Tunneling ionization imaging of photoexcitation of NO by ultrafast laser pulses

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Synopsis Tunneling ionization imaging of photoexcitation of NO has been demonstrated by using ultrafast intense laser pulses (8 fs, 800 nm, 1.1×10^{14} W/cm^2). The distributions of N^+ fragments produced by dissociative ionization of NO starting from the electronically ground ($X^2\Pi$) and excited ($A^2\Sigma^+$) states are in good agreement with theoretical tunneling ionization yields based on WFAT, showing that the fragment anisotropy reflects changes in the outermost molecular orbital by photoexcitation.

Visualization of the highest occupied molecular orbitals (HOMOs) of electronically ground states has been achieved by using the fact that the tunneling ionization rate strongly depends on the molecular alignment (orientation) with respect to the laser polarization direction [1, 2]. The extension to electronically excited states is important for a real-time observation of electron dynamics during chemical reactions. Recently, we investigated the fragment distribution produced by dissociative ionization (DI) of NO, NO $\rightarrow$ $NO^+ + e^- \rightarrow N^+ + O + e^-$, in few-cycle intense laser fields (8 fs, 1.1×10^{14} W/cm^2) from the ground state ($X^2\Pi$, $2\pi^1 3s\sigma^0$) and the excited state ($A^2\Sigma^+$, $2\pi^0 3s\sigma^1$) to demonstrate the applicability of tunneling ionization imaging to the excited states [3].

The output of a Ti:Sapphire laser system (800 nm, 1 kHz) was used to obtain pump DUV pulses (226 nm) tuned to the NO A-X (0, 0) transition and probe few-cycle NIR pulses (800 nm, 8 fs) to induce DI. Those pulses with a pump-probe time delay of 150 ps were focused on the molecular beam of NO by concave mirror (f = 75 mm) in an ultrahigh vacuum chamber. The produced ions were accelerated to a position sensitive detector by four electrodes in a velocity map configuration. The three-dimensional momentum vector of respective ions was obtained from the position (x, y) and the arrival time (t) at the detector. To obtain the net signals from the excited state, an optical chopper (0.5 kHz) was introduced to block the pump pulse in every other shot.

The N^+ produced by DI from the ground state ($X^2\Pi$) via the dissociative state ($c^3\Pi$) of NO^+ show the anisotropic distribution peaked at 45° with respect to the probe laser polarization direction (Fig. 1 (a), (c)). On the other hand, the fragments produced from the excited state ($A^2\Sigma^+$) form a weak peak at 0° (Fig. 1 (b), (d)), reflecting the change in the outermost molecular orbital from 2π to $3s\sigma$. These results show a good agreement with theoretical calculation based on weak-field asymptotic theory (WFAT) [4] under the adiabatic approximation, which naturally includes the effects of the permanent dipole of a heteronuclear diatomic molecule. The present study demonstrates a readout of the electron distribution in the excited states and the change in the distribution by photoexcitation from fragment anisotropy produced by ultrafast intense laser pulses.

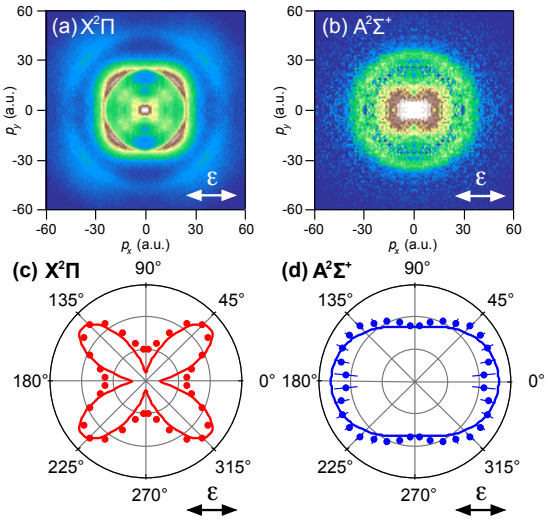

Figure 1. Momentum images of the N^+ fragments produced by DI from (a) $X^2\Pi$ and (b) $A^2\Sigma^+$ states. Polar plots of the fragment angular distributions obtained for (c) $X^2\Pi$ and (d) $A^2\Sigma^+$ states. Solid lines are calculated by WFAT under the adiabatic approximation. The probe laser polarization direction is denoted with ε.

References

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