Sub-Cycle Strong-Field Influences in X-ray Photoionization

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Abstract: We report the first evidence for strong field laser effects in x-ray ionization at LCLS. Experiments using a strong field optical-frequency laser and x-rays have revealed asymmetries in N_2 fragmentation patterns that indicate sub-cycle dynamics

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X-ray/Optical laser pump-probe experiments were carried out in 2009 by the AMO 02709 collaboration at the LCLS x-ray free electron laser at the SLAC National Accelerator Laboratory to probe laser-aligned nitrogen with x rays [1,2]. Intrinsic timing jitter between the x-ray and optical laser pulses prevented direct measurements of the x-ray pulse shape distributions, which were expected to have some pulses shorter than a single optical-frequency laser cycle [2, 3]. We have used a manifold embedding analysis to re-analyze some of the data collected by the AMO 02709 team. Our analysis reveals dissociation patterns with strong asymmetries, indicating sub-cycle x-ray ionization of molecules distorted by the strong laser field.

The experiment was conducted at the LCLS using 1 keV x-rays produced by 20 pC electron bunches (~1-3 fs duration) [1, 2]. An 800 nm optical laser was used to impulsively align an ensemble of ~20K molecular nitrogen, and the relative time of arrival between the x-rays and the optical laser was varied. The ion fragments, about one hundred per x-ray pulse, were collected using an ion time of flight spectrometer (iToF) working in a Wiley McLaren geometry. In the experiments relevant to this study, the arrival time of the laser was scanned over the x-ray arrival time and the ion fragments were collected and recorded for every x-ray laser shot. The ion time of flight traces were subsequently analyzed using a manifold embedding technique that found five reaction channels (RC), each of which had a unique characteristic iToF spectrum [4].

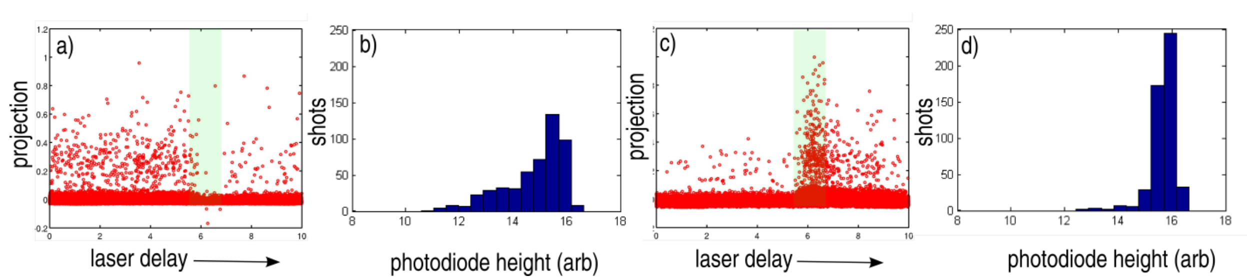

Fig. 1 Characteristics of two Reaction Channels (a) Scatter plot of the relative projection of a reaction channel vs. time where the shaded region indicates time of x-ray/optical time overlap. Larger projections mean more representative spectra were found at those times. (b). Five hundred shot histogram of laser power for a channel that does not correspond to laser power.; (c) Scatter plot of relative projection of another reaction channel vs. time that is more active in the time overlap region (d) Five hundred shot histogram of laser power for this channel shows dependence on laser energy.

As shown in Figure 1(a), one of the channels is composed primarily from data that come when the x-rays preceded the optical laser in time. This suggests that this component primarily contains dynamics that were initiated from x-rays alone and this is confirmed by the relative insensitivity of this component to the optical laser energy as shown in Figure 1(b). In contrast, Figure 1(c) shows a representative channel that is composed of data near x-ray laser time overlap. The data in Figure 1(d) confirms that these RCs are clearly dependent on the magnitude of the optical laser power, and thus possibly contain information about coherent x-ray and optical laser interactions. Figure 2 contains averaged time of flight data composed of individual iToF traces that were found to largely correspond to two different RCs that were active in the time overlap region. The traces contain parent ion peaks that each possess two corresponding side peaks that are composed of ions emitted towards the detector (shorter flight times) and ions emitted away from the detector (longer flight times). Comparing the two channels it is readily apparent that each channel largely consists of ion fragments moving in opposite directions. It has been known for some time that the sub-cycle electric field plays a strong role in atomic and molecular ionization [5,6]. This suggests that the effects of the strong field laser is likely responsible for the observed directional asymmetry in this experiment.

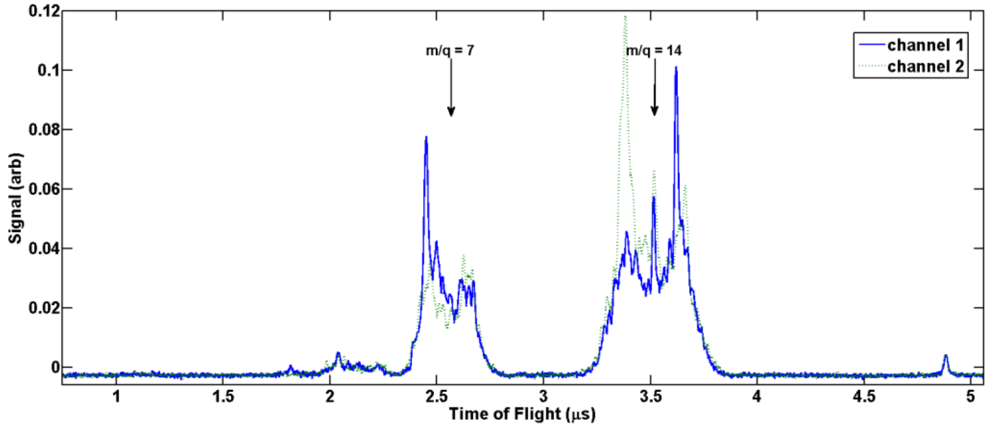

Fig. 2 Averaged IToF spectra that are representative of two reaction channels.

In the experiment, the core excited molecules Auger decayed at an exponential rate with a characteristic decay time of ~8fs. The Auger lifetime is long in comparison to the ~2.7 fs optical cycle period of the strong field laser, and it would mean that any auger decay processes involving many molecules would average over several optical cycles and show little sub-cycle optical field dependence in the resulting ion fragments. Thus, the observed asymmetric dissociation patterns likely occur during the initial x-ray ionization step.

We propose two possible x-ray/optical laser ionization schemes that could exhibit sub-cycle dependence. In one case an atomic site of N_2 is core excited by the short x-ray field in the presence of the strong field optical laser. It is possible that the combined optical and x-ray field can also allow for valence ionization to a dissociative state and the direction of the molecular fragments would depend on the instantaneous field of the optical laser. In another case the field of strong field laser can polarize the valence electron density of the molecule. As the molecule is core excited by the short x-ray pulses a shake event can happen where in addition to ionizing a core electron the x-rays can also excite or ionize a valence electron. This effect is more likely to happen where there is a higher electron density formed by the optical field, and this can also help explain the observed asymmetry. Data analysis is ongoing and other process may contribute

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References

[1] J. P. Cryan, J. M. Glownia et *al.* Phys. Rev. Lett. **105**, 083004 (2010)

[2] J. M. Glownia, J. Cryan, *et al.* Opt. Express **18**, 17620 (2010)

[3] Y. Ding. *et al*. Phys. Rev. Lett. **102**, 254801 (2009)

[4] P. Schwander, C. H. Yoon, A. Ourmazd, and D.Giannakis, arXiv:1109.5286v1 (2011)

[5] T. Seideman, M. Ivanov, and P. Corkum, Phys. Rev. Lett. **75**, 2819 (1995)

[6] G. Paulus et al. Nature **414**, 182 (2001)