Bound-free vacuum uv emissions of $(XeCs)^+$ and $(KrCs)^+$ ionic excimers by relativistic electron beam excitation

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Bound-free vacuum uv emissions from rare-gas alkali ionic excimers were observed by relativistic electron beam pumping of gas mixtures of Xe or Kr with hot cesium vapor. The observed two diffuse emission bands centered at 159 and 131 nm are assigned to be the $2\,^1\Sigma^+ \rightarrow 1\,^1\Sigma^+$ transition of $(XeCs)^+$ and $(KrCs)^+$ ionic excimers, respectively. © *1997 American Institute of Physics.* [S0003-6951(97)02144-X]

It is well known that the ability of atoms to form molecules as a result of the interaction is governed by their electronic structure, and the excitation or ionization of atoms can alter their reactivity. This property of atoms, particularly that of rare gases, is well utilized in high-efficiency excimer laser transitions. The ionic excimer[1,2] extended the concept of the excimer transition in uv region to vacuum and extreme uv (VUV) and (XUV) regions. The idea is that the isoelectronic molecules should have a similar characteristics as neutrals in the optical transition even though those potential energies are much higher. Fluorescences from rare-gas alkali ionic excimers have been reported by charged-particle excitation and laser-produced plasma pumping.[3,4] In particular, as a preliminary step toward achieving practical lasers, VUV emissions from rare-gas alkali ionic excimers $(XeRb)^+$[5–7] and $(KrRb)^+$[6] have been successfully observed by electron beam excitation. Recently, a kinetic model calculation for the gain dynamics of $(XeCs)^+$, which predicted a net gain up to 2% cm^{-1} in the ternary discharge pumping, has been reported.[8] In this letter, we report an observation of VUV emissions from Xe^+Cs and Kr^+Cs excimers by relativistic electron beam pumping.

The experimental setup is similar to that described in the previous research[6,9,10] with the exception of a modification of the guiding magnetic field and alkali vapor cell. The coaxial electron beam has a particle energy of 500 keV, a peak current of 5 kA, a beam diameter of 2 cm, and a triangular-shape pulse duration of 3 ns. The beam was confined by a 2.0 kG axial magnetic field, passed through a low-pressure helium chamber (few Torr) and an inconel foil, and injected into a Cs vapor cell. The active length of the cell is 45 cm. A vertical magnetic field at the end of the active zone was used to deflect the remains of electrons from the axial direction. The emission from the active zone was introduced to a 0.2 m VUV spectrometer through a high-quality LiF window at the forward end of the cell. The spectrum was analyzed by a spectrum multichannel analyzer (SMA) in combination with a model IRY-512 detector and a sodium salicylate scintillator.

The vapor pressure of Cs was varied from 1 to 30 Torr by changing the cell temperature. The atomic number density of the cesium vapor was estimated from vapor pressure curves,[11] and its agreed with the density determined from an absorption experiment at the Cs resonance lines at 455.6 and 459.4 nm.

Figure 1 shows a VUV emission spectrum observed by the electron beam excitation of a Xe/Cs vapor mixture. The xenon pressure was 100 Torr and the cell temperature was 350 °C, corresponding to a Cs vapor pressure of 12 Torr. An approximate continuum structure was observed in the region of 150–165 nm, which was believe to be attributed to the $(XeCs)^+$ ionic excimer fluorescence.[3] The spectrum exhibits one 6-nm-wide peak full width half maximum (FWHM) centered at 159 nm. The spectrum shape and bandwidth, however, is somewhat different from the one previously observed by ion beam pumping.[3,12] Measurements also included the 173 nm region where the Xe_2^* excimer emission was expected. In this region, the Xe_2^* emission was observed at

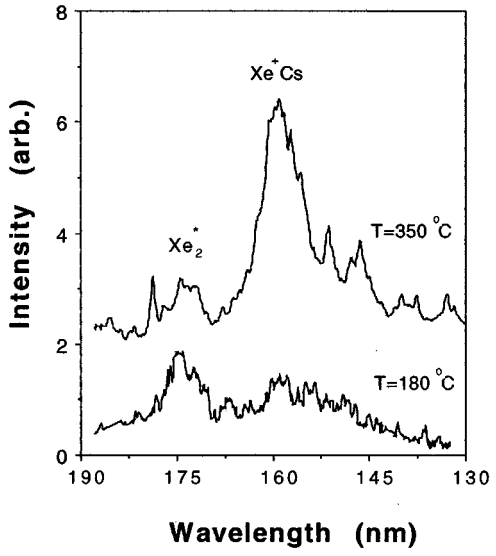

FIG. 1. VUV emission spectrum of the electron-beam-excited Xe/Cs mixture. The cell temperature: 350 °C and the Xe rare gas pressure: 100 Torr.

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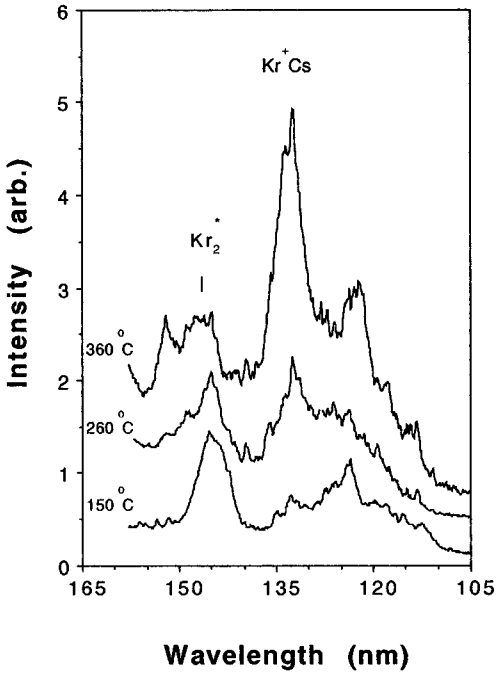

FIG. 2. VUV emission spectrum of the electron-beam-excited Kr/Cs vapor mixture. The cell temperature was changed from 150 to 360 °C, and the Kr rare gas pressure was 100 Torr.

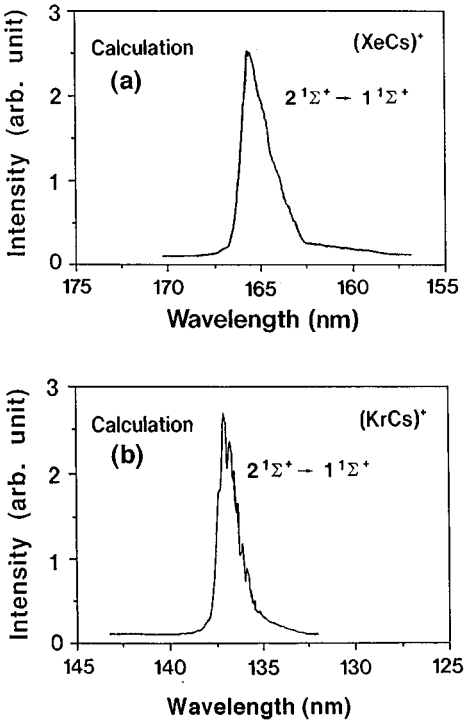

FIG. 3. The simulated spectra for the $2\,^1\Sigma^+ \rightarrow 1\,^1\Sigma^+$ transition of (a) $(XeCs)^+$ and (b) $(KrRb)^+$ by using the modified potential curves, and a linear surprisal distribution (Ref. 6) of vibrational state population.

large Xe number densities and small Cs vapor pressures. For Xe pressures below 100 Torr and alkali pressures above 4 Torr the Xe_2^* signal is almost quenched by the presence of alkali vapor.

The measured VUV spectrum from the electron-beam-excited $(KrCs)^+$ is shown in Fig. 2. The krypton pressure was 100 Torr and the cell temperature was changed from 150 to 360 °C, corresponding to the Cs vapor pressure of 1–20 Torr. The spectrum exhibits a main peak centered at 131 nm with a bandwidth of 5 nm. The spectrum, however, is broader than that of the previous one observed in low pressure experiments by ion beam pumping.[12] The 131 nm diffuse emission peak appeared at a temperature of about 150 °C, and showed an approximate linear increase from 150 to 360 °C. In addition, the weak Kr_2^* emission band at 146 nm was also present at low alkali pressures.

In order to show a comparison of the experimental results with the theory, we calculated the Franck–Condon factors for the $2\,^1\Sigma^+ \rightarrow 1\,^1\Sigma^+$ transition of the $(XeCs)^+$ and $(KrCs)^+$. In the calculations, the *ab initio* potentials of the rare-gas potassium[13] have been used with modification of the transition energy by 0.447 eV[14] to take into account the difference in ionization potentials of Cs and K. The result shows that transitions from the vibrational levels of the $2\,^1\Sigma^+$ state almost hit at the larger internuclear separation region of the shallow ground $1\,^1\Sigma^+$ state where the potential curve is nearly flat. It is expected that this type of potential structures leads to very narrow Franck–Condon vibrational envelopes for the transitions. Figure 3 shows simulated spectra of the $2\,^1\Sigma^+ \rightarrow 1\,^1\Sigma^+$ transition of $(XeCs)^+$ and $(KrCs)^+$ by taking into account the lowest 10 vibrational levels of the upper state. It was clearly exhibited that the $2\,^1\Sigma^+ \rightarrow 1\,^1\Sigma^+$ transition has a main peak at 165 nm with a blue-shaded quasi-continuous feature for $(XeCs)^+$ and a similar continuum centered at 136.5 nm for $(KrCs)^+$. They are in good agreement with the observed results in spectral shapes. Some differences in transition wavelength are due to use of the approximate *ab initio* potentials and average dipole transition moments.

The VUV emission of the $(XeCs)^+$ or $(KrCs)^+$ was also observed when the Ar or Ne was used as a buffer gas. The intensity, however, shows some decrease with the increase of Ne or Ar pressure in the present experiment, especially for the case of argon buffer gas, which is probably caused by the increase of the input electron energy loss in the present longitudinal pumping scheme.

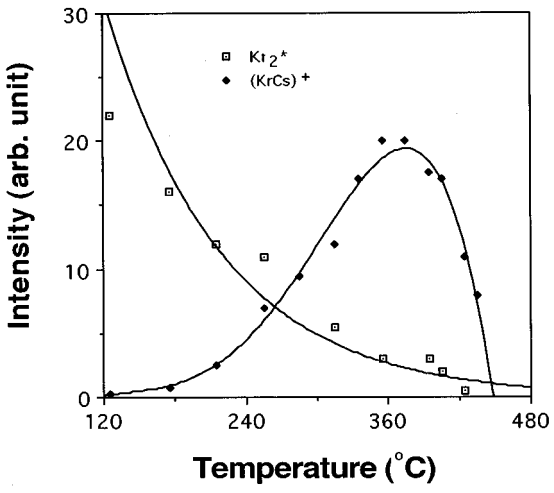

FIG. 4. Observed temperature dependence of the fluorescence of $(KrCs)^+$ ionic quasi-excimers.

Figure 4 shows temperature dependences of the 159 nm emission band of $(KrCs)^+$ and 146 nm band of Kr_2^* dimer in the range of 250–450 °C (corresponding to the Cs vapor pressure of 2–25 Torr). With the increase of cell temperature, the rare-gas dimer Kr_2^* was quenched very fast with the increase of alkali vapor pressure. The intensity of the 159 nm band showed approximately linear increase with the Cs vapor increase and a saturation at high temperatures caused by a self-absorption of Cs vapor or rare-gas Cs molecules. The maximal intensity of the $(KrCs)^+$ fluorescence was observed at 400 °C, which corresponds to about 12 Torr of Cs vapor pressure. Further increase in the Cs density caused an increase in the absorption of the radiation and the fluorescence intensity decreases with the Cs vapor pressure. From Fig. 4 one can see that the larger population density of $(KrCs)^+$ ionic excimer yield, this can be qualitatively supported by the fact that the intensity of $(KrCs)^+$ emission is comparable to the intensity of Kr_2^* in the unheated cell under the same pumping conditions.

The main processes involved in the formation of the ionic excimer, for example of Xe^+Cs, are predicted as follows:

$$Xe^+ + Cs + Rg \rightarrow Xe^+Cs + Rg, \tag{1}$$

$$Xe^* + Cs^+ + Rg \rightarrow Xe^+Cs + Rg, \tag{2}$$

$$Xe_2^+ + Cs \rightarrow Xe^+Cs + Xe, \tag{3}$$

where Rg is Xe or other buffer gases. The formation channel via Xe^+ is predicted to be dominant in the initial stage of the excitation with a rate constant of the order of 10^{-28} $cm^6\,s^{-1}$.[15] The metastable formation channel involving Xe* is conceptually similar to the three-body harpooning reaction in rare-gas halide excimer formation. Such reaction is exothermic for many rare-gas alkali ionic excimers and possesses large rare constants.[8] The formation process (3) is expected to be inefficient under the lower pressure condition. In the case of high pressures, however, the bulk of Xe^+Cs can be formed through the process with a rate constant of 5×10^{-9} $cm^3\,s^{-1}$,[15] which is an analog of the ''harpoon'' reaction between alkali metals and halogens. The main quenching process of the Xe^+Cs occurs as a result of the electron-ion dissociative recombination, $Xe^+Cs + e \rightarrow Xe^* + Cs$, which has a rate constant about 10^{-8} $cm^3\,s^{-1}$.[8]

By using a simplified rate equation describing the temporal behavior of the upper state,[16] in which it is assumed that the electron beam is switched on instantaneously and the pumping power density is 1 MW/cm^3, a maximum Xe^+Cs population of 4×10^{15} cm^{-3} is yielded for a mixture consisting of 3 atm of He, 200 Torr of Xe, and 4 Torr of Cs. This value should be further increased with the increase of rare-gas pressure. When the photoabsorption cross section from the Cs atoms, which was suggested to be about 10^{-19} cm^2[15] is taken into account, a gain coefficient in excess of 0.02 cm^{-1} is estimated for the electron beam pumping of $(XeCs)^+$. This value should be further increased with the increase of rare-gas pressure and pumping power by using a transverse pumping scheme.

In conclusion, the VUV emissions of rare-gas alkali ionic excimers Xe^+Cs and Kr^+Cs (for the first time to our knowledge) produced through reactive kinetics have been observed by the electron beam pumping. A gas mixture of Kr or Xe with a hot vapor of cesium was excited to obtain a diffuse emission band centered at 159 nm from $(XeCs)^+$ and 131 nm from $(KrCs)^+$ molecular ions. Judging from the results of the recent *ab initio* calculation, the observed two diffuse emission bands are assigned to the $2\,^1\Sigma^+ \rightarrow 1\,^1\Sigma^+$ bound-free transition of $(XeCs)^+$ and $(KrCs)^+$ excimers, respectively.

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