Anomalously fast relaxation of metastable states of Ca^+, Eu, and Eu^+ and impact emission from Ca^+, Eu^+, and Sr^+ ions

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An anomalously fast relaxation of metastable states of the Eu atom and of the Ca^+ and Eu^+ ions has been discovered in collisions with He and Ne atoms. An anomalously fast relaxation of metastable states of Ba^+, Ca^+, Eu^+, and Sr^+ has also been discovered in a quasiresonant charge exchange with the like atoms.

It was shown in Ref. 1 that the high-power collisional He + Eu^+ laser operating on $^{7,9}P$-$^7D^0$ transitions operates through an effective relaxation of $^7D^0$ metastable states in collisions with He atoms:

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$$\mathrm{Eu}^+(^7D^0) + \mathrm{He} \rightarrow \mathrm{Eu}^+(J) + \mathrm{He} + (\Delta E > 9kT_g), \quad (1)$$

where k is the Boltzmann constant, and T_g is the gas temperature.

Reaction (1) is unusual in that it has a large cross section ($\sim 10^{-16}$ cm^2) at an energy defect $\Delta E > 0.7$ eV, significantly higher than the thermal energy. A study of the relaxation of the strontium ion has shown that, despite the similarity of the Eu^+ and Sr^+ energy structures, the metastable states of Sr^+ relax only slightly in reaction (1) (Ref. 2). To the best of our knowledge, a rapid relaxation has not been observed in any other metal ions or atoms. The situation supports the classical conclusion of Ref. 3 and other studies that electronic states decay only to a slight extent in collisions with atoms at large values of ΔE.

This letter is the first report of the observation of an anomalously fast relaxation of metastable states of Ca^+ (^2D), Eu ($b\,^8D^0$), Eu(a^8D^0), and Eu^+($^9D^0$) in collisions with He atoms at $\Delta E \gg kT_g$. For the first time, an effective relaxation has also been observed in collisions with Ne atoms and of metastable states of Ba^+, Ca^+, Eu^+, and Sr^+ in a quasiresonant charge exchange with the like atoms.

The deexcitation is studied in the afterglow of a low-current ($i < 3A$) nanosecond ($\tau \sim 50$ ns) discharge by the method of resonant fluorescence.[1] Figure 1 shows the reciprocal relaxation time of the metastable state versus the pressure for the various impurity gases; Fig. 2 shows the results in a plot against the metal vapor pressure. Analysis of these data yields the following cross sections (in units of 10^{-16} cm^2) in collisions with He: 0.25 ± 0.08 for Ca^+ (^2D), 0.5 ± 0.2 for Eu($b\,^8D^0$), 0.11 ± 0.04 for Eu(a^8D^0), and 1.4 ± 0.4 for Eu^+(a^9D^0). The cross sections for the reactions involving neon are, respectively, 0.45 ± 0.12, 0.23 ± 0.07, 0.14 ± 0.04, and 3.4 ± 1.0. The cross sections for the quasiresonant charge exchange turn out to be $\sigma^+(Ba^+(^2D_{5/2}) = (2 \pm 0.4) \times 10^{-14}$ cm^2; $\sigma^+(Ca^+(^2D)) = (1.1 \pm 0.6) \times 10^{-15}$ cm^2; $\sigma^+(Eu^+(^9D^0)) = (4 \pm 1) \times 10^{-14}$ cm^2; and $\sigma^+(Sr^+(^2D)) = (0.5 \pm 0.1) \times 10^{-14}$ cm^2.

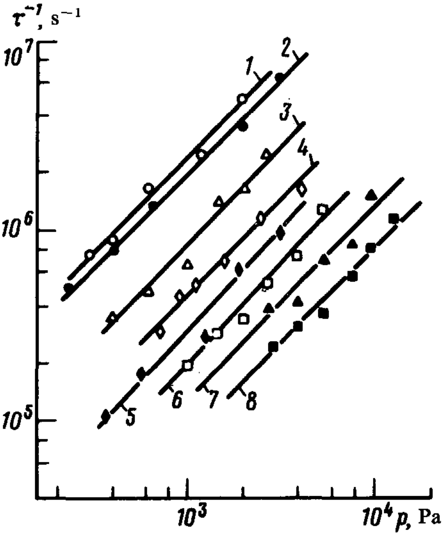

FIG. 1. Reciprocal relaxation time of metastable states versus the He and Ne pressures. 1, 2—Eu^+(a^9D^0); 3, 7—Eu($b\,^9D^0$); 4, 5—Ca^+(^2D); 6, 8—Eu(a^8D^0); 1, 3, 4, 6—with He; 2, 5, 7, 8—with Ne.

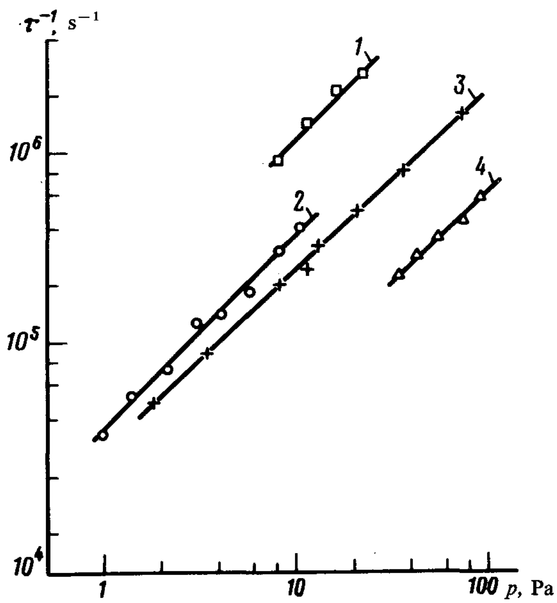

FIG. 2. Reciprocal relaxation time of the metastable states of ions versus the pressure of the metal vapor. 1—Eu^+ ($^9D^0$); 2—Ba^+ ($^2D_{5/2}$); 3—Sr^+ (^2D); 4—Ca^+ (^2D).

The fact that a fast relaxation of the type in (1) also occurs in a quasiresonant charge exchange means that we can add to the list of high-power collisional lasers. Of particular interest is the possibility of using quasiresonant charge exchange for these purposes, since for essentially any excited ion it is possible to select an impurity atom with which quasiresonant charge exchange will be effective.

In the present experiments we studied the possibility of achieving an impact emission from europium and calcium ions, accompanied by a decay of metastable states in a reaction of the type in (1), and from strontium ions, accompanied by the decay of a metastable state in a quasiresonant charge exchange. For this purpose, we pumped a mixture of Ca, Eu or Sr with He with electron beams from an open discharge.[4] These experiments were carried out in a ceramic BeO tube with an active region 4.5 mm in diameter and 5 cm long with an accelerating gap $\delta = 0.2$ mm and a grid with a geometric transparency $\gamma = 0.4$. The electron acceleration was stabilized by using a discharge through a dielectric and a volume resistance.[4]

The fraction of the energy which goes into the resonant states of the Ca^+, Eu^+, and Sr^+ ions during pumping by fast particle beams is small. Accordingly, the $^{7,9}P_j$ levels of Eu^+ and the ^2P_j level of Ca were excited in charge exchange with Zn^+ ions, while the ^2P_j states of Sr^+ were excited in charge exchange with Cd^+ ions. The cross sections measured for these reactions in the present experiments are $\sigma_{Zn^+ + Eu} = (1.6 \pm 1.0) \times 10^{-14}$ cm^2; $\sigma_{Zn^+ + Ca} = (0.8 \pm 0.5) \times 10^{-14}$ cm^2, and $\sigma_{Cd^+ + Sr} = (1.5 \pm 1) \times 10^{-14}$ cm^2. A quasi-cw emission is achieved by virtue of the high repetition frequency (up to 3 MHz) of the pump pulses.

Figure 3 shows the emission power P_2 versus the pump power P_p for emission at various wavelengths: $\lambda = 664.5$ nm in Eu + Zn + He mixtures with the respective pressures 20–40 Pa, 0.5 kPa, and 50–100 kPa; $\lambda = 866$ nm in Ca + Zn + He mixtures

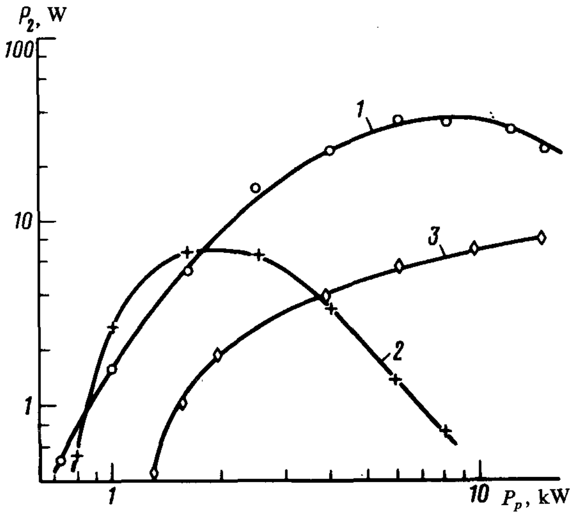

FIG. 3. Emission power versus the pump power in Eu^+ ($\lambda = 664.5$ nm, 1), Ca^+($\lambda = 866$ nm, 2), and Sr^+ ($\lambda = 1033$ nm, 3).

with respective pressures 20–40 Pa, 0.8 kPa, and 50–100 kPa; and $\lambda = 1033$ nm in Sr + Cd + He mixtures with respective pressures of 1, 1.5, and 20 kPa. In these experiments, the pumping was carried out with a train of pulses with a total length of 1–3 ms. Up to the pump power $P_p = 5$ kW, the increase P_p is achieved by raising the voltage across the accelerating gap to 6 kV; increases in the range $P_p = 5$–15 kW are achieved by raising the pulse repetition frequency from 1 to 3 MHz. Over the range of Ca and Eu vapor pressures studied, P_2 varies by no more than 20% at a modulation depth of 10–60%.

In summary, this study has shown that the fast relaxation of the metastable states of metal atoms and ions at energy defects significantly above the thermal energy is not unique to the $^7D^0$ state of the europium ion. There is accordingly the possibility of adding greatly to the list of high-power collisional metal-vapor lasers.

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