Topic number: 5

A 3 quantum cascade laser spectrometer for fast in situ determination of absolute number densities

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Since several decades, the chemical and kinetic phenomena of plasmas containing nitrogen and oxygen have been studied. They play an important role in e. g. pollution abatement from gas exhaust. In former experiments [1], the interaction of rf plasmas with a dielectric material has been of interest in order to understand the catalysed destruction and production of molecules on the surface in the after glow. Additionally, the kinetics of NO_x destruction during an ms dc pulse discharge has been of interest [2] for the verification of modelled results of NO_x containing plasma. In these experiments the temporally behaviour of the concentration of NO [2] or NO and NO_2 [1] has been determined. From the results, the authors suggest the production of other N_xO_y. To investigate especially these phenomena a multi quantum cascade laser (QCL) spectrometer has been designed for extending kinetic studies. It is made of 3 QCL sensitive for NO, NO_2, N_2O and CO. The lasers are combined and the diameter of the laser beams are reduced and guided on the same detector, figure 1. All lasers are working in the Intra Pulse Mode emitting in the infra red region. To avoid signal overlapping a delay generator is used to provide sufficient delays. Figure 2 shows a typical spectrum of all three laser pulses measured with an oscilloscope. All the spectra were analysed using a self made algorithm. In order to obtain absolute species concentration the system was calibrated separately using gas mixtures at known pressure and concentrations.

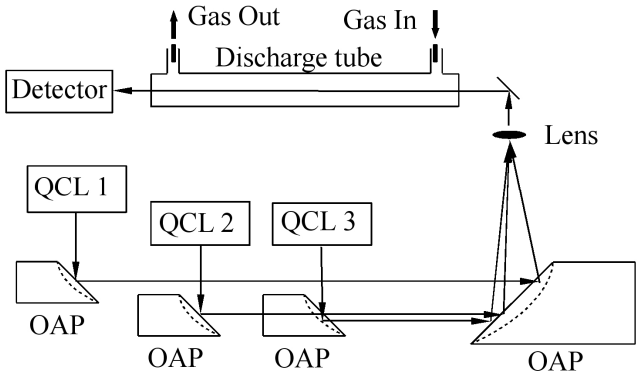

Fig. 1 Principal sketch of the optical arrangement. One inch off axis paraboloid (OAP) mirror parallelises the laser beams. All three are collimated through a lens on the same detector. The figure also shows the dc discharge tube. The two black rectangles in the gas in- and output represent the electrodes.

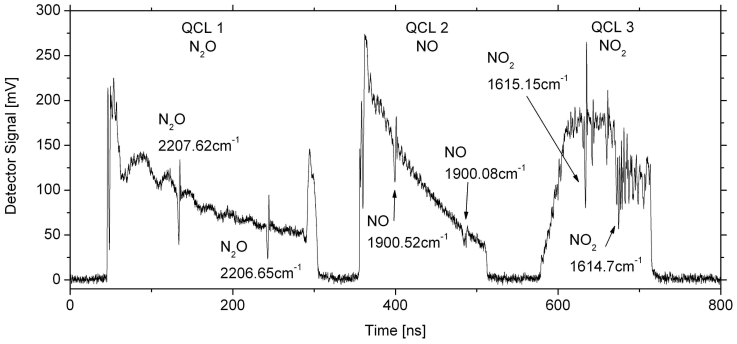

Fig. 2 Pulse pattern of all three lasers working in the Intra Pulse Mode. A delay generator avoids spectral overlapping. Clearly visible is the rapid passage effect occurring for all species, N_2O, NO and NO_2.

In the following, two examples are given of measuring the time depended N_xO_y concentration in a N_2 / O_2 plasma with small N_xO_y admixtures. Both experiments were done under flowing conditions with 10sccm N_2 and 2.5sccm O_2 at a pressure of 1.3mbar. A dc high voltage pulse of 10 ms duration was applied to the discharge tube 8 ms after the start of the spectral measurements. For the first experiment, 1% of N_2O was added. The temporally behaviour of the N_xO_y concentration with a time resolution of 200µs is given in figure 3. For the second experiment, 1% NO_2 was added. Figure 4 shows the time depended N_xO_y concentration during the dc pulse after adding 1% NO_2. The time resolution has been 50µs.

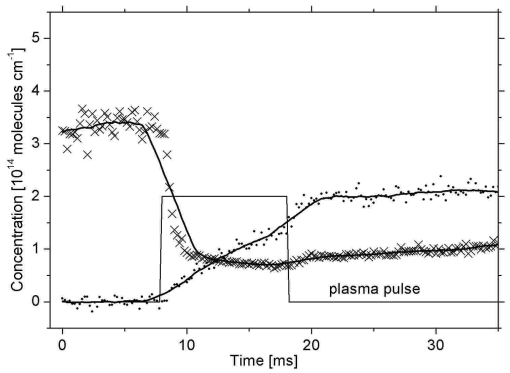

Fig. 3 Time depended behaviour of N_2O (x) and NO (…) during a high voltage dc pulse after an admixture of 1%N_2O. NO_2 was not detectable under such conditions. The pulse started 8ms after spectrum acquisition.

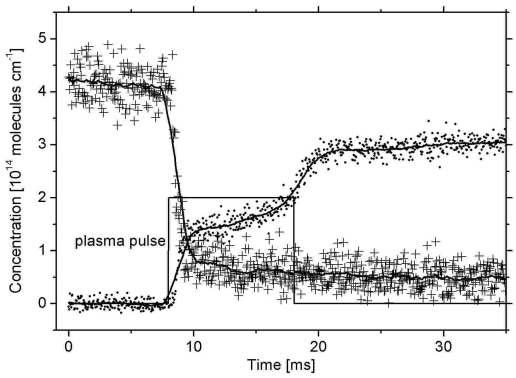

Fig. 4 Time dependency of the NO_2 (+) and NO (…) concentration after adding 1%NO_2. N_2O was not detectable under these conditions.

The first example shows a decreasing of the initial N_2O concentration whereas at the same time an increasing of the NO concentration can be seen. Here, no NO_2 was detectable which indicates that NO_2 was less than the detection limit which was about 3.4 10^{13} molecules cm^{-3}.
On the other hand, figure 4 shows an almost complete reduction of the initial NO_2 concentration within the first view ms of the dc pulse. The NO concentration increases within the pulse duration. The detection limit of N_2O was about 5 10^{12} molecules cm^{-3}. Further studies are in progress to separate surface and volume induced species transformation processes.

Reference

[1] O. Guaitella, M. Hübner, S. Welzel, D. Marinov, J. Röpcke and A. Rousseau, 2010 *PSST* submitted.
[2] S. Welzel, L. Gatilova, J. Röpcke and A. Rousseau, 2007 *PSST* **16** 822