Ro-vibrational excitation of CO_2 measured by quantum cascade laser absorption in a nanosecond pulsed discharge

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Conversion of CO_2 is of growing interest in the context of greenhouse gas abatement and renewable energy exploration. Non-thermal plasmas are promising for an efficient conversion since their unique electron, vibrational, rotational, and gas temperatures allow to focus the discharge energy to the desired channels instead of heating the gas. Specifically, the vibrational excitation of states close to the dissociation threshold level is a more efficient dissociation pathway. In order to gain insight into the excitation and relaxation processes and to validate detailed kinetic models of CO_2 dissociation, temporally resolved measurement of the rovibrational excitation is of great importance in non-thermal and transient discharges.
A continuous wave quantum cascade laser tunable between 2276 and 2290 cm^{-1} and a fast detector (65 MHz) is used for measuring the absorption with high temporal resolution (8 ns) [1]. The chosen wavelength range allows for simultaneous measurement of rotational and vibrational populations and CO_2 concentration. Measurements are performed in a nanosecond pulsed discharge ignited between two parallel electrodes of 20 mm length, 1 mm wide and 1 mm apart. The discharge pressure is around 150 mbar with different N_2:CO_2 mixtures. The voltage pulses (V = 2-3 kV, f = 1 kHz) are about 200 ns long with discharge currents of 10 A. In these types of discharges there is a distinct separation of the timescales of electron heating and afterglow. The different phases can be studied in detail due to the high temporal resolution of the method here. The measurements show that the temperatures determined by the population density of the different states depend on the levels used and that the thermalization to a Boltzmann distribution takes much longer than the discharge duration (fig. 1).

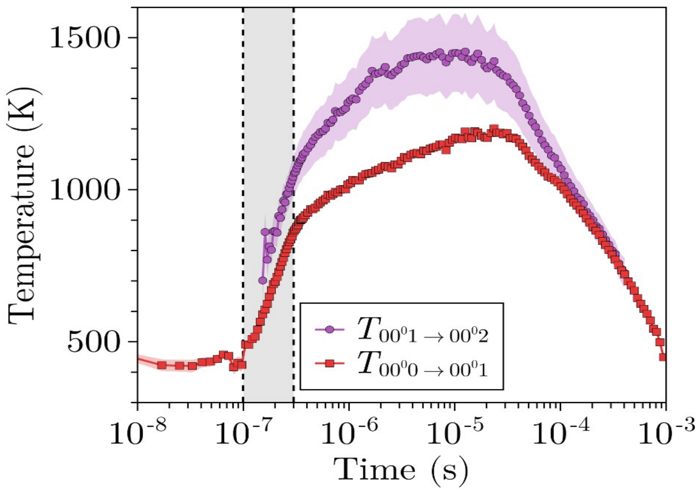

Figure 1. Different vibrational temperature T_3 due to a non-Boltzmann population, the discharge phase is marked in grey.

[1] Yanjun Du et. al. 2021 Journal of Physics D: Applied Physics **54** 365201, ibid 34LT02.

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