**Ultrafast Laser Control of Interatomic-Coulomb-Decay Dynamics**

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Ultrafast Interatomic-Coulomb-Decay (ICD) processes play an important role during an interaction of X-rays and biological samples. Here, we present the first successful experimental demonstration that enables, and precisely times the outcome of an ICD process in an argon dimer, utilizing ultrafast XUV and IR radiation [Fig. 1]. Since the experimental work first identifying the ICD process a decade ago, extensive studies have unraveled its complex character [1-3]. Following X-ray ionization, an ultrafast interaction of the irradiated site with its environment produces a low-energy ICD electron that follows the initial direct-ionized electron. In contrast to the better-known Auger relaxation process where the electron that fills the X-ray ionization hole originates from the same atom, and the decay rate is essentially independent of environment, ICD is intrinsically complex in nature and the low energy electron (LEE) exhibits a broad energy spectrum due to the coupling of the ICD rate with the motion of the molecular nuclear wave-packet. This provides the opportunity for control over the energy and ICD electron ejection site —which could, for example, lead to new approaches for radiation therapy. To develop a better understanding of the potential control over core-relaxation processes, in past work resonant Auger decay was used as an intermediate step and a precursor to ICD. This work allowed us to demonstrate a laser-enabled ICD (LE-ICD) process-- a sophisticated control mechanism over the last step of the relaxation mechanism, involving a valence, Rydberg, and an inner-valence electron transition, allowing us to control bond breaking in the Ar dimer.



Figure 1. Schematically shown the laser-enabled control of ultrafast ICD process in Ar dimer.

REFERENCES

[1] L. S. Cederbaum, J. Zobeley and F. Tarantelli, Phys. Rev. Lett. 79, 4778–4781 (1997).

[2] N. Sisourat, N. Kryzhevoi, P. Kolorenc, et al., Nature Physics 6, 508–511 (2010).

[3] K. Nagaya, D. Lablonskyi, N. V. Golubev, et al., Nat. Communications **7,** 13477 (2016).