Time-resolved resonant soft x-ray scattering with free-electron lasers

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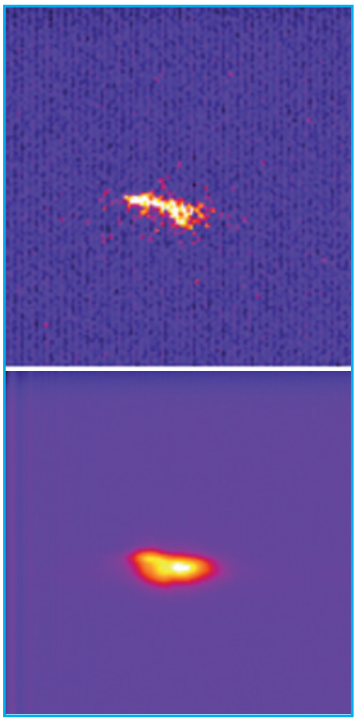
Application Note

New accelerator-based light sources such as the free-electron laser FLASH at DESY in Hamburg produce ultra-short, coherent x-ray pulses with a pulse duration below 100 femtoseconds and extremely high intensity (roughly 10^{12} photons/pulse) [1].



The (0,0,1/2)-superstructure peak of magnetite taken at a photon energy of 529,4 eV with an Andor DO936N-MW-BN Open front end CCD camera

top: single shot image taken with a 40fs-x-ray pulse from FLASH

bottom: integrated signal over more than 1000 pulses

With these very intense x-ray pulses the established x-ray spectroscopic techniques for the investigation of static electronic and magnetic properties of matter can be extended into the time domain. This opens up completely new possibilities for the understanding of structure and, in particular, function of materials. Of great interest is the variety of phase transitions in complex materials such as e.g. metal-insulator transitions, superconductivity or magnetic order-disorder phenomena. The ground state of highly correlated materials is generally determined by a subtle interplay between excitations of the electronic system, the lattice, and the spin and orbital degrees of freedom of the charge carriers, respectively. In these systems very often a controlled excitation out of equilibrium by an IR- or optical pulse and the subsequent relaxation behavior back to the ground state allows to identify the most important degrees of freedom and their coupling.

We have recently performed a pioneering experiment of this kind at FLASH in the framework of a large collaboration with partners from the Helmholtz-Center Berlin, the Stanford Linear Accelerator Center (SLAC), the Universities in Cologne and Augsburg as well as the Max-Planck-Institute for Solid State Research in Stuttgart. There we have investigated the famous Verwey-transition in magnetite (Fe_3O_4) with time-resolved resonant soft x-ray scattering at the oxygen K-edge in a pump-probe experiment [2].

In the insulating low-temperature phase of magnetite charge and orbital order is observed at the oxygen and the iron atoms which results in characteristic superstructure peaks in resonant soft x-rey scattering. After excitation with an infrared laser pulse (1.5 eV, 130 fs) the charge order vanishes on a time scale of less than 300 fs and a transition to the metallic phase is observed.

The very short time scale indicates that this transition cannot be driven simply by phonons, but rather has to be induced by a coupled, polaronic, motion of charge carriers and lattice.

References

[1] W. Ackermann et al., Nature Photonics 1, 336 (2007)
[2] N. Pontius et al., Applied Physics Letters 98, 182504 (2011) and in preparation

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