Particle Accelerators, 1992, Vols. 37–38, pp. 403–407
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XUV EMISSION FROM RARE-GAS/ALKALI IONIC EXCIMERS EXCITED BY HIGH-CURRENT HEAVY-ION BEAMS†

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(Received 3 December 1990)

Mixtures of rare gases with alkali-metal vapor were excited by a high-power argon ion beam in a windowless gas target system. Narrow emission continua were observed in the vacuum ultraviolet region between 60 and 200 nm. The observed emission was assigned to the decay of a new class of excited ionic molecules Rg^+A formed by the combination of a rare-gas ion Rg^+ with an alkali atom A. These excimer-like ions are of great interest for possible short-wavelength lasers, because the ground state consisting of a rare-gas atom and an alkali ion is only weakly bound. The observed decay energies and fine structure splittings agree well with the prediction of calculations. A summary of previous results is presented.

1 INTRODUCTION

High-current ion beams are an ideal tool to achieve high energy density in matter, which is essential for VUV or XUV laser action. Reactions between an ion beam and rare gases result in different ionization and excitation states, as well as in the formation of new molecular states. Excimers that decay by the emission of radiation at short wavelengths are of especially great interest because of their potential application as short-wavelength photon sources. The search for new transitions in the VUV or even XUV region led to new molecules consisting of a rare-gas ion (Rg^+) and an alkali atom (A) which are isoelectronic to the well-known neutral rare-gas excimers (RgRg'). Due to the ionic character of these molecules, the expected transition wavelengths should be shifted to shorter wavelengths[1]. The binding of the excited state is due to the polarization of the alkali atom by the rare gas ion. Almost the total ionization energy of the rare gas minus the ionization energy of the alkali atom can be emitted as radiation in the transition to the ground state. Theoretical considerations of Mantel[2] predict radiative transitions from 190 nm (Xe^+A) to the XUV region at 60 nm (He^+A).

† supported by the BMFT

The ground state of the Rg^+A molecules, consisting of a rare gas atom and an alkali ion, is only weakly bound and dissociates at normal experimental conditions into $Rg^+ A^+$. Therefore Rg^+A may be considered as a quasi excimer state. The shape of the ground state potential curve, with its weak binding, leads to narrow line widths of the emitted continua which results in higher gain in laser experiments and facilitates the application as laser storage media.

2 EXPERIMENTS

The experiments were carried out at the DYNAMITRON accelerator of the Institut für Strahlenphysik at the Stuttgart University, which provided high-dc-current Ar^+ beams. The alkali vapour was produced in a special furnace that could be heated to a temperature corresponding to an alkali vapor pressure of a few hPa (300°C – 750°). The metal vapor was mixed with several kPa of rare gas as a buffer. To prevent oxidation of the highly reactive alkali metals, they were brought into the reaction cell in ampoules, which were broken under vacuum (except for lithium, due to the high melting point). The Ar^+ beam entered the furnace windowless because of its high power density (up to 150 μA at 3.2–3.5 MeV), which no foil could sustain. The pressure difference between furnace and beam line (almost 8 orders of magnitude) was maintained by the 4-stage differential pumping system RHINOCEROS[4]. The ion beam passed windowless through five apertures with diameters of 2–9 mm. These

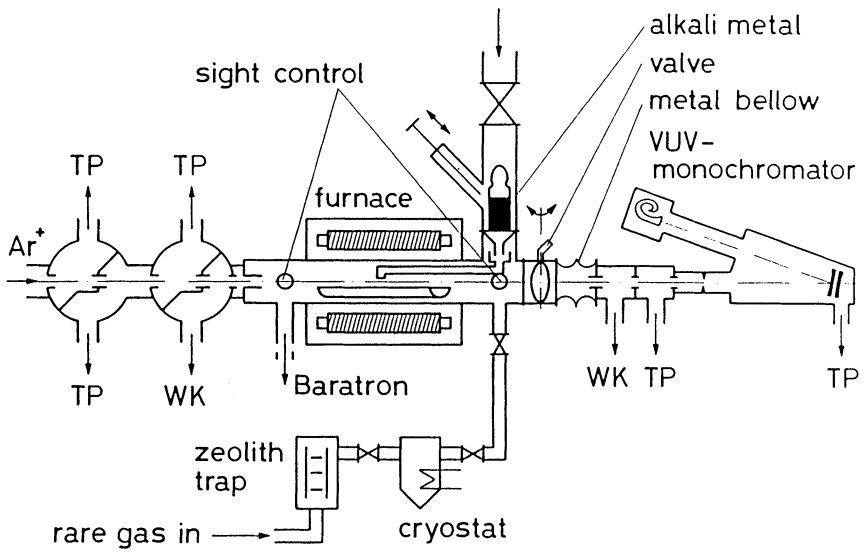

FIGURE 1 Experimental set-up. The rare-gas/alkali mixture at a total pressure of about 100 hPa is excited by an argon ion beam, which is completely stopped in the furnace. The pressure gradient between furnace and beam transport system is maintained by the 4-stage differential pumping system RHINOCEROS. The gas is purified and recirculated. A 1-meter VUV monochromator is connected to the furnace by a 2-stage differential pumping system. The light is detected by a channeltron and data are taken by scanning the wavelength. (WK: Roots pumps; TP: turbomolecular pumps).

small apertures could be used because a beam diameter of approx. 1 mm was achieved by a well-designed beam transport system. The schematic set-up is shown in Figure 1.

The ion beam was stopped completely in the furnace due to the high gas pressure. This allowed the observation of the emitted radiation in the direction of the beam and resulted in higher intensities due to integration along the axis. The radiation was analyzed by a 1-m VUV monochromator (McPherson 225 Nova) with a spectral resolution of 0.03–0.1 nm. Depending on the examined wavelength region, the monochromator was connected to the furnace by either a LiF window (above 110 nm) or windowless through a second differential pumping system, which provided a vacuum of 10^{-3} Pa in the spectrometer. Two different grating- and detector-combinations were used: above 110 nm a MgF_2 coated grating with blaze 150 nm and a solar blind photomultiplier with MgF_2 window (Thorn-EMI 9412 A), below 110 nm a Pt coated grating with blaze 80 nm and a channeltron (Valvo X 914 BL). The spectra have not been corrected for overall detection efficiency.

3 RESULTS

In 22 cases out of 25 examined rare-gas/alkali mixtures, narrow continua were observed[6–8]. The intensity of the emitted radiation was strongly correlated to the temperature of the furnace and therefore to the pressure of the alkali vapor. The observed transition wavelengths are situated between 188.9 nm (Xe^+Li) and 63.8 nm (He^+K) and agree well with the calculations of Mantel and Langhoff[2] using phenomenological potentials. Table 1 lists the experimental results compared with calculated wavelengths and Figure 2 shows the wide range of emission wavelengths in the case of rare-gas-sodium-mixtures. The shift towards shorter wavelengths is due to the higher ionization energy of the lighter rare gases.

Figure 3 shows the spectra obtained by the combination of krypton with different alkali metals. The transition energy rises from the light to the heavy alkalis, which can be explained by the smaller ionization energy of the heavier alkali metals. The

TABLE 1

Observed Transition Wavelengths of the Ionic Rare Gas Alkali Excimers. The Wavelengths are given in nm; in Parentheses are the Values Calculated by Mantel and Langhoff[2] using a Phenomenological Potential

	Li	Na	K	Rb	Cs
He	66.78	65.76	63.82	—	—
	(68.4)	(68.1)			
Ne	80.70	79.40	77.18	76.66	—
	(81.0)	(80.7)	(78.4)	(77.7)	
Ar	124.54	121.30	115.04	113.7	112.26
	(125.0)	(123.5)	(117.3)	(115.6)	(114.3)
Kr	149.75	144.95	135.70	133.85	131.58
	(148.8)	(146.7)	(137.7)	(135.3)	(133.3)
Xe	188.9	182.9	167.38	164.1	160.50
	(189.1)	(185.2)	(170.2)	(166.8)	(162.6)

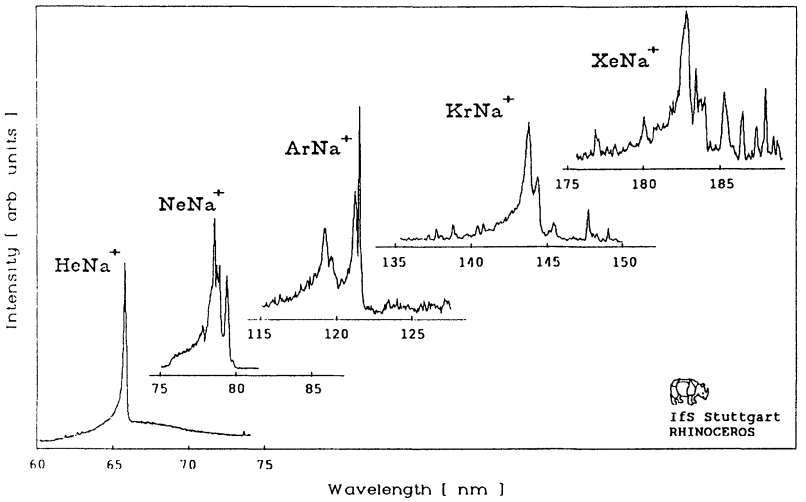

FIGURE 2 Emission spectra of sodium/rare-gas mixtures excited by a 3.2 MeV Ar^+ beam. It can be seen clearly how the transition energy is shifted to shorter wavelengths if one is progressing from heavier to lighter rare gases. This is due to the rising ionization energy of the lighter rare gases.

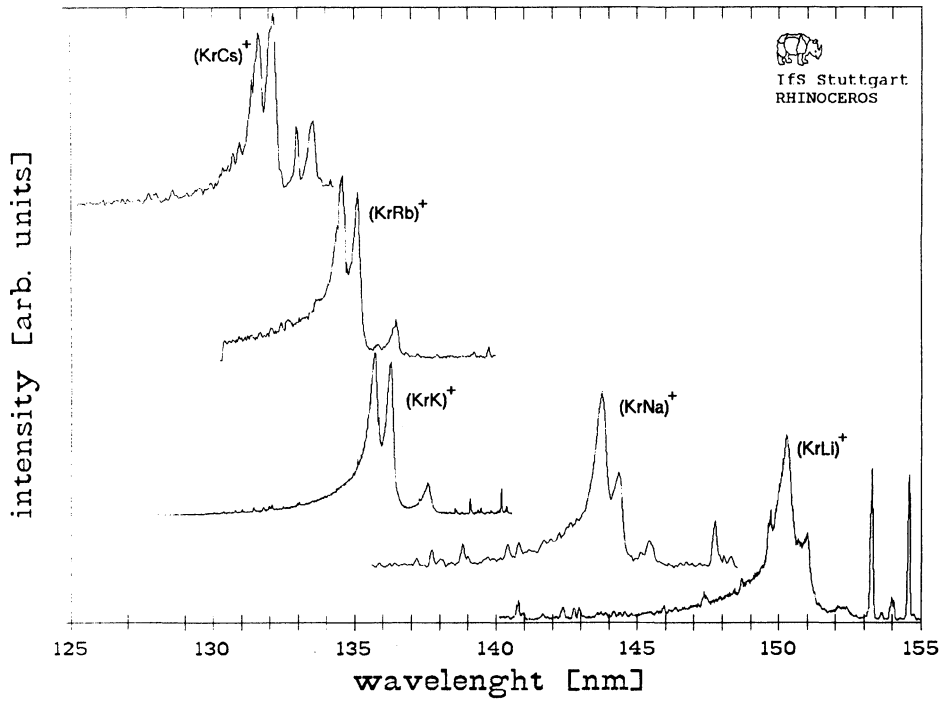

FIGURE 3 Emission spectra of krypton/alkali mixtures excited by a 3.2 MeV Ar^+ beam. With the lower ionization energy of the heavier alkali metals, the transition energy rises and the continua are shifted to shorter wavelengths.

Kr^+A continua show a fine structure which was observed in all rare-gas/alkali continua except of He^+A (see also Figure 2). This structure was explained by Markert[3] and Iwata[5]. The combination of a $Rg^+ {}^2P_{3/2}$ state with an alkali atom in the $^2S_{1/2}$ state results in 5 bound states, a $Rg^+ {}^2P_{1/2}$ state combined with a $A {}^2S_{1/2}$ state leads to a triplet, shifted by the energy difference of the asymptotic states. Due to selection rules for optical transitions, a triplet and a doublet are expected. In the helium system only 2 bound states exist, resulting in a single transition.

The fine structure splittings calculated by Markert could be verified experimentally[7,8]. In a few cases the line intensities were too small to observe all members of a multiplet. For those rare-gas-alkali combinations with no observable transition, the theoretical considerations predict no bound excited states, therefore it can be claimed that the observed group is complete.

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