Observing the Elusive Double-Peak Structure in R-dependent Tunneling Ionization Rate of Hydrogen Molecular Ion

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Abstract: We performed pump-probe experiment on H_2 using intense few-cycle laser pulses and Reaction Microscope detection apparatus. We observe the theoretically predicted [1] double-peak structure in R-dependent tunneling ionization rate for the first time experimentally.

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Tunneling ionization of hydrogen molecular ion by intense femtosecond near-infrared laser pulses exhibits strong dependence on internuclear separation R. It has been shown that rate of such ionization increases dramatically when R reaches certain critical distance R_c (~ 7 atomic units). This phenomenon is known as *enhanced ionization* [2] and it explains kinetic energy release (KER) spectra measured in Coulomb explosion of H_2 by femtosecond laser pulses: that kinetic energy reflects the internuclear distance where the second ionization takes place (R_c).

In terms of physical mechanism, enhanced ionization is explained by appearance of charge-resonance states in field-distorted double-Coulomb-well potential of H_2^+, and therefore it is often referred to as charge-resonance-enhanced ionization or CREI [1]. Similar mechanism is thought to be operational also in larger diatomic, triatomic and even polyatomic molecules.

Theoretical simulations predict that R-dependent ionization rate for H_2^+ should exhibit two well-defined maxima – one at around 7 au and another at around 11 au [1,3]. However, only the first maximum manifests itself in experimental KER spectra. The second maximum was never observed experimentally and various explanations were advanced to explain its elusiveness [4].

Here we present for the first time an experimental observation of full double-peak structure of R-dependent ionization rate for H_2^+, including well-defined second maximum at 12 au. To observe this structure we performed pump-probe measurements on neutral hydrogen molecules using two 6 fs (750 nm central wavelength) laser pulses. Following initial ionization by the pump, we traced KER spectra for the double-proton coincidence channel as a function of pump-probe delay. The experimental time-dependent ionization yield displays two clear maxima at 14 and 22 fs (Figure 1). By modeling dynamics of nuclear wavepackets in H_2^+ we determine that those maxima correspond to internuclear separations of 7 and 12 au in excellent agreement with theoretical predictions.

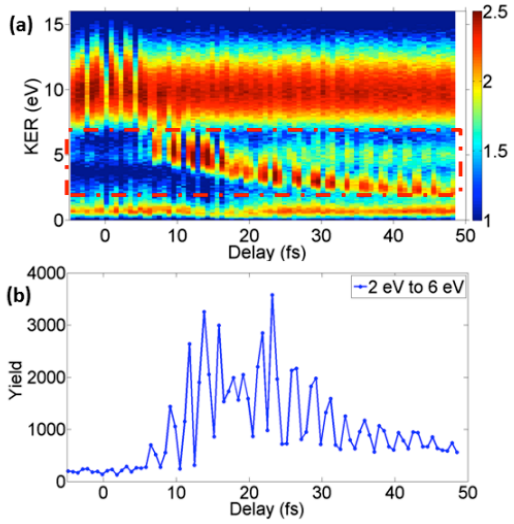

Figure 1. Time-dependent KER spectra (top) and double-proton coincidence integrated yield (bottom) following ionization of H_2 by pump pulse.

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