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Fragment anisotropy on dissociative ionization of NO in two-color intense laser fields

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Synopsis Dissociative ionization of NO, NO $\rightarrow$ $NO^+ + e^- \rightarrow N^+ + O + e^-$, in ω-2ω two-color phase-controlled intense laser fields was investigated by three-dimensional ion momentum imaging. The momentum image of the N^+ ion exhibited anisotropic distributions peaked at 35° against the laser polarization, which show clear left-right asymmetry depending on the ω-2ω relative phase. The present study demonstrates that the molecular orbital can be retrieved by distinguishing the difference between the N and O side from the fragment anisotropy by intense laser pulses.

Visualization of molecular orbitals has been discussed in a number of studies in the last decades by using high-order harmonics generation [1] and Coulomb explosion [2]. The approach based on Coulomb explosion (or dissociative ionization) utilizes the fact that the spatial anisotropy of laser tunneling ionization rate in molecular frame depends on the shape of the outermost molecular orbital, which allows a direct read-out of the shape of molecular orbital from the spatial anisotropy of fragment ion. However, as demonstreted for our previous studies on NO [3], the asymmetry of molecular orbitals cannot be retrieved by a one-color laser pulse having a symmetric electric-field amplitude. To address this problem here, we employed ω-2ω two-color laser fields, where the tunneling ionization becomes an orientation selective process. In the present study, we investigate the dissociative ionization of NO, NO $\rightarrow$ $NO^+ + e^- \rightarrow N^+ + O + e^-$, in ω-2ω two-color intense laser fields using three-dimensional (3D) ion momentum imaging.

The output of a Ti:Sapphire laser system (800 nm, 50 fs) was introduced into β-BBO crystal to obtain 2ω pulses (400 nm, 150 fs). A birefringent α-BBO crystal was placed to compensate the time-delay between ω and 2ω pulses. Both pulses were passed through a dual-wavelength waveplate ($\lambda/2$ for ω, λ for 2ω) to parallel the polarization axis of two beams. The relative phase between ω and 2ω was controlled by a feedback loop using a pair of fused silica wedges. The absolute value of the relative phase was determined by energy distribution of a back-scattered electron [4]. Two-color laser pulses were focused on the molecular beam of NO using a concave mirror (f = 300 mm) in an ultrahigh vacuum chamber. The fragment ions were accelerated to a position sensitive detector by four electrodes.The position (x, y) and the flight time (t) were recorded for each ion to calculate the 3D momentum vector $\mathbf{p} = (p_x, p_y, p_z)$.

The angular distribution of N^+ ion produced by dissociative ionization via $c^3\Pi$ state in two-color intense laser fields ($I_\omega = 4.3 \times 10^{13}$ W/cm^2, $I_{2\omega} = 3.4 \times 10^{12}$ W/cm^2, $E_{2\omega}/E_\omega = 0.28$) is peaked at 35° with respect to the laser polarization direction reflecting the nodal pattern of the 2π outermost molecular orbital of NO as observed in one-color laser fields (Fig. 1). On the other hand, a clear left-right asymmetry was observed, which depends strongly on the ω-2ω relative phase. The obtained result shows that the asymmetry of fragment distribution reflects the difference between N and O sides in the molecular orbital, demonstrating that the asymmetry of the molecular orbital can be retrieved using by two-color intense laser fields.

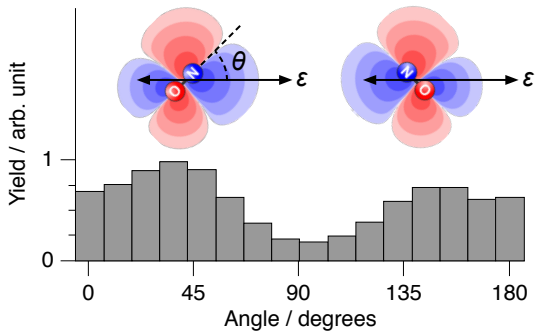

Figure 1. Angular distribution of the N^+ ion produced by dissociative ionization in two-color intense laser fields. The laser polarization denoted by ε.

References

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