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Strong-Field Enhanced Ionization of Water using 6-fs Pulse Pairs

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Abstract: We demonstrate an enhancement in the formation of D_2O^{3+} as a consequence of field-free molecular dynamics following the strong-field multiple ionization of deuterated water via 6-fs 800-nm pulse pairs.

1. Introduction

A significant enhancement exists in the strong-field multiple ionization of water that allows more frequent formation of the trication state (H_2O^{3+}) at certain internuclear geometries [1-3]. This enhancement has been observed to positively correlate with the duration of the ionizing pulse [4]. Models show that longer pulse durations allow the lower charge states (H_2O^+ and H_2O^{2+}) more time to distort under the influence of the strong laser field and reach the critical internuclear geometry necessary to form the trication [5]. Here we find that similar bond distortions occur due to *field-free* interatomic motion following formation of the water dictation (H_2O^{2+}), leading to geometries that favor enhanced trication formation.

2. Experimental Design

These experiments were conducted via a two-step pump-probe ionization process: one 800-nm pulse of 6-fs duration (the pump pulse) doubly ionizes one of the three stable isotopes of water (H_2O/D_2O/HOD), the resulting dictation then evolves without the influence of an external electric field, until a second pulse (the probe pulse), identical to the first, performs the final ionization step to form the trication. After formation of the trication, the molecule undergoes a Coulomb explosion, and the full three-dimensional (3D) momentum of each fragment is measured in coincidence. This measurement allows for the reconstruction of the molecule's internuclear geometry prior to Coulomb explosion. The time delay between the pump and probe pulse was varied in ~1-fs increments over a range of ~110-fs to yield a delay-dependent 3D momentum distribution of the trication. This scheme effectively samples the field-free dynamics of the water dication that occur within the time interval between formation of the dication and formation of the trication. The time-dependent trication yield and 3D momentum distributions together provide detailed insight on the exact conditions necessary to achieve enhanced ionization.

3. Results and Discussion

For the case of D_2O, a clear enhancement in trication formation is observed at pump-probe delays of approximately 20 fs, as seen in the time-resolved trication yield plotted in Fig. 1(a). This enhancement occurs briefly while no less than two distinct internuclear distortions evolve within the molecule: (1) the OD bonds stretch as a function of pump-probe delay to yield a monotonically decreasing kinetic energy release (KER) as seen in Fig. 1(b), and (2) the initially unbent (linear) molecule begins to bend back toward more acute HOH-angles, as seen in Fig. 1(c). These two distortions provide a valuable means of discriminating what conditions are necessary to achieve an enhancement in the production of the water trication. As they develop following field-free dynamics, these results differ significantly from prior understanding of the strong-field enhanced ionization of water. This new understanding may inform novel insights into the strong-field enhanced ionization of larger molecules.

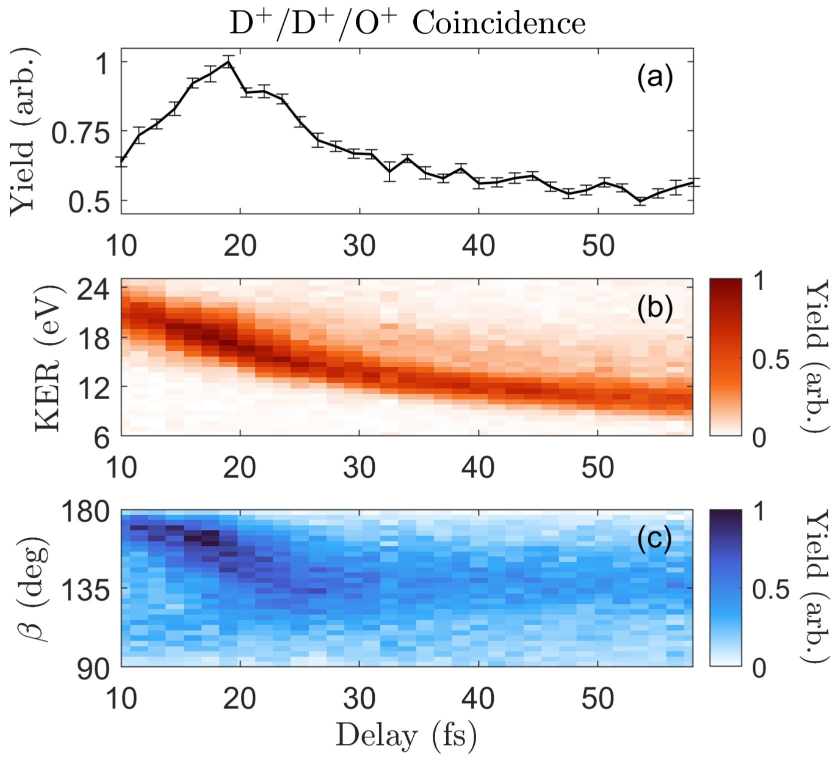

Figure 1: (a) The total yield of $D^+/D^+/O^+$ ions detected in coincidence following the Coulomb explosion of D_2O^{3+} as a function of pump-probe delay. Note the enhancement in yield at a delay of ~20 fs. (b) The total kinetic energy release (KER) in each $D^+/D^+/O^+$ coincidence as a function of pump-probe delay. Note that the mean of the distribution decreases monotonically over delay. (c) The delay-resolved momentum-frame bend-angle, as defined by $\beta = \arccos(\boldsymbol{p}_{\mathbf{D}}^{(1)} \cdot \boldsymbol{p}_{\mathbf{D}}^{(2)})$ where $\boldsymbol{p}_{\mathbf{D+}}^{(1)}$ and $\boldsymbol{p}_{\mathbf{D+}}^{(2)}$ are the 3D momenta of the two deuterium ions caught in each $D^+/D^+/O^+$ coincidence. Note that the majority of molecules are unbent ($\beta \sim 180°$) at early delays (< 20 fs).

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