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Probing proton dynamics in molecules on an attosecond timescale

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A new technique for probing the ultrafast structural rearrangements of light molecules following ionization was demonstrated by our group earlier this year. This technique, termed PACER (**p**robing **a**ttosecond **d**ynamics by **c**hirp **e**ncoded **r**ecollision), interrogates the motion of intramolecular nuclei following ionization via the process of high-harmonic generation (HHG). The strength of harmonic emission on return of the continuum electron wavepacket is weaker the more nuclear motion has occurred in the short time window since ionization. Moreover, since different harmonic orders are emitted at different times, dynamical information is gained by simply recording an harmonic spectrum and examining the change in signal as a function of order (which maps directly to time). Previously the nuclear dynamics of H_2^+ and D_2^+ were reliably determined at a temporal resolution limited by the difference in emission time of successive harmonic orders (roughly 100 as). The time window accessed in the measurement was 0.9 – 1.5 fs following ionization. Enlargement of this time window would be a valuable extension to the PACER technique. To this end, we have now performed a PACER measurement in H_2 and D_2 at longer pump wavelengths (increasing the average electron return time). This work was conducted at the Advanced Laser Light Source facility, using a HE-TOPAS system producing 110 fs pulses in the mid-IR. We observed multiple harmonic orders at pump wavelengths of 1300 nm and 1450 nm using on-target intensities of $< 1 \times 10^{14}$ Wcm^{-2}. Data at the two pump wavelengths was found to be in qualitative agreement with the known nuclear dynamics of the H_2^+ and D_2^+ions to a time 2.1 fs after ionisation. Extension of the time window accessed by the PACER measurement is therefore promising, however, at the pulse durations employed we expect partial alignment of the molecules during the pulse, and therefore the effect of two-centre interference must be carefully examined.