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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 ($\Delta n=2$). 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 am-

plitude 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.

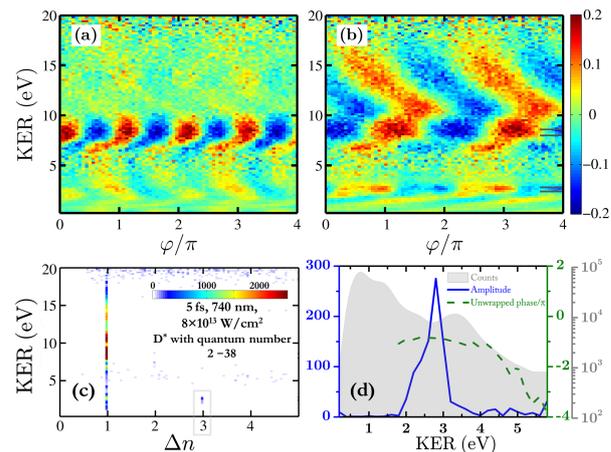


Figure 1. D^* fragment (a) normalized yield and (b) asymmetry as a function of CEP and KER. (c) Fourier transform of the asymmetry map shown in (b) as a function of Δn and KER. (d) Amplitude and phase extracted from (c) for $\Delta n=3$ (Gray: the D^* yield averaged over CEP) as a function of KER. This work was supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. BJ is also supported by DOE-SCGF.

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