Supplemental Material: Coherent control of internal conversion in strong field molecular ionization

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PHOTOELECTRON SPECTRA FOR DIFFERENT PULSE DURATIONS

Fig. 1 shows the photoelectron spectrum measured for CH_2IBr with a short ($\sim$ 10 fs) and long (>30 fs) pulse. The figure illustrates the fact that for a short pulse, the ionization is dominated by D_1 due to resonant enhancement through R_1. However, due to some mixing between R_1 and R_3 at the Franck Condon point, there is some ionization to D_3. This is in contrast to the measurements for a long pulse, which show ionization to D_0, D_3 and D_1, as a result of non-adiabatic dynamics which couple states R_1, R_3 and R_0 [1].
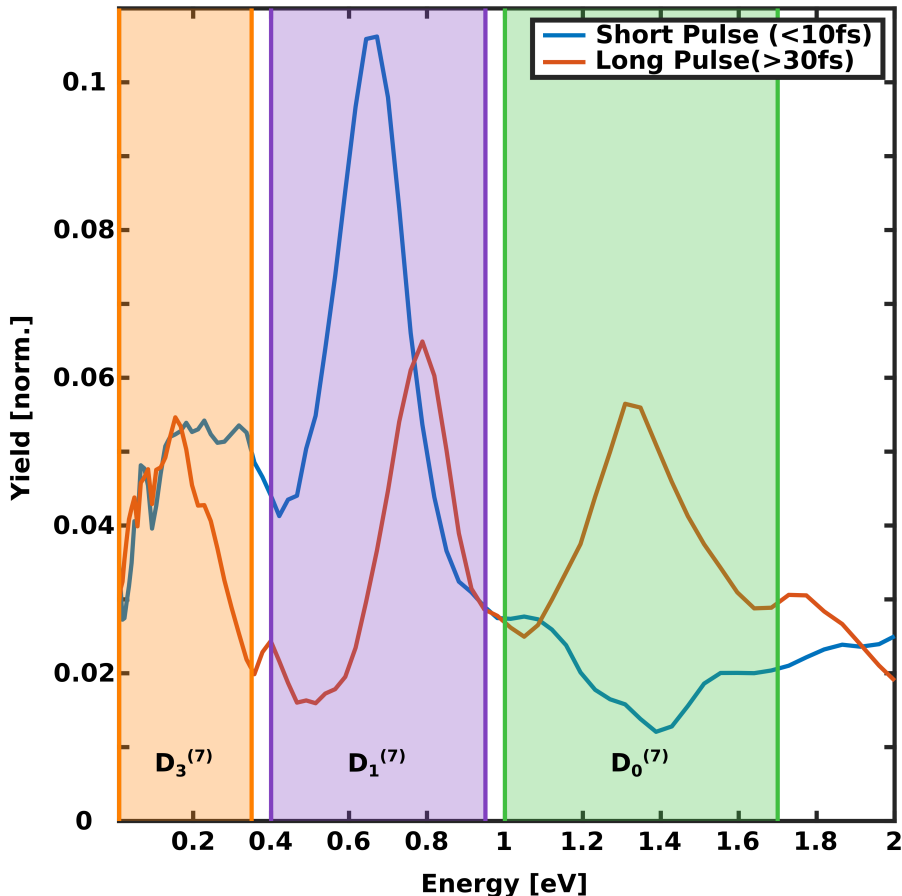
FIG. 1. CH_2IBr photoelectron yield for two different pulse durations normalized to the total number of detected electrons. The three peaks are assigned to ionic states D_3 (orange region), D_1 (purple region), and D_0 (green region) based upon earlier ion-electron coincidence measurements [2].

PHASE SCANS WITH AND WITHOUT INTENSITY COMPENSATION

In this work, we measured the photoelectron yield as a function of pump-probe phase and explored the impact of the pump-probe delay. As mentioned in the main text, the intensity fluctuates as a function of both pump-probe phase and delay which we can compensate for using our pulse shaper. Phase scans with and without intensity compensation are compared in Fig. 2 below. As can be seen from the figure, the phase dependence of the yield for the two ionic states is similar with and without intensity scaling, indicating that the variation in phase is not a simple consequence of the intensity variation.
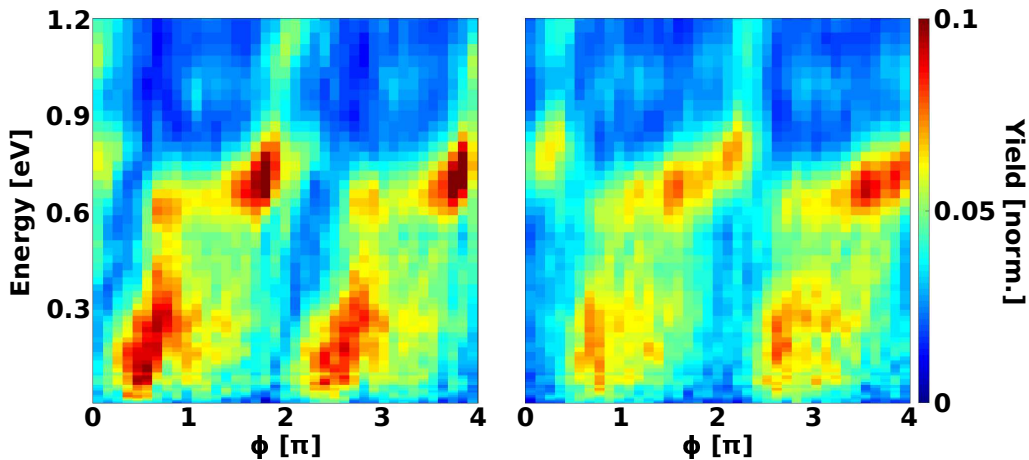

FIG. 2. CH_2IBr photoelectron yield as a function of photoelectron energy and pump-probe phase at a 15fs pump-probe delay. Left figure: no compensation for phase dependent intensity variation. Right figure: intensity variation is compensated, i.e., constant intensity.

SCAN OF PHASELOCKING FREQUENCY

In the main text, we fixed the laser locking frequency to the central wavelength of the laser spectrum, 750 nm or 2.51 rads/fs. Here we investigate the effect of varying the locking frequency by measuring the photoelectron yield as a function of relative pump-probe phase at different locking frequencies. We then construct lineouts of the state yield vs. pump-probe phase and extract the phase offset.

Fig. 3 shows the phase offset for states D_1 (purple) and D_3 (orange) as a function of laser locking frequency at fixed delay $\tau = -15$ fs for CH_2IBr. The measurements are fit to a line of the form:

$$\phi_{offset} = -\tau(\omega_L - \omega_0) + \phi_{molecule} \tag{1}$$

Here ω_0 is the central laser frequency and $\phi_{molecule}$ is the molecular contribution to the measured phase offset.
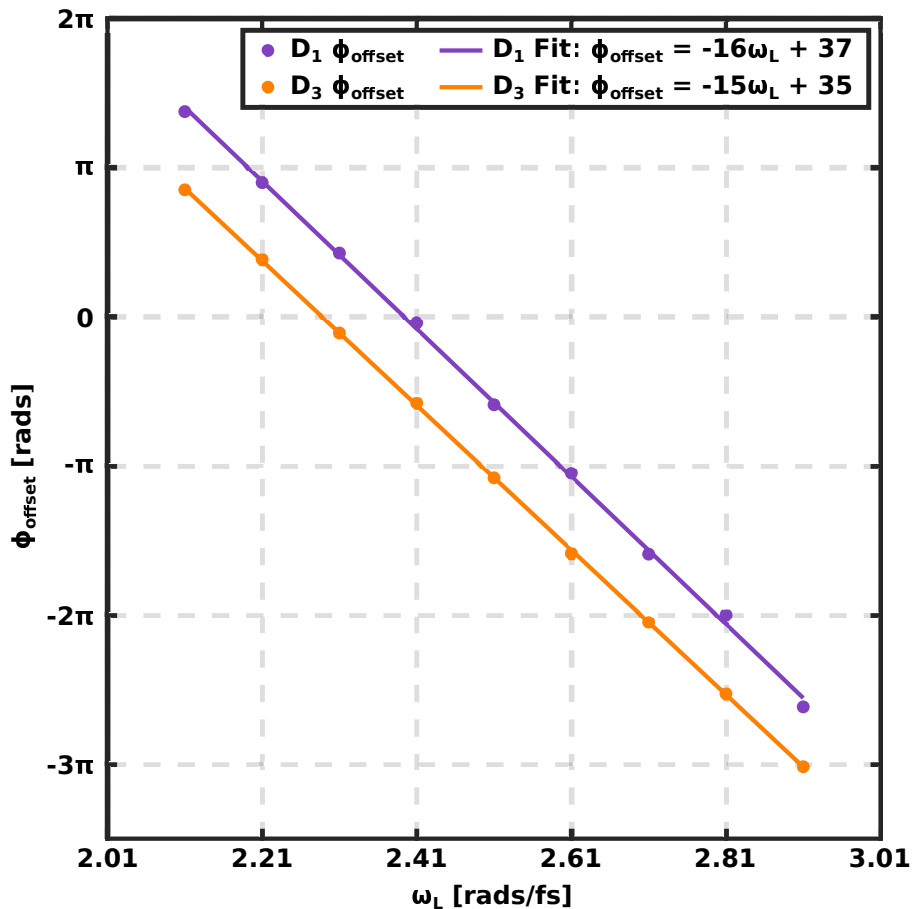

FIG. 3. Phase offset, ϕ_{offset}, as a function of laser locking frequency, ω_L, measured for a pump-probe delay of -15 fs for CH_2IBr. The phase offset is fit to a line with the fit parameters displayed in the legend.

[1] P. Sándor, V. Tagliamonti, A. Zhao, T. Rozgonyi, M. Ruckenbauer, P. Marquetand, and T. Weinacht, Phys. Rev. Lett. **116**, 063002 (2016).
[2] P. Sándor, A. Zhao, T. Rozgonyi, and T. Weinacht, J. Phys. B **47**, 124021 (2014).