Experimental verification of a zero-dimensional model of the kinetics of XeCl* discharges by XeCl*(B)-, XeCl*(C)-, and Xe2Cl*-density measurements

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Abstract. Absolutely calibrated emission spectroscopy has been used to determine the particle number densities of XeCl*(B), XeCl*(C), and Xe2Cl* in a small scale Ne/Xe/HCl discharge with well-defined current and voltage pulses for a wide range of parameters. The measured particle number densities could be reproduced quite well by numerical model calculations using the rate-coefficient values of Quiñones et al. [1] for the quenching of XeCl*(B,C) by Ne, Xe, and 2Xe, but 3.0 x 10^-31 cm^6/s for the formation of Xe2Cl* by (Ne + Xe)-quenching. For the electron quenching, we recommend a rate coefficient value of 3.2 x 10^-8 cm^3/s.

From the equilibrium ratio of the particle number densities of XeCl*(C) and XeCl*(B), the energy separation between these states has been estimated to be 72 ± 33 cm^-1 with the B state placed above the C state.

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Several spatial homogeneous (zero-dimensional) XeCl* excimer laser models [2–14] have been developed for more than a decade. The predictions of these models are usually compared with measurements of the discharge voltage, the discharge current, and the photon pulse of the laser, only. In some cases [6, 9, 15–17], the comparisons of the model calculations have as well been carried out for spectroscopic measurements. As lasers are not built to give easy access to precise measurements of discharge voltage and current and to optical plasma-diagnostic methods, the accuracy of such comparisons is limited.

In order to check the predictions of zero-dimensional models, a small discharge configuration for the generation of homogeneous high-pressure glow discharges was developed [18]. A description of the theoretical model of this discharge configuration containing an extensive reaction kinetics is presented in [19]. In that work, an experimental verification of the ionization and recombination kinetics of Ne/Xe/HCl mixtures is given by comparing the results of model calculations with precise measurements of discharge voltage and current over a wide range of discharge parameters. A further verification of the model by means of comparisons with absorption-spectroscopic measurements of excited-species number densities Xe*, Ne*, Cl*, and H* can be found in [20].

To gain additional insights into the reaction kinetics of XeCl* and to give a further verification of the model, measurements of excited-species number densities XeCl*(B), XeCl*(C), and Xe2Cl* have been performed using absolutely calibrated emission spectroscopy. The formation and the quenching of the upper laser level can be checked by the measurements of XeCl*(B) particle number densities. Additional information about the quenching of XeCl* can be obtained from measurements of XeCl*(C) and Xe2Cl* particle number densities. XeCl* is formed predominantly by Xe+-Cl- recombination in mixtures containing HCl as halogen donor, the comparison of measured XeCl* number densities with model calculations represents a further check of the ionization and recombination kinetics.

The results of the experimental measurements of the particle number densities of XeCl*(B), XeCl*(C), and Xe2Cl* are compared with model calculations over a wide range of parameter variations. The rate-coefficients for XeCl* quenching are discussed based on our simple model of XeCl* taking into account the B and C states only. The influence of vibrational relaxation within these states and of mixing between the vibrational levels of XeCl*(B,v) and XeCl*(C,v') on the results is estimated.

The measurements of XeCl*(B) and XeCl*(C) number densities are utilized as well to estimate the energy separation between the B and C states of XeCl*. The method for the determination of the energy gap is described. Our result is compared with recent values found in literature.

1 Experiments

1.1 Discharge configuration

The discharge configuration has been described previously [18]. It consists of a discharge chamber, an impulse generator to produce a well-defined voltage and current impulse,
The relevant response curves have been measured at those intensities. The signal of a photomultiplier tube (RCA, 931 A), which is linear relative to the incident intensity, is used to calibrate the emission spectra. A low-current carbon arc (current 7.3 A, voltage 70 V) is used to calibrate the emission. A emission bands on the same CCD picture. The broadband XeCl $4^2\Sigma^-\rightarrow1^2\Pi$ emission is recorded by detuning the spectrometer and taking two CCD pictures. However, it is not possible to get information about the spatial distribution of the emission, because the spectrometer has not an entrance slit but a pinhole (diameter 100 μm). The optical axis of the discharge is focussed on this pinhole.

Spatially resolved and time-integrated measurements of the XeCl $B\rightarrow X$ emission around 308 nm are performed with a 1 m spectrometer with a plane ruling ruled at 1200 grooves/mm. Again, the spectra are recorded by the CCD camera. As the entrance slit of the spectrometer lies perpendicular to the optical axis of the discharge, the CCD picture gives information about the lateral and spectral distribution of the XeCl $B\rightarrow X$ emission. A low-current carbon arc (current 7.3 A, voltage 70 V) is used to calibrate the emission spectra absolutely. The CCD camera delivers a standard video signal which is digitized by a real-time digitizing board (Oculus 300, Coreco, 512 x 512 pixel, 8 bit). Due to the good reproducibility of the discharge, it was possible to reduce the noise-to-signal ratio by averaging over up to 30 discharges.

As the carbon-arc spectrum and the discharge spectrum are taken at different exposure times, the relevant linearity of the camera has to be checked. While a satisfactory result is yielded in this case, we found that the DiCAM-2 is not linear relative to the incident intensity. Figure 2 shows a saturation at higher camera signals, i.e., higher incident intensities. The signal of a photomultiplier tube (RCA, 931 A), which is linear over the investigated intensity range, is used as reference signal. The response of the CCD camera has been corrected for the nonlinearity at high intensities. The corrected signal can be described by a logarithmic regression. The saturation depends on the exposure time and the gain of the camera. The behaviour of the camera is similar to the response of a photographic emulsion to radiation. The relevant response curves have been measured at those camera adjustments (gain, exposure time) which are used for recording of the spectra.

For time-resolved detection of the fluorescence, a photomultiplier tube (Burle, 1 P28 B) is placed at the output slit of a 0.3 m monochromator (McPherson, 270). The temporal evolution of the XeCl $B\rightarrow X$ and $C\rightarrow A$ emission is measured.