Coherent Optical Phonons and Parametrically Coupled Magnons Induced by Femtosecond Laser Excitation of the Gd(0001) Surface

A. Melnikov, I. Radu, U. Bovensiepen,* O. Krupin, K. Starke, E. Matthias, and M. Wolf
Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin-Dahlem, Germany
(Received 14 May 2003; published 26 November 2003)

Coherent spin dynamics in the THz domain coupled to a coherent phonon is observed in the time-resolved second harmonic response of the Gd(0001) ferromagnetic metal surface. An LO phonon of 2.9 THz is excited by a transient charge displacement at the surface caused by resonant absorption of a fs laser pulse in the exchange-split surface state. This lattice vibration modulates the interlayer distance inducing a coherent variation of the exchange interaction between spins in adjacent layers. The resulting magnetization dynamics is considered as optical magnon wave packets coupled to the phonon.

DOI: 10.1103/PhysRevLett.91.227403 PACS numbers: 78.47.+p, 63.22.+m, 63.20.Ls, 75.30.Ds

The interaction between spins and their surrounding lattice has always been of central importance for the understanding of magnetization dynamics. In recent years the availability of ultrashort laser pulses elevated the field to a new quality and time-resolved observations of coherent quasiparticle excitations in the picosecond or subpicosecond domain are by now becoming experimental routine. Nevertheless, the investigation of ultrafast processes progressed rather independently for lattice and magnetization dynamics. Concerning the lattice, investigations of optically excited coherent phonons in semiconductors and semimetals developed into a well-established field [1–5], with involved frequencies of typically several THz. In the field of magnetism [6], on the other hand, investigations of coherent spin dynamics were strongly advanced by the demand for fast magnetic switching in magnetic storage devices. Coherent magnetization precession at acoustic spin wave frequencies permits switching rates of several GHz, as demonstrated by experiments using picosecond magnetic field pulses [7–11]. Femtosecond laser pulses enable generation of spin waves at even higher frequencies of about 10 GHz [12] employing a transient magnetic anisotropy field. A considerably faster scheme has been suggested by Bigot *et al.* to occur for *acoustic* modes at 140 GHz [13].

In this Letter, we demonstrate the use of lattice vibrations as a driving mechanism to coherently excite the spin system in the THz regime by modulating the exchange interaction in a ferromagnet. Realization of this concept required (i) a ferromagnetic system which is suitable for optical phonon excitation and (ii) a time-resolved detection technique which is able to distinguish between lattice and spin dynamics. The first requirement is met by Gd, a ferromagnetic hcp metal with two atoms in the primitive unit cell generating the optical phonon mode. Ferromagnetism occurs by indirect exchange coupling of localized $4f$ spins described by the Heisenberg Hamiltonian $H = -\Sigma_{ij} J(\vec{r}_{ij}) \vec{S}_i \cdot \vec{S}_j$, where neighboring spins $\vec{S}_{i,j}$ are separated by r_{ij} and magnetic coupling is mediated by spin polarized conduction electrons. At the Gd(0001) surface there exists a $5d_{z^2}$ surface state which is exchange split into an occupied majority and an unoccupied minority component [14] (see Fig. 1). As shown below, resonant absorption of 1.5 eV photons leads to coherent lattice vibrations perpendicular to the surface plane. Since $J(r_{ij})$ changes sensitively with the interlayer spacing [15], we expect that lattice vibrations of adjacent (0001) planes will result in a concomitant change of their exchange coupling leading to a modulation of magnetization at the phonon frequency. This process should be considerably faster than the incoherent spin-lattice relaxation at ~10 GHz in bulk Gd reported by Vaterlaus *et al.* [16].

The second requirement was satisfied by measuring the second harmonic (SH) response as a function of time. SH generation allows simultaneous detection of electronic and magnetic contributions to the SH field [17] and is extremely surface sensitive. Since the electronic structure varies with lattice vibrations, this technique is well suited for probing both the lattice and spin

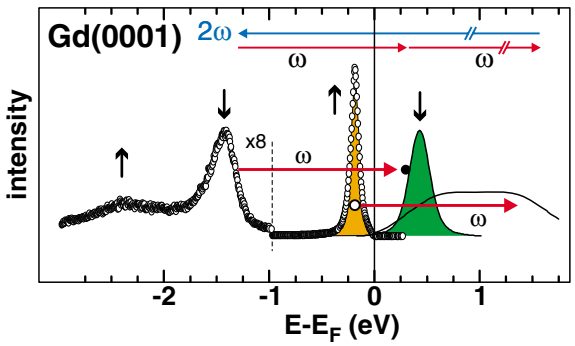

FIG. 1 (color online). Valence electron states in Gd(0001) taken from photoemission ○, inverse photoemission (solid line), and scanning tunneling spectroscopy data above the Fermi level (solid line, filled) [14]. The exchange-split surface state (filled area) appears around the Fermi level in addition to bulk bands. Indicated are the two main absorption channels for 1.52 eV pump photons and the SH probing scheme.

dynamics. SH generation is particularly sensitive to the Gd(0001) surface state 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 (Fig. 1). Thus, the reflected SH is resonantly enhanced by the surface state, as shown in Ref. [18].

The SH intensity reflected from magnetic surfaces is composed of two fields which behave as even or odd with regard to magnetization reversal [17]:

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

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

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

where t_0 denotes a negative delay. Since $E_{\mathrm{odd}} \ll E_{\mathrm{even}}$ and $\cos\varphi \approx 1$, pump-induced variations of even and odd contributions can be separated [19]:

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

and

$$\Delta_{\mathrm{odd}}(t) = \frac{D^{-}(t)}{\sqrt{D^{+}(t)}} - 1 = \frac{E_{\mathrm{odd}}(t)}{E_{\mathrm{odd}}(t_0)} - 1, \quad (4)$$

where $\Delta_{\mathrm{even}}(t)$ and $\Delta_{\mathrm{odd}}(t)$ measure the pump-induced electron and magnetization dynamics, respectively [20].

The experimental scheme is sketched in the inset of Fig. 2(a). Measurements were carried out with 20 nm thick Gd films, grown in ultrahigh vacuum on a $W(110)$ substrate at 330 K with subsequent annealing at 700 K for about 10 min leading to smooth epitaxial ferromagnetic films [22]. The optical response was measured 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 plane of incidence. *P*-polarized laser pulses (35 fs duration, ~800 nm wavelength) from a cavity dumped Ti:sapphire oscillator of about 40 nJ were split 4:1 in pump and fundamental probe pulses. The *P*-polarized SH intensity was recorded by single photon counting.

Typical results for the time dependence of even and odd SH contributions at a temperature of 90 K are displayed in Fig. 2(a) including the variation of the linear reflectivity. Within 3 ps coherent and incoherent processes are clearly discernible. The *linear* reflectivity rises and saturates around 3 ps, reflecting the well known formation of an elevated electron temperature by electron-electron scattering and subsequent equilibration with the lattice by electron-phonon interaction. We attribute the incoherent (nonoscillatory) background of the *even SH* component to these scattering processes in the surface layer, because it follows the general trend of the *linear* reflectivity [23]. Of main interest, however, are the periodic variations in the *even* and *odd SH* components, which are both well resolved in Fig. 2(a), but are not resolved in the linear reflectivity.

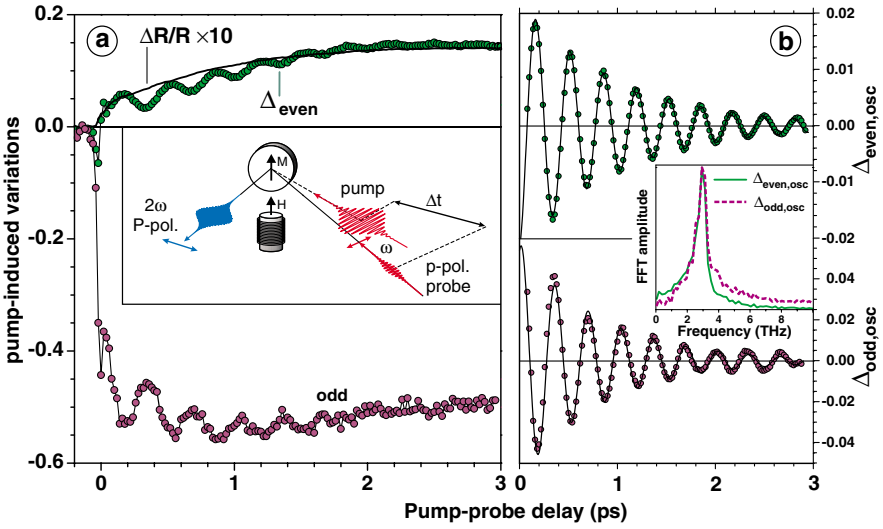

FIG. 2 (color online). (a) Time dependence of the even (upper panel) and odd SH (lower panel) response measured in pump-probe mode for a 20 nm Gd(0001) film at 90 K using 815 nm/35 fs laser pulses. Plotted are the quantities defined in Eqs. (3) and (4). The change of the linear reflectivity is displayed for comparison in the upper panel (thin solid line). The inset shows the experimental scheme of transversal geometry, where the magnetization is oriented perpendicular to the plane of incidence. (b) Coherent part of the even and odd SH fields extracted from (a) after smoothing [21] and subtracting the incoherent background. Solid lines represent a fit to the data. The inset shows the corresponding Fourier transformation.

To extract the frequency the time-averaged background was approximated by a spline function and subtracted from the data. The residual oscillatory part was smoothed [21] leading to the data plotted in Fig. 2(b). A fit by a model of damped oscillations as well as the Fourier transformation (see inset) yields a single frequency of $\nu = (2.9 \pm 0.3)$ THz and a damping time of ~ 1 ps for even and odd fields.

In the following we propose a microscopic mechanism of this coupled lattice and spin excitation. First we discuss the lattice part. The observed frequency matches the optical phonon mode reported in the literature. Calculations for bulk Gd predict a frequency of 3.18 THz for the longitudinal-optical branch at the Γ point (Γ_{3^+} mode) [24]. Since an asymmetric coherent Γ_{3^+} phonon cannot be excited in the bulk [25], we conclude that the observed oscillation originates from a vibration of the surface plane with respect to the underlying bulk. The slightly lower frequency of 2.9 THz compared to bulk calculations is consistent with the reduced coordination of surface atoms.

The displacive excitation of coherent phonons introduced by Ippen and co-workers [3] can serve as a starting point to understand the observed oscillations. In the case of Gd(0001), however, the exchange-split surface state causes pump-induced optical transitions to evolve differently for majority and minority spins (see Fig. 1). While the unoccupied part of the surface state is the final state for minority spin electrons originating from the occupied bulk band, majority spin electrons are excited from the occupied component of the surface state. The photogenerated hole ($\uparrow$) and the excited electron ($\downarrow$) in the surface state have lifetimes which differ by more than a factor of 2, as shown by linewidth measurements [14] ($\Gamma^{\uparrow} = 70$ meV and $\Gamma^{\downarrow} = 175$ meV at 80 K). The lifetime $\tau = \hbar/\Gamma$ of the hole ($\tau^{\uparrow} = 9$ fs) is longer compared to that of the electron ($\tau^{\downarrow} = 4$ fs). Screening of the photohole results in a transient charge redistribution at the surface, which drives the surface plane out of equilibrium along the normal direction. This charge redistribution decays on a 10 fs time scale and leaves the ions in the surface suddenly displaced, which leads to the surface phonon monitored by $\Delta_{\mathrm{even}}(t)$.

The proposed mechanism was verified by an overlayer experiment. Yttrium is a good candidate because it has the same lattice constant and valence electron structure as Gd. Since Y is no ferromagnet, its surface state is not exchange split and located at the Fermi level. Figure 3 shows that the oscillation in the even SH component of bare Gd is damped to half by 1 monolayer (ML) Y and suppressed by 3 ML [26]. Photoemission spectra in the right panel indicate that 1 ML Y reduces the effective exchange splitting since the Y induced peak appears closer to E_F. At 3 ML the Gd peak can hardly be identified. We conclude that excitation of the coherent phonon is unambiguously connected to the exchange splitting of the surface state.

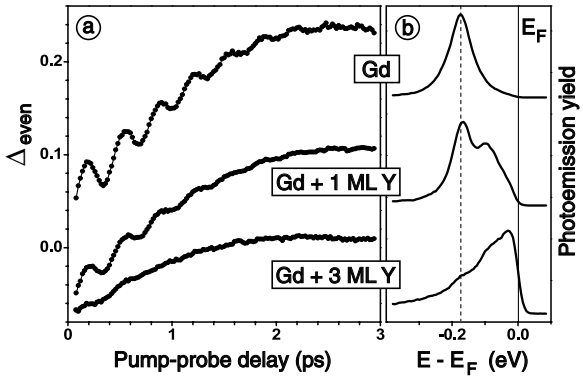

FIG. 3. (a) Quenching of the even SH oscillations by Y overlayers measured at 800 nm. (b) Photoemission spectra of bare Gd(0001) and covered by nominally 1 and 3 ML coverage (photon energy 36 eV).

Now, we turn to the magnetization dynamics. If temperature is increased, the exchange interaction is weakened. Figure 4(a) displays the temperature dependence of the Δ_{even} and Δ_{odd} oscillation amplitudes determined from the fit used in Fig. 2(b). In addition, the variation of the static magnetic SH contrast with temperature is shown, which measures the effective spin polarization at the surface vanishing at the Curie temperature of 293 K. Clearly, both oscillation amplitudes follow the magnetic contrast. As the SH is particularly sensitive to the surface state, this corroborates that the exchange splitting of the surface state is the origin of the coherent response observed in both even and odd SH fields.

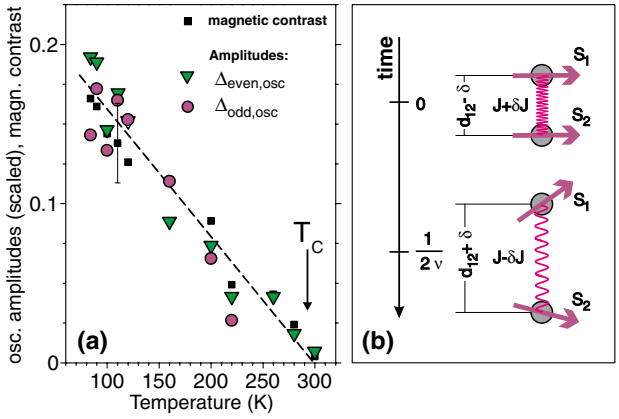

FIG. 4 (color online). (a) Temperature dependence of the static magnetic SH contrast $(I^{\uparrow} - I^{\downarrow})/(I^{\uparrow} + I^{\downarrow})$ and scaled initial amplitudes of the oscillating contributions of Δ_{even} and Δ_{odd} determined from time-dependent data [cf. Fig. 2(b)], measured at 800 nm at different temperatures. The dashed line is to guide the eye. (b) Illustration of a half cycle of coupled lattice and spin oscillations for the magnetic moments of two adjacent Gd atoms in the surface (S_1) and the subsurface layer (S_2), separated by the interlayer distance d_{12} that oscillates with amplitude δ. The initial increase of J, indicated by a maximum of Δ_{odd} in Fig. 2(b), is a consequence of the initial decrease in d_{12} leading to a larger value of J [15].

We propose that the optical phonon modulates the interlayer distance d_{12} at the surface which in turn alters the exchange coupling $J = J(d)$ consistent with density functional calculations [15]. Thus, the degree of magnetic order at the surface is modulated periodically, as illustrated in Fig. 4(b): An increase of the interlayer spacing d_{12} to $d_{12} + \delta$ leads to a decrease of the exchange coupling by δJ to $J - \delta J$ and thus to a lower degree of magnetic order, while half a period later a stronger exchange $J + \delta J$ increases the surface magnetization. This raises the question, whether there should be a finite transfer time between lattice and spin oscillations, which is, however, not observed in the experiment. We believe that this coupling is beyond the time resolution of our experiment, which is plausible as the exchange coupling ($J_{5d-5d} \approx 500$ meV and $J_{5d-4f} \approx 100$ meV) [27] is electronically mediated and much stronger than the observed frequency corresponding to 12 meV.

The above picture requires that the spin system can be excited at 3 THz. The spin wave dispersion along the c axis (Γ-A direction) in bulk Gd exhibits optical magnons with a maximum frequency of 3.5 THz at the Γ point [28]. This frequency will decrease at the surface due to the reduced coordination but may also be altered by the coupling between lattice and spins. Since our observed frequency of 2.9 THz is below the maximum spin wave frequency the spin system is, in fact, able to follow the surface phonon. Thus, overlap of the magnon spectrum with the coherent phonon frequency presents a requirement for such coupled excitation.

We conclude that the modulation of the spin polarization driven by the surface vibration can be viewed as a parametric excitation of the surface magnetization because the exchange interaction $J(d)$ — a parameter of the spin wave — is altered by the phonon. We interpret the magnetic contribution of this coupled excitation as optical magnon wave packets in the sense that the surface magnetization is modulated coherently by $\sim$10%. This new kind of a coupled coherent lattice and spin mode in a ferromagnet requires further studies to achieve a complete understanding and might have the potential to reach THz magnetic switching rates.

In summary, we have observed coherent optical phonons at 3 THz at the ferromagnetic metal surface Gd(0001) at 90 K by employing time-resolved second harmonic generation. The corresponding vibration of the interlayer distance leads to a variation of the exchange interaction and thereby drives parametrically coherent spin excitations at the same frequency in the THz range. This interpretation was supported by quenching the surface vibration by an Y overlayer and by temperature dependent measurements.

This work was supported by DFG through Sfb 290 and BMBF (Contract No. 05 KS1 KEC/2). We thank G. Kaindl for generously providing the UHV equipment.

*Electronic addresses: uwe.bovensiepen@physik.fu-berlin.de; http://www.physik.fu-berlin.de/~femtoweb

[1] S. DeSilvestri *et al.*, Chem. Phys. Lett. **116**, 146 (1985).
[2] G. C. Cho, W. Kütt, and H. Kurz, Phys. Rev. Lett. **65**, 764 (1990).
[3] H. J. Zeiger *et al.*, Phys. Rev. B **45**, 768 (1992).
[4] G. A. Garrett *et al.*, Phys. Rev. Lett. **77**, 3661 (1996).
[5] M. Hase *et al.*, Phys. Rev. Lett. **88**, 067401 (2002).
[6] *Spin Dynamics in Confined Magnetic Structures II*, edited by B. Hillebrands and K. Ounadjela, Topics in Applied Physics Vol. 87 (Springer-Verlag, Berlin, Heidelberg, 2003).
[7] M. Bauer *et al.*, Phys. Rev. B **61**, 3410 (2000).
[8] Y. Acremann *et al.*, Science **290**, 492 (2000).
[9] H. Weber, S. Riesen, and H. C. Siegmann, Science **291**, 1015 (2001).
[10] Th. Gerrits *et al.*, Nature (London) **418**, 509 (2002).
[11] H.W. Schumacher *et al.*, Phys. Rev. Lett. **90**, 017201 (2003); **90**, 017204 (2003).
[12] M. van Kampen *et al.*, Phys. Rev. Lett. **88**, 227201 (2002).
[13] J.-Y. Bigot, L. Guidoni, and V. Halté, in *Ultrafast Phenomena XIII*, edited by R. D. Miller *et al.*, Springer Series in Chemical Physics (Springer-Verlag, Berlin, Heidelberg, 2002), p. 398.
[14] A. Rehbein *et al.*, Phys. Rev. B **67**, 033403 (2003).
[15] I. Turek, J. Kudrnovsky, G. Bihlmayer, and S. Blügel, J. Phys. Condens. Matter **15**, 2771 (2003).
[16] A. Vaterlaus, T. Beutler, and F. Meier, Phys. Rev. Lett. **67**, 3314 (1991).
[17] R.-P. Pan, H. D. Wie, and Y. R. Shen, Phys. Rev. B **39**, 1229 (1989).
[18] A. Melnikov *et al.*, Appl. Phys. B **74**, 723 (2002).
[19] The relative phase φ for $\hbar\omega = 1.52$ eV was measured to be smaller than 15°. Therefore, small pump-induced changes of φ will affect Eq. (1) insignificantly, in accordance with results on Ni reported in A. Melnikov *et al.*, Appl. Phys. B **74**, 735 (2002).
[20] J. Hohlfeld *et al.*, Phys. Rev. Lett. **78**, 4861 (1997).
[21] Smoothing was carried out by polynomial fitting the data in a certain time window for consecutive time delays and averaging the data points at each delay. Polynomial degree and time window were optimized by an autocorrelation criterion.
[22] A. Aspelmeier, F. Gerhardter, and K. Baberschke, J. Magn. Magn. Mater. **132**, 22 (1994).
[23] Fs-optical excitation of a metallic ferromagnet is known to result in an ultrafast loss of magnetization, which is observed in the incoherent part of Δ_{odd} (Ref. [20]).
[24] R. R. Rao and C. S. Menon, J. Phys. Chem. Solids **35**, 425 (1974).
[25] O. Tangney and S. Fahy, Phys. Rev. Lett. **82**, 4340 (1999).
[26] The odd SH contribution of the Y/Gd interface is damped accordingly (not shown).
[27] R. Ahuja *et al.*, Phys. Rev. B **50**, 5147 (1994).
[28] W. C. Koehler *et al.*, Phys. Rev. Lett. **24**, 16 (1970).