Coherent control of antiferromagnetism in NiO

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Coherent control of the antiferromagnetic (AFM) order parameter of NiO is achieved by sequences of optical pump pulses modifying the magnetic anisotropy. The modification allows reversible switching of the AFM order parameter between hard- and easy-axis states. The switching frequency is determined by the magnetic anisotropy only and can therefore be higher than in the case of ferromagnetic spin precession. For NiO frequencies up to 100 GHz are achieved. Possibilities for device application are discussed.

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The potential of exploiting the spin of the electron in addition to its charge for electronic device applications leads to the rapidly evolving field of spintronics.[1] The tunneling magnetoresistance effect and its application in magnetoresistive random access memory (MRAM) is an example of a powerful spintronic effect. Future MRAM devices require spin manipulation rates much greater than 1 GHz so that rapid reversible switching of a magnetic state has become a major challenge.[2] So far, four methods for rapid manipulation of the magnetization of ferromagnets have been described. (i) Specifically designed magnetic-field pulses applied perpendicular to the direction of magnetization initialize spin precession.[2] (ii) Spin-polarized dc currents can change the spin orientation of a ferromagnet via an exchange of spin angular momentum.[3] (iii) Irradiation with laser pulses quenches the magnetization by laser-induced heating above the Curie temperature.[4] (iv) Laser pulses modify the magnetic anisotropy, thus inducing the reorientation and/or precession of spins.[5–7] Up to now, reversible manipulation of the ferromagnetic order parameter has been achieved with methods (i), (ii), and (iv). The time needed for the reversible manipulation of magnetization is determined by the Larmor frequency of spin precession and is thus limited to $\geq$100 ps.

In spite of the important role antiferromagnetism plays in many spintronics applications not much is known about the magnetization dynamics of antiferromagnetic (AFM) compounds. This is mostly due to the absence of a macroscopic magnetization which inhibits direct access to the AFM order parameter. Thus far, time-resolved optically induced spin reorientation or incoherent demagnetization has been observed for the AFM order in $FeBO_3$,[8] orthoferrites,[9] and NiO.[10,11] However, "AFM switching" in the sense of the reversible manipulation of an AFM order parameter has not been reported yet.

In this Brief Report we describe ultrafast all-optical control of the AFM order parameter of NiO by sequential 100 fs laser pulses using optical second-harmonic generation (SHG) as a probe for the AFM state. The first pulse triggers a collective reorientation of Ni^{2+} spins because of photoinduced modification of the magnetic anisotropy. The second pulse enhances or reverses the reorientation, depending on the time lag between the two pump pulses. Since our effect is based on a transient phase transition with collective reorientation of spins, but not on the magnon-related precession and reversal of spins as in the case of the experiments on ferromagnets, spin manipulation in NiO allows us to achieve repetition rates as high as 100 GHz.

The high Néel temperature (T_N=523 K) makes NiO an ideal compound for device applications. An anisotropy energy[12,13] of $D_1=0.10\pm0.01$ meV confines the Ni^{2+} spins to the {111} planes where they are ordered ferromagnetically along the $\langle 11\bar{2}\rangle$ axes. Opposite spin orientation in neighboring {111} planes makes the compound AFM.

The magnetization dynamics of NiO was studied by all-optical pump-and-probe techniques which offer the highest temporal resolution of any experiment. Furthermore, excitation with laser pulses with photon energies of ~1 eV can modify the electronic and the magnetic system more easily than magnetic field pulses which introduce the equivalent of only ~0.1 meV/T. Optical SHG described by the relation

$$P_i(2\omega)=\varepsilon_0\chi^{(2)}_{ijk}(\ell^2)E_j(\omega)H_k(\omega) \quad (1)$$

was used as ultrafast probe for the evolution of the AFM order parameter.[14] Here $\vec{E}(\omega)$ and $\vec{H}(\omega)$ denote the electric and magnetic fields of the incident light wave at frequency ω whereas $\vec{P}(2\omega)$ denotes the nonlinear polarization induced at 2ω. Because of the coupling of $\vec{P}(2\omega)$ to the AFM order parameter ℓ parametrized by the nonlinear susceptibility $\hat{\chi}(\ell^2)$, the intensity $I\propto|\vec{P}(2\omega)|^2$ of the SHG light can be used as a probe for the AFM state of our NiO samples.

The bulk samples were pumped at a repetition rate of 1 kHz with amplified 100 fs light pulses at $\hbar\omega_{pu}=1.55$ eV emitted from a Ti:sapphire laser. The pump pulses possess an energy on the order of 10 μJ and were focused onto an area with a diameter of ~200 μm to produce excitation densities of up to 10^{20} cm^{-3} in NiO. They heat the sample by less than 10 K at low temperature and by less than 1 K at room temperature, which can be neglected for the experiment. The probe pulses were generated by an optical parametric amplifier pumped by the Ti:sapphire laser. The photon energy of the probe pulses was tuned to the *d-d* transitions of the $Ni^{2+}(3d^8)$ ion at $\hbar\omega_{pr}=1.03$ eV in order to achieve large SHG intensities.[14] Pump and probe beams were incident in

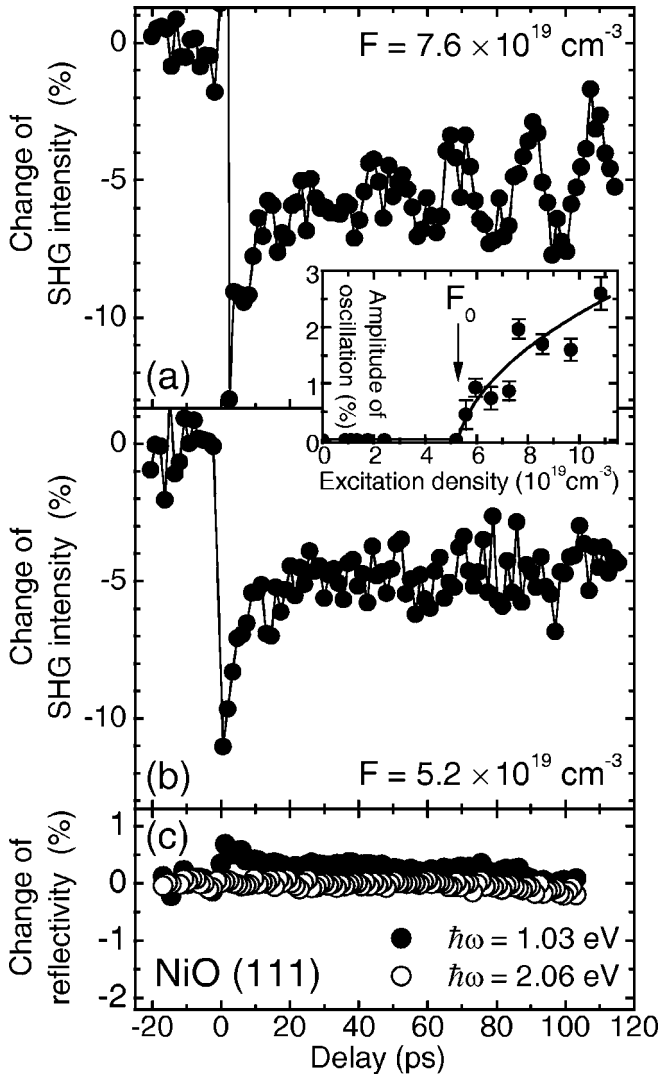

FIG. 1. Change of linear and nonlinear optical responses after photoexcitation with 1.55 eV light pulses at $t=0$ of a (111)-oriented NiO bulk sample at 80 K. (a), (b) Temporal evolution of the intensity change of reflected SHG for two different photon excitation densities F. The inset shows the dependence of the amplitude of the oscillation in (a) and (b) on the excitation density. The line is a guide to the eye. (c) Temporal evolution of the linear reflectivity at ω and 2ω with $\hbar\omega=1.03$ eV as photon energy of the probe pulses.

near-normal incidence onto a polished (111) face of the NiO crystal. The diameter of the pump pulses exceeded that of the probe pulses by a factor of 3. The overlap of pump and probe pulses was verified by maximizing the three-photon difference frequency signal between the probe pulse and at $2\omega_{pu}-\omega_{pr}$ at zero time delay. This process mixes pump and probe waves and therefore requires their overlap to be observed. The reflected SHG of the probe beam was separated from the other light waves by optical filters and detected by a GaAs photomultiplier tube. Initially, experiments were performed below 80 K in a cryostat. However, after no major difference was observed in experiments at room temperature, we refrained from using the cryostat for simplicity and to approach realistic conditions for device applications.

Figure 1 shows the intensity of the magnetic SHG signal as a function of the delay between the pump pulse and the probe pulse for two different photon excitation densities F of the pump beam. In Fig. 1(a) a decrease of the average intensity of the SHG by 7% within 1 ps is observed. It is superimposed by a pronounced oscillation with an amplitude of ±3%. The origin of the oscillation is discussed in detail in Ref. 11 and briefly summarized here for comprehensiveness: The pump pulse changes the magnetic anisotropy with a transient change of the easy direction from $\langle 11\bar{2}\rangle$ to $\langle 111\rangle$. (In supplement to what was said in Ref. 11 we note that the photoinduced modification of anisotropy is plausible because shifts of $3d$ orbital wave functions accompanying the excitation of d-d transitions modify the dipolar and quadrupolar interactions between the Ni^{2+} spins which determine the anisotropy. Further, the same change of the easy direction can be achieved by a magnetic field, which is a relatively weak perturbation.[16]) The transient change of the easy direction leads to a rapid photomediated reorientation of Ni^{2+} spins[15] from the local $[11\bar{2}]$ direction ("$11\bar{2}$ state") to the local [111] direction ("111 state"). Subsequently, the $11\bar{2}$ and 111 states interfere coherently. In the absence of the pump pulse, the spin-orbit interaction mediates the—in terms of a classical picture—slow reversion of the magnetic system to the $11\bar{2}$ ground state. The interference is described by the wave function $\psi=a\psi_{11\bar{2}}+b\psi_{111}\exp(iD_1t/\hbar)$ with $|a|^2+|b|^2=1$ and $\lim_{t\to\infty}b=0$. Thus, the beating frequency is given by the anisotropy energy. Since the SHG process probes the $11\bar{2}$ and 111 states it reproduces their coherent interference in the form of the oscillation of SHG intensity in Fig. 1(a).[11]

Figure 1(b) shows that reduction of the excitation density by 30% quenches the oscillation of the SHG signal. According to the inset of Fig. 1 the threshold F_0 for occurrence of the oscillation is near 5.4×10^{19} cm^{-3}. The threshold value marks the excitation density at which the modification of the anisotropy by the pump pulse lets the energy of the 111 state fall below that of the $11\bar{2}$ state so that the collective rotation of the Ni^{2+} spins and, thus, the oscillation of the SHG signal, are initiated. Because of the transient nature of the modification the reorientation process is not completed so that the amplitude of the oscillation is larger at higher excitation densities from the pump laser.

Figure 1(c) shows the temporal evolution of the linear reflectivity of the probe beam tuned to the photon energies of the fundamental and the SHG waves. At 1.03 eV the excitation by the pump pulse leads to an increase of reflectivity by ~0.3% whereas at 2.06 eV a change of reflectivity is not detected. Along with the threshold behavior and the results of nonlinear optical experiments discussed in Ref. 11, Fig. 1(c) confirms that an emission of phonons cannot be responsible for the periodic modulation of SHG. Their presence usually becomes manifest in an oscillatory modulation of the dielectric function (and thus of the reflectivity[17]) the frequency of which, in the case of acoustic modes, scales linearly with the photon energy. However, none of this behavior is observed in Fig. 1(c).

Note, that the collective reorientation of Ni^{2+} spins cannot be understood as conventional generation of magnons. Magnons describe the coherent oscillation of spins around the

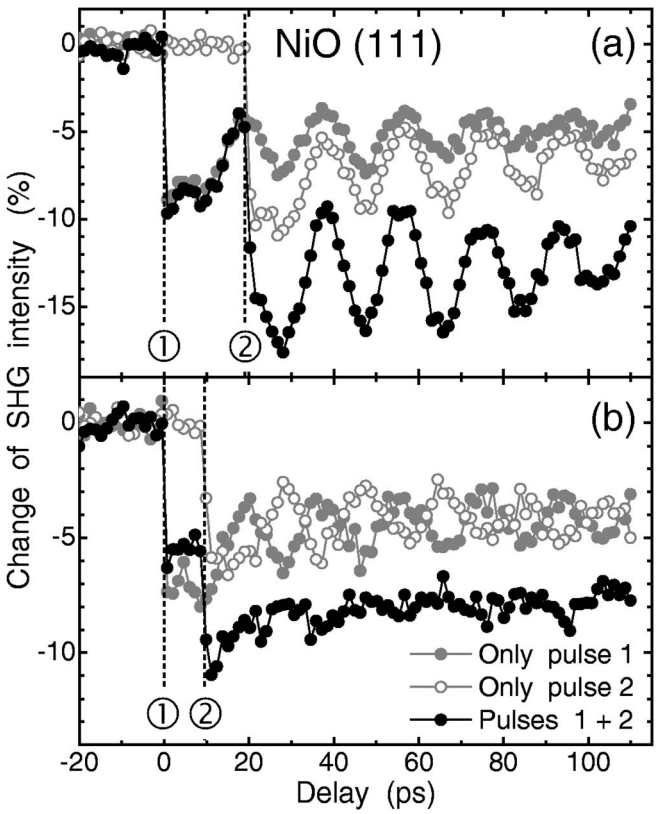

FIG. 2. Change of the intensity of the reflected SHG of a (111)-oriented NiO bulk sample at room temperature for two pump pulses incident at $t=0$ (1) and Δt (2). $\Delta t=$(a) 19 and (b) 9.5 ps.

ground-state orientation. In terms of a classical picture this corresponds to demagnetization but not to a *reorientation* of the order parameter as in the present case.

Contrary to the periodic modulation of SHG intensity the decrease of the average SHG intensity in Figs. 1(a) and 1(b) does not show any threshold behavior. This decrease is due to fluctuations of the Ni^{2+} spins around the $11\bar{2}$ ground state, i.e., the generation of magnons which disperse the excess energy introduced by the pump laser. This is an incoherent process corresponding to an increase of the spin temperature for which the absence of the threshold behavior is natural.

In the classical picture the coherent superposition of the coupled 111 and $11\bar{2}$ states with a gradual transfer of amplitude from the former to the latter corresponds to the rotation of the sublattice magnetization back toward the $[11\bar{2}]$ direction, a process which according to the persistence of the oscillation in Fig. 1(a) clearly exceeds 100 ps. Figures 1(a) and 1(b) thus display an example of active, ultrafast (<1 ps) manipulation of an AFM state followed by passive, slow ($\gg 100$ ps) recovery of the original state. However, reversible switching requires active ultrafast control not only of the transfer to the excited spin state but also of the return to the ground state.

In Fig. 2 the temporal evolution of magnetic SHG after illumination with two pump pulses is shown for different time lags between the pump pulses. Both pump pulses were overlapped by the aforementioned procedure. Therefore they also overlapped with each other, exciting the same region on the sample. As mentioned, the experiments in Fig. 2 were performed at room temperature in order to approach realistic conditions for device applications. Just as in the case of the low-temperature measurements a single pump pulse ("pump 1" incident at $t=0$ or "pump 2" incident at $t=\Delta t$) with an excitation density above the threshold F_0 induces a drop of the average SHG intensity and the pronounced oscillation which is characteristic for the photoinduced $11\bar{2}\rightarrow 111$ reorientation of spins.

A comparison of Figs. 1(a) and 2 reveals that the envelope of the SHG oscillation up to a delay of 110 ps is increasing in the former and decreasing in the latter case. The increase in Fig. 1(a) is not reproducible. It may be related to a small increase of the pump intensity in the progress of data acquisition to which the SHG response is particularly sensitive near the threshold fluence F_0. A survey of all data sets gained at $F>F_0$ at ≤ 80 K shows no increase or decrease of the envelope of the SHG oscillation up to a delay of 110 ps within error limits. However, the decrease of the envelope in Fig. 2 is reproducible and points to a coherence time of the SHG interference at room temperature on the order of 100 ps.

When the time lag separating the two pump pulses is $\Delta t=19$ ps both the average drop of the SHG intensity as well as the amplitude of the oscillation are approximately doubled. The two excitations with the pump wave correspond to independent events during each of which the magnetic anisotropy is modified to the same extent. This case is different from doubling the energy of a single pump pulse. It therefore leads to a dependence of the amplitude of the oscillation on the total excitation energy which is different from that shown in the inset of Fig. 1.

When the time lag between the two pump pulses is reduced to $\Delta t=9.5$ ps the oscillation initiated by the first pulse is stopped by the second pulse. The similarity in the temporal dependence of SHG in Fig. 2(b) after arrival of the second pump pulse and in Fig. 1(b) after arrival of a single pump pulse at an excitation density $F<F_0$ indicates that the quenching of the oscillation is related to the ultrafast reversal of the Ni^{2+} spins to the $11\bar{2}$ ground state. The reversal occurs because the coherent interference of the $11\bar{2}$ and 111 states of the Ni^{2+} spins (expressed by ψ) adds up destructively for the two pump pulses due to the phase shift introduced by the 9.5 ps delay. A similar optical two-pulse manipulation of a magnetic order parameter was reported for ferromagnetic compounds.[6] However, contrary to these previous experiments no remanent oscillation is observed in Fig. 2(b) since the lifetime of the $11\bar{2}\rightarrow 111$ excitation is much longer than the time needed for one $11\bar{2}\rightarrow 111\rightarrow 11\bar{2}$ switching cycle (~ 100 ps versus ~ 10 ps).

Independent of the value of Δt the average drop of SHG intensity is enhanced by the arrival of the second pump pulse. This corroborates the idea that the decrease of average SHG intensity is related to *incoherent* deviations of the Ni^{2+} spins from the $11\bar{2}$ ground state corresponding to a magnon-related decrease of (sublattice) magnetization whereas the oscillation of the SHG intensity is related to *coherent* devia-

tions of the Ni^{2+} spins from the $11\bar{2}$ ground state corresponding to a reorientation of the AFM order parameter.

The time needed for a switching cycle of the AFM order parameter is only determined by the energy separating the magnetic ground state and the photoinduced excited magnetic state, i.e., by the magnetic anisotropy. In NiO this energy corresponds to a manipulation time of 9.5 ps so that in principle AFM switching in this compound can occur at a repetition rate as high as 100 GHz. This is an order of magnitude faster than in related experiments on ferromagnetic systems[2,6] in which the cycle time for the switching process is restricted by the Larmor precession. Note that precession frequencies in AFM compounds are in general much higher than in ferromagnetic compounds[9,18] so that precessional AFM spin reversal can in principle be even faster than the anisotropy-related switching reported here. However, this has not been realized thus far. Further, photomagnetic effects allow one to reorient spins in ferromagnetic systems persistently within much less than 100 ps, but only if the reorientation is small ($<1°$), and reversible switching has not been demonstrated yet.[7]

At present the number of repetitions in our AFM switching experiment is limited by the rate at which the excess energy introduced by the pump pulse can be dissipated. We found that the relation between the amplitude of the coherent SHG oscillation and the incoherent drop of SHG intensity is very sensitive to fine tuning of the photon energy and the pulse shape of the pump wave. Therefore, pulse shaping[19] and frequency tuning of the pump wave in a two-color mixing experiment with two optical parametric oscillators may help to minimize the incoherent part of the excitation. Use of NiO thin films or heterostructures instead of bulk crystals will further modify the performance under repeated switching of the order parameter.

In summary, reversible switching of the AFM order parameter of NiO between hard- and easy-axis states has been achieved by a succession of pump pulses modifying the magnetic anisotropy. Sequences with a limited number of pump pulses can be used to coherently control the orientation of the AFM order parameter at a repetition rate which can be higher than that of ferromagnetic compounds and reaches 100 GHz in NiO.

Experimentally, the AFM state is less accessible than the macroscopic magnetization characterizing any ferromagnetic state. However, the orientation of the AFM order parameter determines many other macroscopic properties such as exchange bias[20,21] or magnetoresistance[22] which are easily detectable. In such systems the higher repetition rates attainable in AFM structures because of the absence of a macroscopic magnetization can be employed to construct spintronics applications with shorter magnetic manipulation times. Moreover, transient photoinduced modification of the magnetic anisotropy with subsequent quantum mechanical interference (quantum beating) is a convenient way to measure the anisotropy energy.

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[1] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[2] *Spin Dynamics in Confined Magnetic Structures II*, edited by B. Hillebrands and K. Ounadjela (Springer, Heidelberg, 2003).
[3] I. N. Krivorotov, N. C. Emley, J. C. Sankey, S. I. Kiselev, D. C. Ralph, and R. A. Buhrman, Science **307**, 228 (2005).
[4] E. Beaurepaire, M. Maret, V. Halté, J. C. Merle, A. Daunois, and J. Y. Bigot, Phys. Rev. B **58**, 12134 (1998).
[5] B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, Phys. Rev. Lett. **85**, 844 (2000).
[6] Q. Zhang, A. V. Nurmikko, A. Anguelouch, G. Xiao, and A. Gupta, Phys. Rev. Lett. **89**, 177402 (2002).
[7] F. Hansteen, A. Kimel, A. Kirilyuk, and Th. Rasing, Phys. Rev. Lett. **95**, 047402 (2005).
[8] A. V. Kimel, R. V. Pisarev, J. Hohlfeld, and Th. Rasing, Phys. Rev. Lett. **89**, 287401 (2002).
[9] A. V. Kimel, A. Kirilyuk, A. Tsvetkov, R. V. Pisarev, and Th. Rasing, Nature (London) **429**, 850 (2004).
[10] G. Ju, A. V. Nurmikko, R. F. C. Farrow, R. F. Marks, M. J. Carey, and B. A. Gurney, Phys. Rev. Lett. **82**, 3705 (1999).
[11] N. P. Duong, T. Satoh, and M. Fiebig, Phys. Rev. Lett. **93**, 117402 (2004).
[12] M. T. Hutchings and E. J. Samuelsen, Phys. Rev. B **6**, 3447 (1972).
[13] H. Kondoh, J. Phys. Soc. Jpn. **15**, 1970 (1960).
[14] M. Fiebig, D. Fröhlich, Th. Lottermoser, V. V. Pavlov, R. V. Pisarev, and H. J. Weber, Phys. Rev. Lett. **87**, 137202 (2001).
[15] R. Gómez-Abal, O. Ney, K. Satitkovitchai, and W. Hübner, Phys. Rev. Lett. **92**, 227402 (2004).
[16] S. Saito, M. Miura, and K. Kurosawa, J. Phys. C **13**, 1513 (1980).
[17] D. Lim, R. D. Averitt, J. Demsar, A. J. Taylor, N. Hur, and S. W. Cheong, Appl. Phys. Lett. **83**, 4800 (2003).
[18] A. F. Andreev and V. I. Marchenko, Usp. Fiz. Nauk **130**, 39 (1980) [Sov. Phys. Usp. **23**, 21 (1980)].
[19] W. S. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993).
[20] C. Binek, *Exchange Bias in Magnetic Heterosystems*, Springer Tracts in Modern Physics Vol. 196 (Springer-Verlag, Heidelberg, 2003), p. 55.
[21] A. E. Berkowitz and K. Takano, J. Magn. Magn. Mater. **200**, 552 (1999).
[22] M. Izumi, T. Manako, Y. Konishi, M. Kawasaki, and Y. Tokura, Phys. Rev. B **61**, 12187 (2000).