The valence band structure of Gadolinium studied with time-resolved photoemission

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Abstract We have studied the response of the exchange split valence bands of ferromagnetic gadolinium to femtosecond laser excitation. We observe a drop of the exchange splitting with a time constant of 0.9 ps but different response times of minority and majority spin bands. Furthermore, even above the Curie temperature there is a finite exchange splitting, which also decreases with laser excitation.

Introduction

It is well established that optical excitation of thin ferromagnetic films leads to demagnetization within a few hundred femtoseconds [1] and even ultrafast optical magnetic switching of ferrimagnets has been demonstrated [2]. However, it remains controversial which microscopic processes are fast enough to provoke femtomagnetism: direct interaction with the laser field [3], scattering among electrons [4], phonons [5], and magnons [6], and/or spin-transport [7]. To establish microscopic models it is necessary to unravel the transient electronic structure [8] and clarify on which timescale the exchange splitting ΔE_{ex} and spin polarization collapses.

To approach these problems we perform time- and angle-resolved photoemission (TR-ARPES). We use laser-driven high-order harmonic generation as an extreme ultraviolet light source [9]. A Ti:Sapphire CPA laser system delivers 1.5 mJ, 40 fs IR pulses at 10 kHz repetition rate, which are focused into a gas cell containing argon at 100 mbar. Individual harmonics are selected by a toroidal grating monochromator (TGM) with a single grating. The time resolution $\tau \sim \lambda / cN$ is primarily determined by the transmitted bandwidth (N is the number of illuminated lines). While the temporal spread is nearly independent of the TGM slit-size, the latter allows tuning of the energy resolution ΔE. For 10 μm slits we obtain $\tau = 100$ fs, $\Delta E = 150$ meV, and $2 \cdot 10^4$ photons/pulse at $h\nu = 35.6$ eV.

Gadolinium valence-band dynamics and exchange splitting

We have studied ultrafast demagnetization of the local-moment ferromagnet gadolinium [10], prepared as epitaxial film of 10 nm thickness on a W(110) substrate. Photoelectrons are detected after a hemispherical energy analyzer. Tuning the TGM to 35.6 eV allows us to map the valence bands of Gd at the Γ-point of the 4th Brillouin zone. Binding energy and exchange splitting of the Δ_2-like Σ bands are shown in Fig.1a. The initial binding energies of 1.39 and 2.25 eV are in agreement with synchrotron measurements conducted at a comparable sample temperature of 100 K [11]. Upon laser excitation (hν = 1.6 eV, s-pol., 300 fs, fluence 1.2 mJ/cm^2) the exchange splitting of the valence bands drops with a time constant of 0.9 ± 0.1 ps. This value coincides with the values observed in MOKE [12] and XMCD [13] measurements. The latter experiment probes the *4f* spin subsystem but was conducted at higher pump fluences (3-5 mJ/cm^2). Comparing ultrafast de- and thermal re-magnetization [10] we conclude that ΔE_{ex} maps the true magnetization. From Fig. 1a it is evident that the minority valence band reacts immediately after laser excitation while the response of its majority counterpart is delayed and is only half as fast. Delay zero was independently determined from the first occurrence of pump-induced hot electrons above the Fermi level E_F. We attribute the instantaneous response of the minority spin band to superdiffusive spin transport [7]. Photoemission probes the first few surface layers and is thus very sensitive to spin currents into the bulk. The delayed response of the majority band can be explained by electron-phonon scattering which increases with increasing lattice temperature [5]. Our data suggest that both spin transport and electron-phonon scattering contribute to ultrafast demagnetization. In addition, the molecular field acting on the spins may delay the response of the majority spin band [14].

It has been a long standing question whether the exchange splitting vanishes at the Curie temperature T_C = 293 K [15]. For the Gd surface state scanning tunnel-

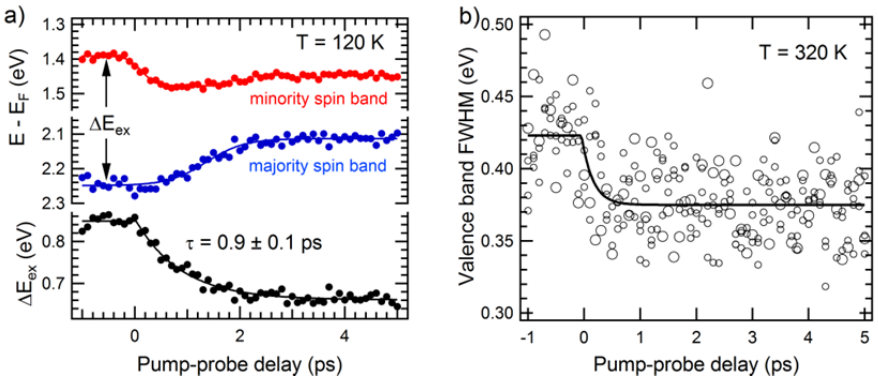

Fig. 1. a) Temporal evolution of the Gd minority and majority spin bands and exchange splitting upon optical excitation. b) Decrease of the valence band's full width at half maximum at 320 K.

ing spectroscopy revealed a sizeable exchange splitting at T_C [16]. However, photoemission measurements of the bulk valence bands remain ambiguous since the minority and majority spin bands coalesce into a single peak due to thermal broadening [11, 15]. In Fig. 1b we show the linewidth of this peak as a function of pump-probe delay at 320 K. Upon laser excitation we observe a small but significant decrease of the linewidth, which can only be explained by a further reduction of ΔE_{ex}. Above T_C there exists in fact a finite exchange splitting, which implies local magnetic order.

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