Vacuum ultraviolet fluorescence of $(XeRb)^+$ produced in an electron-beam-pumped gas mixture

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With a pulsed electron beam a gas mixture of Ar, Xe, and Rb was excited producing $(XeRb)^+$ ionic excimer molecules. To study the formation kinetics the $(XeRb)^+$ fluorescence pulse was measured as a function of the gas composition and the pumping density. From the observed fluorescence signal decay a value of $6\pm1\times10^{-30}$ cm^6/s for the formation rate constant of $(XeRb)^+$ from Xe^+, Ar, and Rb was determined.

Rare-gas alkali ions have excimerlike properties.[1,2] Research is going on to investigate whether they can be used as a laser medium. To achieve high rare gas-alkali ion densities, an intense pumping source is necessary and, therefore, electron-beam pumping is an obvious choice.

Millar *et al.*[3] reported some results on $(XeRb)^+$ and $(XeCs)^+$ produced in a gas mixture excited by an electron beam. Further study on the formation kinetics is done by Schumann *et al.*[4] who gave a kinetic scheme for the $(KrK)^+$ formation in gas mixtures of Kr, K, and He pumped by an electron beam. In this letter, we present a more extensive study of the vacuum ultraviolet $(XeRb)^+$ fluorescence signals from a gas mixture excited by a simple coaxial electron beam device. From the results extracted from the time-resolved fluorescence measurements the formation channel of $(XeRb)^+$ is deduced.

For the experiments, a modified coaxial electron-beam system is used. A description of the standard coaxial electron-beam device is given by Peters *et al.*[5] The anode in the modified coaxial system can be heated in order to vaporize the alkali metal inside. The anode consists of a supporting tube, a thin foil tube, and a heating element. Two windows are cut out of the titanium supporting tube to allow entrance to the electron beam. A thin titanium foil tube is slided over the supporting tube and soldered at the ends. The thin foil tube has an inner diameter of 10 mm and a 25 μm wall thickness. The support and foil tube together form the high pressure cell.

A heating wire is spirally wound around the tube. The wire also reinforces the thin foil tube at high pressures. The hot region in the cell coincides with the windows in the supporting tube. In this way, a cell is formed with a hot and active length of 24 cm and pumped from two sides through the windows. At both end sides a cooling down section of 4-cm long is provided. Here, the temperature is decreased gradually to prevent fog formation by the alkali vapor.

The cathode consists of an aluminum cylinder in which two carbon felt strips are mounted. The aluminum cylinder also works as a heat shield. The coaxial diode is powered through a pulse forming line by a Marx generator. The diode voltage could be varied between 100 and 200 kV. Inside the hot cell the measured peak current density in vacuum varied from 200 to 750 A/cm^2. The pulse shape was triangular with a width of 10 ns [full width at half-maximum (FWHM)].

The temperature distribution over the tube length depends on the heating wire distribution. By adjusting the wire pitch the temperature distribution was kept constant within ±5 °C over the active length in the temperature range of 350 to 410 °C. The electrical input power is only in the order of 35 W.

From the temperature distribution the Rb density was calculated. This density agreed with the density determined from an absorption experiment at the Rb resonance lines at $\lambda=4202$ and 4216 Å. The optical system is adapted for fluorescence experiments in the vacuum ultraviolet.

The $(XeRb)^+$ spectrum was measured by Millar *et al.*[3] and confirmed by Da Xing *et al.*[6] The fluorescence measurements were done at $\lambda=164.5$ nm. The signals contained a small Xe_2^* background for which a correction was made. At $\lambda=172$ nm only Xe_2^* radiation was measured.

In Fig. 1 a typical fluorescence signal is shown together with the current pulse measured inside the tube. The reproducibility of the fluorescence signal from two series of measurements each with a fresh gas fill was within 10%. The peak of the signal is located near the end of the excitation pulse. The signal exhibits a single exponential decay. The risetime varies between 20 and 40 ns and the decay time varies between 10 and 50 ns. For decay times shorter than 15 ns the signal is corrected for the influence of the scintillator decay.

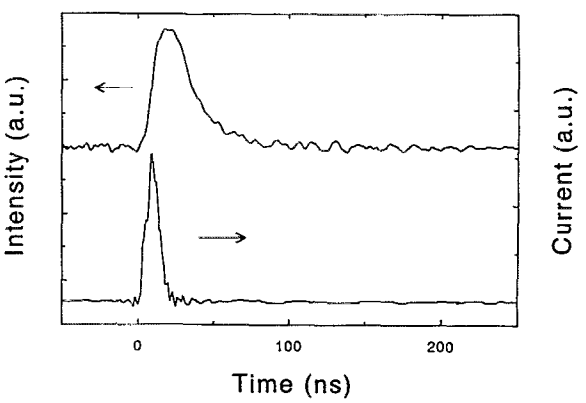

FIG. 1. Temporal behavior of the vacuum ultraviolet fluorescence signal of $(XeRb)^+$ and of the current pulse (measured inside the anode tube).

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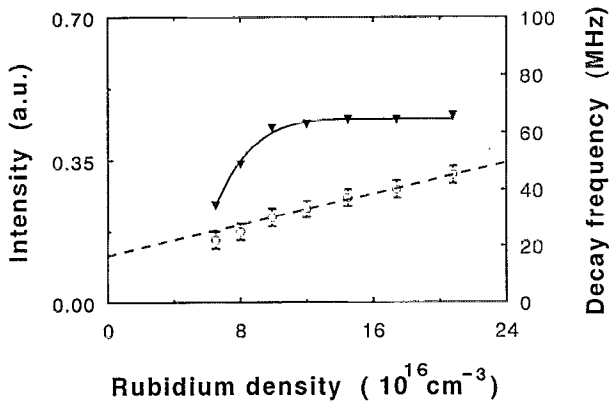

FIG. 2. Fluorescence peak intensity (solid line) and decay frequency (dashed line) as a function of the rubidium density with [Ar]=2.4×10^{19} cm^{-3}, [Xe]=1.0×10^{18} cm^{-3} and a power deposition of 1.2 MW/cm^{-3}.

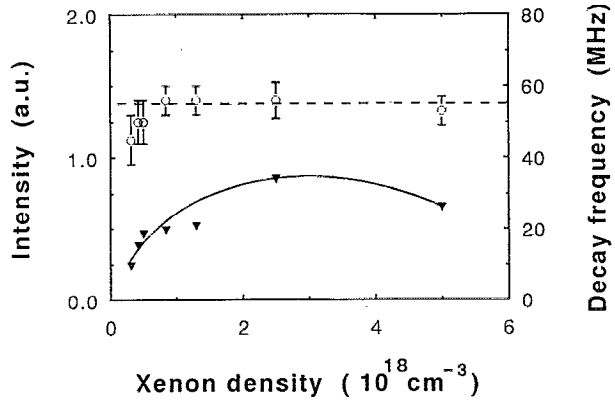

FIG. 4. Fluorescence peak intensity (solid line) and decay frequency (dashed line) as a function of the xenon density with [Ar]=4.7×10^{19} cm^{-3}, [Rb]=9.9×10^{16} cm^{-3} and a power deposition of 3 MW/cm^{-3}.

Figures 2–5 show the dependency of the fluorescence peak intensity and the decay frequency on the Rb, Ar, and the Xe density and the power deposition. Figure 2 shows that if the Rb density is varied, this results in a saturation of the fluorescence peak intensity with increasing Rb density. We also observed that the rise time shortens with increasing Rb density. Figure 2 also shows that the decay frequency increases linearly with the increasing Rb density, indicating that quenching occurs during the decay of the signal. As the Ar density is increased the fluorescence peak also saturates, as shown in Fig. 3. Also, here was observed that the signal rise time shortens with increasing Ar density. The decay frequency shown in Fig. 3 increases linearly with the Ar density. As can be seen in Fig. 4 varying the Xe density, the fluorescence peak intensity shows a maximum at about 3×10^{18} cm^{-3}. At high Xe density the fluorescence is quenched. With increasing Xe density we observed that the signal rise time shortens. At low Xe density the decay frequency seems influenced by slow formation kinetics of a precursor. Above these low Xe densities an increase in the Xe density does not change the decay frequency. It should be mentioned that the values for the fluorescence peak intensity and the decay frequency in Figs. 3 and 4 were averaged over two shots each. In Fig. 5 it is shown that the fluorescence peak intensity decreases slightly with increasing pumping power while the observed rise time and decay frequency do not change as function of the pumping power.

From a theoretical point of view ionic excimers are expected to have short lifetimes in the order of nanoseconds.[7,8] As a consequence, also the effective lifetimes are expected to be short. The effective lifetime is defined as the inverse of the transition probability which results from emission of radiation and quenching. Schumann[4] determined for $(KrK)^+$ an effective lifetime shorter than 3 ns. By analyzing our experimental results we assume that the effective lifetime of $(XeRb)^+$ must be short and that the shape of the measured fluorescence signal only reflects the formation of $(XeRb)^+$ ions.

The $(XeRb)^+$ ionic molecule is formed in a termolecular reaction of Xe^+ and Rb with a third body. The production of Xe^+ stops after the excitation pulse is terminated. The Xe^+ ion is quenched by the formation of $(XeRb)^+$, two electron recombination,[9] Xe_2^+ and $(ArXe)^+$ formation. At elevated temperatures the formation of $(ArXe)^+$ and Xe_2^+ is relatively unimportant since the association reactions with Ar and Xe proceed much slower.[10,11] In this way, the decay of Xe^+ is governed by

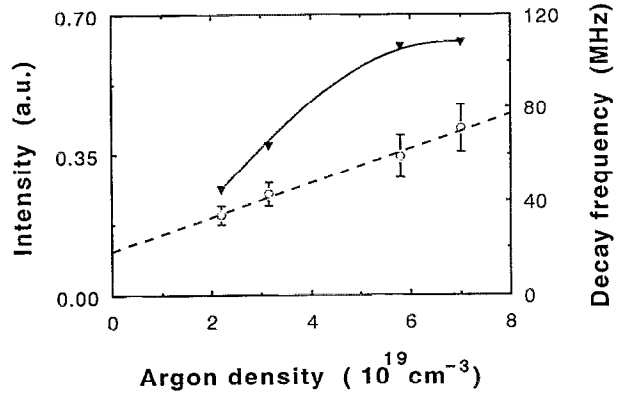

FIG. 3. Fluorescence peak intensity (solid line) and decay frequency (dashed line) as a function of the argon density with [Xe]=5.7×10^{17} cm^{-3}, [Rb]=9.9×10^{16} cm^{-3} and a power deposition of 3 MW/cm^{-3}.

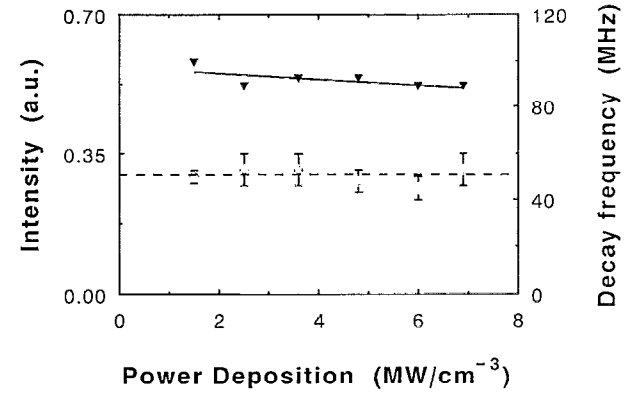

FIG. 5. Fluorescence peak intensity (solid line) and decay frequency (dashed line) as a function of the power deposition with [Ar]=3.3×10^{19} cm^{-3}, [Xe]=3.7×10^{18} cm^{-3} and [Rb]=9.9×10^{16} cm^{-3}.

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two electron recombination and by the formation of $(XeRb)^+$. From the linear dependency of the decay frequencies on the Rb and Ar density as shown in Figs. 2 and 3 we conclude that $(XeRb)^+$ is formed in a termolecular reaction of Xe^+ and Rb with Ar as a third body.

As the Ar and Rb densities increase, the increase in formation rate shows up also in a shorter rise time of the fluorescence signal. Apart from the behavior at a low Xe density the decay frequency is not influenced by an increasing Xe density. So, a termolecular formation channel of $(XeRb)^+$ with Xe as third body is unimportant for our experimental conditions. With increasing Xe density the rise time of the fluorescence signal also shortens indicating that the Xe^+ formation rate is enhanced. In Fig. 4 we see that the fluorescence peak intensity is quenched at higher Xe densities indicating that $(XeRb)^+$ might be quenched directly by Xe.

From the slope of the decay frequency in Figs. 2 and 3 the formation rate constant of $(XeRb)^+$ in the termolecular reaction of Xe^+ with Ar and Rb can be determined at $6\pm1\times10^{-30}$ cm^6/s. The formation rate constant is smaller than an identical rate constant estimated by Basov[2] but in the same range as the formation rate constant for $(KrK)^+$ as determined by Schumann *et al.*[4]

We found no increase of the fluorescence peak intensity with increasing power deposition. The production rate of Xe^+ is expected to increase with increasing power deposition. On the other hand, the quenching rate of Xe^+ increases quadratically due to two-electron recombination. Two-electron recombination quenches also the Ar^+ and Ar_2^+ ions which are the precursors of Xe^+. Moreover, the quenching rate of $(XeRb)^+$ due to dissociative recombination with electrons can increase with higher pumping density. If the production rate of Xe^+ is balanced by the described quenching processes no increase in fluorescence peak intensity is expected.

As mentioned before the decay frequency $1/\tau$ reflects the relative change of the Xe^+ density as follows: $1/\tau = k[Ar][Rb]+k_e[e]^2$. Keeping the Ar and Rb density constant, the decay frequency can vary only with a variation of the second term, which contains the recombination rate constant and the electron density squared.

Surprisingly, the decay frequency remains constant with increasing power deposition, which means the second term has to be constant. The rate constant for two-electron recombination strongly depends on electron temperature. The total electron density consists of the secondary electrons and the electrons produced by Penning ionization of Rb by Xe*. With increasing power deposition the Xe* density increases and by Penning ionization this might result in an increase of the electron temperature and density. As the electron temperature increases, the recombination rate constant decreases. In this way, it is possible that a decrease of the rate constant in the second term is compensated by the increasing electron density which results in the measured decay frequency.

Preliminary results of a kinetic model currently under development show a reasonable agreement between the results of the measurements and the kinetic simulations. To fit the experimental results the effective lifetime of $(XeRb)^+$ has to be shorter than 5 ns.

We have shown that with this modified coaxial electron beam device we are able to pump a gas mixture at high power density thereby using a heated high pressure cell with a flat temperature distribution over a length of 24 cm. We have seen that $(XeRb)^+$ is formed in a termolecular reaction of Xe^+ with Ar and Rb. The formation rate is determined to be $6\pm1\times10^{-30}$ cm^6/s.

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