Electron-ion coincidence laser tunneling ionization imaging of O₂ with auxiliary dissociation pulses

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Intense laser fields ($\sim 10^{14}$ W/cm²) distort electron binding potentials of molecules and induce tunneling ionization. Since the rate is governed by characteristics of molecules, tunneling ionization provides a unique tool to investigate molecular properties, such as distributions of electrons within molecules. Molecular frame photoelectron angular distribution (MFPAD) measurement in circularly polarized intense laser fields is a powerful method for molecular imaging using tunneling ionization [1,2]. On the other hand, since this method requires both ionization and molecular dissociation for electron-ion coincidence detection, it can only be applied to molecules susceptible to dissociation in circularly polarized laser fields. In this study, we demonstrated a new approach, where auxiliary dissociation pulses are introduced to circumvent the difficulty.

The output of Ti:Sapphire laser system (ω , 800 nm, 45 fs) was split into two beams by a beam splitter. One was converted into circularly polarized pulses by a quarter wave plate. The other was introduced into a β -BBO crystal and converted into UV pulses (2ω , 400 nm, 60 fs). The two pulses were introduced into an ultrahigh vacuum chamber and focused on O₂ by a concave mirror. The UV pulses were delayed by 80 fs and blocked every other shot by an optical chopper. The ions and photoelectrons were accelerated toward separate position sensitive detectors by a static electric field and detected in coincidence. Momenta of each particle were calculated from the time of flight (t) and detected positions (x, y).

Figure 1 shows the kinetic energy release (KER) spectra of O₂. The spectrum with the ionization pulses alone $(1.7 \times 10^{14} \text{ W/cm}^2)$ has two peaks, which are observed in the previous study [2]. On the other hand, when introducing the auxiliary pulses (2 ω on), a new peak

emerged. The net KER spectrum (purple) was peaked at 0.7 eV. Since this peak agrees with a previous study for photodissociation (395 nm) of O_2^+ beam [3], it was assigned to the dissociation pathway via ionization from HOMO-1 (π_u). The corresponding MFPAD shows a larger distribution in perpendicular to the molecular axis, supporting the assignment.

^[3] M. Zohrabi et al., Phys. Rev. A 83, 053405 (2011).



Fig. 1: The kinetic energy release spectra of O_2 obtained with 2ω pulse on (green), 2ω pulse off (red), and the net signal (purple).

^[1] A. Staudte et al., Phys. Rev. Lett. 103, 033004 (2009).

^[2] H. Liu et al., Phys. Rev. A 88, 061401(R) (2013).