"BLEACHING" OF MOLECULAR IODINE AND BREAKDOWN INDUCED IN IT BY LASER PULSES

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Submitted July 15, 1969

Zh. Eksp. Teor. Fiz. 58, 438–440 (February, 1970)

Changes in the absorption power of iodine vapor induced by pulses from a Q-switched ruby laser are investigated. It is found that with increase of the intensity from 10^7 to 10^9 W/cm^2 iodine gradually becomes bleached until it becomes totally transparent. At intensities close to the breakdown threshold values, a structure is observed, which consists of a large number of tiny growing points. The breakdown threshold value in iodine is found to be anomalously low.

We investigated the absorptivity of iodine vapor as a function of the intensity of a focused laser beam. It was observed that at intensities from 10^7 to 10^9 W/cm^2 the gas was gradually bleached until it became completely transparent. With further increase of the laser-pulse amplitude, a structure was observed, consisting of a set of small glowing points (Fig. 1a). When the intensity was slightly increased, the structure was transformed into the picture characteristic of breakdown (Figs. 1b, c). The threshold field turned out to be lower by two orders of magnitude than in the case of nonabsorbing gases (e.g., mercury) having approximately the same ionization potential.[1]

In our experiments, a laser pulse of 10 MW power and 50 nsec duration was focused by a lens with focal length 5 cm in the center of a heated cell (T = 420°K) made of stainless steel and filled with iodine vapor. The diameter of the focal spot was 3×10^{-2} cm. In the experiment, we registered simultaneously the incident and transmitted pulses with the aid of FÉU-52 photomultipliers and I2-7 nanosecond oscilloscopes. The laser beam was attenuated with neutral light filters. Much attention was paid to the homogeneity of the beam.

The results of the measurements are shown in Fig. 2 in the form of a plot of the transmissivity of the iodine vapor, i.e., the ratio of the power transmitted through the gas to the incident power, P_{tr}/P_{inc}, against the power of the incident beam (the latter quantity is shown in the figure in relative units, P_{inc}/P_t, where P_t = 28 MW). The experiments were performed at three iodine vapor pressures: 15 (see Fig. 2, squares), 60 (circles), and 200 (triangles) mm Hg, corresponding to transmissivities in the linear-absorption region of 0.8, 0.4, and 0.04, respectively. As follows from the diagrams, the transmissivity of the iodine vapor remains constant up to an intensity of 10^7 W/cm^2. With further increase of the intensity, the transmissivity of the iodine vapor increases, and at intensities on the order of 10^9 W/cm^2 the vapor becomes practically fully transparent at pressures 15 and 60 mm Hg. This means that the bleaching occurred essentially in the entire volume of the light cone. At a pressure of 200 mm Hg, a noticeable increase of the transmission is also observed, but no complete bleaching takes place in this case, owing to the large linear absorption.

Let us proceed to explain the results. The main contribution to the iodine spectrum in the region of ruby-laser emission (λ = 6943 Å) is made at T < 100°K by the system $^1\Sigma_g^+ \rightarrow {}^3\Pi_{1u}$ ($^1\Sigma_g^+$, $^3\Pi_{1u}$ are the ground and first excited electronic states), and the transition oc-

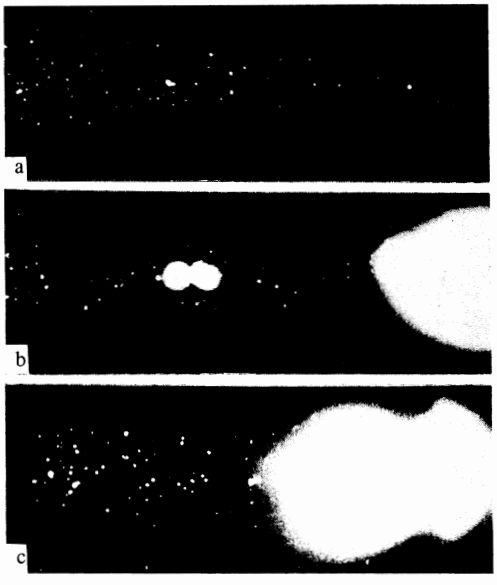

FIG. 1

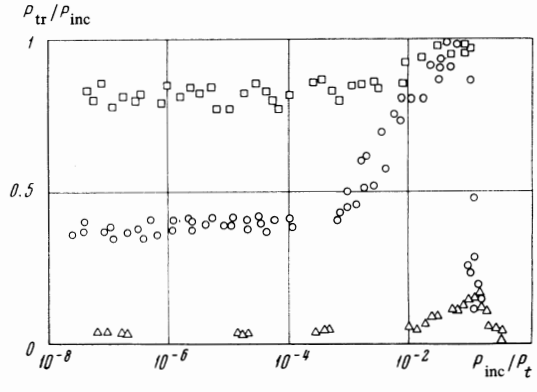

FIG. 2

curs from the zeroth vibrational level of the state $^1\Sigma_g^+$ into the continuum $^3\Pi_{1u}$ (the states $^1\Sigma_g^+$ and $^3\Pi_{1u}$ have a common dissociation limit of 1.54 eV). In addition, when light with λ = 6943 Å is absorbed, a transition is possible from the upper vibrational levels of the state $^1\Sigma_g^+$ to $^3\Pi_{0^+u}$ (where $^3\Pi_{0^+u}$ is the second excited electronic state of the I_2 molecule). The contribution of this transition to the absorption coefficient does not exceed 10–20% under our conditions.[2] Thus, the ruby-laser radiation produces mainly photodissociation of the iodine molecules into atoms in the ground state.

To estimate the intensity of the light at which the "bleaching" begins, we shall consider only the initial stage of the process, when the degree of dissociation of the iodine molecules is small and the recombination rate of the iodine atoms in triple collisions ($K_p = 10^{17}$ cm^6/mole^2 sec) is much smaller than the rate of deactivation of the vibrational levels.

Since the time of vibrational relaxation ($\tau_{vib} = 10^{-7}$ sec) is larger than the duration of the light pulse, at high radiation intensities there can occur a noticeable violation of the equilibrium with respect to the vibrational degrees of freedom, wherein the zeroth vibrational level may turn out to be depleted without an appreciable change in the populations of the upper levels.

If we approximate, for simplicity, the iodine molecule by a two-level model, then the population of the zeroth vibrational level can be estimated from the following kinetic equations:

$$\frac{dN_0}{dt} = -\sigma N_{ph} N_0 + ZP_{10}N_1 - ZP_{01}N_0,$$

$$\frac{d}{dt}(N_0 + N_1) = -\sigma N_{ph} N_0,$$

where σ is the cross section for light absorption (1.5×10^{-19} cm^2), Z is the molecule collision frequency, P_{01} and P_{10} are the probabilities of excitation and deactivation of the first vibrational level ($\sim 10^{-2}$), N_0 and N_1 is the population of the zeroth and first vibrational levels, and N_{ph} is the quantum flux.

From this it is easy to obtain the dependence of the absorption coefficient on the light intensity:

$$\varkappa = \varkappa_0 \left(\frac{k_2 + \sigma N_{ph}}{k_2 - k_1} e^{k_1 t} - \frac{k_1 + \sigma N_{ph}}{k_2 - k_1} e^{k_2 t} \right),$$

$$k_{1,2} = {}^1/_2 \{ \sigma N_{ph} - Z(P_{01} + P_{10}) \pm \pm ([\sigma N_{ph} + Z(P_{01} + P_{10})]^2 - 4ZP_{10}\sigma N_{ph})^{1/2} \},$$

where κ_0 is the absorption coefficient in the linear region. From this formula we find that at a pressure of 60 mm Hg, the "bleaching" begins at an intensity of 10^{25} quanta/cm^2 sec, in satisfactory agreement with experiment.

By way of an explanation of the breakdown structure described above, and also of the low breakdown threshold in iodine vapor, we can advance the following considerations. At high light intensities, photodissociation and excitation of the gas takes place, as a result of which the refractive index in the propagation channel can be appreciably altered.[3] This can lead to intersection of the light beams and consequently to a concentration of energy at individual points. At certain intensities, the energy concentration reaches the threshold value, leading to breakdown at several points.

A similar picture was investigated theoretically for the case of propagation of wave beams in nonlinear media.[4]

The authors are sincerely grateful to Yu. P. Raĭzer for useful discussions and to A. E. Abaliev and Y. A. Lebedev for help with the work.

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Translated by J. G. Adashko
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