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Two-photon absorption using off-resonant excitation with ultrashort X-ray pulses

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Synopsis High energy resolution off-resonant X-ray emission spectroscopy (HEROS) was applied to study one-photon (OPA) and two-photon absorption (TPA) processes in metallic copper generated by ultra-short hard X-ray FEL pulses from the Linac Coherent Light Source (LCLS). The OPA and TPA signals were measured using a multiple-crystal von Hamos diffraction spectrometer allowing observation of both the intermediate and final states in the TPA process. The universal scaling of TPA/OPA cross sections is discussed.

Until recently, the nonlinear interaction of Ångstrom X-ray wavelengths with matter has remained an unexplored regime. It is only within the last few years that the development of X-ray free electron laser sources (XFELs) has permitted the field of nonlinear optical research to be extended into the hard X-ray regime [1]. This has allowed the first observations of processes with very low cross-sections, including double-core hole formation [2], two-photon absorption [3], amplified spontaneous x-ray emission [4], plasma creation [5] and X-ray-optical wave mixing [6].

Among the different mechanisms for nonlinear X-ray interaction with matter, the two-photon absorption (TPA) process exhibits third-order photon-atom interactions. In contrast to one-photon absorption processes, which are determined by dipole-allowed transitions, the TPA mechanism requires excitation between states of the same parity. We have investigated TPA processes for electronic core-levels using off-resonant X-ray emission spectroscopy at an XFEL. The results indicate a strong correlation between one-photon and two-photon mechanisms, with the TPA process being purely sequential and mediated by excitation into a virtual electronic intermediate state.

In the present work, we employed high energy resolution off-resonant X-ray emission spectroscopy [7] (HEROS) to measure one- and two-photon absorption signals (OPA and TPA respectively) in metallic copper generated by ultra-short hard X-ray pulses. The experiment was performed at the Coherent X-ray Imaging instrument at the Linac Coherent Light Source (Menlo Park, USA) XFEL. For x-ray emission detection at around 8040eV ($K\alpha_{1,2}$ x-ray lines, $2p_{3/2}$->1s and $2p_{1/2}$->1s transitions) we employed a spectrometer operated in a multiple-crystal dispersive von Hamos geometry.

We will report on extracted TPA cross-sections and examine the X-ray fluence thresholds at which this process becomes dominant. We find a strong relationship between one-photon and two-photon absorption cross-sections as a function of incoming X-ray energy. Experimental data suggests a universal scaling of TPA/OPA cross-sections, which explains the saturation effects of TPA signal at highest fluences. Thanks to our experimental approach, we track the yields of both the intermediate and final states during the course of the TPA process. The measured cross-section for the second absorption step in TPA has been found to be four orders of magnitude higher than the first absorption event. With the proliferation of experiments using ultrashort, intense X-ray pulses for a variety of measurements at XFELs, measuring TPA cross-sections is critically important for establishing the regimes over which these processes can either be avoided or enhanced. This is especially relevant for X-ray photon energies close to ionization thresholds, where TPA cross-sections can be significant.

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