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Ultrafast laser induced structural dynamics probed by time-resolved photoelectron spectroscopy

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Synopsis This work aims to study laser-induced lattice dynamics when there is strong electron-lattice temperature non-equilibrium for which the material properties are poorly known. The proposed experimental technique is time-resolved photoelectron spectroscopy (Tr-PES). The main challenges of such technique are the superposition of pump and probe spectra and the distortion of the probe spectra due to the interaction with pump-induced electrons, called space charge effect (SCE). We strongly reduced this effect by tuning the 100eV XUV beamline we have developed, enabling us to investigate Copper samples.

When ultra-short laser pulses induce a non equilibrium regime of the electron-lattice temperature, theoretical description is difficult due to the limited knowledge of material properties in these conditions [1], including the electron-phonon coupling factor, the electronic and ionic thermal conductivities, and the electron heat capacity. In this context, time-resolved photoelectron spectroscopy (Tr-PES) experiments may help to improve our understanding of the physical processes and material properties in this regime.

The excitation and relaxation of matter can be diagnosed by studying the time evolution of their PES spectra, which is influenced by the lattice state/temperature. However, such a pump/probe experiment, based on the detection of probe-induced photoelectron spectra, is technically challenging due to the presence of a pump-induced high flux of photoelectrons that can cause a loss of information by overlapping and/or interacting with the probe electrons, distorting the detected spectrum (called space charge effect, SCE) [2].

To prevent the overlap of photoelectron spectra, we have developed a dedicated XUV beamline based on High-order Harmonic Generation [3] that delivers femtosecond pulses with the required photon energy (80 – 100 eV). This beamline will be fully described during the conference.

Thanks to the complete experimental and theoretical investigations on SCE, we have been able to optimize the experimental parameters in order to perform Tr-PES pump/probe experiments on Copper samples where the SCE has been strongly reduced. It allowed us to separate the influence of lattice dynamics on the PES from the SCE, as illustrated Figure 1.

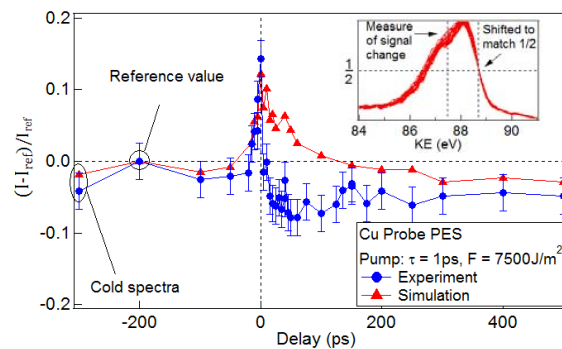


Figure 1. Pump/probe measurements and their corresponding SCE simulations showing the Cu PES evolution. The difference in positive delays is attributed to the PES modification due to lattice dynamics. Inset: spectra normalized and superimposed to match the signal value of $I = 1/2$. The signal I is then measured at $KE = 87.5\text{ eV}$.

References

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