

KINETICS OF THE GENERATION SPECTRUM OF A PHOTODISSOCIATION IODINE LASER

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The generation spectrum of a photodissociation iodine laser is investigated. It is found that the dependence of the frequency composition of the iodine laser radiation on parameters of the active medium, pumping and cavity is defined by a dimensionless combination, δ , of the parameters which characterize the ratio of the relaxation rate between the hyperfine structure sublevels of the $^2P_{1/2}$ excited state to the rate of the stimulated radiative transitions. For small values of the ratio ($\delta \ll 1$) generation involves two, and for large values ($\delta \gg 1$)—one component of the hyperfine structure of the $^2P_{1/2} - ^2P_{3/2}$ working transition. The experimental data agree qualitatively with the conclusions of a theoretical analysis.

THIS article is devoted to the generation kinetics of an iodine laser. We know^[1] that the generation in this laser is on the atomic iodine transition $5^2P_{1/2} - 5^2P_{3/2}$. Both states have hyperfine structures (see Fig. 1). The spectrum of the $5^2P_{1/2} - 5^2P_{3/2}$ transition consists of two groups of lines: $F = 2 \rightarrow F' = 4, 3, 2$ and $F = 2 \rightarrow F' = 3, 2, 1$. In the LS-coupling approximation it is easy to calculate the probabilities $W_{FF'}$ of the corresponding radiative transitions:

Transition $F - F'$:	3-4	3-3	3-2	2-3	2-2	2-1
$W_{FF'}$, sec^{-1} :	5.08	2.20	0.63	2.46	3.07	2.38

Experimental investigations^[1,2] have shown that the generation spectrum has an extremely irregular form. Depending on the experimental conditions, the generation occurs on one, two, and sometimes three of the six hyperfine-structure components, and the number of spectral components can change during the time of one lasing pulse. The causes of this instability and the dependence of the generation spectrum on the laser parameters have remained unexplained to this day.

The present detailed analysis of the specific features of a photodissociation iodine laser provides the answer to this question and indicates method of controlling its spectrum. The latter is important in a number of applications, particularly in nonlinear optics. In essence, the picture of the spectrum is determined (in the absence of a magnetic field¹⁾) by the ratio of the probability of the radiative transition and the frequency of the collisions that lead to the "mixing" of the working sublevels of the excited state. We present also some results of an experimental study of the spectral composition of the iodine-laser generation. Qualitative agreement was observed between the theoretical deductions and the experimental results.

1. Before we proceed to investigate the kinetics of the iodine-laser spectrum, we must analyze the basic processes responsible for the population of the working sublevels. An analysis of the possible types of interactions that occur when an iodine atom collides with the

surrounding particles shows that the mixing of the sublevels inside the ground and excited $^2P_{3/2}$ and $^2P_{1/2}$ states of the iodine proceeds in essentially different fashions.

The point is that collisions with particles having no electronic moment cause hardly any mixing of the sublevels of the excited state $^2P_{1/2}$. By virtue of the Kramers theorem^[3], the probability of a transition between sublevels with angular momentum $J = 1/2$ is equal to zero in the first order of perturbation theory for all interactions of the electrostatic type. The transition becomes possible only in second order of perturbation theory, owing to an admixture of a state with angular momentum $J = 3/2$, and the rate of such transitions is given by $\langle v\sigma \rangle \sim |\Delta E_{1/2, 3/2}|^{-2}$ ^[4]. It can be assumed that for iodine in the state $5^2P_{1/2}$ the mixing rate due to this type of interaction does not exceed $\sim 10^{-13} \text{ cm}^3 \text{ sec}^{-1}$, as in the case of thallium in the $6^2P_{1/2}$ state (the fine splitting of iodine and thallium is practically the same).

The rate of sublevel mixing in excited iodine through exchange interaction (such an interaction leads to mixing of the sublevels of excited iodine in collisions with particles having an electronic moment) is much higher, $\langle v\sigma_{\text{exch}} \rangle \sim 10^{-10} \text{ cm}^3 \text{ sec}^{-1}$, but even this type of interaction does not play a significant role in the mixing of sublevels of the excited state $5^2P_{1/2}$. Under typical experimental conditions, the most effective mixing of the sublevels of the $5^2P_{1/2}$ state occurs at resonant excitation transfer in collisions with iodine atoms in the ground state (the rate of excitation transfer is $\langle v\sigma_{\text{et}} \rangle \approx 1.5 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$).

The Kramers theorem does not forbid any transitions of an iodine atom in the ground state, with angular momentum $J = 3/2$, and the main type of interaction responsible for the large rate of level mixing, is the Van der Waals interaction. An estimate of the mixing rate^[5] yields a value $\langle v\sigma \rangle \approx 0.4 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$, which is practically independent of the type of perturbing particle. The frequency of the corresponding transitions is $\nu(^2P_{3/2}) = N_0 \langle v\sigma \rangle$, where N_0 is the total number of particles in the working mixture.

The other mixing mechanisms considered above play practically no role for the ground state of iodine, since the transition frequencies due to these types of

¹⁾In the presence of a sufficiently strong magnetic field ($H > 100 \text{ G}$), the spectrum becomes much more complicated, owing to differences in the Zeeman splitting of the components. The influence of the magnetic field on the generation spectrum is not considered in the present paper.

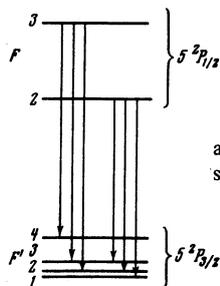


FIG. 1. Hyperfine splitting of the iodine atom levels $5^2P_{1/2}$ and $5^2P_{3/2}$. The arrows show the possible radiative transitions.

interaction are proportional to the concentration of particles with nonzero electronic moment, for example, to the concentration N_I of the iodine atoms in the working mixture, which are always small in comparison with the total number of particles. In a photodissociation iodine laser we usually have $N_I/N_0 \sim 10^{-1}-10^{-3}$.

Thus, the frequency $\nu(^2P_{3/2}) \sim N_0 \langle \nu \sigma \nu \rangle$ of sublevel mixing inside the ground state is much higher than the corresponding value $\nu(^2P_{1/2}) \sim N_I \langle \nu \sigma_{et} \rangle$ for the excited state.

Under typical conditions of a photodissociative iodine laser, the ground-state sublevel mixing frequency $\nu(^2P_{3/2})$ is much larger than the probability of the stimulated transitions

$$\nu(^2P_{3/2}) \gg W_{stim}. \quad (1)$$

Thus, for example, at a radiation intensity $I \sim 10^5$ W/cm² and a pressure ~ 0.1 atm we have $\nu(^2P_{3/2}) \sim 10^9$ sec⁻¹ and $W_{stim} \sim I \sigma_{stim} / \hbar \omega \sim 10^8$ sec⁻¹ (on the basis of the experimentally measured line width^[6], we estimate the gain cross section at $\sigma_{stim} \sim 10^{-18}$ cm²). This means that the ground-state sublevel populations become almost instantaneously proportional to their statistical weights during the generation process, and all that varies in time is the total population of the $5^2P_{3/2}$ state. This circumstance makes it possible to assume the lower level to be common for all the spectral components. Naturally, in this case the lasing can occur only on the two most intense transitions, 3-4 and 2-2 (the gain is lower than the threshold value for all other lines).

Relation (1) can be violated only under highly specific conditions, namely, in the case of low active-mixture pressures and at very high pump intensity. Generation on a large number of lines is then possible.

Atomic iodine in the ground state builds up during the generation process²⁾, and the frequency of the transitions between the sublevels of the excited state $5^2P_{1/2}$ increases in time and can be either higher or lower than the probability of the stimulated emission. As will be shown below, in the former case the lasing spectrum consists of two lines, 3-4 and 2-2, and in the latter case the generation is only on the single component 3-4, which is the strongest of all six components. When investigating the generation kinetics of an iodine laser, it suffices to take into account the change of population of two sublevels of the excited state, assuming the lower level to be common to both lines, i.e., we can use a simple three-level model (see Fig. 2).

2. Within the framework of the assumed model, we can describe the generation kinetics with the aid of the

²⁾Photodissociation produces only excited iodine [7].

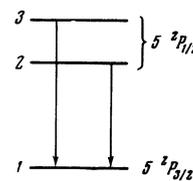


FIG. 2

following system of rate equations:

$$\dot{N}_{31} = f - k \frac{g_1}{g_3} N_1 (N_{31} - N_{21}) - c \sigma_{31} \left(1 + \frac{g_2}{g_1} \right) N_{31} n_{31}^{ph} - c \sigma_{21} \frac{g_2}{g_1} N_{21} n_{21}^{ph} + \gamma N_1, \quad (2)$$

$$\dot{N}_{21} = f + k \frac{g_1}{g_2} N_1 (N_{31} - N_{21}) - c \sigma_{31} \frac{g_3}{g_1} N_{31} n_{31}^{ph} - c \sigma_{21} \left(1 + \frac{g_2}{g_1} \right) N_{21} n_{21}^{ph} + \gamma N_1, \quad (3)$$

$$\dot{N}_1 = c \sigma_{31} \frac{g_3}{g_1} N_{31} n_{31}^{ph} + c \sigma_{21} \frac{g_2}{g_1} N_{21} n_{21}^{ph} - \gamma N_1, \quad (4)$$

$$\dot{n}_{31}^{ph} = -R n_{31}^{ph} + g_3 c \sigma_{31} N_{31} n_{31}^{ph}, \quad (5)$$

$$\dot{n}_{21}^{ph} = -R n_{21}^{ph} + g_2 c \sigma_{21} N_{21} n_{21}^{ph}. \quad (6)$$

We have used here the following notation:

$$N_{31} = \frac{n_3}{g_3} - \frac{n_1}{g_1}, \quad N_{21} = \frac{n_2}{g_2} - \frac{n_1}{g_1}, \quad N_1 = \frac{n_1}{g_1}, \quad (7)$$

n_i is the population, g_i is the statistical weight of the i -th level ($i = 1, 2, 3$; $g_1 = 24$, $g_2 = 5$, $g_3 = 7$); n_{31}^{ph} and n_{21}^{ph} are the photon densities corresponding to the transitions 3-1 ("strong" line) and 2-1 ("weak" line)³⁾; σ_{31} and σ_{21} are the corresponding effective gain cross sections; the pumping rate f for one nondegenerate state is given by

$$f = \frac{N_m}{g_2 + g_3} \int \sigma_{\omega} P_{\omega} d\omega [\text{cm}^{-3} \cdot \text{sec}^{-1}],$$

where N_m is the concentration of the active medium, σ_{ω} is the absorption cross section, P_{ω} is the spectral density of the pump-photon flux [cm⁻²]; $k = \langle \nu \sigma_{et} \rangle (g_2 + g_3) / g_2 g_3$, where $\langle \nu \sigma_{et} \rangle$ is the rate of resonant excitation transfer; $1/R$ is the lifetime of the photon in the resonator; c is the speed of light; γ is the relaxation rate of the lower iodine level [sec⁻¹].

The system (2)-(6) takes into account only the most effective excited-state hyperfine-structure sublevel-mixing mechanism, namely mixing at resonant excitation transfer. Quenching of the excited iodine does not exert a noticeable influence on the spectrum, since the ground state is populated under generation conditions mainly as a result of induced radiative transitions. In this connection, the quenching processes are disregarded in Eqs. (2)-(6). The question of the relaxation of the ground state of iodine is of interest in itself. Up to now, the only such process was assumed to be the recombination of iodine atoms into the I_2 molecule. This reaction calls for participation of a third body, so that at the usual working-gas pressure, ~ 0.1 atm, it is relatively slow and hardly changes the population of the lower level of the iodine atom during one generation pulse. Recent detailed investigations of the reactions accompanying the photodissociation of CF_3I molecules^[8] have shown that there exists a very rapid recombination process $CF_3 + I \rightarrow CF_3I$, and that such a

³⁾We recall that the 3-1 transition corresponds to the most intense hfs line $F = 3 \rightarrow F' = 4$, and the 2-1 transition corresponds to $F = 2 \rightarrow F' = 2$.

reaction can lead to an appreciable relaxation of the lower iodine level $5^2P_{3/2}$. So far, however, such investigations were performed only for the CF_3I molecule. Therefore the question of the relaxation of the lower level calls for a separate analysis for each concrete active medium. To describe phenomenologically the relaxation of the lower level, we confine ourselves in the present article to introduction of a term $-\gamma N_1$ in Eq. (4), without specifying the value of γ , and investigate in such a formulation the two cases $\gamma = 0$ and $\gamma \neq 0$.

3. We consider first the case of negligibly small relaxation of the lower level, $\gamma = 0$. We assume for simplicity that the pump intensity is independent of the time. The threshold values of the inversion for the transitions 3-1 ("strong" line) and 2-1 ("weak" line) are equal to

$$N_{31}^{\text{thr}} = R / g_3 c \sigma_{31}, \quad N_{21}^{\text{thr}} = R / g_2 c \sigma_{21}. \quad (8)$$

The ratio $N_{21}^{\text{thr}} / N_{31}^{\text{thr}}$ varies with pressure in the range⁴⁾

$$1.3 < N_{21}^{\text{thr}} / N_{31}^{\text{thr}} = g_3 \sigma_{31} / g_2 \sigma_{21} < 2.3. \quad (9)$$

The threshold condition is therefore satisfied earlier for the 3-1 transition than for the 2-1 transition. Generation on the "strong" line begins to develop starting with the instant of time

$$T_{31}^{\text{thr}} = N_{31}^{\text{thr}} / f \quad (10)$$

Estimates show that in most cases of practical interest the generation development time is $\tau_0 \sim 5 \sqrt{T_{31}^{\text{thr}} / R} \ll T_{31}^{\text{thr}}$,⁵⁾ i.e., it can be assumed that a quasistationary generation regime on the transition 3-1 is established starting with the instant T_{31}^{thr} . In this case $n_{21}^{\text{ph}} = 0$ and Eq. (6) should be discarded. It is easy to show that in the quasistationary regime the generation on the 3-1 transition differs little from the threshold value, $|N_{31} - N_{31}^{\text{thr}}| \sim N_{31}^{\text{thr}} / RT_{31}^{\text{thr}} \ll N_{31}^{\text{thr}}$, since in practice we always have $RT_{31}^{\text{thr}} \gg 1$. We therefore put $N_{31} \approx N_{31}^{\text{thr}}$ and $N_{31} = 0$, and seek quasistationary solutions for $N_{21}(t)$, $N_1(t)$ and $n_{31}^{\text{ph}}(t)$ from Eqs. (2)-(4). These equations have an exact integral:

$$g_3 N_{31} + g_2 N_{21} + (g_1 + g_2 + g_3) N_1 = (g_2 + g_3) f t. \quad (11)$$

Further, eliminating n_{31}^{ph} (under the condition $\dot{N}_{31} \approx 0$), we obtain an equation for the inversion N_{21} on the weak transition 2-1:

$$\frac{d}{dt} \left(\frac{N_{21}}{N_{31}^{\text{thr}}} \right) = \frac{g_1}{T_{31}^{\text{thr}} (g_1 + g_2)} \left\{ 1 + \delta \frac{N_{21}}{N_{31}^{\text{thr}}} \left[\frac{N_{21}}{N_{31}^{\text{thr}}} - \frac{g_2 + g_3}{g_2} \left(\frac{t}{T_{31}^{\text{thr}}} - 1 \right) \right] \right\}, \quad (12)$$

$$\delta = \frac{k}{f} \left(\frac{R}{g_3 c \sigma_{31}} \right)^2 = k N_{31}^{\text{thr}} T_{31}^{\text{thr}} \sim \frac{\nu(^2P_{3/2})}{W_{\text{stim}}} \quad (13)$$

⁴⁾ $g_3 \sigma_{21} / g_2 \sigma_{31} = 2.3$ at low pressures. At higher pressures the hfs components $F = 2 \rightarrow F' = 2$ and $F = 2 \rightarrow F' = 1$ overlap, it is necessary to use their summary probability to calculate the gain cross section for the "weak" line, and $g_3 \sigma_{31} / g_2 \sigma_{21} = 1.3$.

⁵⁾ The generation intensity increases from the level of the spontaneous emission $n_{\text{sp}}^{\text{ph}}$ to the quasistationary value $n_{\text{st}}^{\text{ph}}$ approximately like $n_{\text{ph}} = n_{\text{sp}}^{\text{ph}} \exp [Rt^2 / 2T_{31}^{\text{thr}}]$, i.e., within a time $\tau_0 \approx 2[(T_{31}^{\text{thr}}/R) \ln(n_{\text{st}}^{\text{ph}}/n_{\text{sp}}^{\text{ph}})]^{1/2}$.

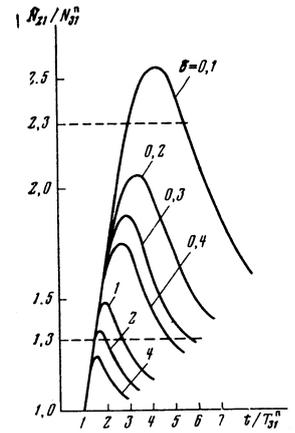


FIG. 3. Plot of $N_{21}/N_{31}^{\text{thr}}$ vs. t/T_{31}^{thr} at different values of δ . $N_{31}^{\text{thr}} = 2.3$ and 1.3 at low and high pressures, respectively.

with the initial condition

$$N_{21}(t = T_{31}^{\text{thr}}) / N_{31}^{\text{thr}} = 1. \quad (14)$$

Numerical solutions of (12) for different values of the parameter δ are shown in Fig. 3. It is seen from these solutions at sufficiently large values⁶⁾ of δ the threshold value of the inversion on the "weak" transition 2-1 is not reached, and no generation occurs on this line. This means that the rapid mixing of the sublevels 3 and 2 equalizes their population, as a result of the accumulation of the iodine in the ground state. In other words, the rate of transfer of the atom from the sublevel 2 to the sublevel 3, followed by emission at the frequency of the 3-1 transition, is larger at large values of δ than the rate of pumping to the sublevel 2.

At small δ , the inversion N_{21} increases almost linearly and at the instant of time

$$t = T_{21}^{\text{thr}} \approx T_{31}^{\text{thr}} + \frac{g_1 + g_3}{g_1} \frac{N_{31}^{\text{thr}} - N_{31}}{f} \quad (15)$$

it reaches the threshold value $N_{21} = N_{21}^{\text{thr}}$. Generation on the weak line sets in at this instant, and a quasistationary regime in which both components are generated is established, with $N_{21} \approx N_{21}^{\text{thr}}$ and $N_{31} \approx N_{31}^{\text{thr}}$. When the iodine in the ground state is accumulated, the frequency of the transitions between the sublevels 2 and 3 increases. After a time interval⁷⁾

$$\Delta t \approx \frac{g_2}{g_2 + g_3} \frac{1}{k(N_{21}^{\text{thr}} - N_{31}^{\text{thr}})} - \frac{g_1 + g_2 + g_3}{g_2 + g_3} \frac{N_1(T_{21}^{\text{thr}})}{f} \quad (16)$$

the sublevel-mixing frequency becomes so large that generation on the "weak" line 2-1 is quenched, N_{21} at $t > T_{21}^{\text{thr}} + \Delta t$ drops below the threshold value, $N_{21}(t) < N_{21}^{\text{thr}}$, and approaches the value N_{31}^{thr} . As a result, stable generation on one strong line is established at $t > T_{21}^{\text{thr}} + \Delta t$.

We have considered above a case in which the entire resonator is filled with the active medium. If the resonator length is L and the length of the active volume is l , it is necessary to make the following substitutions in all the obtained formulas: $N^{\text{thr}} \rightarrow N^{\text{thr}} L/l$, $T^{\text{thr}} \rightarrow T^{\text{thr}} L/l$, and

$$\delta = \frac{k}{f} \left(\frac{R}{g_3 c \sigma_{31}} \right)^2 \left(\frac{L}{l} \right)^2. \quad (17)$$

⁶⁾ At large values of δ , the frequency of mixing of the excited-state sublevels exceeds the probability of the induced radiative transitions (see (13)).

⁷⁾ At $\delta \ll 1$, the second term of (16) is negligibly small.

Thus, the character of the generation spectrum of a photodissociation laser is determined mainly by the value of the dimensionless parameter δ , which depends on the pump f , on the composition and pressure of the working mixture, on the cross section σ_{31} , and on the properties of the resonator (R). The most complicated is the dependence on the partial pressures of the working mixture. In the general case the dependence of δ on the pressure of the active medium is nonmonotonic, this being connected with the following circumstance. The pumping rate f in a photodissociation laser is proportional to the pressure of the active medium N_M , and the cross section is $\sigma_{31} \sim 1/\Delta\omega$, where $\Delta\omega$ is the width of the gain contour and depends on the pressure of both the active medium N_M and the buffer gas N_B . At sufficiently low working-mixture pressures, the line width is determined by the Doppler effect and does not depend on the pressure at all, so that in this region the dependence of the parameter δ on the pressure N_M is determined entirely by the quantity $f \sim N_M$, and in accord with (17) the value of δ decreases with increasing pressure of the active medium N_M . Starting with a certain pressure, however, the line width begins to be determined by the impact broadening mechanism, and therefore depends linearly on the pressure of the active medium (as well as on the pressure of the buffer gas). In this pressure range, δ increases with increasing pressure. It is possible to realize both cases in the experiments, and this is why the photodissociation iodine laser operates in a very wide range of pressures from several Torr to hundreds of Torr. Under conditions when the parameters of the pump lamp and of the resonator are fixed, δ increases with increasing buffer-gas pressure, the time of generation on the weak line Δt decreases, and starting with a certain pressure there can exist only a single-frequency generation regime⁸⁾.

4. The presence of a noticeable relaxation of the ground state of iodine can lead to stabilization of the two-frequency stabilization regime. After a time $\sim 1/\gamma$ from the onset of generation on the strong line, a stationary frequency of the mixing of sublevels 2 and 3 is established:

$$v_{st} = kf(g_2 + g_3) / \gamma.$$

From the solution of the system (2)-(6) under the conditions of the stationary two-frequency regime it is easily seen that simultaneous generation on the two lines is possible if the following condition is satisfied:

$$\eta = v_{st} T_{s1}^{thr} \frac{1}{g_2} \left(\frac{N_{21}^n}{N_{s1}^n} - 1 \right) < 1.$$

At $\eta > 1$, lasing is possible only on the strong line. However, a more detailed discussion of the spectrum in the presence of relaxation of the lower iodine level will not be presented here, for this calls for knowledge of the concrete relaxation mechanisms in different photodissociation iodine lasers, and these have not yet been sufficiently studied to date.

We note that in the case of very rapid relaxation under the condition $\gamma/W_{stim} \gg 1$, the concentration

⁸⁾At a large value of N_B/N_M the mixing of excited levels by the molecules of the buffer gas can become significant (see Sec. 1).

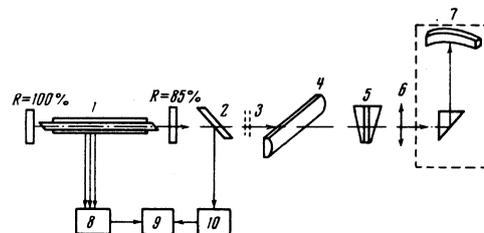


FIG. 4. Block diagram of experimental setup: 1—coaxial flash lamp with internal cell diameter 8 mm and length 600 mm; 2—plane-parallel glass plate; 3—IKS-3 filter; 4—cylindrical lens with focal length $F = 170$ mm; 5—Fabry-Perot interferometer with gap 3 mm between mirrors; 6—objective of SFR-2M high-speed camera; 7—SFR-2M camera operating in the slit-scanning mode with a luminor insert for the registration of radiation with $\lambda = 1.315\mu$; 8, 10—photocells F-7 and F-111, respectively; 9—OK-17M oscilloscope.

of the iodine in the ground state is low and an important role may be assumed by the mixing of the excited-state sublevels by exchange interaction in collisions of two excited iodine atoms.

5. We present below experimental data on the variation of the generation spectrum of a photodissociation iodine laser with C_3F_7I molecules and a weak magnetic field as the buffer-gas (xenon) pressure is increased.

A block diagram of the experimental setup used in the study is shown in Fig. 1. The radiation from laser 1, the working medium of which was in the internal cavity of a coaxial flash lamp^[9], was focused by cylindrical lens 4 on a Fabry-Perot interferometer. The interference pattern, localized in the focal plane of the cylindrical lens, was time-scanned with a high-speed camera on the SFR-2M type and registered on a luminor, from which it was subsequently photographed on an ordinary A-2 film.

The shape and duration of the pump and lasing pulses were registered respectively with type F-7 and F-11 photocells, the signals from which were fed to an OK-17M two-beam oscilloscope.

The magnetic field was measured at different points of the laser cell. The measurements were made with a magnetic probe comprising an inductance coil with four turns of 5 mm diameter. The probe axis was oriented in various fashions relative to the cell axis. The measurements have shown that the magnetic field in the cell does not exceed 40 G, corresponding to the sensitivity limit of the measuring apparatus (this probe and the OK-17M oscilloscope).

Figure 5 shows the variation of the generation spectrum with variation of the buffer gas (Xe) pressure. The interference patterns show that when the active laser medium is pure C_3F_7I at 15 Torr there is a simultaneous lasing at two hfs lines of the atomic transition (Fig. 5a). When xenon with partial pressure 120 Torr is added (Fig. 5b), the lasing on the 3-4 transition begins earlier than on the 2-2 transition, and ends later. When the pressure of the working medium is atmospheric (Fig. 5c), the generation is on the 3-4 line only. The generation energy in all three cases was approximately 1 J and the pulse duration 20 μ sec.

As seen from the experimental data, when the pressure of the buffer gas is increased the generation goes from a two-frequency to a one-frequency regime. Such

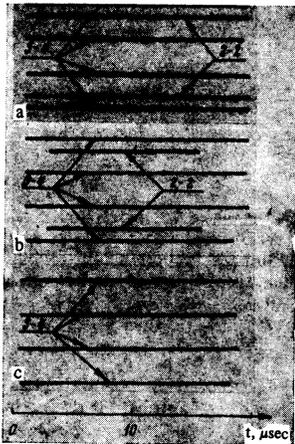


FIG. 5. Time scan of the generation spectrum: a— C_3F_7I pressure 15 Torr; b—partial C_3F_7I and xenon pressures 15 and 120 Torr, respectively; c—partial pressures 15 and 750 Torr.

a behavior of the generation spectrum agrees qualitatively with the theoretical analysis presented above. With increasing buffer-gas pressure, the line width increases, i.e., the cross section and the probability of induced emission decrease. The parameter δ [see (17)] is increased thereby, and at sufficiently large values of δ , as shown above, there should be no generation on the 2-2 transition at all. It is difficult to present in this case an accurate quantitative comparison of theory and experiment, since the pump intensity at the instant of onset of generation is determined with a very large error. Additional experimental data are needed for a quantitative comparison of theory and experiment.

6. A recent paper by Campbell and Kasper^[10] is devoted to the generation spectrum of a bromine photodissociation laser using CF_3Br molecules. They have observed that generation on the transition 2-3 occurs almost simultaneously with generation on the transitions 1-1 and 1-2 of the bromine atom, in spite of the appreciable difference in gain. It is concluded on this basis that the level $F = 1$ of the $4^3P_{1/2}$ state of the bromine atom is predominantly populated in the photodissociation of the CF_3Br molecule. These assumptions by Campbell and Kasper that one of the sublevels of the hyperfine structure is selectively populated in photodissociation has no physical foundation whatever. To explain the observed picture of the spectrum it suffices to assume the presence of a magnetic field (of which

no mention is made in^[10]). The presence of even a rather weak magnetic field (on the order of several hundred gauss)⁹⁾ greatly alters the form of the generation spectrum. Owing to the different values of the Zeeman splittings of the lines 2-3, on the one hand, and 1-1 and 1-2, on the other, the gains for these transitions may turn out to be quite close. (We note that in the iodine laser, in the presence of a magnetic field, the gain for the weak transition 2-2 can be larger than for the strong transition 3-4, as a result of which lasing can set in earlier on the 2-2 transition.)

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¹V. S. Zuev, V. A. Katulin, and O. Yu. Nosach, Proc. Internat. Conf. "Lasers and Their Application," Dresden, E. Germany, June 10, 1970, Part 19, p. 1261.

²V. N. Volkov and I. G. Zubarev, *Kratkie soobshcheniya po fizike* No. 10, 10 (1970).

³L. D. Landau and E. M. Lifshitz, *Kvantovaya mekhanika* (Quantum Mechanics, Fizmatgiz, 1963 [Addison-Wesley, 1965]).

⁴H. M. Gibbs, G. G. Churchill, T. R. Marschall, J. F. Papp, and F. A. Franz, *Phys. Rev. Lett.* **25**, 263 (1970).

⁵A. N. Okunevich and V. I. Perel', *Zh. Eksp. Teor. Fiz.* **58**, 666 (1970) [*Sov. Phys.-JETP* **31**, 356 (1970)].

⁶V. S. Zuev, V. A. Katulin, V. Yu. Nosach, and O. Yu. Nosach, *ibid.* **62**, 1673 (1972) [**35**, 870 (1972)].

⁷V. Yu. Zaleskii, *ibid.* **61**, 892 (1971) [**34**, 474 (1972)].

⁸T. L. Andreeva, S. V. Kuznetsova, A. I. Maslov, I. I. Sobel'man, and V. N. Sorokin, *ZhETF Pis. Red.* **13**, 631 (1971) [*JETP Lett.* **13**, 449 (1971)].

⁹V. N. Volkov, V. S. Zuev, and V. A. Katulin, *Zh. Prikl. Spekt.* No. 2, 190 (1970).

¹⁰J. D. Campbell and J. V. V. Kasper, *Chem. Phys. Lett.* **10**, 436 (1971).