Separating Exchange Splitting from Spin Mixing in Gadolinium by Femtosecond Laser Excitation

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The ferro- to paramagnetic phase transition of gadolinium caused heated debates in the 1990s. While some experiments found classical Stoner-like band shifts [1], others observed the spin-mixing behavior proposed for rare-earth metals like Gd [2]. The description of the magnetic band structure is equally challenging for density-functional [3] as well as many-body theory [4]. Driving the phase transition by an ultrafast laser pulse is one way to study this puzzling phenomenon from a different point of view, since it allows us to disentangle interactions by the different timescales they occur on. Especially Gd shows several timescales for magnetic reactions accessible by different experimental techniques [5-7]. To resolve the puzzle of these different timescales ranging from the sub-picosecond to the 100-ps regime, we set up an experiment for spin-, time- and angle-resolved photoemission spectroscopy (STARPES). We find, that STARPES is able to probe all previously measured timescales simultaneously and can thus explain the differences.

In laser photoemission, we measure the binding energy and spin polarization of the majority-spin Gd(0001) d_{z^2} surface state. The magnetic phase transition is driven by femtosecond laser pump pulses with a fluence of 8 mJ/cm² and photon energy of 1.56 eV generated in a 300 kHz Ti:Sapphire regenerative amplifier. Frequency-quadrupled pulses of 6.2 eV photon energy are used for photoemission. Spin resolution is achieved by means of an exchange scattering spin detector.

After the ultrafast excitation of hot electrons by the femtosecond laser pulse, we find a fast sub-picosecond reduction of the surface states' exchange splitting, while the spin polarization is reduced much slower on a timescale of 15 picoseconds. This reveals a magnetic non-equilibrium in the surface state resulting from the indirect exchange interaction in 4*f* magnets [8].

The authors thank the Helmholtz Virtual Institute Dynamics in Multidimensional Landscapes for financial support.

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