

## Intra-atomic exchange in ultrafast magnetism – the Gd puzzle

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The exchange interaction is the defining element in the formation of magnetic order in atoms and solids. Therefore the role of intra- and interatomic exchange during ultrafast magnetization dynamics needs to be explored. In the atomic magnetism of lanthanide metals localized 4f and itinerant 5d orbitals contribute to the overall magnetic moment. In general it is assumed that the intra-atomic exchange coupling is fast enough to be treated as an instantaneous process.

We studied the magnetization dynamics of the lanthanide metals gadolinium [1] and terbium by time-resolved photoemission employing femtosecond higher-order harmonic vacuum-ultraviolet pulses [2]. We prepare lanthanide films of 5 - 10 nm thickness on a W(110) substrate by evaporation of Gd and Tb in ultrahigh vacuum. Recording in parallel the 4f magnetic linear dichroism in the angle-resolved photoelectron distribution and the exchange splitting of the 5d minority and majority spin valence bands we observe distinct spin dynamics in Gd, which indicate a breakdown of the intra-atomic exchange upon femtosecond laser excitation. While the exchange splitting reduces with a time constant of 800 fs the 4f MLD signal shows a significantly slower 13 ps decay. An orbital-resolved Heisenberg model [3] explains well the state-dependent two timescales of magnetization dynamics in Gd metal, which differ by one order of magnitude.

Spin-, time-, energy-, and angle-resolved photoemission measurements of the occupied Gd surface-state using 6.2-eV laser pulses corroborate these findings. While the peak position of the d-derived surface state mimics the femtosecond dynamics of the 5d exchange splitting, the spin polarization of the surface state follows the dynamics of the 4f magnetic linear dichroism and drops only slightly. The latter is very different from spin-mixing observed in thermal equilibrium for increasing sample temperature.

However, recent X-ray magnetic circular dichroism measurements probing the unoccupied 4f states via the  $M_5$  and  $M_4$  absorption edges show again an ultrafast, femtosecond response. We performed these measurements in reflection geometry on an analogous Gd/W(110) sample-system thereby ruling out different preparations, fluences, and sample temperatures as the cause of the different MLD and XMCD dynamics. These puzzling results point to different dynamics of 4f orbital and spin momenta.

Due to its much stronger spin-lattice coupling, Tb shows a distinctly different magnetization dynamics, yet consistent in all experiments.

[1] Robert Carley *et al.*, Phys. Rev. Lett. **109** (2012) 057401.

[2] Frietsch *et al.*, Rev. Sci. Instrum. **84** (2013) 075106.

[3] Soenke Wienholdt *et al.*, Phys. Rev. B **88** (2013) 020406(R).