



Picosecond magnetization dynamics of the Gd(0001) surface

A. Melnikov, U. Bovensiepen^{*}, I. Radu, O. Krupin,
K. Starke, E. Matthias, and M. Wolf

Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin, Germany

Elsevier use only: Received date here; revised date here; accepted date here

Abstract

The magneto-induced fraction of the optical second harmonic reflected from Gd(0001) surfaces was studied in pump-probe experiments as a function of temperature. The magnetic contrast in thermal equilibrium and at 1 ps pump probe delay show a similar temperature dependence, but at 1 ps delay the magnetic contrast is reduced to 30% due to ultrafast demagnetization. Compared to lattice cooling the magnetization recovery is significantly slower within the first 150 ps and, therefore, does not obey the typical M(T) behavior in this time range. This is attributed to the localized character of the 4f electrons carrying the large magnetic moment in Gd. © 2003 Elsevier Science. All rights reserved

Keywords: second harmonic generation, optical pump-probe-experiments, magnetization dynamics

PACS: 78.47.+p, 42.65.Ky, 75.40.Gb

Demagnetization of a ferromagnetic metal by femtosecond laser pulses has been observed to occur on a subpicosecond time scale [1,2,3]. Generation of an excited electron distribution in the Ni conduction band which carries the magnetic moment in itinerant ferromagnets has been shown to be the origin of this phenomenon [2]. Since the elementary mechanism of this ultrafast demagnetization is still under discussion, it is also interesting to compare the pump induced magnetization dynamics of an itinerant ferromagnetic material like Ni with that of localized magnetic moments as in Gd. In the latter, the optically excited electrons of the 5d6s conduction band mediate the ferromagnetic coupling of the localized 4f moments. Thus, the absorbed pump energy has to be transferred from the conduction band to these localized states, which suggests different timescales for magnetization and electron dynamics in case of Gd. Up to now, Vaterlaus et al. [4] reported a spin-lattice-equilibration time of about 100 ps. Below we will show that the magnetization of the Gd(0001) surface can be reduced on a subpicosecond time scale like for band ferromagnets, but the magnetization recovery up to 100 ps proceeds significantly delayed compared to the electron and lattice relaxation dynamics.

Gd films of 20 nm thickness were grown on W(110) at 330 K and annealed to 700 K for smooth epitaxial ferromagnetic films [5]. The second harmonic (SH) intensity was measured in saturation at 500 Oe along the easy axis of magnetization in the film plane oriented perpendicular to the optical plane of incidence (transversal configuration). For pump probe measurements laser pulses of 35 fs duration at 1.5 eV photon energy and 40 nJ pulse energy were split 4:1 in pump and fundamental probe beams. For a fundamental photon energy of 1.5 eV SHG evolves resonantly enhanced by the unoccupied component of the exchange split surface state of Gd(0001), which makes the SH response a particularly surface sensitive probe [6]. The second harmonic intensity for opposite magnetization direction is composed of two fields which behave even or odd with regard to magnetization inversion as shown by Pan et al. [7]: $I^{\uparrow\downarrow}(t)=[E_{\text{even}}(t) \pm E_{\text{odd}}(t)]^2$.

In static measurements the data are represented by the magnetic contrast $[I^{\uparrow\downarrow}]/[I^{\uparrow\uparrow}]$. In time-resolved experiments the normalized intensities for opposite fields are given by $D_{\pm}(t)=[I^{\uparrow\downarrow}(t) \pm I^{\downarrow\downarrow}(t)]/[I^{\uparrow\downarrow}(t_0) \pm I^{\downarrow\downarrow}(t_0)]$, t_0 denotes a negative delay, *i.e.* without pump pulse. Pump-

^{*} Corresponding author. Tel.: +49-30-838-53340; fax: +49-30-838-56059; e-mail: uwe.bovensiepen@physik.fu-berlin.de

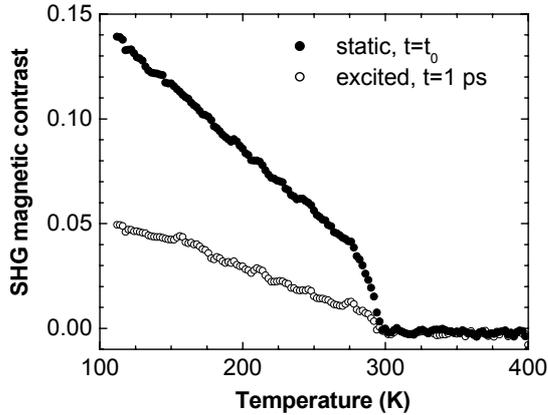


Fig. 1: Temperature dependent magnetic SH contrast $[I^{-1}]/[I^+I^-]$ in thermal equilibrium ($t=t_0$) and at a pump probe delay of 1 ps. Both data sets were recorded simultaneously with the pump beam blocked in case of the static measurement.

induced variations $\Delta(t)$ of the even and odd SH field can be expressed by [8]:

$$\Delta_{\text{even}}(t) = \sqrt{D_+(t)} - 1 = [E_{\text{even}}(t)/E_{\text{even}}(t_0)] - 1$$

$$\Delta_{\text{odd}}(t) = [D_-(t)] / \sqrt{D_+(t)} - 1 = [E_{\text{odd}}(t)/E_{\text{odd}}(t_0)] - 1$$

Fig. 1 displays the temperature dependent magnetic SH contrast in equilibrium, which shows a quasi-linear behavior up to 270 K followed by the critical behavior near the Curie temperature. It agrees with earlier investigations with secondary electron emission [9]. Note that we observe no indication of an enhanced surface Curie temperature. The open circles represent the magnetic contrast 1 ps after pump pulse excitation. In this case, the magnetic contrast shows a similar temperature dependence but the contrast is reduced to one third of the equilibrium value. This result implies that photon induced demagnetization occurs in Gd faster than 1 ps. The mechanism is pump pulse excitation of conduction electrons which weakens the exchange interaction of neighboring magnetic moments and thus results in a reduction of magnetic order.

The temporal evolution of Δ_{even} and Δ_{odd} within several 100 ps is depicted in Fig. 2. Δ_{even} represents the electron dynamics following the pump pulse. Excited electrons thermalize on a subpicosecond timescale within the electron gas and reach equilibrium with the surrounding lattice in a few picoseconds. The plotted time range is too large to resolve such ultrafast processes, but the peak in Δ_{even} resembles maximum electron temperature. Here, we focus on the later time domain where electrons and lattice cool in equilibrium. Thermal diffusion into the film bulk and the substrate takes the excess energy out of the detection volume limited to the surface. The time scale of this process is governed by the thermal conductivity of 10.5 J/(msK) for Gd.

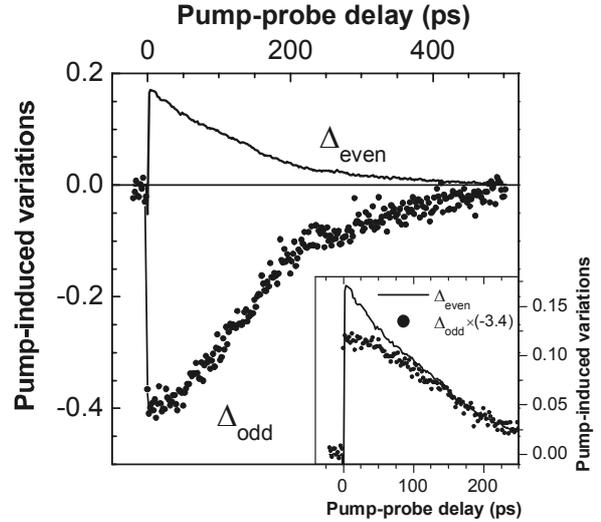


Fig. 2: Temporal evolution of Δ_{even} and Δ_{odd} at 84 K; the inset shows Δ_{odd} inverted and normalized to Δ_{even} at 200 ps.

The odd contribution represents the magnetization dynamics which for times longer than 150 ps follows the one of electrons and lattice, which agrees with the reported spin-lattice relaxation time [4]. A detailed inspection for shorter delays, as shown in the inset, reveals a considerably slower relaxation of the magnetization. This observation reflects the spin inertia of the $5d_z^2$ surface state probed with SHG and confirms the localized character of this state against the bulk [10].

In summary, we have observed femtosecond-laser induced demagnetization of Gd to occur in less than 1 ps, as is also found for itinerant magnets. The recovery of magnetization agrees in general with the spin-lattice relaxation, however, for times <150 ps the spin dynamics is significantly slower than the lattice cooling.

- [1] E. Beaurepaire, J.C. Merle, A. Daunois, J.-Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996).
- [2] J. Hohlfeld, E. Matthias, R. Knorren, K.H. Bennemann, Phys. Rev. Lett. **78**, 4861 (1997).
- [3] B. Koopmans, M. van Kampen, J.T. Kohlhepp, W.J.M. de Jonge, Phys. Rev. Lett. **85**, 844, (2000).
- [4] A. Vaterlaus, T. Beutler, F. Meier, Phys. Rev. Lett. **67**, 3314 (1991).
- [5] A. Aspelmeier, F. Gerhardter, K. Baberschke, J. Magn. Magn. Mat. **132**, 22 (1994).
- [6] A. Melnikov et al., Appl. Phys. B **74**, 723 (2002).
- [7] R.-P. Pan, H.D. Wie, Y.R. Shen, Phys. Rev. B **39**, 1229 (1989).
- [8] From an spectroscopic point of view, the odd SH field probes the spin polarization of the Gd surface state, which is a good measure for the magnetization of the surface because the 4f magnetic moments are coupled by direct exchange interaction to the probed $5d_z^2$ surface state.
- [9] C.S. Arnold and D.P. Pappas, Phys. Rev. Lett. **85**, 5202 (2000).
- [10] Ph. Kurz, G. Bihlmayer, S. Blügel, J. Phys.: Condens. Matter **14**, 6353 (2002)