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Emission of vacuum ultraviolet radiation from neon excimers excited by a heavy ion beam

W. Krötz, A. Ulrich, B. Busch, G. Ribitzki, and J. Wieser
Technische Universität München, Physik-Department E12, D-8046 Garching, Federal Republic of Germany

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The vacuum ultraviolet emission of neon excited with a pulsed 100 MeV $^{32}S^{9+}$ ion beam from the Munich Tandem van de Graaff accelerator was studied at pressures between 1.8 and 96.1 kPa. In the wavelength range between 70 and 110 nm the first, second, and third excimer continua were observed. From time- and pressure-dependent studies of the third continuum emission at a wavelength of 99 nm, rate coefficients $k_2 = (3.6 \pm 0.3) \times 10^{-13}$ cm^3/s for the bimolecular reaction $Ne^{2+} + Ne \rightarrow 2Ne^+$ and $k_3 = (2.84 \pm 0.09) \times 10^{-31}$ cm^6/s for the termolecular reaction $Ne^{2+} + 2Ne \rightarrow (Ne^{2+}Ne)^{2+} + Ne$ were determined.

The excitation of rare gases leads to the emission of molecular continua in the vacuum ultraviolet (VUV) wavelength region. The so-called first and second continua have been studied using different excitation methods like electron and proton beams or α particles. Laser action has been observed on the second continuum of the heavier rare gases at about 130, 150, and 170 nm, in Ar, Kr, and Xe, respectively.[1]

The first and second continua originate from transitions between the lowest bound state and the repulsive ground state in neutral excimer molecules. Fewer studies have been made on the third continua, which appear at longer wavelengths than the second continua and the origin is still under discussion. In general, the excimer continua of neon are less studied because they are emitted at wavelengths shorter than the cutoff wavelength of LiF windows ~ 105 nm.

We are studying the light emission from matter excited by heavy ion beams.[2] Here we report on measurements of the VUV emission of neon excimers excited by 100 MeV ^{32}S ions.

Ne gas targets in the pressure range between 1.8 and 96.1 kPa were excited with a pulsed 100 MeV $^{32}S^{9+}$ from the Munich Tandem van de Graaff accelerator. The target cell was separated from the vacuum in the accelerator beam line by a 1.1 mg/cm^2 Ti entrance foil, leading to an energy loss of 12.2 MeV for the ^{32}S ions.[3] The beam pulses had a width of 2 ns and a repetition rate of 78 kHz. The average beam current was 5.6 nA (particle), which led roughly to 6 μJ excitation energy for each beam pulse.

In two experiments, light emission from neon was studied using a 2.2 m grazing incidence stepping monochromator with a channeltron detector. At pressures between 9.6 and 96.1 kPa, the ^{32}S beam was stopped in a 60-cm-long target cell and the monochromator was placed in the forward direction. For lower pressures between 1.8 and 19.2 kPa, a short 17-cm-long gas target cell was used. In this case, the monochromator, placed at an angle of 20° with respect to the beam axis, accepted light from a 1.5-cm-long region 11.5 cm behind the Ti entrance foil. The experimental setup for the experiments at higher target gas pressures is schematically shown in Fig. 1.

A differential pumping stage was used to separate the vacuum in the monochromator from the gas target. The pumping stage had three apertures 1 mm in diameter within a distance of 2.5 cm. The distance between the last aperture and the entrance slit of the monochromator was 21 cm for the experiments at high pressures and 3 cm at low pressures.

The main impurities in the target gas were specified as follows: He$\leqslant$10.0 ppm, $N_2 \leqslant 5.0$ ppm, $H_2O \leqslant 2.0$ ppm, $O_2 \leqslant 1.0$ ppm, total $\leqslant 20$ ppm. The monochromator was operated at an angle of incidence of 86° and the reflectivity of the grating was optimized for a wavelength of 87.3 nm. Time-resolved measurements of light emission were performed using the time-to-amplitude conversion method. A start signal for the electronics correlated with the incident beam pulse was derived from a capactive probe in the accelerator beam line.

A time-integrated spectrum of neon at a pressure of 38.5 kPa is shown in Fig. 2. The spectrum was corrected for the detection efficiency of the channeltron detector, but not for the spectral response function of the grazing incidence monochromator.

The first and second Ne_2 excimer continua at 73 and 83 nm, respectively, were observed. An additional emission band, which is referred to as third continuum in literature,[4,5] had its maximum intensity at a wavelength of 99 nm. Taking into account an estimated spectral response function of the monochromator, the intensity of the third excimer continuum is about half the intensity of the second continuum.

Time spectra were taken at Ne gas pressures between 1.8 and 96.1 kPa and at a wavelength of 99 nm to study the formation and decay kinetics of the third continuum. The experimental results for pressures of 9.6, 18.6, 38.5, and 96.1

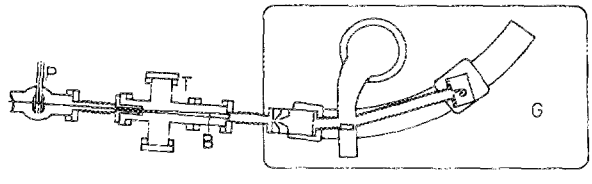

FIG. 1. Schematic view of the experimental setup for measurements at pressures between 9.6 and 96.1 kPa. The ^{32}S ion beam (B) comes from the left, passes a capacitive probe (P), and is stopped in the Ne gas target (T), which is separated from the vacuum in the accelerator beam line by a 1.1 mg/cm^2 Ti entrance window. The VUV emission below 125 nm is studied with a grazing incidence monochromator (G). A differential pumping stage is installed between the target cell and the monochromator.

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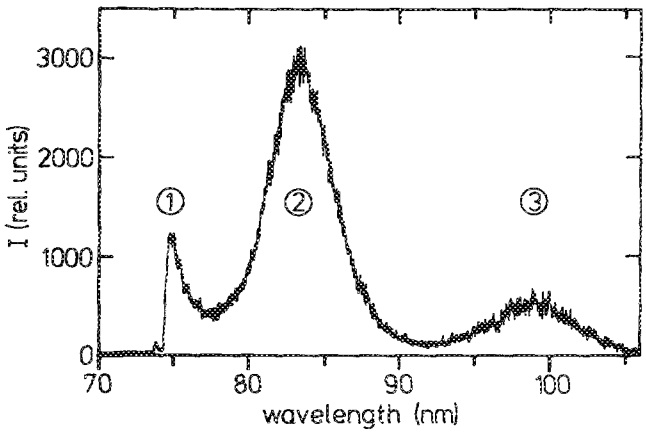

FIG. 2. Time-integrated emission spectrum of Ne at a pressure of 38.5 kPa. The first (1), second (2), and third (3) excimer continua were observed. The spectrum is corrected for the detection efficiency of the channeltron detector, but is not for the spectral response function of the monochromator.

kPa, which were recorded with the monochromator in forward direction, are shown in Fig. 3. Due to a shorter range, and, therefore, a shorter time of flight of the ^{32}S projectiles the observed rise times shorten at higher pressure. As soon as the ^{32}S ions are stopped in the Ne gas, the decaying light emission can be fitted by a single exponential decay function with a time constant $\tau(p)$ for each target gas pressure p. The time constants are summarized in Table I.

The time-integrated light emission of the first, second, and third continuum excited by protons of 4 MeV particle energy was studied by Stewart *et al.*[4] and the results were compared with those from gas discharge excitation. They concluded from impurity-dependent quenching studies that the second and third continua may have different precursors or may be produced by different kinetic processes.

Leichner[6] studied the time-dependent emission of the first and second continuum excited by 250 KeV electrons. He observed a weak continuum in the 100 nm region at pressures < 40 kPa.

Recently Langhoff[5] explained the third excimer continua by an emission during the decay of the $(Ne^{2+}Ne)^{2+}$ molecular ion to the repulsive ground state $Ne^+ + Ne^+$:

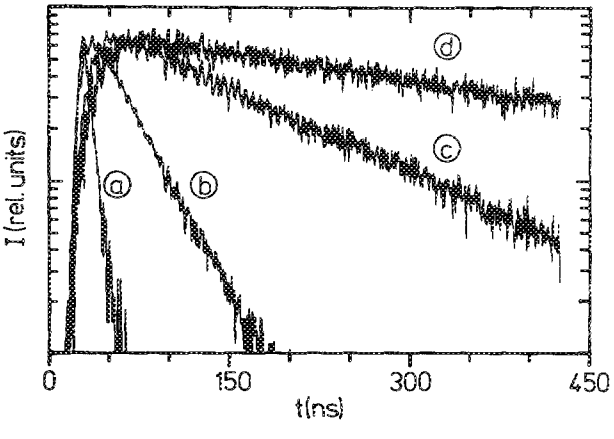

FIG. 3. Time spectra of the third excimer continuum in Ne at a wavelength of 99 nm. The spectra were taken with the setup shown in Fig. 1 at target gas pressures of (a) 96.1 kPa, (b) 38.5 kPa, (c) 18.6 kPa, and (d) 9.6 kPa.

TABLE I. Pressure dependence of the decay time constant $\tau(p)$ of the third continuum emission in neon at 99 nm. The results shown at the left side of the table were taken with the monochromator in the forward direction. For the results at the right side, the monochromator was placed at an angle of 20° with respect to the ion beam axis.

p(kPa)	$\tau(p)$(ns)	p(kPa)	$\tau(p)$(ns)
96.1	5.88 ± 0.04	19.2	119.1 ± 7.1
38.5	33.3 ± 1.1	9.6	435 ± 57
18.6	131.6 ± 1.7	4.7	1430 ± 200
9.6	385 ± 15	1.8	4170 ± 20

$$(Ne^{2+}Ne)^{2+} \xrightarrow{1/\tau_0} 2Ne^+ + h\nu. \quad (1)$$

The doubly charged diatomic molecular ion $(Ne^{2+}Ne)^{2+}$ is assumed to be formed in the following termolecular reaction:

$$Ne^{2+} + 2Ne \xrightarrow{k_3} Ne_2^{2+} + Ne, \quad (2)$$

where the Ne^{2+} ions are in the ^3P ground state. The most probable loss process for Ne^{2+} ions is the bimolecular charge transfer reaction:

$$Ne^{2+} + Ne \xrightarrow{k_2} Ne^+ + Ne^+. \quad (3)$$

The rate constants for the bimolecular and termolecular reactions are indicated by k_2 and k_3, respectively.

The high intensity of the third continuum in the spectrum shown in Fig. 2 compared with the intensity for electron and proton excitation[4,6] supports this kinetic scheme, as the cross section for the production of Ne^{2+} is large for heavy ion excitation.[7]

By solving the rate equation for processes (1), (2), and (3), the time dependence of the emitted light pulse can be described by the following function:

$$I(t) = A \exp(-t/\tau_0) + B \exp\{-t/\tau(p)\}, \quad (4)$$

with the pressure-independent time constant τ_0 of reaction (1) and a time constant $\tau(p)$, which is the inverse of $k_2[Ne] + k_3[Ne]^2$. The square brackets indicate the density of the neon atoms. Taking into account that the pressure is directly proportional to the density of neon atoms, this relation can be written as

$$(\tau(p)p)^{-1} = k_2 \times 2.48 \times 10^{17}\ \text{kPa}^{-1}\ \text{cm}^{-3} + k_3 \times p \times 6.15 \times 10^{34}\ \text{kPa}^{-2}\ \text{cm}^{-6} \quad (5)$$

for a temperature of 292 K.

A fit to the experimental data shows that τ_0 must be short compared with the smallest value of $\tau(p) = 5.9$ ns observed in our experiment at a pressure of $p = 96.1$ kPa. A lifetime τ_0 shorter than 5 ns for the third continuum in neon seems reasonable from the systematic behavior of values τ_0 obtained by Birot *et al.*[8] and Millet *et al.*[9] for the heavier rare gases.

The reciprocal of the product of the decay constant $\tau(p)$ and pressure p from our experiments is plotted versus the pressure p in Fig. 4. By a least-squares fit of Eq. (5) to the data points, rate constants $k_2 = (3.6 \pm 0.3) \times 10^{-13}$ cm^3/s and $k_3 = (2.84 \pm 0.09) \times 10^{-31}$ cm^6/s were obtained. To

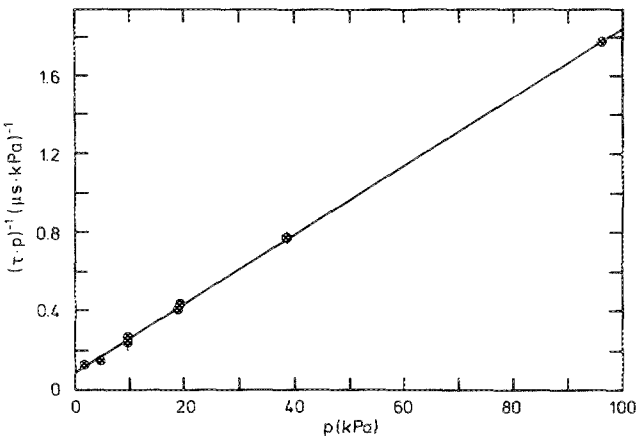

FIG. 4. Inverse of $(\tau(p)\times p)$ is plotted vs the pressure p of the neon target in the pressure range between 1.8 and 96.1 kPa at an emission wavelength of 99 nm. Where no error bar is shown, the experimental error was smaller than the data point indicated. The straight line is a least-squares fit to the data.

our knowledge this is the first measurement of these rate constants for neon by studying the light emission of the third neon excimer continuum directly.

A comparison can only be made with measurements at substantially lower pressure. De Hoog and Oskam[10] determined the rate constant k_2 for reaction (3) by studying the time dependence of the density of Ne^{2+} and Ne^{+} ions during the decay period of a neon plasma at pressures ranging from 18.7 to 133.3 Pa. Johnsen and Biondi[11] used a drift-tube mass-spectrometer apparatus at pressures between 13.3 and 173.3 Pa to measure state-selective charge-transfer rate coefficients k_2 and k_3 for reactions (2) and (3) at thermal energy (300 K). For the rate constant k_2 we find a significant deviation from the values given in the literature. Our measurement of the rate constant k_3 is on the other hand in good agreement with the value of Johnsen and Biondi.[11] A comparison of the results is shown in Table II.

TABLE II. Comparison of rate constants k_2 and k_3 for the two- and three-body partial charge transfer reactions of Ne^{2+} in Ne, obtained in different experiments.

Pressure range	State (Ne^{2+})	k_2 (10^{-14} cm^3/s)	k_3 (10^{-31} cm^6/s)	Author
13.3–173.3 Pa	^3P	2.1 ± 0.2	5.1 ± 1	Johnsen and Biondi[a]
	^1D	1.9 ± 0.2	3.5 ± 0.5	
	^1S	2.7 ± 0.3	3.5 ± 0.5	
18.7–133.3 Pa	···	9 ± 2	···	De Hoog and Oskam[b]
1.8–96.1 kPa	^3P	36 ± 3	2.84 ± 0.09	This paper

[a] Reference 11.
[b] Reference 10.

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