Ultrafast dynamics of a coherent phonon-magnon mode at the Gd(0001) surface

Uwe Bovensiepen and Alexey V. Melnikov

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

ABSTRACT

The Gd(0001) surface is investigated by femtosecond pump-probe experiments using laser pulses at 740–860 nm wave length. By non-linear optical second harmonic generation a coherent phonon-magnon mode at a frequency of 3 THz is observed which is excited through the exchange-split surface state. In parallel, electron-electron and electron-phonon interaction and their magnetic counterparts lead to incoherent dynamics of the electron, lattice, and spin subsystems. Variation of the optical wave length shows that for longer wave lengths up to 860 nm the coherent mode is excited predominantly while for shorter ones ($\geq$ 740 nm) incoherent contributions are favored. This presents a strong indication that depopulation of the occupied surface state component drives the coherent excitation. We find identical time scales for damping of the coherent mode and for electron-lattice equilibration which identifies electron-phonon scattering as an important relaxation channel for the coherent contribution. Increasing the temperature results in faster damping indicating that scattering of coherent phonons with thermal ones is an active relaxation channel as well.

Keywords: ultrafast phenomena, time-resolved optical spectroscopy, coherent excitations, non-equilibrium dynamics

1. INTRODUCTION

Phonon-magnon scattering is considered as a major relaxation mechanism of excitations in a ferromagnet.[1] For low energy excitations the linear phonon dispersion relation and the quadratic one of magnons lead to a crossover region where magnons and phonons exist with the same wave vector. The true normal modes of the system are admixtures of spin waves and phonons which in a quasi particle picture can be considered as an one-phonon one-magnon scattering process. However, quantitatively the amplitudes of these admixtures are determined by the coupling strength of the spin and lattice excitations which has been attributed to magneto-elastic effects based on spin-orbit interaction.[2]

In rare earth ferromagnets the magnetic spin moment is dominated by the strongly localized 4f electrons ($\mu_{Gd}^{4f} = 7\mu_B$; $\mu_{Tb}^{4f} = 6\mu_B$). Itinerant contributions are generated by spin polarization of the valence band through 4f electrons. The itinerant magnetic moment μ^{5d6s} is about one order smaller and is responsible for ferromagnetic order through indirect exchange coupling. Inelastic neutron diffraction studies showed that for Tb with its 4f orbital moment of L=3 crossing of magnon and phonon dispersion curves is avoided and mode mixing occurs.[3] However, for Gd the 4f orbital moment and, accordingly, spin-orbit coupling vanishes due to a half filled 4f shell. The observation of an absence of avoided crossings in the Gd dispersion curves has been taken as evidence that phonon-magnon interaction is generated by spin-orbit coupling.[2]

In the present contribution we follow a conceptionally different approach to study the interaction of phonons and magnons. A coherent optical phonon is launched by optical excitation with a femtosecond laser pulse at the Gd(0001) surface which modulates in the time domain optical and electronic properties at the phonon frequency of 3 THz. We find a quasi instantaneous coupling of the lattice vibration to the magnetization which oscillates at the same frequency. The well defined phonon mode enables a detailed investigation of the spin dynamics coupled to this coherent lattice vibration and allows us to discuss excitation and coupling mechanisms.

Further author information: www.physik.fu-berlin.de/∼femtoweb
E-mail: uwe.bovensiepen@physik.fu-berlin.de, telephone: +49 30 838 53340

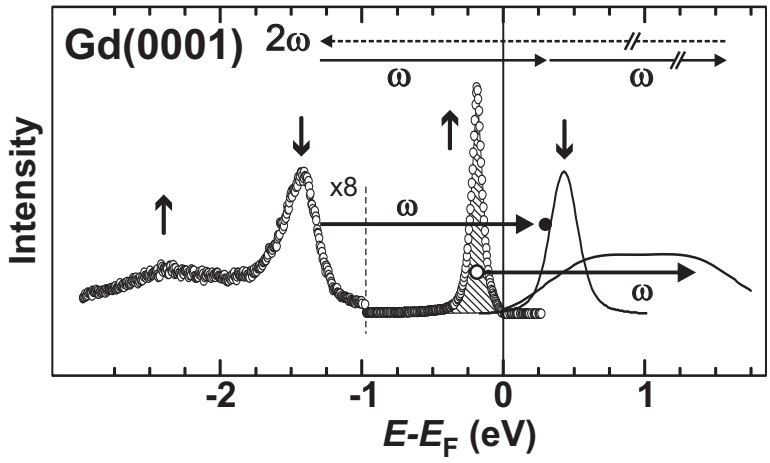

Figure 1. Valence electron states of Gd(0001) at the $\overline{\Gamma}$-point for 90 K taken from photoemission, inverse photoemission, and scanning tunneling spectroscopy data.[18] The exchange-split $5d_{z^2}$ surface state appears around the Fermi level with its majority (↑) and minority (↓) contribution in addition to bulk bands. Indicated are the two main absorption channels for 1.52 eV pump photons and the SH probing scheme.

Coherent optical phonons represent a well-established field in case of semiconductors and semimetals.[4–8] The involved phonon frequencies are typically several THz. For metals, on the other hand, coherent phonons have been observed only recently for the Gd(0001) surface[9] and for several 3d transition metals.[10]

Investigation of coherent spin dynamics in ferromagnets has been strongly advanced by the demand for fast magnetic switching in data storage devices. Coherent acoustic spin waves permit switching rates of a few GHz, as demonstrated by experiments using picosecond magnetic field pulses.[11–14] Ultrashort laser pulses increase the time-resolution to the femtosecond range and optically excited magnetic excitations at higher frequencies in the GHz regime have been studied.[15,16]

At the Gd(0001) surface we recently succeeded to excite coherent spin dynamics at a considerably higher frequency of 3 THz by coupling of the magnetization to an optically induced lattice vibration.[9] This approach required a time-resolved and surface sensitive probe which is able to distinguish between spin and phonon excitations. Detection of the optical second harmonic (SH) in pump-probe experiments fulfills this requirement because it is intrinsically surface sensitive and consists of contributions which are even and odd with respect to magnetization reversal.[17]

In our optical experiment the electronic structure of the valence band is investigated. As illustrated in Fig. 1, it exhibits exchange split bulk states and an exchange split localized $5d_{z^2}$ surface state[19] with an occupied majority (↑) and unoccupied minority (↓) component around E_F. In an earlier work we reported that SH generation (SHG) proceeds resonantly enhanced through the unoccupied surface state component because the fundamental photon energy of 1.5 eV nearly matches the energetic separation of the minority bulk band and the unoccupied surface state component[20] which make the non-linear optical probe particularly surface sensitive. Note that it has been shown for equilibrium conditions that at finite temperatures spin-mixing of these exchange-split components lowers the spin-polarization of the surface.[21] Moreover, the exchange splitting decreases with increasing temperature but remains finite at 0.4 eV at the Curie temperature of 293 K while the spin polarization is lost.[21,22]

2. EXPERIMENTAL

Besides its extreme surface sensitivity, SHG allows for the simultaneous detection of electronic and magnetic contributions. Since the electronic structure varies with lattice vibrations this technique is well suited for probing both the lattice and the spin dynamics. The SH intensity reflected from the magnetic surface is composed of two fields which behave even or odd with regard to magnetization reversal[17]:

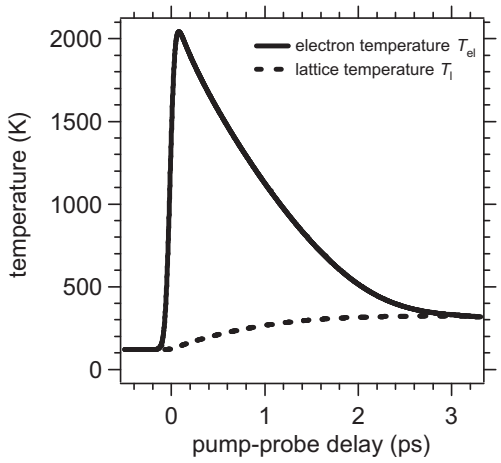

Figure 2. Transient evolution of electron and lattice temperature in Gd after optical excitation with 1.5 eV photon energy at a fluence of 1 mJ/cm^2 according to a two-temperature model. Model parameters have been optimized to describe the time-dependence of the electronic distribution function observed with time-resolved photoemission.[23, 24] The initial rise of the electronic temperature T_{el} is followed by a decay which is accompanied by an increasing temperature of the lattice T_l due to electron-phonon interaction. After 3 ps an equilibrium between electrons and phonons has been established.

$$I^{\uparrow,\downarrow}(2\omega) \propto E^2_{even}(2\omega) + E^2_{odd}(2\omega) \pm 2 \cdot E_{even}(2\omega)E_{odd}(2\omega) \cdot \cos(\phi). \tag{1}$$

The arrows indicate opposite magnetic field directions and ϕ is the relative phase between even and odd SH fields. In time-resolved experiments pump-induced variations are expressed by

$$D^{\pm}(t) = \frac{I^{\uparrow}(t) \pm I^{\downarrow}(t)}{I^{\uparrow}(t_0) \pm I^{\downarrow}(t_0)}, \tag{2}$$

where t_0 denotes a negative delay, i.e. before the pump pulse has arrived. Since $E_{odd} \ll E_{even}$ and $\cos(\phi) \approx 1$, the pump-induced variations Δ can be determined for E_{odd} and E_{even} separately:

$$\Delta_{even}(t) = \sqrt{D^+(t)} - 1 = \frac{E_{even}(t)}{E_{even}(t_0)} - 1; \quad \Delta_{odd}(t) = \frac{D^-(t)}{\sqrt{D^+(t)}} - 1 = \frac{E_{odd}(t)}{E_{odd}(t_0)} - 1. \tag{3}$$

The time-dependent data will be presented in terms of $\Delta_{even}(t)$ and $\Delta_{odd}(t)$ which measure the pump-induced electron/lattice and magnetization dynamics, respectively. The experimental scheme is sketched in the inset of Fig. 3a. Measurements were carried out with 20 nm thick Gd films, grown epitaxially in ultrahigh vacuum (base pressure $\approx 3 \cdot 10^{-11}$ *mbar*) on a W(110) substrate at 330 K with subsequent annealing at 700 K which leads to smooth ferromagnetic films. The optical response was measured *in situ* in a transversal magneto-optical configuration with films saturated in a magnetic field of 500 Oe applied along the easy axis of magnetization oriented in the film plane perpendicular to the optical plane of incidence. P-polarized laser pulses of 35 fs duration at 740-860 nm wavelength were produced by a cavity dumped Ti:sapphire oscillator. Output pulses with pulse energy of 42 nJ were split 4:1 in pump and fundamental probe pulses. The P-polarized SH intensity was separated from the background of the fundamental wave length by a monochromator and recorded by single photon counting.

3. RESULTS AND DISCUSSION

Absorption of a fs-laser pulse in a metal generates hot electron-hole pairs. Under typical excitation densities of $\sim$1 mJ/cm^2 a thermalized electron gas is formed within the first several 100 fs through electron-electron

scattering. The respective temperatures of the electron gas T_{el} are much higher than in thermal equilibrium. Due to electron-phonon interaction heat is transferred subsequently from the electronic to the lattice subsystem which takes several picoseconds. In addition spatial redistribution of the excitation energy leads to ballistic and diffusive transport effects.[23, 25, 26] In a simplified picture neglecting non-thermalized electrons and interaction with further quasiparticles the dynamics can be described by the two-temperature model (2TM) where the electron and lattice subsystems are characterized by two temperatures T_{el} and T_l. The dynamical evolution of these temperatures is described by the electron and lattice heat capacities which are coupled by electron-phonon interaction. We have investigated the transient evolution of T_{el} for Gd(0001) with time-resolved photoemission by analysis of the Fermi-Dirac distribution as described for the case of Ru in detail in Ref. 23. In Fig. 2 we show the evolution of T_{el} and T_l according to a 2TM simulation which is optimized to reproduce the transient distribution function measured by photoemission. The electron temperature peaks at a delay of 100 fs and decays to 300 K while simultaneously the lattice temperature increases from the static temperature of 120 K to 300 K. Thus both subsystems have reached their equilibrium after 3 ps. Further details on the time-dependent photoemission studies on Gd are subject to a future publication,[24] and data presented here serve as an independent measurement of incoherent electron and lattice dynamics which will be important to analyze the transient optical data presented in the following.

An exemplary set of time-dependent SH optical data are given in Fig. 3. We start the description in the top of panel (a) which displays the pump induced variation of the even SH part and the linear reflectivity. Within the first 3 ps the pump-induced variation Δ_{even} presents oscillations with an amplitude of several percent of the equilibrium value superimposed on a monotonous increase. The latter is the signature of incoherent electron-electron and electron-phonon scattering discussed in Fig. 2. On the other hand the intense oscillations are not obvious for the linear reflectivity $\Delta R/R$ which suggests, owing to the surface sensitivity of SHG, that the oscillating part arises at the surface.[27] As seen from the bottom panel of Fig. 3a, we find a comparable behavior for the spin dynamics measured by the odd SH component. The instantaneous drop is generated by ultrafast demagnetization due to spin-dependent scattering processes. Relaxation back to thermal equilibrium occurs on time scales of spin-lattice relaxation within several 100 ps, as reported in Refs. 28, 29. Interestingly, an oscillating contribution is observed for the spin dynamics as well. The oscillating contribution after subtraction of the incoherent part is shown for Δ_{even} and Δ_{odd} in Fig. 3b. For clarity, data have been averaged as described in Ref. 9. Fourier transformation shown as an inset identifies a frequency of 2.9±0.3 THz for the even and odd contribution. The oscillations have been fitted by an exponentially damped periodic function including a frequency increasing linearly with time as discussed in Ref. 30.

In the following part we focus on the origin of these coherent dynamics. The phonon dispersion which has been calculated by Rao and Menon for bulk Gd[31] presents a longitudinal optical (LO) phonon along [0001] with a frequency of 3.15 THz which is close to the one observed in our experiment. Thus, we interpret the oscillations in $\Delta_{even}(t)$ to occur from a coherent optical phonon at the Gd(0001) surface excited by the fs-laser pulse. The slightly smaller frequency compared to the calculated value might well result from the lower coordination number at the surface compared to bulk. The vibration symmetry of the LO derived mode at the surface is such that the (0001) surface plane vibrates with respect to the underlying bulk. Since we are sensitive to lattice excitations by the even SH field, the oscillations in Δ_{even} are therefore attributed to coherent phonons.

3.1. Coupling of lattice and spin quasi-particle excitations

In the next step we proceed to the coherent magnetic dynamics observed in Δ_{odd}. Since this oscillating variation presents the same frequency and a constant phase shift with respect to Δ_{even}, we conclude that these coherent lattice and spin dynamics present a coupled mode. Therefore, we need to consider a reasonable coupling between the two subsystems. Spin-orbit interaction presents a possible candidate as discussed in section 1. However, in case of Gd it is weak (16 μeV for bulk, see Ref. 32), and is thus unlikely to transfer the surface vibration to the spins coherently. Since the amplitude of the magnetic oscillations does not show a built-up time within our time resolution and follows the lattice directly after the excitation, we expect a stronger interaction to be responsible. Eventually the relevant coupling strength even changes during the periodic variation of the lattice spacing along the surface normal. We consider the exchange interaction J to mediate the phonon-magnon coupling and to drive the ultrafast coherent spin dynamics. The strength of J is determined by the wave function overlap of neighboring sites. Due to the d_{z^2} symmetry of the surface state probed predominantly by SHG, the exchange

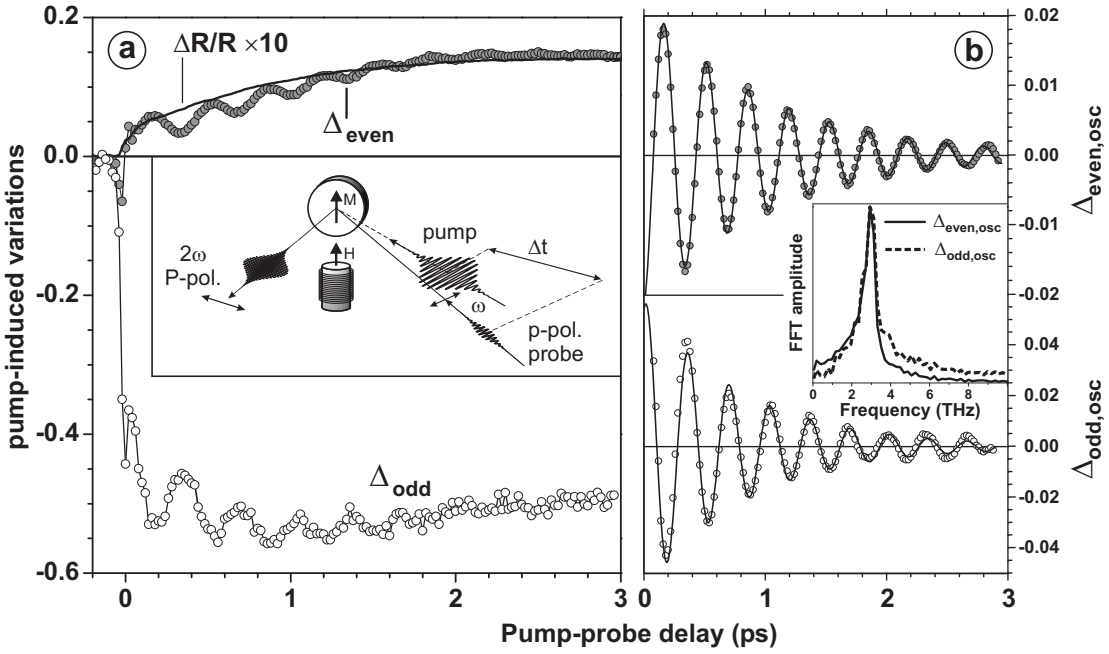

Figure 3. (a) Time dependence of the even (upper panel) and odd SH (lower panel) response measured as a function of pump-probe delay for a Gd(0001) film of 20 nm thickness at T=90 K using 815 nm/35 fs laser pulses. Plotted are the quantities defined in Eq. 3. The linear reflectivity $\Delta R/R$ is displayed for comparison in the upper panel by the solid line. The inset shows the experimental scheme of the transversal magneto-optical geometry where the magnetization is oriented perpendicular to the optical plane of incidence. (b) Coherent part of the even and odd SH fields depicted in (a) after smoothing and subtracting of the incoherent part. Solid lines represent a fit to the data. In the inset, Fourier transformations of both oscillating contributions are given.

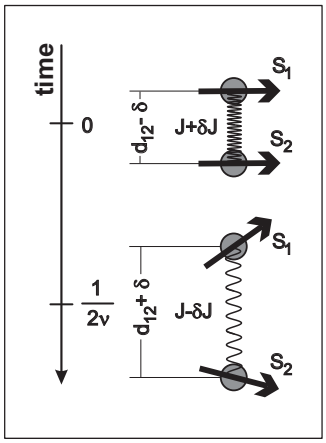

Figure 4. Illustration of the parametric coupling mechanism between the distance of surface to subsurface layer d_{12} and individual spins $S_{1,2}$. An increase in the lattice spacing by δ lowers the exchange by δJ, and the spin orientation can deviate from the parallel alignment. An increase in J enhances the spin ordering, respectively.

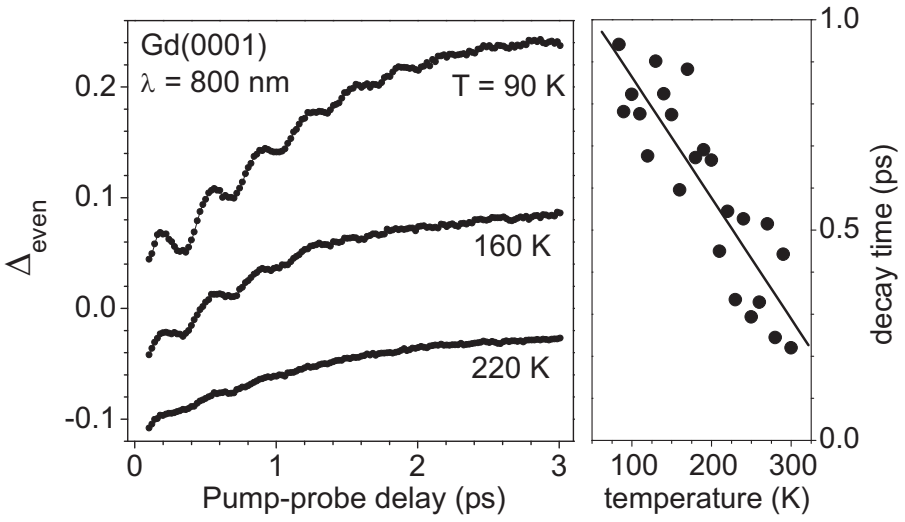

Figure 5. Left panel: Transient pump-induced variation of the even SH field Δ_{even} for different static temperatures. Right panel: Damping time of the oscillating even SH component determined from the fit to $\Delta_{even,osc}$ (c.f. Fig. 3) as a function of temperature. The solid line guides the eye.

strength along the surface normal will depend on the lattice spacing between the surface and the subsurface plane, i.e. $J = J(d_{12})$. This spacing d_{12} is varied coherently through the coherent phonon by a length δ. Consequently, J is enhanced and weakened by an amount δJ, depending whether d_{12} is smaller or larger than its value in thermal equilibrium. In a Heisenberg picture, neighboring spins in the first and second layer are due to this periodic change in J more or less aligned, as is illustrated in Fig. 4. An experimentally observable quantity showing this variation is the surface spin polarization which is thus expected to vary periodically. This agrees well with the experimental observation in the dynamical evolution of Δ_{odd} (see Fig. 3). Furthermore, we consider this coupling to be parametric because J, which is a parameter of the spin ordering and also of the spin wave spectrum, is varied as a function of time. Thereby, we have identified a new phonon-magnon interaction mechanism at the Gd surface which does not originate from spin-orbit coupling but from a coherent variation of the exchange interaction.

3.2. Damping of the coherent excitation

Various decay channels for the coherent surface mode are to be considered.[33] (a) phonon-phonon scattering: A coherent phonon will loose its phase if it scatters with a thermal phonon and will no longer contribute to the coherent mode. (b) Electron-phonon interaction is responsible for the heat-transfer from the electron gas excited by the fs-laser pulse to the lattice as discussed above (c.f. Fig. 2) and thus we consider the inverse process – interaction of coherent phonons with the electron system – as a further decay channel. For separation of these two contributions we carried out temperature-dependent studies depicted in Fig. 5. As visible from the data for Δ_{even}, the coherent lattice dynamics are affected by an increase in temperature in two ways. The initial oscillation amplitude is reduced which is attributed to changes in the excitation probability (see Ref. 9). A fit of the oscillating contribution introduced above (Fig. 3) shows a decrease of the damping time by a factor of four if the temperature is raised from 90 to 300 K. Thus, scattering of coherent phonons with thermal ones represents a significant contribution to damping of the coherent mode. On the other hand at 90 K the incoherent electron-phonon interaction proceeds on the same time scale of 0.9 ps as the damping. This has already been pointed out in an earlier paper[30] and is corroborated in the present paper by the very comparable time constant of the increase in the transient lattice temperature analyzed by the 2TM based on time-resolved photoemission (c.f. Fig. 2). This means that at low temperatures electron-phonon interaction is the dominant contribution to damping. Thus, one might expect that the damping time saturates at low temperature. However, the finite temperature range of the presented data hinder a more detailed analysis at present.

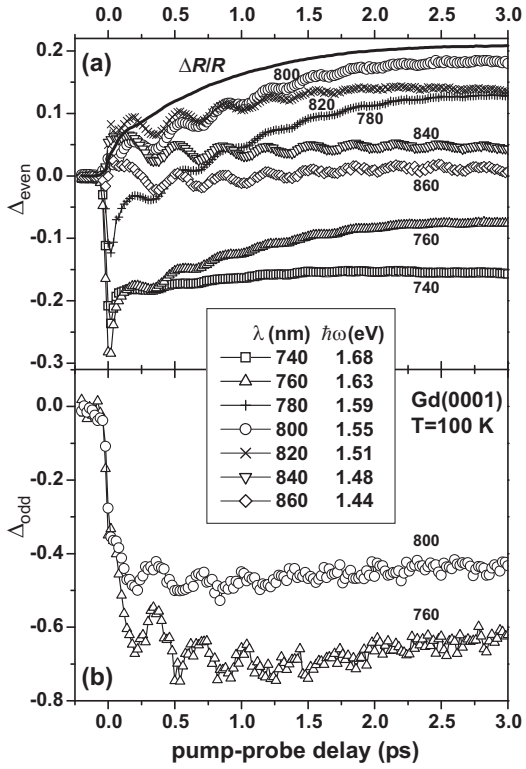

Figure 6. Pump-induced variations of SH fields during the first 3 ps for various wave lengths between 740 and 860 nm. All data are normalized to zero at negative delays. (a) The even SH field presents a well discernible spectral dependence of the coherent phonon contribution and of the monotonous increase after the electron system has thermalized at delays > 200 fs. The solid line represents the dependence of the linear reflectivity which is independent on wave length. (b) The odd field is a measure of the spin polarization at the Gd(0001) surface which consists of coherent and incoherent contributions as well. The dominant spectral dependence here is found in the minimum value of Δ_{odd}.

3.3. Optical excitation of the coherent mode

Finally we turn to the excitation mechanism of the coupled coherent mode. Following the above presentation of the time-dependent data, relaxation through thermal (i.e. incoherent) and coherent phonons present competing processes subsequent to excitation of the electronic system. Understanding which optical excitation in the rather complex electronic structure shown in Fig. 1 favors the coherent or the incoherent relaxation might further the insight into the underlying principles. For this purpose we have varied the laser wave length. Respective time-dependent optical response curves are depicted for Δ_{even} and Δ_{odd} in Fig. 6. The linear reflectivity $\Delta R/R$ is independent on the laser wave length. Due to the pronounced surface sensitivity of SHG this observation confirms that the surface state, which acts as an intermediate level (see Fig. 1) in the nonlinear optical process, determines the spectral variation of the transient response rather than initial or final bulk states probed by $\Delta R/R$.

The spectral dependence of the time-dependent even SH response shown in the upper panel of Fig. 6 contains two trends for the incoherent and the coherent contribution with opposite wave length dependencies. While with increasing wave length the amplitude of the coherent contribution increases, the opposite holds for the incoherent part. For longer wave length an increasing reduction of the incoherent pump induced change which is quantified by the absolute value of the pump-induced variation at 3 ps compared to negative delays[34] is found. That means with higher photon energy incoherent contributions are excited predominantly, while for smaller $h\nu$ the coherent mode is enhanced. If we consider the electronic structure of the Gd(0001) surface, we find that with smaller photon energy the efficiency of excitations in the minority channel is reduced because the resonance condition

from the bulk minority to the surface minority state is less fulfilled. The excitation probability of the majority channel is assumed to present a considerably weaker spectral dependence as it starts from the occupied surface state component into the energetically rather broad unoccupied bulk band.

This pronounced spectral dependence suggests the following scenario to explain the observed behavior. For *longer* wave length the majority excitation channel is dominant. It creates a reduction in the spin polarization and of the electron density at the surface (see Fig. 1). Both remain on time scales of their respective relaxation processes which are in case of electrons given by the screening time on the order of fs (i.e. within the pulse duration). The screening results in an effective charge redistribution in the near surface region which relaxes by e-e scattering among surface and bulk states leading to a hot distribution equilibrated among surfaces and bulk. Decay of the excited spin density might evolve on slower time scale by spin-dependent scattering. In the case of *shorter* wave length the minority and majority excitation channels are both active. That means the spin density at the surface is reduced further compared to longer wave length because holes in the majority surface state component and electrons in the minority component lower both the spin polarization at the surface. This is corroborated by the observation of a higher loss of spin polarization for the shorter wave length seen in Fig. 6. Considering the transient charge distribution at the surface, electrons are excited to the unoccupied surface state component and, at the same time, holes are created in the occupied part by excitation into unoccupied bulk states. Thus, a weaker excess charge is generated and a correspondingly smaller charge redistribution is required for screening near the surface. In our earlier publications[9,30] we have explained the excitation of coherent phonons by such a process which is driven by charge redistribution.

Alternatively to this charge driven mechanism, we consider that the pump-induced variation of the surface spin polarization, which is lowered upon laser excitation by more than 50% (see Fig. 6), drives the coherent relaxation. For a ferromagnetic system the screened ion potential is spin-dependent as has been investigated by analysis of the extended X-ray absorption fine structure of Gd.[35] This work reports a considerable spin-dependent variation of the local screened ion potential on the order of half of the exchange interaction. This means that the ion lattice is coupled to the spin system, and it would be well conceivable that the coupled coherent phonon-magnon mode is generated by a change of the surface spin polarization.

Nevertheless, we favor a charge driven process due to the following argument based on the spectral dependence of the dynamics depicted in Fig. 6. As has been pointed out in this subsection the coherent mode is excited more efficiently by laser pulses with longer wave length. This asks for an excitation process that presents a comparable trend in the spectral dependence. If the loss of spin-polarization generates the driving force for the coherent contribution in the dynamics, the oscillation amplitude would increase for shorter laser wave length. However, the opposite is observed which favors a charge driven process. From Fig. 6 and the above discussion we conclude that the uncompensated excess charge and the resulting screening which is dominant for longer wave length generates the driving force. For shorter wave length, the excess charge is compensated by holes created in the occupied surface state component which might suppress the coherent mode. Albeit this leads to a self-consistent model for the excitation of the coherent mode, theoretical studies of lattice parameters as a function of electron density at the surface and bulk as well as the spin polarization would certainly promote a microscopic understanding of the underlying principles.

4. CONCLUSION

Periodic variations of the transient optical second-harmonic response at a frequency of 3 THz observed on the metallic and ferromagnetic Gd(0001) surface have been identified as coherent phonons derived from an LO phonon in the bulk. Exploiting the different symmetry of magnetic and lattice excitations enabled us to separate spin and lattice dynamics in a single experiment by detecting the second harmonic contributions which are even and odd with respect to magnetization reversal. The spin dynamics present a coherent contribution as well which occurs at the same frequency as the transient variations induced by the coherent phonon part. Thus, we conclude that a coupled coherent phonon-mode is excited optically at the Gd surface. From a certain perspective this efficient phonon-magnon coupling is unexpected because in Gd the phonon-magnon coupling is considered to be very weak due to the small spin-orbit interaction. We propose a different coupling mechanism that is driven through the dependence of the exchange interaction on the lattice spacing varied by the coherent phonon. Damping of the coherent phonon mode occurs through phonon-electron scattering and interaction of coherent phonons with

thermal ones. The excitation is mediated by optically induced electronic excitations among surface and bulk states and the according charge redistribution in the region near the surface leading to a transient change in the lattice constant. This conclusion is based on the laser wave length dependence in the pump-probe experiment (740–860 nm) which shows that in case of shorter wave length incoherent processes and for longer wave length coherent ones are excited predominantly.

ACKNOWLEDGMENTS

It is our pleasure to thank M. Wolf, M. Lisowski, P. Loukakos, I. Radu, K. Starke, G. Kaindl, and E. Matthias for their continuous support and contributions. We acknowledge funding by the Deutsche Forschungsgemeinschaft through SPP 1133.

REFERENCES

1. C. W. Haas and H. B. Callen, *Ferromagnetic relaxation and resonance line width*, vol. 1 of *Magnetism*. Academic Press, 1963.
2. B. Coqblin, *The electronic structure of rare-earth metals and alloys*, Academic Press, London and New York, 1977.
3. S. H. Liu *Phys. Rev. Lett.* **29**, p. 793, 1972.
4. Y. M. Chang, L. Xu, and H. W. K. Tom *Phys. Rev. Lett.* **78**, p. 4649, 1997.
5. H. J. Zeiger, J. Vidal, T. K. Cheng, E. P. Ippen, G. Dresselhaus, and M. S. Dresselhaus *Phys. Rev. B* **45**, p. 768, 1992.
6. G. A. Garrett, T. F. Albrecht, J. F. Whitaker, and R. Merlin *Phys. Rev. Lett.* **77**, p. 3661, 1996.
7. A. I. Lobad and A. J. Taylor *Phys. Rev. B* **64**, p. 180301, 2001.
8. M. Hase, M. Kitajima, S. Nakashima, and K. Mizoguchi *Phys. Rev. Lett* **88**, p. 067401, 2002.
9. A. Melnikov, I. Radu, U. Bovensiepen, O. Krupin, K. Starke, E. Matthias, and M. Wolf *Phys. Rev. Lett.* **91**, p. 227403, 2003.
10. M. Hase, K. Ishioka, J. Demsar, K. Ushida, and M. Kitajima to be published.
11. Y. Acremann, C. H. Back, M. Buess, O. Portmann, A. Vaterlaus, D. Pescia, and H. Melchior *Science* **290**, p. 492, 2000.
12. T. Gerrits, H. A. M. van den Berg, J. Hohlfeld, L. Bär, and T. Rasing *Nature* **418**, p. 509, 2002.
13. H. Weber, S. Riesen, and H. C. Siegmann *Science* **291**, p. 1015, 2001.
14. H. W. Schumacher, C. Chappert, P. Crozat, R. C. Sousa, P. P. Freitas, J. Miltat, J. Fassbender, and B. Hillebrands *Phys. Rev. Lett* **90**, p. 017201, 2003.
15. M. van Kampen, C. Josza, J. T. Kohlhepp, P. LeClair, L. Lagae, W. J. M. de Jonge, and B. Koopmans *Phys. Rev. Lett.* **88**, p. 227201, 2002.
16. J.-Y. Bigot, L. Guidoni, and V. Halté, *Ultrafast Phenomena XIII*, p. 398. Springer Series in Chemical Physics, Springer, 2002.
17. R.-P. Pan, H. D. Wei, and Y. R. Shen *Phys. Rev. B* **39**, p. 1229, 1989.
18. A. Rehbein, D. Wegener, G. Kaindl, and A. Bauer *Phys. Rev. B* **67**, p. 033403, 2003.
19. P. Kurz, G. Bihlmayer, and S. Blügel *J. Phys.: Condens. Mat.* **14**, p. 6353, 2002.
20. A. Melnikov, O. Krupin, U. Bovensiepen, K. Starke, M. Wolf, and E. Matthias *Appl. Phys. B* **74**, p. 723, 2002.
21. K. Maiti, M. C. Malagoli, A. Dallmeyer, and C. Carbone *Phys. Rev. Lett.* **88**, p. 167205, 2002.
22. M. Bode, M. Getzlaff, S. Heinze, R. Pascal, and R. Wiesendanger *Appl. Phys. A* **66**, p. S121, 1998.
23. M. Lisowski, P. A. Loukakos, U. Bovensiepen, J. Stähler, C. Gahl, and M. Wolf *Appl. Phys. A* **78**, p. 165, 2004.
24. M. Lisowski to be published.
25. J. Hohlfeld, S.-S. Wellershoff, J. Güdde, U. Conrad, V. Jähnke, and E. Matthias *Chem. Phys.* **251**, p. 237, 1999.

26. N. D. Fatti, C. Viosin, M. Achermann, S. Tzortzakis, D. Christofilos, and F. Valleé *Phys. Rev. B* **61**, p. 16956, 2000.
27. Note that a much weaker oscillating contribution has been observed also for the linear reflectivity which is attributed to the coupling of the surface to the bulk, as discussed in Ref. 30.
28. A. Vaterlaus, T. Beutler, and F. Meier *Phys. Rev. Lett.* **67**, p. 3314, 1991.
29. A. Melnikov, U. Bovensiepen, I. Radu, O.Krupin, K. Starke, E. Matthias, and M. Wolf *J. Magn. Magn. Mat.* **272-276**, p. 1001, 2004.
30. U. Bovensiepen, A. Melnikov, I. Radu, O. Krupin, K. Starke, M. Wolf, and E. Matthias *Phys. Rev. B* **69**, p. 2004, 2004.
31. R. R. Rao and C. S. Menon *J. Phys. Chem. Solids* **35**, p. 425, 1974.
32. M. Colarieti, S. I. Simak, R. Ahuja, L. Nordström, O. Eriksson, D. Åberg, S. Edvardsson, and M. S. S. Brooks *Phys. Rev. Lett.* **91**, p. 157201, 2003.
33. Here, we focus on the damping of the lattice part. A discussion including the spin dynamics is out of the scope of this paper. We are aware that inelastic and elastic scattering processes lead to depopulation and dephasing, respectively. However, since both result in a reduction of the oscillation amplitude we do not separate between elastic and inelastic processes for the present phenomenological discussion.
34. A. Melnikov, I. Radu, U. Bovensiepen, K. Starke, M. Wolf, and E. Matthias *J. Opt. Soc. Am. B* **in press**, 2005.
35. H. Wende *Rep. Prog. Phys.* **67**, p. 2105, 2004.