Observation and coherent control of single-photon laser-enabled Auger decay using the free-electron laser FERMI

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Synopsis The correlation-driven ultrafast dynamics of holes created through light-matter interaction is central to a variety of physical, chemical, and biological processes. The theoretically predicted process of single-photon laser enabled Auger decay (spLEAD) is one whose magnitude depends on the extent of electron correlation. Here, we report the first observation of spLEAD in Neon by highly sensitive homodyne spectroscopy with coherent, bichromatic free-electron laser pulses. At the same time, we demonstrate coherent control over the angular distribution of the emitted electrons. The experimental results are well supported by the theoretical modeling.

The Auger process has played an important part in modern physics, particularly surface science, because it is by far the strongest decay channel for core holes of light elements. However, it is forbidden when the energy of the final dication state is higher than that of the initial ion.

It was recently discovered that such states could decay through a novel process called laser enabled Auger decay (LEAD) [1], by absorbing multiple photons from a strong infrared field. Another class of LEAD, single-photon LEAD (spLEAD), has been theoretically predicted [2] but so far never observed. spLEAD is allowed only in systems with electron correlation. Therefore, it can potentially provide much new information such as correlation-induced ultrafast charge migration in molecules [2].

In this work, we report the first observation of spLEAD, and moreover we show that the emission can be coherently controlled. The spLEAD takes place for the 2s-hole state in Ne^+:

$$2s^1 2p^6 + \omega \rightarrow 2s^2 2p^4 + e^-, \quad \text{to give } {}^1\mathrm{S}, {}^1\mathrm{D}. \quad (1)$$

The 2s-hole state is prepared by absorption of a single photon by the ionic ground state (thus, ω is tuned to the 2s-2p transition energy 26.9 eV):

$$2s^2 2p^5 + \omega \rightarrow 2s^1 2p^6. \quad (2)$$

At the same time, we directly ionize the ground state Ne^+ by the second harmonic (2ω):

$$2s^2 2p^5 + 2\omega \rightarrow 2s^2 2p^4 + e^-, \text{ to give } {}^1\mathrm{S}, {}^1\mathrm{D}, {}^3\mathrm{P}. \quad (3)$$

We used the FERMI light source to produce intense bichromatic radiation with controllable phase [3]. We have detected the inteference between path (2)+(1) and path (3) though the oscillation in asymmetry of photoelectron angular distribution as a function of the ω-2ω relative phase (Fig. 1). The strong modulation in ^{1}S (not shown) and ^{1}D, and its absence in ^{3}P unambiguously demonstrates the detection of spLEAD (1) as well as its coherent control. The oscillation amplitude for ^{1}D (2.3 %) is consistent with the value (1.9 %) estimated from our theoretical modeling.

This work was supported by MEXT, JST, JSPS, and IMRAM.

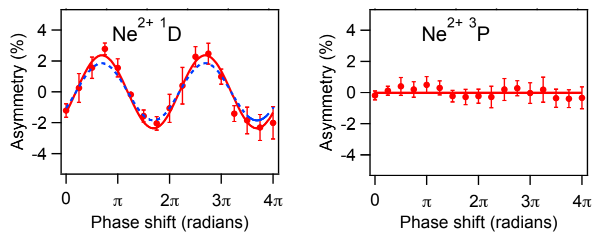

Figure 1. Asymmetry of photoelectron angular distributions vs. ω-2ω relative phase for final ionic states ^{1}D and ^{3}P of Ne^{2+}. Circles and solid curve: measurement (phase is not absolute), dashed curve: theory.

References

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