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Probing the interplay between electron and nuclear dynamics at attosecond timescale

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The development of attosecond photoelectron spectroscopy based on two-color (*XUV* or, extreme-ultraviolet and *NIR*, or, near-infrared) interferometric techniques [1] has enabled studying the dispersion dynamics of electron wave-packets in atoms [2], molecules [3] and solids [4]. In a recent experiment, we have demonstrated that it is possible to simultaneously combine high spectral and temporal resolution [5], thus paving the path towards probing fundamental processes in molecular systems where the electronic motion can be coupled to the nuclear degrees of freedom following photoionization. One manifestation of such an interplay between the electron and nuclear dynamics can be visible in a shape resonance which appears as a strong photon energy dependence of the vibrational branching ratios, a direct departure from the Franck-Condon principle [6].

Here, we investigate the shape resonance in photoionization of the nitrogen (N_2) molecule at attosecond timescale. We performed an interferometric measurement in the region of the shape resonance, with photon energies ranging from 25 to 40 eV. A magnetic bottle electron spectrometer was used to detect the photoelectrons in a vibrationally resolved manner from both the X- and A-states in the N_2^+ ion. As the delay between the XUV and NIR pulses was varied, the amplitude of the sidebands (XUV±NIR) from two adjacent harmonics oscillated. The variations in amplitude of the sidebands, in particular, their relative phase-shifts were used to extract the photoionization time-delays. The measured photoionization time-delay differences between $v = 0$ and $v = 1$ vibrational levels for the X-state are found to change drastically as a function of the photon energy, compared to those for the A-state. Unlike in previous studies close to the ionization threshold of N_2 [7], in our case, following photoionization at higher energies, the departure of the photoelectron can be hindered due to the existence of a centrifugal barrier in the effective molecular potential. It leads to the photoelectron being temporarily trapped close to the ionic core. In this case, the electronic and vibrational modes in the molecule can no longer be considered separate from each other. Our study provides a direct way to probe the timescale at which this coupling between the electronic and nuclear degrees of freedom can occur within a molecular system.

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