COMPARISON OF THE LIFETIMES AND EXCITATION RATES OF THE $4p^2D^0_{5/2}$ AND $4p^4D^0_{5/2}$ LEVELS IN IONIZED ARGON

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Excitation and deactivation of the upper laser levels of the 4p configuration in ionized argon were studied experimentally at current densities between 190 and 500 A/cm² and pressures between 0.2 and 1.5 Torr. It is found that quenching collisions with electrons play an important role in the deactivation of the upper laser levels.

1. INTRODUCTION

THE progress in the development of lasers based on ionic transitions in inert gases has attracted interest in the investigation of the processes of formation of the inverted population under conditions of a strong-current low-pressure discharge. This type of discharge is among those least investigated, and differs in principle from the weak-current low-pressure discharges and strong-current high-pressure discharges.

Serious experimental difficulties arise when attempts are made to determine the basic parameters of the plasma of a strong-current low-pressure discharge and to determine the characteristics of the elementary processes that govern the mechanism of inverted-population production. The known methods of investigating a low-temperature plasma, which have given good account of themselves in the study of weak-current low-pressure discharges or strong-current high-pressure discharges, are either unsuitable or ineffective in this case. In such a situation, progress in experimental research of the population-inversion mechanism is determined by the reliability of the theoretical analysis of the conditions under which the measurements are performed, by the possibility of improving the known methods of determining the plasma parameters, and the possibilities of developing new effective methods.

The inverted population of the generating transition is maintained by the difference between the lifetimes or rates of population of the upper and lower and working levels. Calculations presented in[1] have shown that the rates of population of the configurations 4p and 4s in ionized argon are approximately the same, and the probability of radiative depopulation of the configuration 4s is approximately 25 times larger than the probability of such a depopulation for the 4p configuration. This gives grounds for assuming that the inversion between the levels of the 4p and 4s configurations in ionized argon is maintained by the difference between the probabilities of the radiative decay of these levels.

Another important question, however, still remains unanswered: what causes the saturation of the inversion when the electric pumping is increased? Saturation of the output power of an argon laser with increasing discharge current was observed experimentally in[2]. This phenomenon can be adequately explained within the framework of the available experimental and theoretical data concerning the parameters of the strong-current low-pressure discharge plasma. As was correctly noted in[2], at the estimated electron densities, 10^{13}–10^{14} cm^{-3}, the influence of the dragging of the radiation cannot be appreciable, and consequently it does not determine the saturation of the output parameters of the ionic laser.

To determine the causes of the change of the population of the laser level under different discharge conditions, it is necessary to evaluate separately the contributions of the processes of excitation and deactivation of the ion. The purpose of the present investigation was to make separate measurements of the rates of population and the lifetimes of the upper laser levels of ionized argon as functions of the discharge conditions. The character of the relations obtained in this case should reveal the processes that lead to saturation of the inverted population. These processes become manifest before the lowering of the output power of the laser takes place.

2. MEASUREMENT PROCEDURE AND EXPERIMENTAL SETUP

The employed procedure is based on the effective saturation of the gain of the active medium in the laser's own radiation field. We have shown earlier[3,4] that the output power T of the stimulated emission is determined by the following relation:

$$P = \frac{\Delta I_{sp}}{I_{sp}{}^0} \frac{\tau_3}{\delta} lSW_2\hbar\Omega_0, \quad (1)$$

$\Delta I_{sp} = I^0_{sp} - I_{sp}$; I^0_{sp} and I_{sp} are the intensities of the spontaneous emission of the working level in the absence and in the presence of generation, respectively; S is the cross section of the tube with the active medium; τ_3 is the transmission of the mirror; δ is the loss in the resonator; W_2 is the number of acts of excitation of the upper laser level per unit time and per unit volume (electric pumping); $\hbar\Omega_0$ is the quantum energy of the given working transition; l is the length of the active medium.

It follows from (1) that the rate of population of the upper level is expressed in terms of the ratio $\xi = \Delta I_{sp}/I^0_{sp}$, which we shall henceforth call the relative modulation of the intensity of the spectral line of the working transition, and in terms of the generation power P. The relative modulation ξ characterizes the efficiency of conversion of the electric-pump energy in the

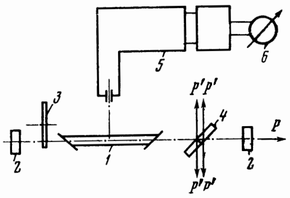

FIG. 1. Schematic diagram of experimental setup: 1—cell with active medium; 2—mirrors; 3—modulator; 4—glass plate; 5—monochromator; 6—galvanometer.

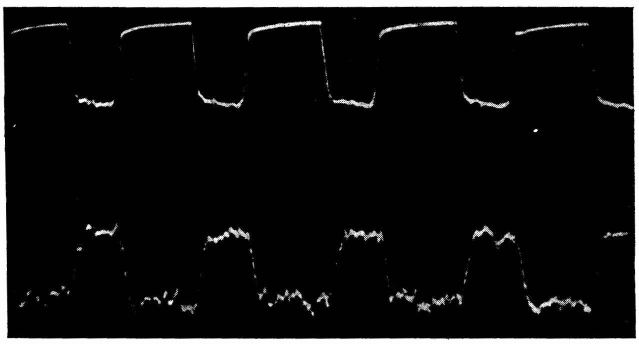

FIG. 2. Oscillograms of the pulse amplitude proportional to P (upper trace) and of the pulse amplitude proportional to ΔI_{sp} (lower trace).

active medium into coherent-radiation energy. For absolute measurements of the electric pumping, it is necessary in addition to know the volume Sl of the active medium and the ratio τ_3/δ.

In the stationary regime, the level population is determined by the pumping W_2 and by the total decay probability A_{22}. Taking this into account, and also the fact that the level lifetime is $\tau_{22} = I/A_{22}$, we obtain from (1)

$$\tau = \frac{\Delta I_{sp}}{P}\frac{\tau_3}{\delta}\frac{lS}{A_{21}}. \qquad (2)$$

The experimental setup is shown in Fig. 1. The output generation power was measured in the direction of the laser optical axis, and the intensity of the spontaneous emission in the presence and in the absence of generation were registered in a direction perpendicular to the axis. A mechanical modulator introduced into the resonator (interruption frequency ~15 kHz) has made it possible to display P and ΔI_{sp} on an oscilloscope screen simultaneously (see Fig. 2). I^0_{sp} was determined by additional measurements. In addition, provisions were made for registering all these quantities with the aid of a galvanometer.

To increase the density of the stimulated emission that leads to modulation of the level population, mirrors with transmission ~0.2% at the generation frequencies were installed in the resonator. Direct measurements of τ_3 and δ in such a resonator do not ensure the required accuracy of the absolute values of W_2, and we have therefore developed a procedure for directly measuring the ratio τ_3/δ. As seen from Fig. 1, a plate was inserted into the resonator at a certain angle to the axis; this plate introduced additional losses δ_{pl} as a result of Fresnel reflection by the faces. The interference of the reflected rays was excluded, and the absorption loss in the plate was negligible. If the plate angle is close to the Brewster angle, we have (taking into account the fact that the reflection coefficient of the mirror is > 99%)

$$\delta_{pl}/\tau_3 = 4P'/P, \qquad (3)$$

where P′ is the power reflected from the plate in one beam.

After determining the values of P and ξ without and with the plate in the resonator, we obtain from (1)

$$\frac{\tau_3}{\delta} = \frac{P_1/P_2 - \xi_1/\xi_2}{4(P'/P_2)(\xi_1/\xi_2)}. \qquad (4)$$

For the resonator used in our experiments, $\tau_3/\delta = 0.04 \pm 0.005$.

The procedure ensured an experimental accuracy on the order of ±15% in the determination of the absolute values of the rate of population of the level W_2, and ±5% in the determination of the relative lifetime variation. The output generation power was determined with a bolometer meter with accuracy ±5%.

3. MEASUREMENT RESULTS AND DISCUSSION

We investigated the processes of excitation and deexcitation of the two upper laser levels that determine the most intense generation lines in ionized argon, namely $\lambda = 4880$ Å (transition $4p^2D^0_{5/2} \to 4s^2P_{3/2}$) and $\lambda = 5145$ Å (transition $4p^4D^0_{5/2} \to 4s^2P_{3/2}$). The object of the study was an argon laser with a capillary having an inside diameter 2 mm, a discharge length 280 mm and a distance between outside mirrors 1100 mm at working pressures 0.2—1.5 Torr and a discharge current from 6 to 15 A.

Figure 3 shows the dependence of the lifetimes of the levels $4p^2D^0_{5/2}$ and $4p^4D^0_{5/2}$ on the discharge current i_d at a working pressure p = 0.7 Torr, while Fig. 4 shows the dependence of the lifetime on the pressure at a discharge current 10 A. In relative units, the plots for each of the indicated levels coincide. When the current density increases from 192 to 450 A/cm² (the current increases from 6 to 14 A), the lifetime decreases by approximately 50%. When the pressure is raised from 0.2 to 0.8 Torr, the lifetime decreases by approximately 60%.

Attention must be called to the fact that the relative variation of the lifetime with increasing current coincides for the doublet and quartet terms, although the probability of the radiative decay of these levels differs by a factor of 1.5[5]. It follows therefore that the probability of nonradiative deactivation of the level $4p^4D^0_{5/2}$ is approximately 1.5 times larger than that of $4p^2D^0_{5/2}$.

On the basis of the prevalent notions concerning the

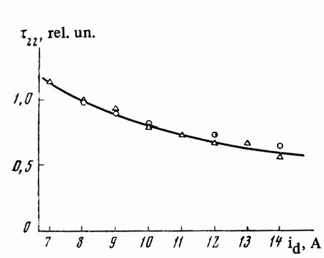

FIG. 3

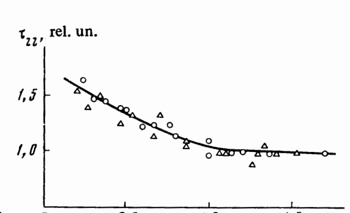

FIG. 4

FIG. 3. Dependence of the level lifetime on the discharge current i_d at constant gas pressure: Δ—level $4p^2D^0_{5/2}$, p = 0.73 Torr, O—level $4p^4D^0_{5/2}$, p = 0.67 Torr.

FIG. 4. Dependence of the level lifetime on the pressure at a discharge current 10A. Δ—level $4p^2D^0_{5/2}$, O—level $4p^4D^0_{5/2}$.

character of the variation of the discharge parameters with increasing current density and pressure[6-8], we can conclude from the obtained dependence of τ_{22} on the discharge conditions that the nonradiative decay of the upper levels is determined by electronic quenching processes. Indeed, the electron concentration increases with increasing current density and with increasing pressure, and this is accompanied by an increase in the number of the quenching collisions and by an appropriate decrease of the lifetime. To be sure, the same consequences result from quenching collisions between the ions and the ions, but estimates show that the frequency of the electron-ion collisions is larger by one order of magnitude than that of ion-ion collisions.

The mechanism deactivation of the excited ions, due to their drift to the wall, does not agree qualitatively with the experimental dependence of the lifetime on the pressure, while the mechanism of deactivation as the result of collisions with neutral atoms does not agree with the current dependence of τ_{22}, since the concentration of the neutral particles decreases with increasing current, and the lifetime should therefore increase.

Assuming that the electron concentration in the discharge is $n_e = 5 \times 10^{13}$ cm^{-3} [6], let us estimate the rate constants of the deactivation of the investigated levels:

$$\langle \sigma_{\text{quen}} V_e \rangle_{\text{quart}} = 10^{-6}\ \text{cm}^3 \cdot \text{sec}^{-1}; \quad \langle \sigma_{\text{quen}} V_e \rangle_{\text{doubl}} = 6 \cdot 10^{-7}\ \text{cm}^3 \cdot \text{sec}^{-1}.$$

Figure 5 shows the dependence of the rate of excitation of the doublet and quartet levels of the 4p configuration on the discharge current at constant pressure. With increasing discharge current, the electric pumping to the upper laser levels increases. This is connected, on the one hand, with the increase of the total number of electrons in the discharge, and on the other with the increase of the fraction of the fast electrons. Up to the maximum current densities attained by us (500 A/cm^2), we observed no symptoms or tendencies of saturation of the rate of population of the upper levels.

The intersection of the plots obtained at two different pressures, seen in Fig. 5, means that the rate of level population has an optimum pressure. The measured pressure dependences of W_2 (for the level $4p^2D_{5/2}$) at three fixed values of the current are shown in Fig. 6. The dependence at 8 A actually has a maximum, but with increasing current this maximum shifts towards higher pressures, going beyond 1.5 Torr. The maximum in the pressure dependence of W_2, and its shift, can be attributed to the fact that the increase of the pressure in the case of a constant current is accompanied by an increase of the total number of electrons and by a simultaneous decrease of the fraction of the fast electrons participating in the level-excitation processes. With increasing current, first, a decrease takes place in the gas density in the discharge channel, and second, the fraction of the fast electrons increases, and this causes the observed shift of the maximum of W_2 towards higher pressures.

The table lists the known published data on the rate of population of the upper laser levels of ArII, and the last column gives the results of the present investigation. There is good agreement between our results and the calculations of[6] as well as the experimental data of[9]. There is a discrepancy between our data and those of[5] by a factor of 4, and the reasons for this discrepancy are not clear.

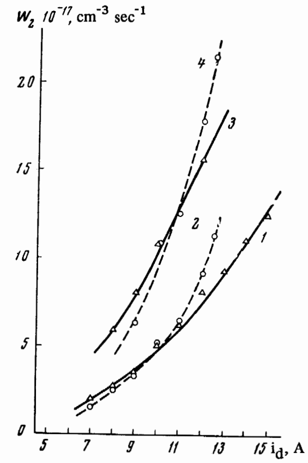

FIG. 5

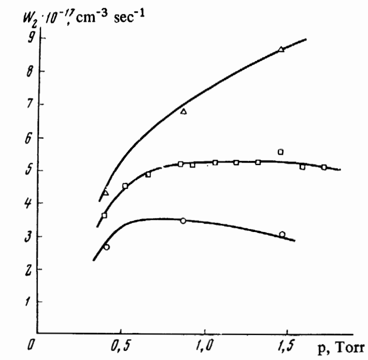

FIG. 6

FIG. 5. Dependence of the rates of excitation of the doublet and quartet levels of the configuration 4p on the discharge current at a constant pressure: 1—level $4p^2D^0_{5/2}$, p = 0.73 Torr; 2—level $4p^2D^0_{5/2}$, p = 1.07 Torr; 3—level $4p^4D^0_{5/2}$, p = 0.67 Torr; 4—level $4p^4D^0_{5/2}$, p = 1.33 Torr.

FIG. 6. Dependence of W_2 on the pressure at a constant current: ○—i_d = 8 A; □—i_d = 10 A; Δ—i_d = 12 A.

Source	p, Torr	j, A/cm^2	level	$W_2 \cdot 10^{-17}$, cm^{-3} sec^{-1},	$W_2 \cdot 10^{-17}$, cm^{-3} sec^{-1}, present work
[6]	0.5	100—500	$4p^2D^0_{5/2}$*	2.25—9.17	3±0,5 j=318 a/cm^2
[9]	0.2	220	$4p^2D^0_{5/2}$	1.8±0.1	1.4±0.2
			$4p^4D^0_{5/2}$	2.7±0.1	3.2±0.5
[5]	0.3	160	$4p^2D^0_{5/2}$	5.5±1.0	1.0±0.15
			$4p^4D^0_{5/2}$	8.0±1.5	2.3±0.3

*The calculation was carried out for the 4p configuration as a whole. It is assumed that the distribution of the population rate within the configuration is proportional to the statistical weights of the levels.

As seen from Fig. 5, the rate of population of the quartet level is approximately two times larger than the rate of population of the doublet level. This circumstance explains why a more rapid increase of the intensity of the 5145 Å line compared with the 4880 Å line is observed with increasing current. It should be borne in mind that, owing to the intercombination character of the transition, the gain for 5145 Å is much smaller than for 4880 Å. At small excesses of the gain of the active medium over threshold, the main contribution to the output power of the laser is given by the 4880 Å line. With increasing gain, the relative modulation ξ in the medium tends to saturate for both transitions, and the intensity of the output radiation, in accordance with (1), is determined only by the electric pumping to the upper level. Therefore the radiation power in the 5145 Å line should become equalized with the power in the 4880 Å line when the current density is increased, as is indeed observed in the experiment.

It was indicated earlier that according to[1] the rates of population of the upper and lower laser levels in ArII are approximately the same. We have established experimentally that the electric pumping to the upper level increases monotonically with increasing current density. In these investigations, the only possible causes of the decrease of the inverted population with intensification of the discharge are the processes that lead to a change in the probability of the depopulation of the working levels. As shown by approximate calculations, at electron densities $10^{13}-10^{14}$ cm^{-3} and at real optical discharge depths, the dragging of the radiation cannot cause a noticeable increase in the lifetime of the lower level. At the same time, the presented experimental data indicate that electronic quenching causes a noticeable change in the probability of the depopulation of the upper level. With increasing electron density, the contribution of the quenching collisions should increase, and this unavoidably leads to a limitation and then to a decrease of the inverted population. For the lower level, the probability of the radiative decay is much higher than for the upper level, and the contribution of the quenching collisions can be relatively small here.

It should be noted that the mechanism that limits the growth of the inversion becomes manifest before the occurrence of noticeable symptoms of saturation of the output generation power. At the present time there are no experimental data on the character of the change of the electric pumping at current densities above 500 A/cm^2. Nonetheless, the results allow us to expect deactivation of the excited ions as the result of quenching collisions with the electrons to be one of the main causes of the saturation of the output parameters of ionic lasers.

4. CONCLUSIONS

1. An experimental procedure was developed for separately determining the contributions made by the processes of excitation and deactivation to the populations of the working levels of an inverted transition.

2. Experimental data were obtained on the dependence of the rate of population and of the lifetime on the current and on the pressure for the two upper laser levels of the 4p configuration of ArII. An analysis of these data has shown that: a) the electric pumping to the levels increases monotonically with increasing current density, up to 500 A/cm^2; b) quenching collisions with electrons play an important role in the deactivation of the levels.

3. Comparisons were made of the rates of population and of the lifetimes of the doublet and quartet levels at different discharge regimes. These comparisons show the causes of the redistribution of the generation power among the 4880 Å and 5145 Å lines with increasing current density.

4. An important and perhaps decisive role in the saturation of the output parameters of an ion laser is played by processes that reduce the inversion; these processes are connected with nonradiative deactivation of the upper laser levels as the result of collisions with electrons.

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