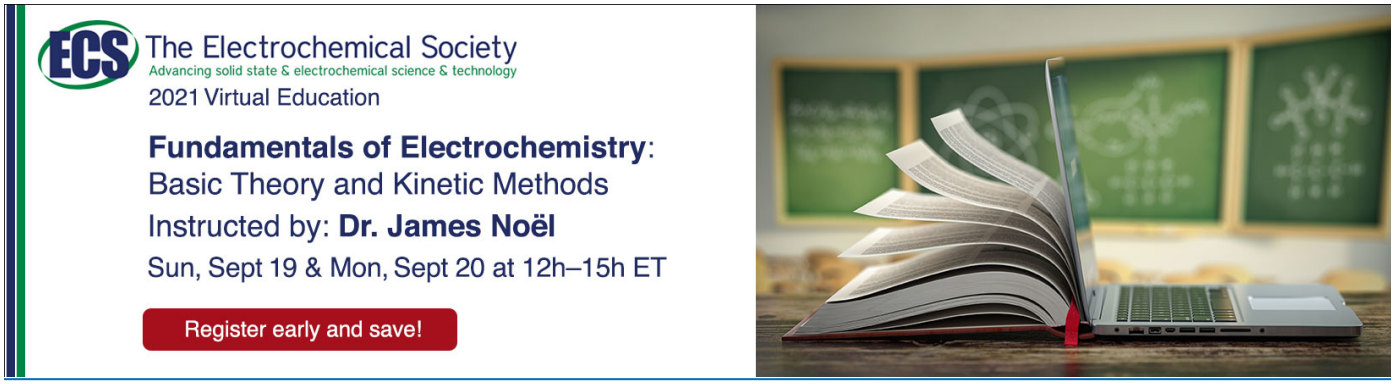
X-ray two-photon absorption with high fluence XFEL pulses



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X-ray two-photon absorption with high fluence XFEL pulses

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Synopsis. We report on nonlinear interaction of solid Fe with intense femtosecond hard x-ray free-electron laser (XFEL) pulses. The experiment was performed at the CXI end-station of the Linac Coherent Light Source (LCLS) by means of high-resolution x-ray emission spectroscopy. The focused x-ray beam provided extreme fluence of ~10^5 photons/Å^2. Two-photon absorption leading to K-shell hollow atom formation and to single K-shell ionization of solid Fe was investigated.

X-ray free electron laser (XFEL) facilities, with unprecedentedly high peak power densities reaching ~10^{20} W/cm^2, have paved the way to study nonlinear phenomena in the x-ray regime [1-6]. In this work we explored nonlinear interaction of high-fluence hard x-ray femtosecond pulses with solid Fe. Single and double K-shell electron ionization processes resulting from two-photon absorption were observed.

The experiment was carried out at the CXI end-station of the Linac Coherent Light Source (Menlo Park, USA) XFEL by means of the high energy resolution x-ray emission technique. The XFEL beam of ~5x10^{11} x-rays/pulse and pulse energy of 0.6 mJ was focused on a metallic Fe sample. The ultra-focused x-ray beam provided extreme fluence of ~10^5 photons/Å^2. Moving the sample out of the focus along the beam allowed varying the fluence. For the Fe Kα ($K^{-1}\rightarrow L^{-1}$) and Kα^h ($K^{-2}\rightarrow K^{-1}L^{-1}$) radiative transitions measurements the bent crystal von Hamos x-ray spectrometer of PSI [7] installed at CXI and equipped with the CSPAD detector developed at SLAC was employed. The K x-ray emission spectra were collected at photon beam energies below the Fe K-shell single- and double-ionization thresholds for the two-photon single ionization and double ionization processes, respectively.

For illustration, the probability of double K-hole formation *via* sequential absorption of two photons *versus* x-ray fluence is shown in figure 1. We observe a ~60-fold increase in the production probability of Fe hollow-atoms as compared to single-photon double ionization mediated by K-shell electron-electron correlations [8]. The cross-sections for double-K-hole formation and two-photon single K-shell ionization were derived from the x-ray fluence dependence of the measured x-ray emission intensities. For the two-photon single ionization process a square dependence of the Kα signal was found. This is the first observation of K-shell double core-hole creation following sequential photon absorption, and two-photon single K-shell ionization for metallic Fe.

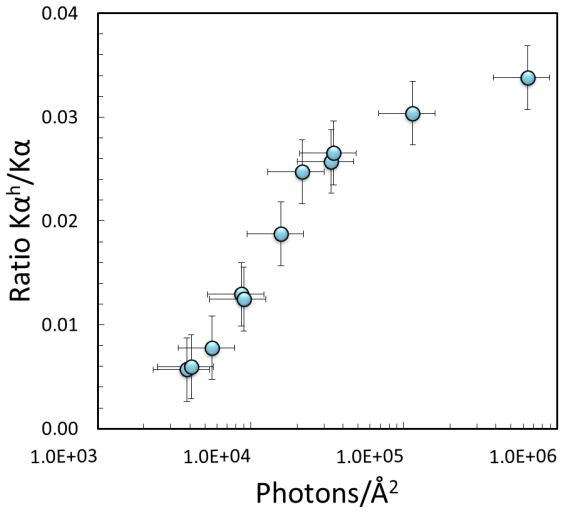

Figure 1. Probability of double K-hole formation *via* sequential absorption of two photons for Fe as a function of x-ray fluence. The data were derived from the Kα^h ($K^{-2}\rightarrow K^{-1}L^{-1}$) to Kα ($K^{-1}\rightarrow L^{-1}$) intensity ratios. The x-ray pulse energy was 7.6 keV and the duration 30 fs.

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