.* **18**, 303 (1973)].
- <sup>16</sup>L. S. Vasilenko, V. P. Chebotaev, and A. V. Shishaev, *ibid.*, **12**, 161 (1970) [**12**, 113 (1970)].

D. N. Mirlin. Optical Studies of Surface Vibrations in Ionic Dielectrics and Semiconductors. This paper sets forth the results of a joint study made with V. V. Bryksin, Yu. M. Gerbshteĭn, and I. I. Reshina. A spectroscopic method was used to observe surface optical vibrations in crystals and investigate them in detail. In accordance with the phenomenological treatment, such vibrations arise in the frequency range between  $\omega_{TO}$  and  $\omega_{LO}$ —the limiting frequencies of transverse and longitudinal optical phonons—where the dielectric constant is negative (the range of "residual rays"). The studies were made on a series of ionic dielectrics and semiconductors with the structure of NaCl, CaF<sub>2</sub>, and TiO<sub>2</sub>, and on InSb and  $\alpha$ -SiO<sub>2</sub>. The experimental part of the work was done using a modification of the disturbed total internal reflection (DTIR) method, which made it possible to investigate the absorption in a nonradiative region of the spectrum, i.e., at  $k_x > \omega/c$  (where  $k_x$  is the wave vector of the surface vibrations). DTIR spectra were calculated for the configuration of the experiment. Dispersion relations for the surface vibrations were obtained experimentally for the first time, and the influence of anharmonicity on their characteristics was studied. It was shown that satisfactory agreement between the calculated and observed frequencies requires consistent consideration of the anharmonic contribution to the dielectric constant of the crystal. Splitting of the surface-vibration frequencies was detected in thin films: in this case, two surface-phonon branches were observed. Mixed surface plasma-phonon modes were investigated in degenerate semiconductors, dispersion and concentration dependences of the frequencies were recorded, and the damping of the surface plasmons was measured. (In InSb, it was found to be 2–3 times stronger than the damping of bulk modes.)

The influence of anisotropy on the conditions under which surface vibrations appear and on their characteristics was investigated. The existence of two types of surface-vibration branches in uniaxial crystals was established experimentally: one of them exists only in the polariton region of the spectrum and has no analog in isotropic crystals. As the  $k_x$  of these "anomalous" surface excitations increases, they mix with the bulk spectrum and attenuate.

Surface waves at the boundary between two dielectrics were also investigated, and extinction of these waves at a boundary with a metal was observed. "Boundary" plasmon-phonon modes at metal (semiconductor)-dielectric boundaries were studied. Various manifestations of the surface phonons can be expected in study of transport phenomena in thin films, in the surface layers of single crystals, in multilayered structures, in tunnel spectroscopy, etc. In particular, surface-phonon manifestations can be expected in Raman spectroscopy of semiconductors with excitation beyond the edge of the

fundamental band, where the light penetrates only to a small depth.

The basic results discussed in the paper were set forth in the following articles:

V. V. Bryksin, Yu. M. Gerbshteĭn, and D. N. Mirlin, *Fiz. Tverd. Tela* **13**, 2125 (1971); **14**, 543 (1972) [*Sov. Phys.-Solid State* **13**, 1779 (1972); **14**, 453 (1972)]; *Phys. Stat. Sol.* **B51**, 901 (1972).

V. V. Bryksin, D. N. Mirlin, and I. I. Reshina, *ZhETF Pis. Red.* **16**, 445 (1972) [*JETP Lett.* **16**, 315 (1972)]; *Fiz. Tverd. Tela* **15**, 1118 (1973) [*Sov. Phys.-Solid State* **15**, 760 (1973)]; *Sol. State Comm.* **11**, 695 (1972).

I. I. Reshina, Yu. M. Gerbshteĭn, and D. N. Mirlin, *Fiz. Tverd. Tela* **14**, 1280 (1972) [*Sov. Phys.-Solid State* **14**, 1104 (1972)].

G. A. Askar'yan, V. A. Namiot, and M. S. Rabinovich. Use of Ultracompression of Matter by Light Reaction Pressure to Obtain Microcritical Masses of Fissile Elements, Ultrastrong Magnetic Fields, and Particle Acceleration. The possibility of obtaining very high pressures by vaporizing metal<sup>[1]</sup> with a powerful light or charged-particle flux ( $p \approx I/v$ , where  $I$  is the power density of the incident radiation causing vaporization and  $v$  is the outflow velocity of the matter) has recently been put to use to obtain ultracompression<sup>[2]</sup>—an increase in the density of matter by hundreds and thousands of times—under quasismooth (without shock-wave formation) isostatic compression by a vaporization pressure  $p \approx 10^{11}$ – $10^{12}$  atm. At such densities and pressures, matter behaves like a degenerate electron gas whose pressure is determined by quantum motion of the electrons:  $p \sim n_e \epsilon$ , where we have from the indeterminacy principle:  $\epsilon \approx (\Delta P)^2/2m \approx \hbar^2/2m(\Delta x)^2 \approx \hbar^2 n_e^{2/3}/2m$  and the pressure  $p \approx \hbar^2 n_e^{5/3}/2m$ , i.e., the effective adiabatic exponent  $\gamma = 5/3$  unless the total number of electrons changes appreciably.

The paper<sup>[2]</sup> proposed the use of ultracompression to lower the threshold for initiation of controlled thermonuclear fusion and to increase its efficiency. We shall consider other aspects of ultracompression—the formation of microcritical masses of fissile elements<sup>[3]</sup>, ultrastrong magnetic fields<sup>[3]</sup>, and acceleration of particles<sup>[3]</sup>.

1. Microcritical masses of ultracompressed fissile elements. Back in 1943, Neddermeier (a reference to his work appeared only recently, in<sup>[4]</sup>) took note of the possibility of lowering the critical masses of fissile elements by explosive compression, but modest blast pressures did not open the possibilities inherent in ultracompression, which produces critical dimensions and masses so small that they can be accommodated in the small regions occupied by concentrated high-density radiation.

In fact, the critical dimension  $R_{cr} \approx l_f \sim 1/n_i$ , while the critical mass  $M_{cr} \sim n_i R_{cr}^3 \sim 1/n_i^2$ , whence it follows that even hundredfold density increases reduce the critical mass by a factor in the tens of thousands. The concentration of the nuclei then reaches  $n_i \approx 10^{25}$  cm<sup>-3</sup>, which corresponds to an ionization multiplicity  $Z_{eff} \approx 10$ . Ultracompression permits the use of an ultradense reflecting layer to reduce the critical size still further. The equation describing the development of the neutron avalanche is