Long-wavelength Laser-driven Ultrafast Magnetization Dynamics in Gadolinium

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Introduction

Manipulating magnetic order by laser irradiation is promising for applications and fundamental science. A non-equilibrium distribution of hot electrons excited by a femtosecond laser pulse dissipates energy to the lattice and spin system, leading to transient states with unexplored properties [1]. Among the many experimental probes of magnetization dynamics [2-7], time- and angle-resolved photoemission spectroscopy (tr-ARPES) is a particularly direct one.

The magnetism of gadolinium derives from its half-filled 4f electronic shell, which gives rise to a large, atomically localized magnetic moment. Indirect exchange coupling via the valence band aligns the moments to produce a ferromagnet. Gadolinium exhibits, therefore, both itinerant and localized magnetism, making it an ideal test system for studies in femtosecond magnetization dynamics.

Our previous tr-ARPES work on gadolinium has revealed magnetic non-equilibrium in the valence bands [8, 9], between the valence band and 4f systems [10], and in the surface state [11]. We revisited this topic in an Artemis beam time in November 2015 to study the influence of reducing the pump photon energy on the different dynamics.

Magnetization Dynamics in the Valence Bands

Our experiment made use of the Artemis time-conserving monochromator for high-order harmonics. By tuning the photon energy to 36 eV, we measured tr-ARPES spectra with the time-of-flight (ToF) electron kinetic energy analyser from thin films of gadolinium grown on a W(110) single-crystal substrate. The ferromagnetic gadolinium was excited using femtosecond pump pulses at 1300 nm. The experiments yielded a number of unexpected results summarized in Figures 1 and 2.

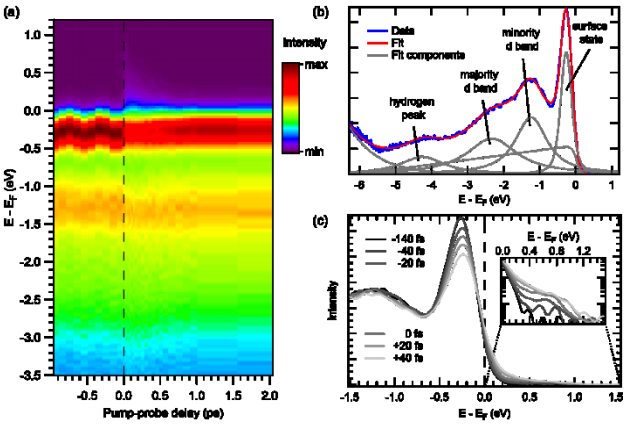

Figure 1. (a) Raw tr-ARPES at the Γ point of Gd(0001) at 40 K taken with the Artemis ToF apparatus. The pump and probe photon energies were 0.95 eV (1300 nm) and 36 eV, respectively. Depletion of the surface state and excitation of electrons above the Fermi level by the pump pulse can be clearly seen (also shown in selected spectra around t = 0 in (c)). Large oscillations of the binding energy are observed before t = 0. **(b)** Fit of the unpumped spectrum (at t < 0). The spectrum is decomposed into contributions from the surface state, the minority- and majority-spin valence bands, a peak arising from hydrogen contamination, and an exponential- and a linear background. To visualize the Fermi function, it is multiplied with the linear background. **(c)** Laser-excited non-equilibrium electrons thermalizing into a Fermi distribution around t = 0.

Pumping at 1300 nm induces a sizable redistribution of the electronic population around the Fermi level (see Fig. 1(a) and (c)). The thermalization of the hot electron population with the

lattice leads to demagnetization in the valence bands, which is reflected in the decreasing exchange splitting (Fig. 2(b)). This is in line with our previous measurements at a pump wavelength of 800 nm [9]. However, the dynamics in the individual bands differ from our previous observations. Here we find a simultaneous shift of the valence bands to higher binding energies on a very fast timescale (< 100 fs). Furthermore, we find a clear increase of the exchange splitting at t = 0, which is in line with MOKE experiments [3]. We attribute it to coherent dynamics between 5d and 4f states while the laser-excited non-equilibrium electronic distribution persists.

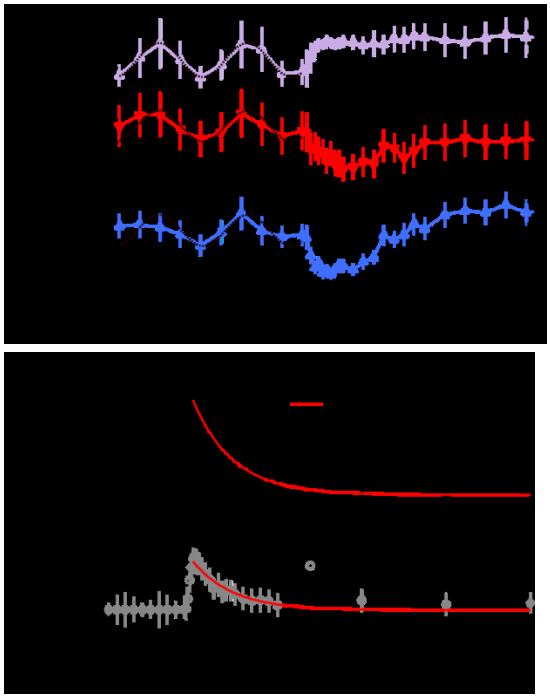

Figure 2 (a) Temporal evolution of the binding energy of the spectral features of gadolinium in Fig. 1(b). The dotted line is a fit to the oscillations in the surface state binding energy. It reveals an amplitude of 34.5 meV and a periodicity of 440 fs. **(b)** Ultrafast magnetization dynamics of gadolinium revealed by changes in the exchange splitting after laser excitation at 0.95 eV. The increase just after t = 0 can be attributed to the non-equilibrium electron distribution formed by the pump pulse. The electronic temperature extracted from the fitting procedure is shown in grey. An exponential fit to the decreasing exchange splitting results in a time constant of 0.47±0.02 ps, which is on the same timescale as the equilibration of the electronic temperature with that of the lattice.

Ponderomotive Oscillations

Perhaps the most visible feature of the data is an oscillation in the binding energy at t < 0. It has been attributed to ponderomotive acceleration of the outgoing photoelectrons by the transient grating formed by interference between the incident and reflected parts of the pump pulse [12]. The oscillations extend to positive delays (see E_F in Fig. 1(a)), albeit with much reduced amplitude. The origin of the large magnitude of the oscillations in Fig. 2(a) remains to be clarified.

Conclusions

The direct measurement of the time-dependent band structure with the time resolution available in the Artemis facility, in combination with the tunable pump energy revealed new aspects of non-equilibrium magnetism in the valence band structure of gadolinium. In particular, we found an *increase* of the d-band exchange splitting within the first 100 fs after laser excitation generated by a shift of its majority spin component to higher binding energy. Further investigations will help to clarify whether this new observation is the signature of coherent excitation of the exchange coupled 4f5d spins.

Furthermore, we observed an extremely pronounced ponderomotive oscillation of the photoelectrons in the laser field. According to the model presented in [12], the amplitude and frequency of the oscillations depend on the pump laser wavelength. We plan to verify this in another experiment at Artemis with varying the pump laser wavelength, which would be equally interesting for further investigations of the non-equilibrium magnetization dynamics.

Acknowledgements

The authors gratefully acknowledge financial support from Laserlab Europe under Application Number 15120039.

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