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# Resolving the dynamics of valence-shell electrons and nuclei through laser-induced diffraction and holography

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**Synopsis** We have studied a coupled electronic-nuclear wave packet in nitric oxide using time-resolved strong-field photoelectron holography and rescattering. We show that the electronic dynamics mainly appears in the holographic structures whereas nuclear motion strongly modulates the angular distribution of the rescattered photoelectrons.

The interaction of intense laser pulses with molecules has opened several new avenues for imaging ultrafast dynamics. Strong-field photoelectron holography has been used to measure static atoms [1, 2] and molecules [3] whereas rescattering has additionally revealed first evidence of nuclear motion [4]. Here, we transpose the powerful methods of photoelectron holography and rescattering to observing electronic dynamics and its coupling to nuclear dynamics.

First, we show that the electronic dynamics modulate the contrast of the holographic pattern which is found to probe the lateral-momentum distribution of the photoelectron wave packet at the instant of ionization. Second, the electronic dynamics is shown to neither modify the structure of the holographic pattern nor the angular distribution of the rescattered electrons, providing a clean separation of nuclear dynamics occurring simultaneously to the electronic dynamics. Third, owing to this separation, the nuclear dynamics is recorded by rescattering in a background-free manner.

We use impulsive stimulated Raman scattering (ISRS) to prepare a coupled electronic and rotational wave packet in the neutral NO molecule. The wave packet is probed by strong-field ionization and the photoions and photoelectrons are detected by a velocity-map-imaging spectrometer. An overview of the experimental results is given in Fig. 1. Figure 1a shows the normalized $NO^+$ yield as a function of the pump-probe delay. The rapid regular oscillation originates from the electronic wave packet whereas the two features around 5 and 10 ps correspond to the quarter and half rotational revivals, respectively. Figure 1b shows a cut through the three-dimensional photoelectron momentum distribution recorded at a delay of 1.56 ps. Figures 1c and 1d show normalized differences of photoelectron momentum distributions recorded at maxima and minima of the photoelectron (or, equivalently, $NO^+$) yield. The electronic dynamics (Fig. 1c) appears as a pronounced modulation of a holographic pattern whereas the nuclear dynamics manifests itself by modifying the angular distribution of the rescattered photoelectrons.

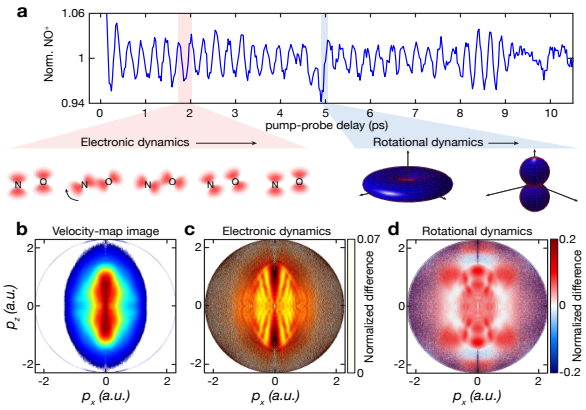


**Figure 1. Resolving electronic and nuclear dynamics through holography and rescattering.** (**a**) Time-dependent normalized $NO^+$ signal and illustration of the prepared electronic and rotational dynamics, (**b**) photoelectron momentum distribution of excited NO molecules recorded at a delay of 1.56 ps, (**c**) normalized difference of momentum distributions at maxima and minima of the $NO^+$ signal dominated by electronic dynamics (1.55 and 1.72 ps, respectively), (**d**) normalized difference of momentum distributions at maxima and minima of the $NO^+$ signal dominated by rotational dynamics (4.92 and 1.50 ps, respectively).

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