Exploring ultrafast spin and electronic structure dynamics with femtosecond time-resolved x-ray spectroscopy

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(Invited)

X-ray absorption spectroscopy (XAS) at core level absorption edges in the soft x-ray range gives element- and orbital-specific information on the electronic structure, while x-ray magnetic circular dichroism (XMCD) probes spin and orbital moments in ferromagnets. By employing femtosecond (fs) pulsed x-ray sources such as free electron lasers and synchrotron femtoslicing sources [1], we now not only have access to the properties in thermal equilibrium, but can probe non-equilibrium states of condensed matter on the femto- to picosecond (ps) timescales relevant to fs laser-induced electronic, spin, and lattice excitations and their relaxation.

In the rare earth ferromagnets Gd and Tb, most of the magnetic moment is carried by the 4*f* orbitals localized at the ion cores. After fs laser excitation and electron-phonon relaxation, the rate of the resulting demagnetization is linked to the strength of spin-lattice coupling in the respective rare earth, which depends on the anisotropy of the 4*f* orbitals, leading to faster dynamics with a demagnetization time constant of approx. 8 ps in Tb with a $4f^8$ configuration (L=3) compared to about 40 ps in Gd ($4f^7$, L=0) [2]. By alloying Gd and Tb, we can systematically tune this demagnetization time. From element-sensitive XMCD measurements at the M5 edges we derive that demagnetization occurs for Gd in $Gd_{0.6}Tb_{0.4}$ on the same timescale as for Tb, indicating an enhanced coupling of the Gd 4*f* magnetic moments to the lattice via indirect interatomic exchange (RKKY) interaction with the Tb 4*f* moments, while a transient difference in the amount of demagnetization between Gd and Tb is attributed to a limited efficiency of this interatomic exchange coupling [3].

I will furthermore present first results on fs time-resolved XAS of the Fe-pnictide high temperature superconductor $BaFe_2(AsP)_2$, where we observe pump-induced changes at the Fe L_3 edge that are described by the interplay of a transient shift in energy and broadening. This complex behavior will be discussed in the context of electronic and phononic excitations in the Fe-pnictides and compared to similar observations in weakly correlated materials, *i.e.* metals.

References:

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