New Interpretation of the Third Rare Gas Excimer Continua ◇

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Particle beam excitation of dense rare gases leads to broadband light emission in the vacuum ultraviolet and ultraviolet spectral region. Time integrated emission is dominated by the so called second continuum which can be assigned to the radiative decay of the lowest lying states of neutral excimer molecules. The first continuum on the short- and the so called left turning point region (LTP) on the long wavelength side of the second continuum can be assigned to transitions from high lying vibrational levels of the neutral excimers.

A broad continuum at longer wavelengths, however, could still not be unambiguously assigned. It is often referred to as the third excimer continuum. fig. 1 shows emission spectra of 500mbar argon, excited by a pulsed 100MeV ^{32}S beam from the Tandem accelerator. The third continuum shows, by far, the most intense light emission if time resolved light detection is used following pulsed excitation. Light emitted at different times, following the 2ns beam pulse excitation, and normalized to a 4ns time interval, was detected in these spectra, as indicated in the figure.

It is agreed in all interpretations of the third continua published so far that this emission is due to transitions of electronically highly excited excimer states. One assignment attributes the third continuum to transitions from highly excited, singly ionized dimers [1] (asymptotically corresponding to the ground state of an ion R^+ and an excited state R^*) to the R_2^+ ground state. Another assignment interprets the third continua being due to radiative decay of doubly ionized dimers $R_2^{2+} \rightarrow 2R^+$ [2]. Trimer molecules may contribute to the emission as well in both cases.

Up to now, the complex structure (see fig. 1) of the third continuum spectral features, measured in various experiments, has been described as follows: Either, yet unspecified shifts and broadening effects are assumed as a reason for the variations in the shape of the third continuum with time and gas density in the case of the R_2^{+*} approach, or different emission wavelengths due to spin-orbit coupling in the R_2^{2+} hypothesis. None of these interpretations, however, can consistently explain the temporal behaviour of experimental data, obtained so far.

Here, a new, detailed study was performed using pulsed heavy ion beam excitation of argon at various gas densities with a pulse width of 2ns. Spectra were recorded in the time intervals of 0-4, 4-8, 8-16, 16-32, and 32-128ns, following the onset of ion beam excitation of the gas target. Light intensity was normalized to 4ns time intervals observed for a quantitative comparison of the shape of the emission spectra.

A new approach is used to describe the origin of the third continuum radiation, using the high quality data obtained at the Munich Tandem accelerator. It is based on the idea that the complex structure of the third continuum is due to a combination of optical transitions from different excited species, radiating at different times. Consequently, an attempt was made to fit the complex spectra, which were observed for gas densities between 230 and 1500mbar and time intervals between 0-4ns and 32-128ns, with a limited number of regular excimer bands of about 15nm spectral width. Indeed, it could be shown that 6 bands in the third continuum region are suffient for reproducing 16 spectra observed for various conditions; (actually 9 gaussian curves have been used to fit the entire spectrum, including the second continuum and the LTP region). The central wavelengths are listed in the insert in fig.1.

In this approach, we would tentatively assign the emission observed at 188nm, occuring at early times and low pressures, to the doubly charged dimers R_2^{2+} described in ref. [2] since only one rapid gas kinetic process is necessary to populate the upper levels of these transitions. The emission at 199nm, occuring later than the 188nm emission could then be attributend to the R_3^{2+} excimer. The R_3^{2+} potential curves, however, are crossed by those of a (R^{+*} + R) + R^+ configuration [3], and a transition to the precursers of the 3rd continuum, as proposed by Boichenko [1] will occur. The emission at 211nm is therefore attributed to Ar_2^{+*} transitions. Emission bands at 224nm and 252nm, observed at late times and/or high pressures might then be due to trimer and cluster decay, respectively, which are invoked as a reason for the red shift at late times and high gas densities in all interpretations of the third continua discussed in the literature.

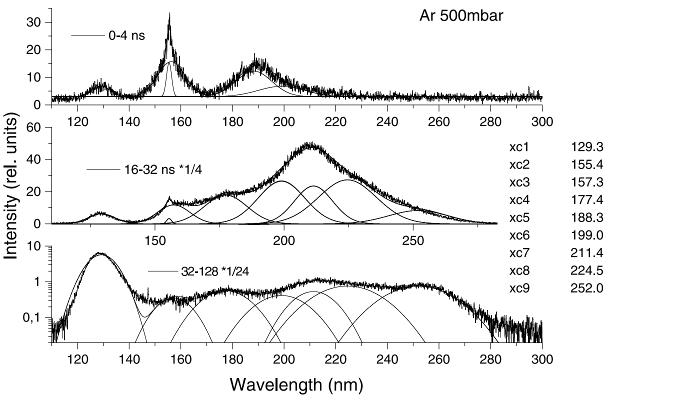

Fig. 1: Wavelength spectra of 500mbar Argon are shown in three different time windows after a 2ns S-beam pulse excitation. The spectra can be reproduced by gaussian curves, representing the decay of different excited species. The center-wavelengths of the emission bands are indicated in the plot (xc1-9 in nm). Keeping the position and width, all spectra at pressures ranging from 230mbar to 1500mabr and time intervals reaching from 0-4ns after excitation until 32-128ns can be described. The emission at 129.3nm and 155.4nm are the second continuum and the LTP region, respectively.

References

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