Rotationally resolved desorption of NO from graphite at FLASH

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Femtosecond laser desorption from surfaces has so far been studied in the visible and UV spectral region. At XUV photon energies new processes might be initiated not accessible to the lower photon energies. Besides the study of the basic processes these results might also be of interest for the understanding of light-induced molecule formation on interstellar carbonaceous dust particles in the vicinity of star formation regions.
We present results of the NO desorption from highly oriented pyrolytic graphite (HOPG). For the first time rotationally resolved molecular spectra are recorded after desorption with femtosecond pulses from FLASH at 38 eV and 57 eV photon energy. The FEL pulses showed a pulse energy of up to 40 μJ. The weakly focussed $\hat{p}$-polarized radiation strikes the surface at an angle of incidence of 67.5° relative to the surface normal. Due to the oblique incidence the 200 x 300 μm^2 ellipsoidal beam produces a 0.46 mm^2 ellipsoidal spot on the graphite. An average pulse energy of 25 μJ resulting in a fluence of 5.4 mJ/cm^2 is applied. No damage of the graphite was observed during the whole run. The desorbed molecules are detected by a UV laser tunable around 227 nm. A scan across the γ-bands of NO from 227.1 nm to 223.7 nm yields the rovibrational population distribution of the desorbed NO molecules in the vibrational ground and first excited states $v' = 0$ and 1 shown in figure 1.
In figure 2 the degeneracy corrected logarithm of the state population is plotted against the internal

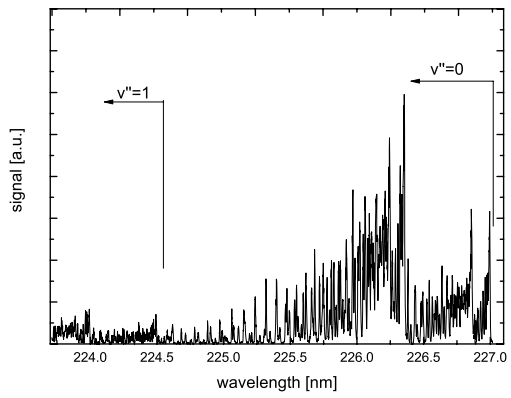

Figure 1: The γ-bands of NO around 227 nm recorded at a photon energy of 57.1 eV.

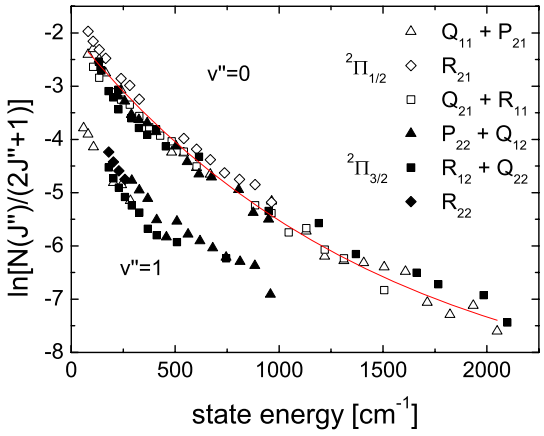

Figure 2: The rovibrational population distribution of neutral NO molecules recorde at a pump-probe delay of 24 μs (approximately 300 m/s) at a photon energy of $h\nu = 57.1$ eV.

energy in form of a Boltzmann plot. The open symbols correspond to levels in the $^2\Pi_{1/2}$ state while the filled symbols correspond to those in $^2\Pi_{3/2}$. The rotational population is highly non-thermal. Low rotational states with a rotational energy below 500 cm^{-1} are fitted by a rotational temperature

of about 290 K while higher rotational states can be fitted to a temperature of 690 K. The non-thermal rotational population yields an averaged rotational energy of $< E_{rot} >= 38.6$ meV, a vibrational energy of $< E_{vib} >= 136$ meV and an electronic excitation of $< E_{el} >= 3.9$ meV.
For a linear dependence of the number of desorbing NO molecules, N_{des}, on the desorption laser flux a desorption cross section can be derived in the form of

$$N_{des} \propto exp - \frac{\sigma N_{phot}}{A} \tag{1}$$

where σ denotes the desorption cross section, N_{pho} the number of photons, and A the aqrea of the desorption laser spot. The desorption yield recorded at 38.8 eV as a function of accumulatively applied photons is displayed in figure 3. A slightly non linear desorption yield of neutral NO is

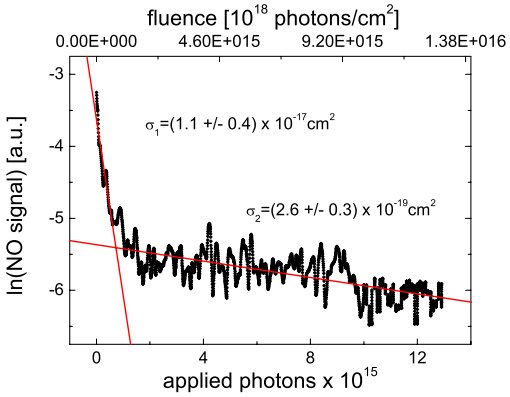

Figure 3: The desorption yield recorded on the bandhead of the P_{11} branch ($J'' = 7.5 - 10.5$) at a photon energy of $h\nu = 38.8$ eV.

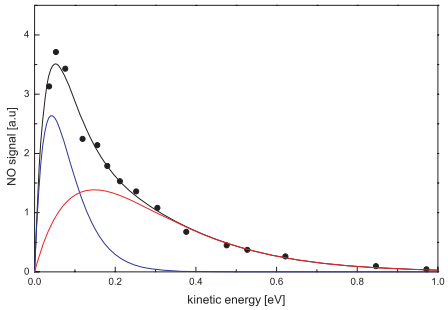

Figure 4: The kinetic energy of neutral NO melecules desorbed at $h\nu = 57.1$ eV ($^2\Pi_{3/2}$, $v'' = 0$, $J'' = 11.5$).

observed with a slope of $m = 1.4 \pm 0.2$. Linearizing the yield dependence the double exponential decay yield for low photon fluxes a desorption cross section of $\sigma_1 = (1.1 \pm 0.4) \times 10^{-17}$ cm^2 and for high photon fluxes $\sigma_2 = (2.6 \pm 0.3) \times 10^{-19}$ cm^2.
The velocity distribution shown in figure 4 yields translationally hot molecules. The distribution can be fitted by the sum of two Maxwell-Boltzmann distributions of $T = 490$ K and $T = 1720$ K. The average kinetic energy amounts to $< E_{kin} >= 170$ meV.