A temperature study of a low pressure, pulsed dc plasma using quantum cascade laser absorption spectroscopy (QCLAS)

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The gas temperature is measured in a pulsed N_2 plasma with a duration of some ms using absorption spectroscopy in the mid-infrared spectral range with pulsed quantum cascade lasers (QCL). For the determination of the temperature the intensity ratio of two absorption structures of NO used as a probe gas has been evaluated. A calibration routine based on simulated spectra has been developed in order to compensate the influences of non-linear absorption phenomena. This procedure has been validated for a heated gas cell, where the gas temperature was known.

The destruction of volatile organic compounds (VOC) from an air stream has become a more interesting topic in order to fulfil latest environmental regulations. An important parameter for these studies is the gas temperature. It influences the rate of chemical reactions as well as the diffusion of the particles toward the wall. The latter becomes important in plasma catalyst coupling experiments. Consequently, the temperature of a plasma has to be known.

In this contribution, a method is introduced which applies QCLAS to the measurement of the temperature of NO handled as a probe gas. This has been done to demonstrate the principal possibility to measure the temperature using QCLAS despite the strong disturbance of the absorption structures. Such phenomena are caused by non-linear effects, e.g. rapid passage effect or saturation effects, due to the high tuning rate and intensity of the probing laser.

The measurements of the temperature have been carried out in a Pyrex tube reactor with an inner diameter of 20mm and a length of 60cm. The ends of the tube were closed with Kalium Bromide (KBr) windows, figure 1, [1]. The experiments have been done under static gas conditions in a closed reactor. The used power supply creates HV pulses with a rise time in the kV/µs range. The high voltage was measured with a HV probe (Tektronix, P6015A). The current has been detected by the current proportional voltage drop over a measurement resistor. Both signals have been detected by an oscilloscope (LeCroy, 500MHz, 1GS/s).

A commercially available Q-MACS system (neoplas-control) has been used with a three pass alignment. The laser pulses have been detected by a fast detector with a rise time of 3ns (neoplas-control, IRDM600A, bandwidth: 600MHz, rise time: 2ns). The pulse duration of the quantum cascade laser (QCL) was 150ns with a pulse repetition frequency of 30kHz. The internal thermal expansion of the QCL leads to a slightly tuning of the emitted laser radiation. This allows a scan over the spectral range of interest. An example of the detected spectrum can be seen in figure 2. One can see a single line absorption structure of NO at 1900.52cm^{-1}, $NO(X_{3/2}, \upsilon = 0) \rightarrow NO(X_{3/2}, \upsilon = 1)$: R6.5, and a double line absorption structure of NO at 1900.08cm^{-1}, $NO(X_{1/2}, \upsilon = 0) \rightarrow NO(X_{1/2}, \upsilon = 1)$: R6.5, of a gas mixture of 1% NO in N_2 at 1.3mbar total pressure with an absorption length of L = 180 cm, i.e. three pass alignment. Thus, the detection limit was about 4×10^{13} molecules cm^{-3}.

The gas temperature has been determined from the absorption spectrum applying the line ratio method [2]. The intensity of an absorption structure for a constant concentration depends on the gas temperature. Therefore, the ratio of the intensity of two absorption structures, figure 2, can be used to study the gas temperature. However, due to the distortion of the absorption line, figure 2, this approach needs an additional calibration routine. Therefore, the same spectral range has been simulated for different gas temperatures using the HITRAN [3] database. Afterwards, the line ratio of the simulated spectra has been calculated in the same way as for the measured spectra. Subsequently, a calibration factor has been determined in order to ensure that the measured line ratio is equal to the simulated line ratio. Finally, one obtains a calibration curve temperature as a function of the measured line ratio. This approach has been validated in a heated reference gas cell, where the gas temperature was known.

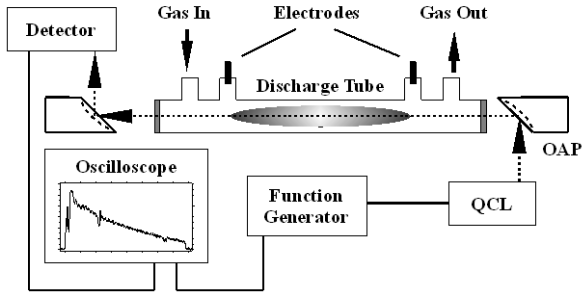

Fig. 1: Sketch of the experimental setup. The IR radiation passed the tube reactor three times. The T-shaped electrode holders allow spectroscopic studies in the homogenous part of the dc plasma [1].

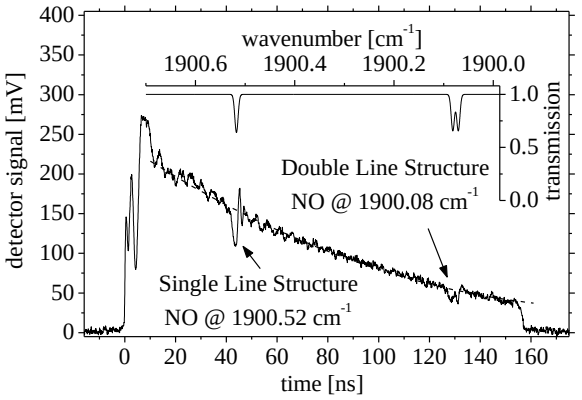

Fig. 2: NO spectrum (bottom) measured by QCLAS and a simulated spectrum (embedded diagram) using the HITRAN database [3] both for a gas mixture of 1% NO diluted in N_2 [1].

As an example, the line ratio method has been applied to a pulsed dc plasma. A gas mixture of about 1% in N_2 at 1.3mbar initial pressure has been used. The HV pulses have been applied for 5ms. The applied high voltage was varied in order to obtain several mean plasma currents, ranging from 25mA to 150mA. The injected electrical power ranges from 0.2J up to 1.5J. Figure 3 shows the temperature as a function of the electrical energy.

Using absorption spectroscopy, one integrates the line intensities along the line of sight. In the tube reactor used here, cold parts remain close to the windows. Consequently, the temperature of the plasma might be slightly higher. These corrections, however, have been neglected.

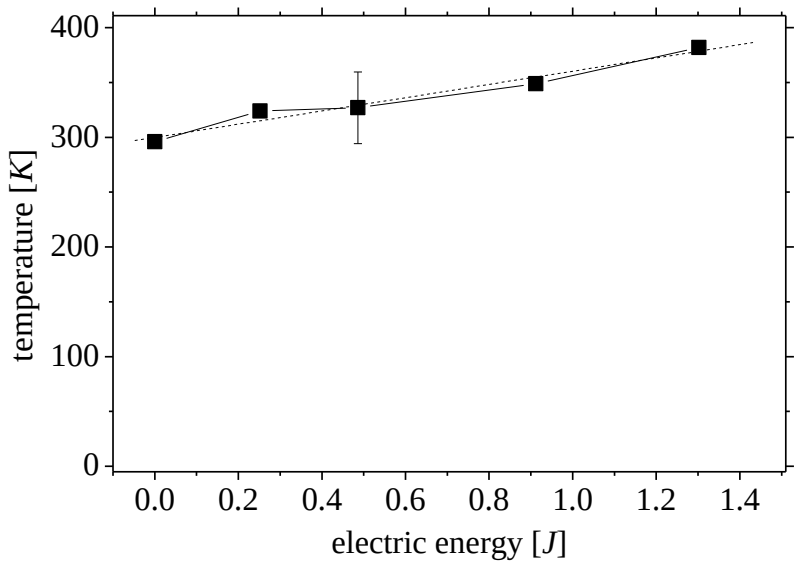

Fig. 3: Final NO temperature of a pulsed dc plasma measured using the line ratio method. The electric energy corresponds to a mean pulse current of 25mA, 50mA, 100mA and 150mA, respectively. The dashed curve shows a linear fit.

We have shown that QCLAS is suitable to measure the gas temperature of NO as a probe gas. This has been possible despite the strong distortion of the absorption structure at low pressure conditions.

References

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