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INVESTIGATION OF THE OPTICAL INHOMOGENEITIES OF THE ACTIVE MEDIUM OF A CF_3I PHOTODISSOCIATION LASER

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The time dependence of the refractive index during photodissociation is demonstrated by an interferometer technique. It is shown that a shock wave appears in the medium contaminated with the photodissociation products. The wave is due to evaporation (resulting from absorption of the pumping light) of molecular iodine deposited on the cell walls. It is shown that the time delay between generation and the pumping pulses depends weakly on the pressure (at high values of the latter). This effect is ascribed to an increase of the spontaneous emission linewidth and naturally leads to an increase of the generation threshold.

INTRODUCTION

THE generation of stimulated radiation in pulsed photodissociation of CF_3I was first obtained by Kasper and Pimental[1]

The photodissociation reaction and the secondary processes accompanying it[2] lead to a change of the concentration of the medium, and consequently of its refractive index. The change of the refractive index (i.e., of the optical length of the resonator), which can be local and varies in time, exerts a strong influence on the spectral composition, on the directivity pattern, and on the temporal characteristics of the laser emission[3]. An investigation of the optical inhomogeneities produced in substances during the course of photodissociation laser pumping is the subject of the present paper.

EXPERIMENTAL PROCEDURE

To investigate the refractive-index inhomogeneities, we used an interferometric method. A block diagram of the experimental setup is shown in Fig. 1. The radiation of an He-Ne laser (LG-35, λ = 6328 Å), operating in the $TEM_{00}q$ mode, passes through telescope 2, in the focus of which is placed an optical shutter (Kerr cell) 3, and is incident on a Mach-Zender interferometer made up of mirrors A, B, C, D during the time of a single pulse of duration from 400 to 1200 μsec.

A linear electric pulse is applied to the Kerr shutter from the modulator 4, which is synchronized with the ignition of the lamps. Prior to the experiment, the

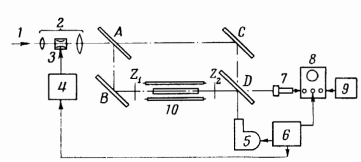

FIG. 1. Block diagram of experimental setup: 1—beam from He—Ne laser, 2—telescopic system, 3—Kerr cell, 4—modulator, 5—SFR camera, 6—control block fro SFR, 7—FÉU-61 photomultiplier, 8—oscilloscope, 9—audiogenerator, 10—photodissociation laser, A, B, C, D—Mach-Zender interferometer, Z_1, Z_2—flat mirrors.

10 mm diameter, with windows set at the Brewster angle, with flat mirrors Z_1 and Z_2. The mirror Z_1 had a dielectric coating with a reflection coefficient ~ 99% at the generation wavelength of CF_3I (λ = 1.32 μ).

The nonlocalized interference pattern, the bands of which were perpendicular to the optical axis of the laser cell, was time-photographed through an interference filter (with bandwidth 20 Å) by means of an SFR camera 5, operating in the slit-scanning mode (the camera slit was perpendicular to the fringes of the interference pattern and cut out part of the band in the direction of the cell diameter).

The shape and duration of the pump light pulse in the absorption band of CF_3I (~ 2700 Å) was determined by measuring the optical signal from the lamps with the aid of a DMR-4 monochromator, an FÉU-18 photomultiplier, and an oscilloscope (not shown in Fig. 1). The shape and duration of the photodissociation laser generation pulse was registered with FÉU-61 photomultiplier 7 with oscilloscope 8, and a scale signal duration and shape of the illuminating-light pulse are monitored with the aid of an FÉU-51 photomultiplier and an oscilloscope (not shown in the figure). In the BD arm of the interferometer is placed a laser 10, whose operation is based on molecular photodissociation. The laser 10 consists of a two-lamp elliptic illuminator (using IFP-2000 lamps), a quartz cell of from the audiogenerator 9 was applied to the second channel of the oscilloscope.

The measurements of the optical inhomogeneities of the active medium and of the generation pulse were carried out alternately under similar experimental conditions.

EXPERIMENTAL RESULTS AND DISCUSSION

1. The form of the interference patterns when the cell is illuminated with the first and second flashes of the lamps is shown in Figs. 2a and b (the cell was illuminated the second time without refilling with gas).

As seen from Fig. 2a, after the first illumination, the interference bands begin to bend practically simultaneously, i.e., the refractive index of the medium increases. After a certain time, the refractive index reaches a maximum and then decreases. (We shall henceforth consider only the part of the interference pattern corresponding to the time interval 0–100 μsec.)

The observed behavior of the interference bands can be connected with photodissociation and with processes that accompany it. On the basis of Bouguer's law with allowance for absorption in photolysis, we calculated the ratio of the pump intensity at the center and on the periphery of the cell (I_{center}/I_{per}) as a function of the pressure. For the pressures 15, 60, and 120 Torr, this ratio was respectively 0.97, 0.86, and 0.6, i.e., it changed little in a wide range of pressures of CF_3I. The pump intensity at the center of the cell differed little from the intensity at the sidewall. The latter explains the simultaneous bending of the interference-pattern fringes.

The change of the refractive index is due to the change of the composition of the gas. It can be readily understood here that during the initial stage, when the secondary processes can be neglected (the formation of I_2 and C_2F_6), the change of the optical path length expressed in fractions of the fringe is directly proportional to the compensation of the photodissociated CF_3I molecules.

Further elimination of the gas leads to the appearance of extraneous processes, principal among which is the formation of the molecules I_2, C_2, F_6, and pyrolysis of the CF_3I molecules. The pyrolysis of the CF_3I is manifest in a change of the refraction, the same

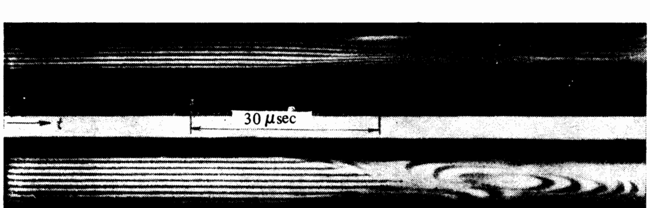

FIG. 2. Interference patterns—time scanned: a—for the first flash of the lamp and b—for the second.

as photodissociation. The formation of I_2 and C_2F_6 can lead to a decrease of the refractive index (a process inverse to photodissociation). However, no sharp decrease of the refractive index should be observed, since photodissociation also takes place at the same time. Experiment, on the other hand, shows a rapid decrease of the refractive index (following the maximum) (Fig. 2a). The latter, apparently, may be connected with the anomalous dispersion of the I_2 molecules, the absorption band of which captures the 6328A line of the He-Ne laser. However, a thorough understanding of this effect calls for additional investigations.

After the second irradiation of the cell with the light, the type of bending of the fringes changes appreciably. As shown by Fig. 2b, in this case a shock wave is observed. To explain the reason for the appearance of the shock wave, the following tests were performed.

1. A glass tube, which cuts off the ultraviolet region of the spectrum, corresponding to the CF_3I absorption band, was placed over the cell. No bending of the fringes was observed after the first illumination of the cell with the pump light. The glass tube was then removed, the first pump flash was applied, after which the glass tube was put on and the cell illuminated for a second time. A shock wave was observed. Thus, the shock wave is connected with absorption of the visible spectrum of the radiation from the lamps by the photodissociation products. We note that these photodissociation products should be localized on the periphery of the cell (uniform distribution of the molecules of the products over the volume, just as in the case of pure photodissociation, should lead to a uniform change of the refractive index). The latter remark leads us to the conclusion that the shock wave is connected with the I_2 molecules that settle on the walls of the cell. Molecular iodine, when exposed to light, absorbs quanta with wavelengths 4,000–6500 Å and dissociates into atomic iodine, producing a density jump on the periphery of the cell, and this in turn causes the shock wave.

Similar results are obtained by preliminary illumination of the cell with daylight during an appreciable time interval (about one day).

2. At a fixed pump power, we plotted the form of the interference patterns and the generation flash as functions of the pressure. (All the results pertain here to the first flash.) The interference patterns were used to plot the dependence of the optical path length (in percentage of the fringe shift S) against the time. The results are shown in Fig. 3.

As seen from the figure, with increasing pressure (30, 67, and 120 mm Hg), the rate of change of the refractive index increases (the fringes begin to bend earlier and shift more strongly); this corresponds to the general notions concerning the photodissociation and agrees with the expression (1) given above. On the other hand, the generation pulse also becomes more powerful, but it begins at approximately the same time. As seen from Fig. 3, the start of the generation pulse corresponds to different changes of the refractive index, i.e., to different concentrations of the excited iodine (assuming that during the initial stage, at least prior to the start of the generation, the secondary

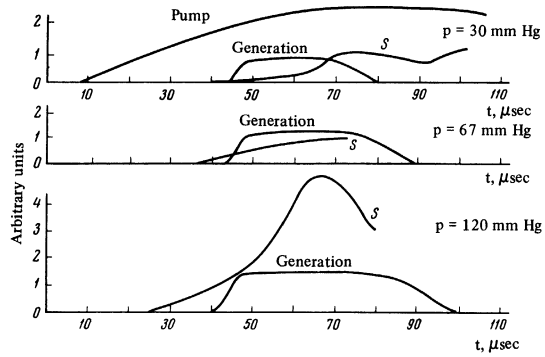

FIG. 3. Generation pulses and shift of interference fringes (S) at different pressures of CF_3I and at fixed pumping.

processes can be neglected). The start of the generation, as is well known, corresponds to the condition of equality of the gain to the loss.

The gain K_0 in photodissociation prior to the start of the generation was determined by the expression

$$K_0=\left[\frac{h\nu}{cL\Delta\nu_D}B_{ik}\right]N_{I*}, \qquad (1)$$

where L is the length of the active part, $\Delta\nu_D$ is the width of the spontaneous-emission line on which the generation takes place, B_{ik} is the Einstein coefficient, and N_{I*} is the concentration of the excited iodine.

Expression (1) has been written out under the assumption that prior to the start of the generation there are no iodine atoms in the ground state. Thus, the course of S(t) in the initial section coincides with the course of $N_{I*}(t)$. Consequently, with increasing pressure, the generation starts at larger concentrations of the excited iodine (larger inverted population), and this can be due to two causes.

1. The loss in the gas may increase with increasing pressure. If this loss were due to absorption by an unknown impurity (and not by the decay products of CF_3I), then the generation delay time would depend on the pressure at small pressures. This dependence appears when the loss in the gas becomes comparable with the loss in the mirrors (the generation delay should increase with decreasing pressure. The dependence of the generation delay time (t_0) on the working-gas pressure (p) at a fixed pump is shown in Fig. 4 for different mirror transmissions (τ). As seen from the figure, the region of transition to the linear dependence shifts towards larger pressures with increasing mirror transmission. In the intermediate region, the losses in the medium and the losses to transmission of the mirrors should be approximately equal within the framework of this hypothesis, and the losses as functions of the pressure can be determined from the transition regions shown in Fig. 4. The losses determined in this manner increase almost linearly with pressure.

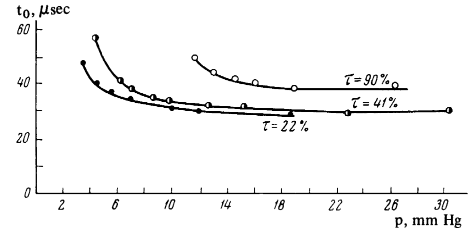

FIG. 4. Dependence of the generation delay time t_0 on the working-gas pressure at a fixed pump for different mirror transmissions.

The absolute value of the losses turn out to be very large. For example, at 14 mm Hg, the loss coefficient is $K_0 \approx 0.2\ cm^{-1}$, which naturally cannot be the case, for otherwise the length of the active medium would be strongly limited. Thus, for example, at $K_0 \approx 0.23\ cm^{-1}$, corresponding to a gain of 100 dB/m, the limiting length should be ~ 2 cm.

2. With increasing pressure, the line width of the spontaneous emission, on which generation takes place, may increase. This effect should lead to a decrease of the gain and consequently to an increase of the generation threshold. The absence of a dependence of the generation delay on the gas pressure (at large pressures), as seen from formula (1), indicates in this case that the line width should increase approximately in proportion to the pressure.

It should be noted that the form of the dependences of the generation delay on the pressure (Fig. 4) does not contradict this hypothesis. Indeed, at significantly low pressures, so long as the width of the shock contour does not exceed the Doppler width, the generation delay time will decrease with increasing pressure. With increasing mirror transmission coefficient, the threshold increases, and this should cause an increase of the slowly varying value of t_0, corresponding to large pressures.

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