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Carrier-envelope phase control over fragmentation of H_2^+ and D_2

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Synopsis We demonstrate control over fragmentation of H_2^+ and D_2 molecules via the carrier-envelope phase of sub-5 fs laser pulses. Moreover, we attribute our findings to interferences between different pathways involving different net numbers of photons [1], revealing "high-order" pathways and the importance of the bandwidth.

The carrier-envelope phase (CEP) of ultrashort laser pulses is a known control knob for molecular fragmentation [2]. Recently, we performed a detailed study of CEP dependences in H_2^+ dissociation [3] and Rydberg D^* production upon fragmentation of D_2.

In our first experiment, an H_2^+ ion-beam target, produced in an ion source, interacts with sub-5 fs laser pulses. The CEP of every laser shot is tagged using a stereo-ATI CEP meter [4]. We observe CEP-dependent asymmetries in the emission direction of H^+ fragments relative to the laser polarization. The CEP dependence of these asymmetries match that predicted by the general CEP theory [1] and are due to interference of odd and even photon number pathways. To further our understanding, the measurements are quantitatively compared to state-of-the-art *ab initio* calculations that include nuclear rotation and intensity averaging [3].

In the second experiment, the CEP dependence in the fragmentation of D_2 molecules into D^* has been studied using ultrashort linearly-polarized laser pulses. The total dissociation yield of D^* fragments exhibits an oscillatory behavior as a function of CEP, as shown in Fig. 1(a). These oscillations result from interference between photon pathways differing by net two photons (Δn=2). The difference in the emission direction (asymmetry) of these D^* fragments along the laser polarization, shown in Fig. 1(b), reveals a similar CEP dependence to those seen in H_2^+ [3]. It is noted that these CEP dependences vary with the specific band of excited states of the D^* fragments that were measured.

The asymmetry map possesses higher-order oscillations related to a higher net number of photons involved in the interference pathways. We employ a Fourier analysis method to extract information linked to these pathways, as shown in Fig. 1(c). Lastly, for the first time, the amplitude and phase of "higher-order" pathways, specifically those differing by net three photons, are revealed and plotted as a function of kinetic-energy release (KER) in Fig. 1(d).

The broad spectral bandwidth of the laser pulses plays a crucial rule in these CEP dependences. This importance is demonstrated by utilizing chirped pulses of the same spectral bandwidth as our 5 fs FTL laser pulses. Clear CEP dependences persist for pulses chirped up to 20 fs.

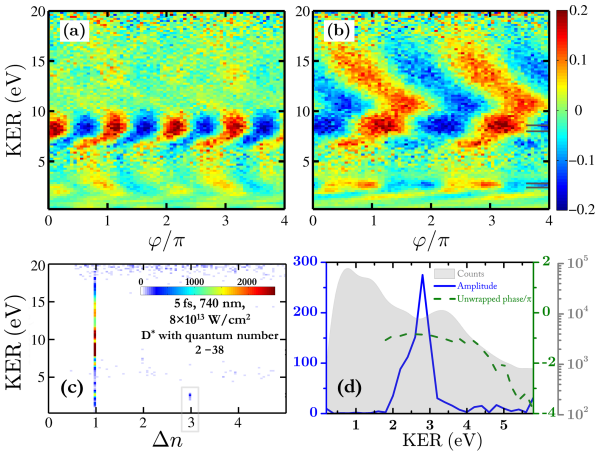

Figure 1. D^* fragment (a) normalized yield and (b) asymmetry as a function of CEP and KER. (c) Fourier transform of the asymmetry map shown in (b) as a function of Δn and KER. (d) Amplitude and phase extracted from (c) for Δn=3 (Gray: the D^* yield averaged over CEP) as a function of KER.

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