Helium-initiated inelastic channel for the collision between the $5d|3/2|_1^0$ and $5d|7/2|_3^0$ states of excited Xe

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The existence of a helium-initiated inelastic channel for the collision between the excited states of Xe has been demonstrated experimentally for the first time.

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Metastable atoms play an important role in processes occurring in an excited gas and in weakly ionized plasma and determine the operating conditions of gas lasers.[1–3] Bennett[4] and Linford[5] attempted to use the mechanism of collisional transfer between the levels of the $5d$ group in Xe to account for the anomalously large gain in laser transitions to this group. The energy gap ΔE (1500 cm^{-1} - 4000

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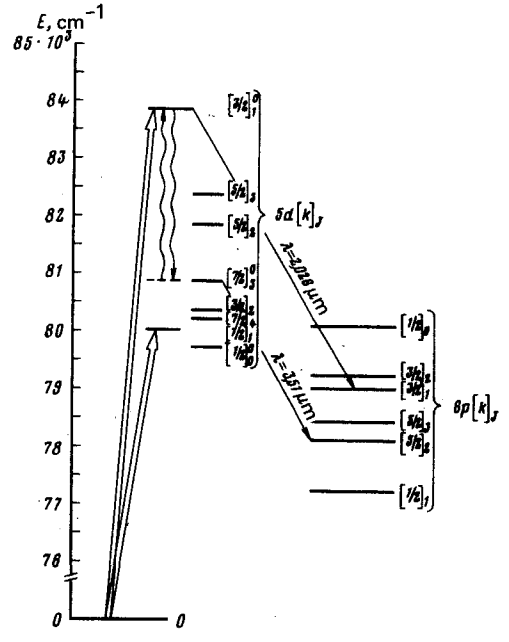

FIG. 1. Energy-level diagram of the $5d$-$6p$ multiplet.

cm^{-1}) between the levels is the main argument against the use of this mechanism.[4-6] Arguments were presented in Ref. 7 in favor of incorporating such transfers in the lasing at the $5d[3/2]_1^0 - 6p[3/2]_1$ transition and it was shown[7] that the obtained experimental results can be understood in terms of the mechanism mentioned above.

In this letter we are reporting the observation of an inelastic collision channel between the excited $5d[3/2]_1^0$ and $5d[7/2]_3^0$ states of Xe in the reactions (see Fig. 1)

$$\mathrm{Xe}\,5d[7/2]^{\circ}_{3} + \mathrm{He} + 2920\ \mathrm{cm}^{-1} \leftrightarrow \mathrm{Xe}\,5d[3/2]^{\circ}_{1} + \mathrm{He}. \qquad (1)$$

The experimental arrangement, shown in Fig. 2, consists of a laser (with a disperse LiF prism) that operates in a multimode regime simultaneously at $\lambda = 2.026$ μm ($5d[3/2]_1^0 - 6p[3/2]_1$) and $\lambda = 3.51$ μm ($5d[7/2]_3^0 - 6p[5/2]_2$). A disk shutter inserted in one of the arms, arm 1, for example, modulated the laser radiation with $\lambda = 3.51$ μm. Figure 3 is a plot of the amplitude of the signal I_1^M corresponding to the variable component of the intensity of the laser with $\lambda = 2.026$ μm as a function of the pressure of the buffer gas when the adjacent arm (1) is modulated. As expected, I_1^M increases with the He pressure. The maximum value of I_1^M obtained by us was equal to 4% of the total lasing power, consistent with the estimates obtained before the experiment. The presence of a modulation signal proportional to the transfer rate between optically unbound levels demonstrates unambiguously the existence of a nonadiabatic channel. This channel is possible only if there is a quasi-intersection U of the $5d[3/2]_1^0$ and $5d[7/2]_3^0$ levels in the collision with helium.[2] The effect of quasi-intersection of the lower levels of the $6p$ group can be ignored be-

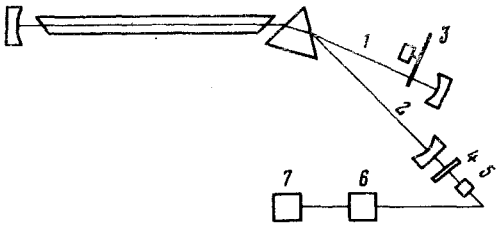

FIG. 2. Experimental setup. 1–Laser with $\lambda = 3.5$ μm; 2–laser with $\lambda = 2.026$ μm; 3–shutter; 4–filter; 5–photoresistance; 6–U2-8 selective amplifier; 7–recorder.

cause of the large difference between the radiative decay times (more than a factor of 30).[8]

The ratio of the modulation component I_1^M (Se) in pure Xe at a pressure 4 Pa and I_1^M (Xe + He) in an He-Xe mixture at the same partial pressure of Xe (see Fig. 3) is lower,

$$\frac{I_1^M(\mathrm{Xe})}{I_1^M(\mathrm{Xe}+\mathrm{He})} < \frac{1}{10}. \tag{2}$$

Since the main parameters of Xe discharge change slightly[9] as a result of addition of helium at the pressures used in the experiments, the component for the collisions with electrons in the transfer channel is small. A simple comparison of the possible reaction rates is consistent with (2).

Let us estimate the reaction rate ν in (1) using a method similar to that in Ref. 7

$$\nu = N_1 N_2 < v\sigma > \approx N_1 N_2^{ne} \bar{v}_p \bar{\sigma} \approx 10^{15} \ \mathrm{sec}^{-1}. \tag{3}$$

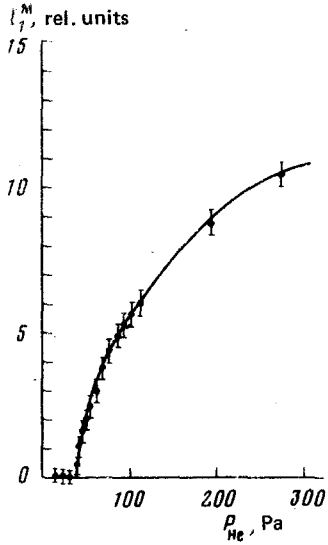

FIG. 3.

We used the following parameters for the estimate: The density of metastable $5d$ Xe atoms $N_1 = 10^{10}$ cm^{-3}, the density of the nonequilibrium part $N_2^{ne} = 5 \times 10^{14}$ cm^{-3} (10^{-2} of the equilibrium density N_2 of helium atoms), the relative collision rate $\bar{v}_p = 1.8 \times 10^6$ cm/sec, and the cross section of the reactions (1) $\bar{\sigma} = 10^{-14}$ cm^2. The justification of their choice for this system is given in Ref. 7.

Here we have taken advantage of the fact that only the nonequilibrium part of the distribution function of helium with the threshold energy $\Delta E \approx m_{He}\bar{v}_p^2/2$ participates effectively in the multiplet mixing of the $5d$ group. The sources that maintain this nonequilibrium are the reactions (1) and other types of reactions that occur in the discharge of an He-Xe mixture; primarily the dissociative recombination must be separated from them.[10]

An estimate of the transfer rate ν_e involving the electrons in the processes such as (1) gives

$$\nu_e = N_1 N_e < v_e \sigma_e > \lesssim 10^{13} \text{ sec}^{-1} \tag{4}$$

for $\bar{v}_e = 10^8$ cm/sec, $N_e = 10^{10}$ cm^{-3}, and $\bar{\sigma}_e = 10^{-14}$ cm^2. We chose the maximum value of N_e for a glow discharge at $\approx$12-mA current used in the experiment. A comparison of the results of estimates (2) and (3) with q, the rate of electron pumping from the $5p\ ^{61}S_0$ ground state of Xe to the $5d[3/2]_1^0$ level, shows[7] that $\nu/q \approx 1$ and $\nu_e/\nu \approx 0.01$, i.e., channel (1) is the most important channel. It follows from this that the transfer is caused by the process (1) and that there is a crossing U of the $5d[3/2]_1^0$ and $5d[7/2]_3^0$ levels of the $5d$-He Xe quasimolecule.

In summary, we note that, although only one of the multiplet-mixing channels has so far been identified, we can still assert that such channels are real and that they determine the kinetics of the processes in an Xe-He laser.

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