論文 / 著書情報
Article / Book Information

Title	Ultrafast Manipulation of Antiferromagnetism of NiO
Authors	Nguyen Phuc Duong, Takuya Satoh, Manfred Fiebig
Citation	Physical Review Letters, Vol. 93, Issue 11, pp. 117402-1-4
Pub. date	2004, 9
Pub. version	http://dx.doi.org/10.1103/PhysRevLett.93.117402
Copyright	(C) 2004 American Physical Society

Ultrafast Manipulation of Antiferromagnetism of NiO

N. P. Duong,[1] T. Satoh,[1,2] and M. Fiebig[1,*]

[1]*Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin, Germany*

[2]*Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan*

(Received 12 May 2004; published 10 September 2004)

Photoexcitation of antiferromagnetic NiO leads to ultrafast reorientation of Ni^{2+} spins due to change of the magnetic anisotropy. Recovery of the magnetic ground state occurs as coherent oscillation of the antiferromagnetic order parameter between hard- and easy-axis states manifesting itself as quantum beating. The coherence time is ~1 ns with the beating frequency being determined by the anisotropy energy.

DOI: 10.1103/PhysRevLett.93.117402 PACS numbers: 78.47.+p, 42.65.Ky, 75.30.Gw, 75.50.Ee

The question of how fast the magnetic state of a medium can be changed has far-reaching consequences for the future of data storage. Ultrafast control of the magnetic order parameter is therefore a key issue of the rapidly evolving field of spin electronics. Specifically defined magnetic-field pulses allow repeated magnetic switching on the sub-ns time scale [1]. In 1996 optical experiments surmounted the time scale of spin-lattice relaxation by revealing magnetization dynamics below 1 ps, at which electrons, spins, and lattice are regarded as partly decoupled systems with individual temperatures [2]. Although a wealth of data on ultrafast spin dynamics of magnetically doped semiconductors and metals has been accumulated up to date [1,3,4], studies on antiferromagnetic (AFM) systems are restricted to solitary cases [5–8]. Yet, AFM systems offer inherent advantages for ultrafast magnetization dynamics: The absence of a macroscopic magnetization makes AFM compounds candidates for extremely rapid spin manipulation since conservation of angular momentum does not affect the dynamical properties [9,10]. Further, many AFM compounds are insulators with a characteristic sequence of discrete states. The associated long coherence times and localization of magnetic moments are favorable attributes in quantum computing. Practical interest in AFM compounds based on their use as pinning or spacer layer in modern electronic devices and the close relation between antiferromagnetism and high-correlation phenomena like colossal magnetoresistance and high-temperature superconductivity supplement the contemporary interest in the AFM state [11] and make a better understanding of its dynamical properties indispensable.

In this Letter we report the magnetization dynamics of AFM NiO after excitation with an intense optical 100 fs pump pulse using second and third harmonic generation as probes for the AFM and crystallographic sublattices, respectively. An increase of spin temperature and reorientation of the AFM order parameter $\vec{\ell}$ in the presence of the pump pulse is observed. Recovery of the magnetic ground state can be described as oscillation of $\vec{\ell}$ between the hard- and easy-axis states, their interference leading to pronounced quantum beating with a coherence time of ~1 ns. Whereas in many previous experiments on antiferromagnets [5–7] the *quenching* dynamics of the AFM state was investigated and probed indirectly via an accompanying ferromagnetic component or a modification of band structure, the present work demonstrates *manipulation* of the order parameter of a pure antiferromagnet observed by a magneto-optical process with direct coupling to the order parameter.

Recent interest in NiO was triggered by its attempted use as exchange-bias compound in magnetic read-write heads which is favored by the high Néel temperature $T_N = 523$ K. The optical properties of NiO were attributed to transitions within the $3d^8$ levels as sketched in Fig. 1(a) [12–14]. Paramagnetic NiO possesses NaCl-type cubic structure (point group $m3m$). Below T_N the Ni^{2+} spins are ordered ferromagnetically in {111} planes where they lie along $\langle 11\bar{2}\rangle$ axes [15]. The in-plane magnetization alternates along the stacking direction so that the crystal is AFM with 24 possible domains. Magnetostrictive deformation of the unit cell along the stacking and spin directions reduce the magnetic symmetry to $2/m$ [12]. The Ni^{2+} spins are fixed in the $\langle 11\bar{2}\rangle$ direction by mag-

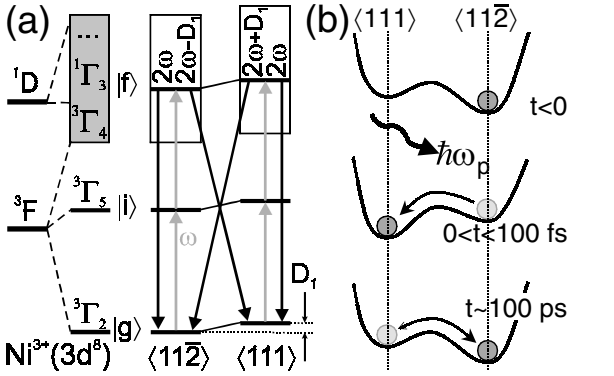

FIG. 1. (a) Electronic states of $Ni^{2+}(3d^8)$ for NiO spins along $\langle 11\bar{2}\rangle$ and $\langle 111\rangle$ axes and possible routes for second harmonic generation (SHG). (b) Schematic evolution of the dynamic antiferromagnetic phase transition triggered by the pump pulse at $\hbar\omega_p$ (see the text).

netic anisotropy which is parametrized by the anisotropy constants $\hbar D_1 = (0.10 \pm 0.01)$ meV for the $\langle 111 \rangle$ axes and $\hbar D_2 = 0.005$ meV for the $\langle 1\bar{1}0 \rangle$ axes which makes NiO an easy-plane system [15,16].

We studied the magnetization dynamics of NiO by optical pump-and-probe techniques which offer the highest temporal resolution of any experiment. During the last decade nonlinear optical processes were developed into an exceptionally powerful tool for investigation of the magnetic properties of matter [3,17]. When light waves with frequencies ω_i are incident on a crystal, harmonic generation from the electromagnetic fields $\vec{E}(\omega_i)$ and $\vec{H}(\omega_i)$ can induce a polarization at any frequency which is a linear combination of the frequencies of the incident waves. While linear optical effects like Faraday and Kerr rotation measure the absolute magnetization and are therefore unsuitable for probing AFM compounds, the coupling of nonlinear optical processes to the AFM order parameter is especially effective [18–20]. For NiO it was shown that magnetic-dipole-type SHG [14,21,22]

$$P_i(2\omega) = \epsilon_0 \chi^{(2)}_{ijk}(\vec{\ell}^2) E_j(\omega) H_k(\omega) \qquad (1)$$

is present below T_N. Figure 1(a) shows that the SHG process is made up by transitions $|g\rangle \rightarrow |i\rangle$, $|i\rangle \rightarrow |f\rangle$, and $|f\rangle \rightarrow |g\rangle$ which are all resonant. The nonlinear susceptibility $\hat{\chi}^{(2)}(\vec{\ell}^2)$ mediates the coupling between the incident light at frequency ω and the induced polarization at frequency 2ω. It couples quadratically to the AFM order parameter, so that SHG constitutes a direct probe of the AFM state [14].

Contributions to SHG from $\hat{\chi}^{(2)}(0)$ were not observed [14]. Therefore electric-dipole-type third harmonic generation (THG) [21,22]

$$P_i(3\omega) = \epsilon_0 \chi^{(3)}_{ijkl}(0) E_j(\omega) E_k(\omega) E_l(\omega) \qquad (2)$$

is employed as a probe of the "spinless" crystallographic lattice.

For the experiment (111)-oriented polished bulk samples were prepared from single crystals grown by the flame-fusion method. Brownish samples with domains in the order of ~ 1 μm and greenish samples with domains in the order of ~ 100 μm produced by oxygen annealing were used. Samples were investigated in the 5–300 K range. They were excited with amplified 100 fs pump-light pulses at $\hbar\omega_p = 1.55$ eV emitted from a Ti:sapphire laser. Harmonic generation with 100 fs pulses ($\hbar\omega = 0.82 - 1.06$ eV), which were emitted from an optical parametric amplifier pumped by the Ti:sapphire laser, was employed for probing spin and lattice dynamics. Pump and probe beams irradiated the same (111) face of the sample at near-normal incidence. Wave plates and polarizers were applied to set the polarization of the incoming light and analyze the polarization of the reflected signal light at 2ω or 3ω. The SHG and THG light was separated from the incident beams by optical filters and detected by a GaAs photomultiplier tube. Spectroscopy data were normalized with respect to the intensity of the incident probe beam.

In Fig. 2 we compare SHG transmission and reflection spectra in order to identify the electronic states probed by SHG. The transmission spectrum exhibits a peak at 2.05 eV which is due to the two-photon transition from the $^3\Gamma_2$ ground state to the $^1\Gamma_3/^3\Gamma_4$ excited state of the $Ni^{2+}(3d^8)$ ion which is resonance enhanced by the $^3\Gamma_2 \rightarrow {}^3\Gamma_5$ single-photon transition as depicted in Fig. 1(a) [14]. The reflection spectrum displays the same transitions as the transmission spectrum while new surface-bound transitions predicted for the 1.7 eV range [10] were not observed. The magnetic SHG signal thus probes exclusively bulk spin dynamics while being insensitive to surface-bound magnetization processes.

Figure 3 shows the temporal evolution of the change of reflected intensities of SHG and THG after excitation with the pump pulse. Based on Fig. 2 the photon energy of the probe beam was fixed at $\hbar\omega = 1.03$ eV. Within <1 ps a steplike decrease of magnetic SHG by $\langle \Delta I \rangle \simeq 10\%$ is observed. It is followed by a pronounced oscillation which persists for >100 ps and is observed in the whole investigated temperature range with only its frequency decreasing by 5% between 5 and 300 K. Independent of polarization the steplike decrease and oscillation of SHG were observed for all SHG tensor components $\chi^{(2)}_{ijk}(\vec{\ell}^2)$ which was verified by probing different domains on a sample with large domains. The universality confirms that manipulation of the magnetic order parameter, which enters all components of $\hat{\chi}^{(2)}$ in the same way, forms the basis of the observed effects [23].

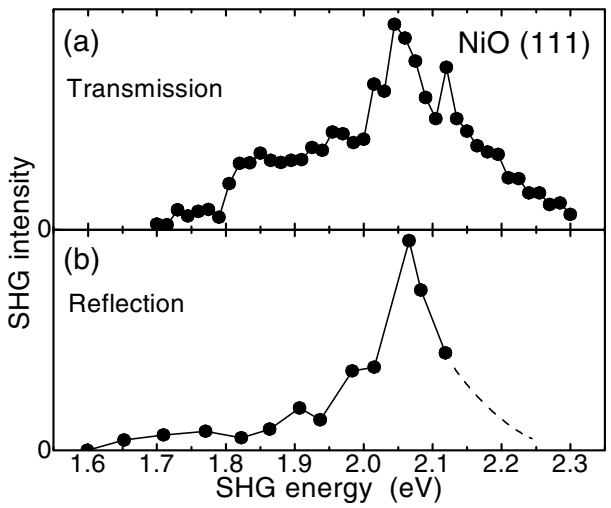

FIG. 2. Normalized SHG spectrum of NiO measured at room temperature with light incident along the [111] axis. (a) Transmission data taken with the 3 ns laser system described in Ref. [14]. (b) Reflection data taken with 100 fs laser pulses.

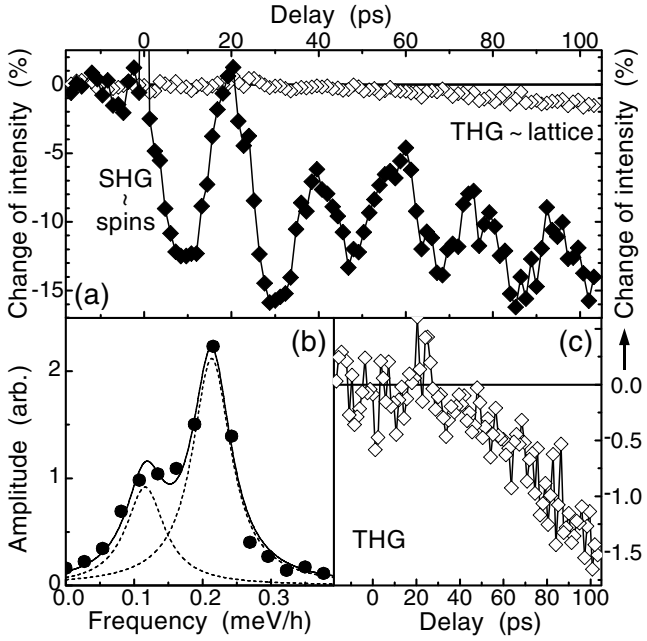

FIG. 3. (a) Change of reflected SHG and THG intensities in dependence of delay t between pump beam ($\hbar\omega_p = 1.55$ eV) and probe beam ($\hbar\omega = 1.03$ eV) at 6 K for a photonic excitation density of 1.02×10^{20} cm^{-3}. (b) Fourier transform of the SHG data after subtraction of the steplike decrease at $t = 0$. Dashed and straight lines: fitted spectral contributions and envelope. (c) THG signal from (a).

Because of the large thickness of samples interference effects from multiple reflection of acoustic phonons as observed in magnetic heterostructures [24] cannot explain the remarkable oscillation of "magnetic" SHG in Fig. 3. For the same reason unpinning effects [9,25] are ruled out. Typical phonon and magnon frequencies differ by 1 order of magnitude [26]. Since the change of linear reflectivity by the pump pulse was found to be of the order of 10^{-3} trivial modulation of the SHG signal by variation of the refractive index at ω or 2ω is also excluded.

$\langle \Delta I \rangle$ is 1 order of magnitude too large to account for demagnetization as a consequence of lattice heating from the pump pulse, which amounts to ≤ 10 K at 80 K [12]. We therefore conclude that $\langle \Delta I \rangle$ originates in an increase of spin temperature which occurs independent of the (presumably fast) increase of lattice temperature. This is corroborated by the evolution of the "nonmagnetic" THG signal which reproduces none of the dynamic properties of the SHG signal. THG simply displays a small continuous decrease which may be due to magnetostrictive modification of $\hat{\chi}^{(3)}(0)$ [12].

Magnetic anisotropy is determined by dipolar and quadrupolar interactions between the Ni^{2+} spins [27,28]. It is therefore easily modified by the shift of $3d$ orbital wave functions accompanying the excitation of $d - d$ transitions by the pump pulse. According to our model this leads to a change of easy direction from $\langle 11\bar{2} \rangle$ ("$11\bar{2}$ phase") to $\langle 111 \rangle$ ("111 phase") as a consequence of which a photoinduced reorientation of the AFM order parameter $\vec{\ell}$ occurs. A photoinduced change of magnetic anisotropy or structure at a similar excitation density was already reported for various transition-metal compounds [5,7,8,29]. For NiO manipulation of $\vec{\ell}$ within 10 fs by an intense laser field was also predicted [10], and a static change of the easy axis was already observed [30]. The proposed spin reorientation cannot be directly observed since with quadratic coupling of SHG to the AFM order parameter selection rules are the same for the $11\bar{2}$ and 111 phases. However, as will be seen our model of a dynamic AFM phase transition is in excellent agreement with the characteristic oscillation of the SHG signal in Fig. 3.

For recovery of the $11\bar{2}$ phase the subpicosecond spin-reorientation processes mediated by the laser field [10] are no longer available so that we should be able to temporally resolve the rotation of Ni^{2+} spins from the 111 phase back to the $11\bar{2}$ ground state. Quantum mechanically the rotation corresponds to a superposition of the coupled (see below) $11\bar{2}$ and 111 eigenstates. Physically their coherent interference corresponds to the oscillation of the system between the $11\bar{2}$ and 111 states. The beating frequency of the interference is determined by the energy separating the eigenstates, i.e., the anisotropy energy $\hbar D_1 = (0.10 \pm 0.01)$ meV. Figure 1(a) depicts that SHG in NiO with populated 111 and $11\bar{2}$ states can lead to emission of coherent light waves at $2\omega + D_1$, 2ω, and $2\omega - D_1$, the interference of which would lead to quantum beats at frequencies D_1 and $2D_1$. The model is in outstanding agreement with the observation of spectral components at energies 0.116 meV and 2×0.107 meV in the Fourier

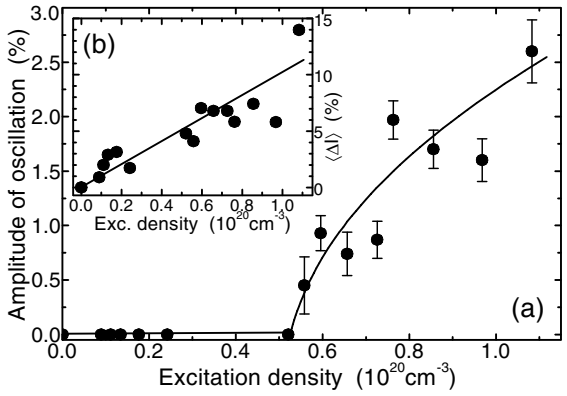

FIG. 4. Dependence of antiferromagnetic spin dynamics on photonic excitation density at 80 K. The data points were measured in random sequence in order to exclude systematic errors. (a) Amplitude of the coherent oscillation in the change ΔI of magnetic SHG in Fig. 3(a) fitted in the 50–100 ps interval. (b) Average drop $\langle \Delta I \rangle$ of magnetic SHG in Fig. 3(a). Lines are guides to the eye.

transform of $\Delta I(t)$ in Fig. 3(b). After 100 ps damping of the interference is not yet noticeable, implying a coherence time of ~ 1 ns in good agreement with the decoupled behavior of the lattice-sensitive THG signal in Fig. 3(c). Presumably coupling of the $11\overline{2}$ and 111 states in Fig. 1(a) is mediated by spin-orbit interaction for which lifetimes of $\gtrsim 100$ ps are a typical value. Note that in the case of "classical" quantum beats, as, e.g., in semiconductor quantum wells [31] the beating states decay simultaneously and are no longer populated after the system has returned to its ground state. In the present case one of the beating states (the $11\overline{2}$ state) is the ground state, and the recovery occurs as a transfer of averaged population density between the beating states due to their coupling.

The most striking evidence for a photoinduced AFM phase transition is provided by Fig. 4 which shows the dependence of the photoinduced change of magnetic SHG on the photonic excitation density. The steplike decrease and the oscillation of ΔI display an entirely different behavior which immediately confirms their different origin. $\langle \Delta I \rangle$ increases continuously with the excitation density which reflects the continuous increase of spin temperature with laser fluence. However, the amplitude of the oscillation exhibits a pronounced threshold behavior. Only if the photon fluence of the pump beam exceeds a value of 0.52×10^{20} cm^{-3} is the oscillation of ΔI observed. The threshold value marks the fluence at which the photoinduced modification of magnetic anisotropy exceeds the critical value where a new global minimum for $\langle 111 \rangle$ orientation of the Ni^{2+} spins is formed. The threshold is indicative of a collective process in the form of a photoinduced phase transition with ultrafast reorientation of antiferromagnetic order in NiO.

In summary, ultrafast manipulation of AFM order in NiO was observed using SHG as a probe for the dynamics of the AFM order parameter. $d - d$ transitions excited by a 100 fs pump pulse modify the magnetic anisotropy, thus leading to photoinduced reorientation of the Ni^{2+} spins from the $\langle 11\overline{2} \rangle$ into the $\langle 111 \rangle$ direction. The process is evidenced by subsequent recovery of the $11\overline{2}$ ground state via coherent oscillation of the Ni^{2+} spins between the $11\overline{2}$ and 111 eigenstates with a lifetime in the nanosecond range. The coupling is presumably guided by spin-orbit interaction and leads to pronounced quantum beating between the eigenstates. With an appropriately designed sequence of pump pulses repetitive switching of Ni spins between the $11\overline{2}$ and 111 phases as discussed in Ref. [10] is realistic. With application of such a kind of experiment to Ni/NiO heterostructures ultrafast manipulation of exchange bias becomes possible which opens up interesting opportunities for device construction.

Subsidy from the SPP 1133 of the Deutsche Forschungsgemeinschaft and discussions with K. Reimann, M. Wörner, and W. Hübner are appreciated. M. F. thanks T. Elsässer for continuous support.

*Electronic address: fiebig@mbi-berlin.de

[1] *Spin Dynamics in Confined Magnetic Structures II*, edited by B. Hillebrands and K. Ounadjela (Springer, Heidelberg, 2003).
[2] E. Beaurepaire, J. C. Merle, A. Daunois, and J. Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996).
[3] *Nonlinear Optics in Metals*, edited by K. H. Bennemann (Clarendon Press, Oxford, 1998).
[4] S. A. Wolf *et al.*, Science **294,**, 1488 (2001).
[5] M. Fiebig, K. Miyano, Y. Tomioka, and Y. Tokura, Appl. Phys. B **71**, 211 (2000).
[6] A. V. Kimel, R. V. Pisarev, J. Hohlfeld, and Th. Rasing, Phys. Rev. Lett. **89**, 287401 (2002).
[7] S. Tomimoto, S. Miyasaka, T. Ogasawara, H. Okamoto, and Y. Tokura, Phys. Rev. B **68**, 035106 (2003).
[8] A. V. Kimel, A. Kirilyuk, A. Tsvetkov, R. V. Pisarev, and Th. Rasing, Nature (London) **429**, 850 (2004).
[9] B. Koopmans, in Ref. [1], p. 253.
[10] R. Gómez-Abal, O. Ney, K. Satitkovitchai, and W. Hübner, Phys. Rev. Lett. **92**, 227402 (2004).
[11] *Magnetism Beyond 2000*, edited by A. J. Freeman and S. D. Bader (North-Holland, Amsterdam, 1999).
[12] *Numerical Data and Functional Relationships*, edited by K. H. Hellwege and O. Madelung, Landolt-Börnstein, New Series, Group III, Vol. 17g (Springer-Verlag, Berlin, New York, 1984).
[13] T. Tsuboi and W. Kleemann, J. Phys. Condens. Matter **6**, 8625 (1994).
[14] M. Fiebig *et al.*, Phys. Rev. Lett. **87**, 137202 (2001).
[15] M. T. Hutchings and E. J. Samuelsen, Phys. Rev. B **6**, 3447 (1972).
[16] H. Kondoh, J. Phys. Soc. Jpn. **15**, 1970 (1960).
[17] A. Kirilyuk, J. Phys. D **35**, R189 (2002).
[18] M. Fiebig, D. Fröhlich, B. B. Krichevtsov, and R. V. Pisarev, Phys. Rev. Lett. **73**, 2127 (1994).
[19] M. Fiebig *et al.*, Phys. Rev. Lett. **84**, 5620 (2000).
[20] M. Fiebig *et al.*, Phys. Rev. Lett. **86**, 6002 (2001).
[21] P. S. Pershan, Phys. Rev. **130**, 919 (1963).
[22] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).
[23] B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, Phys. Rev. Lett. **85**, 844 (2000).
[24] C. A. C. Bosco, A. Azevedo, and L. H. Acioli, Phys. Rev. B **66**, 125406 (2002).
[25] G. Ju *et al.*, Phys. Rev. Lett. **82**, 3705 (1999).
[26] M. J. Massey, N. H. Chen, J. W. Allen, and R. Merlin, Phys. Rev. B **42**, 8776 (1990).
[27] A. J. Sievers and M. Tinkham, Phys. Rev. **129**, 1566 (1963).
[28] J. I. Kaplan, J. Chem. Phys. **22**, 1709 (1954).
[29] M. Fiebig, D. Fröhlich, Th. Lottermoser, and R. V. Pisarev, Phys. Rev. B **65**, 224421 (2002).
[30] W. G. Park, U. Mick, and E. Kisker, in *Verhandlungen der Deutschen Physikalischen Gesellschaft* (DPG, Bad Honnef, 2004), Ser. VI, Vol. 39, Abstract No. MA 13.43 (in English).
[31] E. O. Göbel *et al.*, Phys. Rev. Lett. **64**, 1801 (1990).