Strong-field physics in the molecular frame

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Abstract: Laser-aligned carbonyl-sulfide molecules were strong-field ionized by using mid-infrared light. Investigating the strong-field effects in the molecular frame allowed to add novel facets to the understanding of the intrinsic nature of strong-field physics.

1. Introduction

The interaction of strong laser fields with matter intrinsically enables the imaging of transient dynamics with extremely high spatiotemporal resolution [1]. This paradigm of photophysics has grown into new emerging research areas, ranging from attosecond science to laser-induced electron diffraction, providing new insight into atoms, molecules and, more recently, condensed matter. Also, the earliest moments of strong-field interactions have attracted attention for capturing the intrinsic nature of strong-field physics. While pioneering attosecond science experiments and molecular-frame measurements [2,3] revealed non-trivial spatiotemporal features in electron tunneling, these initial conditions are generally considered a weak perturbation. Here, we investigated strong-field ionization of strongly aligned molecules. We demonstrated that the electron dynamics is properly understood only in the molecular frame. Our findings have strong impact on the interpretation of self-diffraction experiments, where the photoelectron momentum distribution is used to retrieve molecular structures. Furthermore, we extracted the encoding of the time-energy relation in molecular-frame photoelectron distributions, which provided a new way of probing the molecular potential with sub-femtosecond resolution and accessing a deeper understanding of electron tunnelling.

2. Results and discussion

Fig.1 shows the experiment. An ultracold ensemble of carbonyl sulfide (OCS) molecules was adiabatically aligned in the laboratory frame, with $< cos^2\theta_{2D} >= 0.9$, by using a linearly polarized, 500 ps laser pulse, centered at 800 nm.

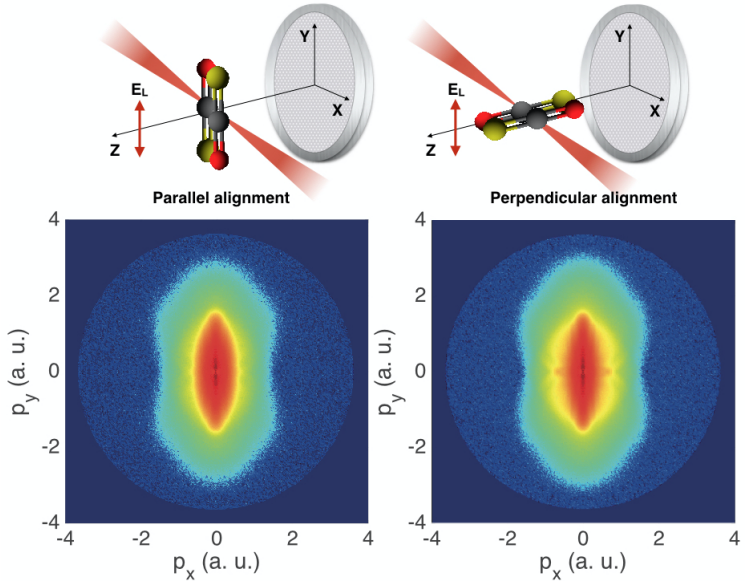

Fig.1: Sketch of the experimental arrangement with alignment of OCS molecules in the laboratory frame, parallel and perpendicular to the Y axis. The ionizing laser is linearly polarized along the Y axis and the detection is in the XY plane. The molecular-frame angle-resolved photoelectron spectra are projected onto a 2D detector in a velocity map imaging spectrometer.

The molecules were aligned in two different configurations, shown in Fig.1, with the molecular axis along the Y and Z axes, named parallel and perpendicular alignment, respectively. A second laser pulse, centered at 1300 nm, with a duration of 65 fs, and a peak intensity of $8 \cdot 10^{13}$ W/cm^2, was used to singly ionize the OCS molecules. The ionizing laser pulse, $\mathbf{E}_L$ in Fig.1, was linearly polarized along the Y axis. The produced molecular-frame angle-resolved photoelectron spectra (MF-ARPES) were recorded in a velocity map imaging (VMI) spectrometer with its detector parallel to the XY plane.

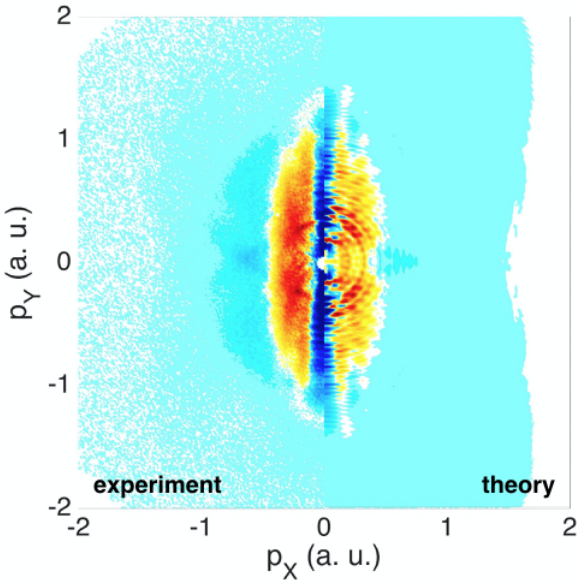

Fig.2: Measured (left) and computed (right) differential momentum distribution (parallel − perpendicular), respectively.

We performed a differential analysis by subtracting the two experimental spectra from each other. In Fig.2 (left half) the relative normalized difference, parallel minus perpendicular, is reported. A strong depletion along the vertical axis and two transversely offset broad lines of positive yield appear as main features. The depletion along the centerline is due to the node along the molecular axis of the degenerate Π highest occupied molecular orbital (HOMO) of OCS; it represents a forbidden direction of electron ejection [4]. Here, the node and the transverse momentum were captured with unprecedented resolution. To unravel the experimental observations, state-of-the-art calculations were performed using time-dependent density-functional theory (TDDFT). The right half of Fig.2 shows the normalized difference for the computed spectra, in really good agreement with the experimental data. In our work we used the result depicted in Fig.2 as a starting point to investigate the electron dynamics following the strong-field interaction. To do this, we discussed the seminal statements of strong-field physics by introducing an extended simple-man model.

Our result has a strong impact in the interpretation of self diffraction experiments and, in principle, allows one to achieve a deeper understanding of electron tunnelling.

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