

Ultrafast magnetization dynamics of Gd(0001): Bulk vs. surface

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(Dated: March 9, 2011)

Abstract

Ultrafast laser-induced demagnetization of Gd(0001) has been investigated by magneto-induced optical second harmonic generation and the magneto-optical Kerr effect which facilitate a comparison of surface and bulk dynamics. We observe pronounced differences in the transient changes of the surface and bulk sensitive magneto-optical signals which we attribute to transfer of optically excited, spin-polarized carriers between surface and bulk states of the Gd(0001) film. A fluence dependent analysis of the bulk magnetization dynamics results in a weak variation of the demagnetization time constant, which starts at about 700 fs and increases by 10% within a fluence variation up to 1 mJ/cm². We compare these results with fluence dependent changes in the transient energy density calculated by the two temperature model. The determined characteristic times of excess energy transfer from the electron system to the lattice, which is mediated by e-ph scattering, range from 0.2 - 0.6 ps. Such a more pronounced fluence dependent change in the characteristic time compared to the observed rather weakly varying demagnetization times suggests a more advanced description of the optically excited state than by the two-temperature model.

PACS numbers: 78.47.J-, 71.38.-k, 75.70.Ak, 78.70.Dm

I. INTRODUCTION

The understanding of spin dynamics in ferromagnets is widely based on radio frequency spectroscopy¹ and inelastic scattering techniques employing photons³ or neutrons². These experimental methods provide frequencies and decay rates of magnetic excitations by the line position and line width, respectively. More recently time domain techniques that employ short laser pulses have been established in a considerable number of laboratories and provide a complementary approach to analyze spin dynamics⁴⁻⁶. In pump-probe experiments the magnetization dynamics is driven by an external stimulus like a laser or magnetic field pulse and is probed by a second, time-delayed laser pulse. Using femtosecond laser pulses, which are nowadays routinely provided by Ti:Sapphire laser systems, the field of femtosecond magnetization dynamics has developed enormously. One reason for the interest in this approach might be that it is not only complementary to the established tools for magnetization dynamics, but that the non-equilibrium state and the respective dynamical magnetization processes have become accessible for systematic experiments⁶.

A number of novel phenomena like laser-induced demagnetization on sub-picosecond time scales^{7,8}, laser-driven formation of ferromagnetic order⁹, and magnetization reversal by individual laser pulses¹⁰ demonstrates the potential for spin manipulation in solid state materials on ultrafast time scales. At the same time, these phenomena challenge our understanding of ferromagnetism and ask for an appropriate description of the observed dynamics.

Beside the investigation of multi-constituent materials with a potential for novel phenomena, it is essential to develop insight into the more general laser-driven dynamics to establish this understanding. Finally, the meanwhile well established sub-picosecond demagnetization of elemental metallic ferromagnets is still controversial because the underlying elementary interactions remain under discussion. It has been shown recently that descriptions based on phonon-mediated spin-flip scattering (Elliott-Yafet type)^{11,12}, electron-spin-flip scattering (Stoner excitation)¹³, and ballistic transport of spin-polarized charge carriers¹⁴ can be used to describe rather similar experimental data of femtosecond demagnetization in Ni films. In the phonon-mediated process the magnetic moment is considered to be transferred directly to the lattice through phonons with appropriate symmetry obeying angular momentum conservation. In the electronic spin-flip scattering process the magnetic moment is not transferred within the elementary scattering process itself, but is dissipated by secondary

low energy spin excitations, which in their sum lead to demagnetization¹³. In the transport concept¹⁴ the spin polarization is basically taken out of the ferromagnet into a substrate or, alternatively, out of the experimental observation window. Therefore, the current understanding is not conclusive and a discrimination of these above processes is important at this stage.

Our efforts have concentrated on rare earth ferromagnets and in particular on Gadolinium which is considered to be a model system for a Heisenberg ferromagnet with a magnetic moment localized at the ion core. We have employed in earlier studies time-resolved surface sensitive techniques which are optical magneto-induced second harmonic generation (SHG) and photoelectron spectroscopy, to analyze the excited state and its relaxation. The response to the laser excitation is characterized by a coherent phonon-magnon mode localized at the surface and incoherent dynamics of optically excited electron-hole pairs, lattice vibrations, and magnetic excitations. A comprehensive overview is given in Refs.^{15,16}.

Here we present experimental results obtained by the femtosecond time-resolved magneto-optical Kerr effect (MOKE) on Gd(0001) films to analyze the magnetization dynamics in the bulk part of the Gd film, which we compare with the surface sensitive SHG signal. We find pronounced differences during the first 2 ps which we attribute to transfer of spin-polarized carriers between the surface and bulk part of the film. Moreover, we study the fluence dependence of the demagnetization process and compare it with predictions based on the two-temperature model.

II. EXPERIMENTAL ASPECTS

The experiments were carried out at a setup that combines an ultrahigh vacuum (UHV) chamber and a femtosecond (fs) Ti:Sapphire laser. For details see¹⁷. Gd was evaporated from an electron beam heated crucible under UHV conditions onto a W(110) substrate at 300 K. Annealing to 700 K subsequent to the deposition results in smooth epitaxial films¹⁸. Here we studied films with 20 nm thickness. Pump-probe experiments were performed at 50 K equilibrium temperature of the sample which was cooled by a liquid He cryostat. Laser pulses of 35 fs duration and 40 nJ energy were generated by a cavity dumped Ti:Sapphire oscillator at 800 nm central wave length with a repetition rate of 1.52 MHz. For pump-probe experiments the laser output was divided at a ratio 4:1. To detect the p-polarized second

harmonic (SH) generated by the p-polarized probe pulse a monochromator selecting 400 nm and a photomultiplier were used. To analyze the MOKE a balance detection scheme was used to measure the polarization rotation of the probe pulse θ in the longitudinal MOKE geometry. The magneto-optical signals were analyzed by a lock-in amplifier for MOKE and a single photon counter for SHG to detect the differential signals for open and blocked pump in opposite saturation magnetic fields ($H_s^{\uparrow,\downarrow}$) as a function of pump-probe delay t . Formally negative delays $t < 0$ denote the state before the optical excitation has occurred. For MOKE we show below

$$\frac{\Delta\theta(t)}{\theta_0} = \frac{\theta(t)}{\theta_0} - 1 = \frac{\vartheta(H_s^\uparrow, t) - \vartheta(H_s^\downarrow, t)}{\vartheta(H_s^\uparrow, t < 0) - \vartheta(H_s^\downarrow, t < 0)} - 1 \quad (1)$$

where ϑ is the angle between the linear polarization of the reflected probe pulse and a fixed reference polarization, θ is the magneto-optical Kerr rotation. $\Delta\theta$ is the time-dependent change in the Kerr rotation which represents the magnetization of the bulk part of the Gd film.

SHG is detected in the transversal geometry leading to magneto-induced changes in the SH intensity.

$$I_{2\omega}^{\uparrow,\downarrow} \propto (E_{2\omega}^{\text{even}})^2 + (E_{2\omega}^{\text{odd}})^2 \pm 2E_{2\omega}^{\text{even}}E_{2\omega}^{\text{odd}} \cos \phi. \quad (2)$$

Here $E_{2\omega}^{\text{even}}$ and $E_{2\omega}^{\text{odd}}$ are the SHG optical fields which behave as even or odd with respect to reversal of the magnetization M . The phase between these two field contributions is ϕ which is smaller than 15° and weakly time-dependent¹⁵. Since $E_{2\omega}^{\text{odd}} \propto M$ we plot below

$$\Delta_{2\omega}^{\text{odd}}(t) \approx \frac{E_{2\omega}^{\text{odd}}(t) - E_{2\omega}^{\text{odd}}(t < 0)}{E_{2\omega}^{\text{odd}}(t < 0)} \approx \frac{M(t) - M(t < 0)}{M(t < 0)} = \frac{\Delta M(t)}{M_0} \quad (3)$$

which represents the time-dependence of the surface sensitive magneto-optical signal.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The article presents results of time-resolved SHG and MOKE. The first section focuses on the comparison of surface and bulk sensitive detection of the transient magnetic state in Gd(0001). The second section contains a detailed study of time-resolved MOKE and reports on the fluence dependence of the bulk demagnetization.

A. Surface and bulk demagnetization dynamics

We start by comparing the transient SHG and MOKE signals that probe the surface and bulk of the Gd(0001) film, respectively. In Fig. 1, top panel, we show $\Delta_{\text{odd}}^{2\omega}$ (solid line, left axis, surface) and $\Delta\theta/\theta_0$ (dotted line, right axis, bulk) taken at nominally identical experimental conditions. Both signals indicate a pronounced laser-induced demagnetization, but their transient behavior is very different. The magnetic SHG signal is reduced within the laser pulse duration of 35 fs by 0.5 and exhibits on that level oscillations that are damped out within 3 ps. The signal starts to recover after 1.5 ps and returns to its value before optical excitation after several 100 ps (not shown, see Ref.¹⁹). The oscillatory part has been explained before as a coupled phonon-magnon mode localized to the surface^{15,16}. It is therefore expected, that it is not found in the bulk signal. The MOKE data can be described by a continuous reduction that will be fitted by a single exponential decay below and tends to saturate at a delay of 3 ps, before it demagnetizes further, not shown see Ref. ²⁰. Overall, the bulk sensitive signal is reduced on a much slower time scale than the surface one and is in agreement with femtosecond x-ray magnetic dichroism studies²⁰ if the different excitation densities are considered. We therefore conclude that the time-resolved MOKE signal probes the time-dependent magnetization of the film. We note here that a detailed comparison of MOKE ellipticity and rotation points to time-dependent variations of the magneto-optical constants at delays where the excess energy resides predominantly in the electronic system, as will be discussed in a forthcoming publication²¹.

To explain the difference between the surface and bulk demagnetization it is informative to inspect the electronic structure at the surface and to take transfer processes into account. Fig. 1, bottom, depicts several data sets which in their combination represent the electronic valence states of epitaxial Gd(0001) films. For a ferromagnetically ordered situation the states are exchange-split due to intra-atomic exchange interaction with the strong magnetic moment of the half filled $4f$ shell. At the $\bar{\Gamma}$ -point the bulk $5d$ -states appear at 1.4 eV (minority) and 2.4 eV (majority) binding energy and disperse towards E_F with increasing k_{\parallel} ²². The majority component of the exchange-split $5d_{z^2}$ -surface state is dominantly occupied and the minority one unoccupied. The system also contains unoccupied exchange-split bulk states²³.

For linear polarized light, within the dipole approximation, optical transitions proceed

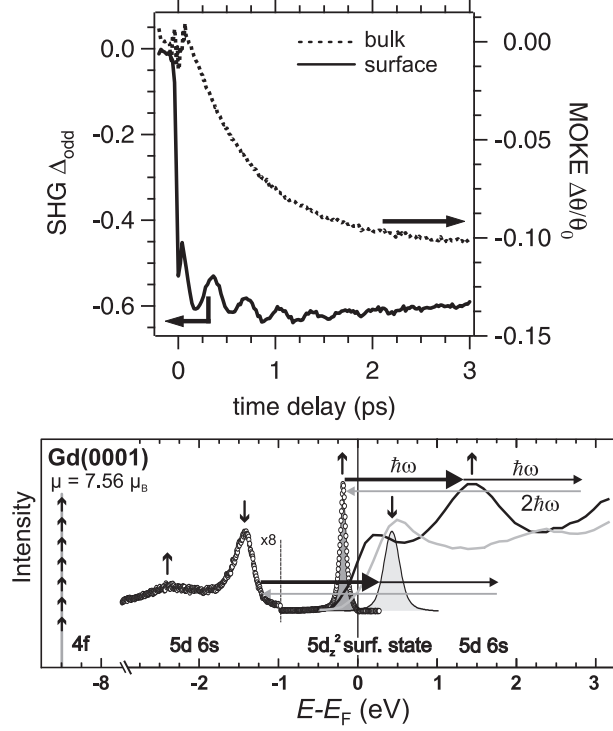


FIG. 1: Top panel: Time-dependent magneto-optical signals measured on 20 nm thick Gd(0001) films, which were grown epitaxially on W(110). The solid line shows the pump-induced change of the magnetic SHG contribution $\Delta_{2\omega}^{\text{odd}}$ sensitive to the surface. Similar results were published before^{15,24,25}. The dotted line depicts the pump-induced MOKE polarization rotation $\Delta\theta$ normalized to the static rotation θ_0 . The bottom panel is reprinted from Ref.¹⁵ and shows the valence electron states of Gd(0001) taken from normal direction photoemission (circles), inverse photoemission (solid lines), and scanning tunneling spectroscopy data above the Fermi level (solid line, filled)²⁶. The exchange-split surface state (filled area) appears around the Fermi level. Vertical arrows represent the majority and minority character of the electronic states. Indicated are the two main absorption channels for 1.5 eV pump photons and the resonant second harmonic probing scheme.

among occupied and unoccupied electronic states in (bulk) valence bands with the same spin character. In case of the excitation by the 800 nm (or 1.55 eV) pump laser pulses, an additional excitation channel becomes available at the surface because of resonant transitions coupling the bulk and the localized surface electronic states, see Fig. 1, bottom. Therefore a difference of the surface sensitive SHG response with respect to the bulk sensitive MOKE can be expected. Similarly to a charge-transfer excitation across molecular interfaces²⁷, such

surface-bulk resonances lead to effective electron transfer between surface and bulk which modify the transient population of the minority and majority components of the surface state. The population of the initially occupied majority surface state component is reduced due to the surface-to-bulk spin-up electron transfer, see Fig. 1, bottom. At the same time, the population of the initially unoccupied minority surface state component increases due to the bulk-to-surface spin-down electron transfer, which can alternatively be viewed as a surface-to-bulk spin-down hole transfer. The carriers in the surface state which are transferred to bulk bands represent initially a wave packet at the surface which spreads and propagates into the bulk material with its Fermi velocity v_F of about 1 nm/fs²⁸. Thus, a spin-polarized current which propagates from the surface to the bulk is optically excited. In the vicinity of the surface this current propagates ballistically and consists of electron and hole contributions. Spin-down holes have the same spin polarization as spin-up electrons. Therefore, the spin components of these two contributions will add up. If the charge of electrons and holes is in sum zero we conclude that a net spin current between surface and bulk is excited. It is likely that the efficiency of electronic transitions in spin-up and spin-down channels are different (Fig. 1, bottom) which will lead to a prevalence of either the electron or hole component of the spin-polarized current. Note that this does not necessarily imply charging of the surface because a transient charge imbalance will be screened by electron rearrangement on the time scale of the inverse plasma frequency²⁹. Note that on the basis of these considerations an effect due to optically excited transfer of spin polarization between surface and bulk should proceed within few femtoseconds, i.e. well within the time resolution of the SHG experiment.

As seen in Fig. 1, top panel, the pronounced initial drop in the surface sensitive magneto-optical signal occurs within the experimental time resolution and clearly faster than the bulk demagnetization. Therefore, we explain the pronounced difference in the bulk and surface demagnetization times by spin transfer between surface and bulk of the film. The time scale at which the magnetic SHG signal $\Delta_{\text{odd}}^{2\omega}$ changes from 0 to -0.5 at time zero is in agreement with a ballistic character of these transport effects.

The pump-induced reduction of the relative magneto-optical signals differ by a factor of six. We consider the resonant surface excitation among bulk and the surface states near $\bar{\Gamma}$, see Fig. 1, as essential for an explanation of the pronounced effect at the surface.

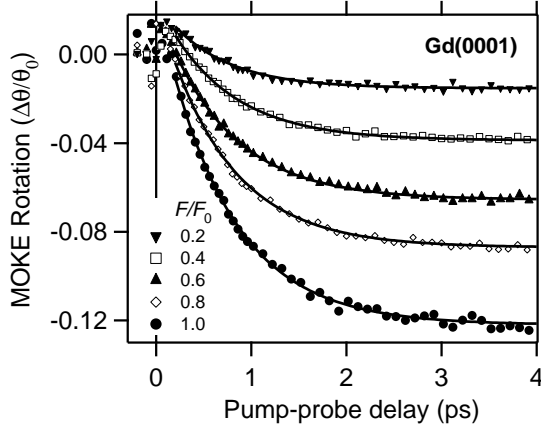


FIG. 2: Symbols indicate experimental data of time-resolved MOKE for different relative pump fluences F/F_0 , with $F_0 \approx 1 \text{ mJ/cm}^2$. The solid lines are fits considering a single exponential decay.

B. Fluence dependent demagnetization dynamics in the bulk

The setup used for the present study allowed in addition a more detailed analysis of the bulk demagnetization phenomenon. Using a combination of half wave plate and Glan-Thomson polarizer the absorbed fluence has been reduced step wise from the maximum value $F/F_0 = 1$, F_0 was determined to be $1.0 \pm 0.3 \text{ mJ/cm}^2$. Fig. 2 shows representative time-resolved MOKE curves for different F/F_0 up to delay times of 4 ps. The values at 4 ps decrease linearly with F/F_0 . This ensures that we are analyzing a low excitation density regime reasonably far away from a full demagnetization of the sample, where magnetic fluctuations would contribute to the ultrafast magnetization dynamics^{13,21}. Near time zero the data feature a less clear behavior and an effectively positive contribution in $\Delta\theta/\theta_0$. The time delay at which the signal crosses the zero line shifts with increasing fluence closer to time zero. While for $F/F_0 = 0.2$ the amplitude of the positive and negative contributions are comparable to each other, the negative one dominates for larger F/F_0 . In this study we focus on the pronounced demagnetization dynamics, i.e. at the fluences where the negative contribution dominates. The origin of the positive contributions are currently under discussion since they could originate from transient changes in the magneto-optical constants or the spin polarization of the $5d$ electrons.

All data were fitted by a single exponential time dependence with fixed time zero and variable $\Delta\theta/\theta_0$ at 0 and 4 ps. The obtained fits are shown in Fig. 2 and describe the

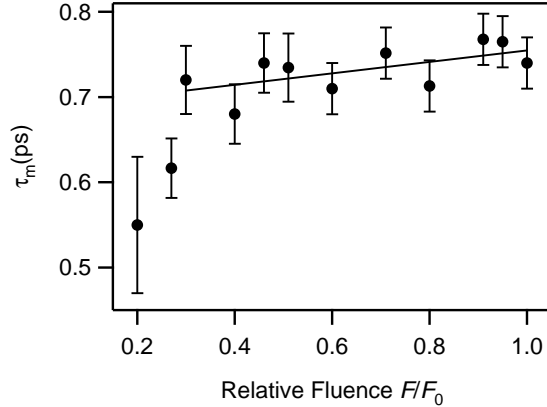


FIG. 3: Characteristic times determined from the decay times in the single exponential fitting of $\Delta\theta/\theta_0$ as a function of relative fluence. The line is a linear fit to the data with $F/F_0 \geq 0.30$

experimental data well. Fig. 3 depicts the time constants τ_m determined by the fitting procedure. The numbers range from about 0.5 to 0.8 ps with a trend towards larger times for higher F/F_0 . The smallest τ_m are obtained for the lowest $F/F_0 = 0.20$ and 0.27 and the respective τ_m are out of the weak linear increase observed for $F/F_0 \geq 0.30$. We already mentioned that for such small F/F_0 the positive signal near time zero is comparable in size to the demagnetization observed at later delays. It is therefore well possible that the obtained τ_m is influenced by the processes that are responsible for the positive $\Delta\theta/\theta_0$ and we refrain from further conclusions based on τ_m obtained for $F/F_0 < 0.30$.

We fitted the values for $F/F_0 \geq 0.30$ by a linear dependence. The result is shown in Fig. 3 by a line. We find that the demagnetization time increases by 67 ± 30 fs within $F/F_0 = 1$ with a nominal zero fluence limit of $\tau_m = 690 \pm 20$ fs.

For itinerant ferromagnets like Ni or Co an observed increase in τ_m with fluence in combination with a finite value at zero fluence was explained by model descriptions based on Elliott-Yafet scattering¹² and Stoner excitations¹³. The case of Gd has been discussed in Ref.¹². It features two separate demagnetization times in agreement with the experimental observation²⁰. These two time scales are attributed (i) to cooling of the hot electron system through e-ph coupling for the fast time scale, which is discussed in the present article, and (ii) to the demagnetization determined by the equilibrium spin-flip probability, which is weaker in Gd than for $3d$ transition metal ferromagnets and sets the slower timescale observed in Refs.^{20,30,31}.

This description predicts a fluence dependence of the ultrafast demagnetization of Gd that is determined by the increase of the transient lattice temperature T_1 . We have calculated $T_e(t)$ and $T_1(t)$ for different fluence by the well known two-temperature model³² in refined versions, which were published earlier^{33,34}; regarding its application to Gd(0001) see¹⁷. Considering a variation in fluence following the experimentally investigated range we find a pronounced variation of the time scale on which $T_e(t)$ and $T_1(t)$ equilibrate (not shown). This change in time scale is at a first glance more pronounced than the changes found for the demagnetization time in Fig. 3. However, a systematic analysis of the time scale at which $T_e(t)$ and $T_1(t)$ are changed is non-trivial since they vary in a non-exponential way. Therefore, we turn to a discussion of the excess energy ϵ of the electronic system which is related to the electron temperature through $\epsilon = \gamma T_e(t)^2$, with γ being the linear parameter in the temperature dependent specific heat of the electron system. Fig. 4 shows the results obtained for $\epsilon(t)$ with F/F_0 varying between 0.2 and 1. The transient behavior can be described by a single exponential that represents energy transfer from the electron system to the lattice. The exponential time scales were determined with 100–800 fs and are plotted in the inset of Fig. 4. The energy transfer time shifts to larger values for higher fluence and changes by three times in the investigated fluence range. Consequently the energy content of the lattice changes with these varying time constants.

The comparison between time-dependent demagnetization curves and the time evolution of excess energy emphasizes two points which lead to the conclusion that the explanation of the ultrafast demagnetization in Gd solely through the time-dependent electron or lattice temperature is incomplete. At first we argue that the range of the energy transfer times τ_{e-ph} found in the investigated fluence range is with 0.2–0.6 ps considerably broader than the range observed for τ_m which is about 0.65–0.8 ps (see Figs. 3,4). Second, and maybe more general, the demagnetization can be described by a simple exponential time dependence similar to the transient energy density. In contrast, the transient electron and lattice temperatures follow a more complicated evolution. Moreover, spin fluctuations which present a sizeable energy content of the system in particular in excited magnetic systems³⁵, might be essential to take into account.

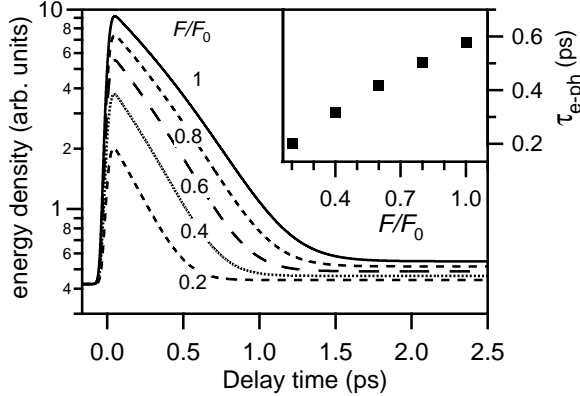


FIG. 4: Main panel: Time-dependent excess energy density for different relative fluence, which was calculated by the two-temperature model. The inset shows the characteristic times τ_{e-ph} of energy transfer from the electronic system to the lattice mediated by electron-phonon scattering which was determined by single exponential decay to the excess energy density in time interval from 100 to 800 fs.

C. Discussion

Our pump-probe analysis of fs laser-induced magnetization dynamics in Gd(0001) revealed two qualitatively different mechanisms, which both change transiently the magnetic moment per atom. The one mechanism is transfer of angular momentum from the spin system to another degree of freedom of the sample, which is finally the lattice. In this case the magnetic moment of the sample as a whole is reduced. Such mechanisms are discussed in the literature^{12,13} to offer consistent descriptions of ultrafast demagnetization as observed in several time-resolved magneto-optical studies, see e.g. Ref. ⁷, and x-ray magnetic-circular dichroism (XMCD) experiments⁸ for Ni. The observed demagnetization time of about 700 fs determined for Gd(0001) can be explained by relaxation of the optically excited electrons by interactions with phonons or spin waves which subsequently interact with the lattice. On a more microscopic level the demagnetization process in Ni and Gd must be different, because in a transition metal the magnetic moment is formed in the same band which is also optically excited. In a lanthanide like Gd the dominant part of the magnetic moment is localized near the ion core and resides in the $4f$ level, which is not primarily optically excited²⁰. To achieve significant demagnetization the excess energy generated by the laser excitation must be transferred to the $4f$ electrons. While details of this process will be discussed in a

forthcoming publication, we note here that optically excited $5d$ electrons are coupled to the localized $4f$ electrons through intraatomic exchange interaction. Ultrafast changes of the magnetization, which are investigated by MOKE, can be expected to be dominated by the $5d$ contribution to the magnetic moment. The femtosecond XMCD study performed at the Gd M_5 edge probes the magnetic moment of the $4f$ levels directly²⁰. A comparison of the time scales of demagnetization obtained in both these experiments suggests that the $4f$ and $5d$ contribution to the magnetic moment are essentially strongly coupled on ultrafast time scales accessible here and demagnetize as a total magnetic moment.

The second mechanism investigated here is spin transfer which we probe by means of the exchange-split spin polarized $5d_{z^2}$ surface state of Gd(0001). Here the magnetic moment integrated over the sample remains constant, but is redistributed between different parts of the sample, in our case among surface and bulk electronic states. We explain this process to originate from the optical transitions that couple the exchange-split surface and bulk states. In the literature a similar scenario has been discussed recently as a potential explanation for bulk demagnetization in Ni¹⁴. In this theoretical study the authors consider the optically excited transport of hot carriers in metallic samples which redistribute the magnetic moment from the ferromagnetic layer into a para- or diamagnetic substrate. Such transport effects of hot carriers are known since early investigations of femtosecond electron dynamics in metallic layers²⁸, however, to what fraction they are indeed responsible for the ultrafast demagnetization remains an interesting question which is to be clarified in future investigations.

IV. CONCLUSIONS

By combining ultrafast bulk and surface sensitive magneto-optical techniques we have analyzed the femtosecond laser-induced demagnetization of epitaxial Gd(0001) films. In the bulk the demagnetization occurs with a characteristic time of about 0.7 ps. Considering that the surface sensitive signal changes within the laser pulse duration of 35 fs and taking resonant optical transitions between valence electronic surface and bulk states into account, we attribute this ultrafast change to transfer of spin-polarized charge carriers between surface and bulk states. Variation of the pump fluence up to 1 mJ/cm² shows a weak increase in the bulk demagnetization time of about 70 fs/(mJ/cm²). A comparison with the fluence

dependence expected from the energy transfer among electrons and the lattice revealed that albeit there is qualitative agreement in the increasing trend of demagnetization time such description remains incomplete due to pronounced quantitative variations.

Acknowledgments

We are grateful for the continuous support by M. Wolf and acknowledge fruitful discussions with O. Chubykalo-Fesenko. This work was supported by the Deutsche Forschungsgemeinschaft through ME3570/1 and by the HEC-DAAD.

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