Spin Oscillations in Antiferromagnetic NiO Triggered by Circularly Polarized Light

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Coherent spin oscillations were non-thermally induced by circularly polarized pulses in fully compensated antiferromagnetic NiO. This effect is attributed to an entirely new mechanism of the action, on the spins, of the effective magnetic field generated by an inverse Faraday effect. The novelty of this mechanism is that spin oscillations are driven by the time derivative of the effective magnetic field acting even on “pure” antiferromagnets with zero net magnetic moment in the ground state. The measured frequencies (1.07 THz and 140 GHz) of the spin oscillations correspond to the out-of-plane and in-plane modes of antiferromagnetic magnons.

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All-optical magnetization switching has been extensively studied in recent years. In 1996, demagnetization within 1 ps was discovered by irradiating ferromagnetic nickel with femtosecond laser pulses [11]. This pioneering finding has stimulated intense theoretical and experimental investigations. Many of the experiments on so-called “ultrafast magnetism” can be interpreted in terms of laser-induced heating, which is already exploited technologically in the form of heat-assisted magnetic recording [2]. However, this is a relatively slow process since the recording rate is limited by thermal diffusion. Thus, magnetization control beyond the limit of such thermal control is highly desirable.

A typical form of non-thermal magnetization control is the inverse Faraday effect (IFE). The IFE was predicted by Pitaevskii [3] and was demonstrated in non-absorbing media by van der Ziel et al. [4]. Due to this effect, circularly polarized light induces magnetization that can be described as a light-induced effective magnetic field acting on the body. A pump-probe technique with sub-picosecond time resolution has revealed transient IFE at zero time delay in itinerant ferromagnets [5-8]. However, evidence of the spin-related contribution has been an issue because no impact was observed after the temporal overlap of the pump and probe pulses [9].

The dynamic properties of antiferromagnets (AFMs) are rapidly gaining importance [10,11]. These compounds can display inherently faster spin dynamics than ferromagnetic compounds [10,12], and offer the advantage that the spin oscillation frequency extends into the subTHz and THz regime. In addition, ultrafast manipulation of the antiferromagnetic order parameter may be employed for ultrafast control of the magnetization of an adjacent ferromagnet via the exchange-bias effect. Recently spin precession caused by the IFE has been reported for ferrite-garnets [13] and for canted AFMs [11,14], which possess non-zero net magnetic moment caused by the Dzyaloshinskii–Moriya interaction. The presence of this magnetic moment is an important issue for the recently proposed mechanism of inertia-driven excitation of spin oscillations in canted AFMs [15]. On the other hand, a fully non-thermal control of spin oscillations has not been demonstrated yet in “pure” AFMs, having a fully compensated magnetic moment ($\vec{M} = 0$) in the ground state.

Here we report the first observation of coherent spin oscillations in a fully compensated ($\vec{M} = 0$) AFM NiO in a pump-probe experiment. The oscillations consisted of 1.07 THz and 140 GHz frequency components, which are assigned to out-of-plane and in-plane modes of antiferromagnetic spin oscillations. The sign of the oscillation was reversed with the reversal of the circularly polarized pump helicity. This is interpreted within the σ-model approach as a direct action of the time derivative of the impulsive magnetic field generated by a circularly polarized pulse via the IFE on the zero-magnetization AFM. This mechanism (discussed in Refs. 16,17 but never observed before) opens a novel way for the ultrafast effective control of spins in compensated AFMs.

NiO is one of the most promising exchange-bias AFMs because of its simple structure and room-temperature antiferromagnetism. Therefore, the investigation of the time-resolved responses of the electric, magnetic, and optical properties of NiO could play an important role for applications of ultrafast optical switching, and in fundamental research. The sub-picosecond spin reorientation in NiO has been demonstrated by modifying the magnetocrystalline anisotropy with linearly polarized light [18]. However, this process depends on a resonant optical excitation and is thus limited by thermal effects.

Above the Néel temperature ($T_N = 523 \text{K}$), NiO has an NaCl-type cubic structure (point group: $m\bar{3}m$). Below $T_N$, NiO has antiferromagnetic order. The Ni$^{2+}$ spins align ferromagnetically along the $\{110\}$ axes in $\{111\}$ planes with antiferromagnetic coupling in between adjacent $\{111\}$ planes [19]. Exchange striction leads to a contraction of the cubic unit cell along the $\{111\}$ axes and reduces the crystallographic symmetry to $\overline{3}m$. This gives rise to four-types of twin (T) do-
mains. The deformation is accompanied by magnetic birefringence between the \{111\} plane and the \{111\} direction. NiO is a charge-transfer insulator with a 4 eV band gap. The intragap optical transition in the mid-infrared to visible region is ascribed to the electric-dipole forbidden \textit{d}-\textit{d} transitions of the Ni\textsuperscript{2+} (3\textit{d}\textsuperscript{8}) electrons. In particular, a \textsuperscript{1}Γ\textsubscript{2} → \textsuperscript{3}Γ\textsubscript{4} transition centered at 700 nm and a \textsuperscript{3}Γ\textsubscript{2} → \textsuperscript{3}Γ\textsubscript{3} transition centered at 1150 nm have been identified \cite{20}.

A NiO single crystal was grown by a floating-zone method. The bulk sample was polished into \{111\}-oriented platelets with lateral dimensions of a few millimeters and a thickness of \(\simeq 100 \mu\text{m}\). As-grown samples possessed \(\text{T}\) domains with a lateral size of \(< 1 \mu\text{m}\). To obtain \(\text{T}\) domains of 0.1–1 mm, the platelets were annealed in an argon-oxygen mixture with small oxygen partial pressure at 1400°C \cite{21}. By rotating a polarizer and analyzer in the cross-Nicol configuration, four types of \(\text{T}\) domains with a size of \(~500 \mu\text{m}\) were distinguished. For the pump–probe measurement, we selected a single \(\text{T}\) domain with the \{11\overline{1}\} plane different from the sample surface \{111\}, as shown in Fig. 1(a).

The temporal evolution of the polarization rotation and transmission were measured with a pump–probe setup (Fig. 1(b)). The sample was in a cryostat at 77 K with no external magnetic field. Linearly polarized light from a mode-locked Ti:sapphire laser with a wavelength of 792 nm, a pulse width of 120 fs, and a repetition rate of 1 kHz was used as the probe. Circularly polarized optical pulses with a wavelength of 1280 nm, generated by an optical parametric amplifier, were used as the pump. The pump and probe beams were focused on the sample surface to spots of about 100 \(\mu\text{m}\) and 40 \(\mu\text{m}\), respectively. The pump fluence was 10 mJ/cm\(^2\), which corresponds to the absorption of about one photon per \(10^5\) Ni\textsuperscript{2+} ions. The probe beam fell on the sample at normal incidence, whereas the pump beam was incident at an angle of 7°. The transmitted probe beam was divided into two orthogonally polarized components by a Wollaston prism, and each beam was detected with a Si photodiode to obtain the polarization rotation and the transmission change.

To clarify the spin-related contribution, we examined the impact after photo-excitation with different time delays. Figures 2(a,b) show the polarization rotation and the transmission change, respectively, versus the time delay between the probe beam and the pump beam. The inset in Fig. 2 shows the polarization rotation of the probe beam near zero-delay. Here the signal is compared for \(\sigma_+\) and \(\sigma_-\) polarized pump beams at fixed laser fluence. In Fig. 2 two processes can be distinguished: (1) a fast (practically instantaneous) change of the polarization rotation within the time of the pulse action (in the inset); and (2) damped oscillations of the polarization rotation which persists for much longer times (upper frame).

In regime (1), for short time delays (\(< 1 \text{ ps}\)), the rotation exceeded 20 mrad when the pump and the probe beams overlapped temporally. The full width at half-maximum of the signal was about 200 fs, which reflects the duration of the pump and probe pulses. In regime (2), the slowly damped oscillations of the signal (with a signal amplitude much lower than for short times) were observed at times longer than 10 ps. For both time intervals, the sign of the signal changes with reversal of the pump helicity, which is a clear indication of the non-thermal origin of the effect. Note the significant difference in the amplitude of the rotation angle observed at these two time scales (\(< 0.2 \text{ ps}\) and \(\geq 1 \text{ ps}\)) that likely reflects the difference of the mechanisms responsible for them.

Process (1) can be considered as a typical example of so-called \textit{femtomagnetic effects}, arising at times much shorter than the thermalization time \cite{23}. An adequate description of this regime involves either a direct transfer of photon angular momentum to the medium or a photo-enhanced transfer between orbital and spin momenta \cite{8,13,22,24}. For our compound, the ground state \(\textsuperscript{1}Γ\textsubscript{2}\) of the Ni\textsuperscript{2+} (3\textit{d}\textsuperscript{8}) ion, the orbital momentum is quenched due to orbital non-degeneracy. In the virtually excited state, the orbital momentum is \(\pm 1\) depending on the helicity \(\sigma_\pm\) of the pump beam, which leads to the appearance of a transient magnetization. Recently, \textit{ab initio} calculations \cite{25} of an ultrafast laser-induced spin switch in NiO has demonstrated the possibility of inducing a spin magnetic moment at tens of femtoseconds that results in \textit{instantaneous} magneto-optical effects in this material.

In process (2), the sign of the oscillations in the polarization rotation changes with reversal of the pump helicity, indicating that the oscillation is triggered by non-thermal photo-excitations. No oscillation is observed in the transmission, indicating that the oscillation in the rotation is magnetic in origin. The damped oscillations are fitted well with

![FIG. 1:](Image)
For such AFMs, the antiferromagnetic vector $\vec{L} = \vec{M}_1 - \vec{M}_2$ is the principal dynamical variable. Within the $\sigma$-model, the equation for the normalized (unit) antiferromagnetic vector $\vec{l} = \vec{L}/|\vec{L}|$ can be written through the variation of the Lagrangian $[16, 26]$ $\mathcal{L}[\vec{l}] = \mathcal{L}_0 + \mathcal{L}_{\text{int}}$, where $\mathcal{L}_0$ describes the free oscillations of the spin system:

$$\mathcal{L}_0 = \frac{\hbar}{2\gamma H_{\text{ex}}} \left( \frac{d\vec{l}}{dt} \right)^2 - w(\vec{l}), \quad w(\vec{l}) = g\mu_B(H_{a1}\vec{l}_1^2 + H_{a2}\vec{l}_2^2), \quad (1)$$

$\mathcal{L}_{\text{int}}$ determines the action of the light. The magnetization $\vec{M} = \vec{M}_1 + \vec{M}_2 = 2M_0\vec{m}$ is a slave variable and can be written in terms of the vector $\vec{l}$ and its time derivative:

$$H_{\text{ex}}\vec{m} = [\vec{H} - \vec{l}(H \cdot \vec{l})] + \frac{1}{\gamma} \left( \frac{d\vec{l}}{dt} \times \vec{l} \right). \quad (2)$$

Here, the first term determines the canting of the sublattices, caused by the effective magnetic field $\vec{H}$, and the second term describes the dynamic contribution $[16, 26]$. The value of the Lagrangian is presented per one spin. $\gamma = g\mu_B/\hbar$ the gyromagnetic ratio, $g$ the Landé factor, $\mu_B$ the Bohr magneton, $H_{\text{ex}} = zSJ/g\mu_B$ the exchange field of AFM, and $z = 6$ is the number of next-nearest neighbors. For NiO, $J=221$ K, which for $S=1$ gives $\gamma H_{\text{ex}} = zSJ/\hbar = 27.4$ THz. We used the simplest form of the biaxial anisotropy $w(\vec{l})$, written in terms of the out-of-plane anisotropy field $H_{a1}$ and much smaller in-plane anisotropy field $H_{a2}$ $[19]$.

Within the $\sigma$-model, the action of the circularly polarized light can be described by an effective magnetic field $\vec{H}(t) \propto (\vec{E} \times \vec{E}')$, corresponding to the IFE; in this case

$$\mathcal{L}_{\text{int}} = -\frac{\hbar}{H_{\text{ex}}} \left( \vec{H} \cdot (\vec{l} \times \frac{d\vec{l}}{dt}) \right). \quad (3)$$

The variation of $\mathcal{L}[\vec{l}]$ gives the dynamical equations for $\vec{l}$. In linear approximation over the deviation from the ground state ($\vec{l}_{\text{ground}}$ is parallel to the z-axis, see Fig. 1), they read

$$\frac{d^2l_x}{dt^2} + \omega_1^2l_x = \gamma \frac{dH_x}{dt}, \quad \frac{d^2l_y}{dt^2} + \omega_2^2l_y = -\gamma \frac{dH_z}{dt}, \quad (4)$$

where $\omega_1 = \sqrt{2H_{\text{ex}}H_{a1}}$ and $\omega_2 = \sqrt{2H_{\text{ex}}H_{a2}}$ are the frequencies of the out-of-plane and in-plane antiferromagnetic spin oscillations, respectively, and we omitted the dissipation terms. For a short enough pulse ($\omega_{1,2}\Delta t \ll 1$), Eq. (4) describes a quite universal behavior for AFM $[17]$. Namely, after the pulse action, for times $t \gg \Delta t$, the spin dynamics exhibits free oscillations with frequencies $\omega_{1,2}$ and amplitudes $l_{x,y}(t=0) = a_{1,2}$ determined by the form of the pulse:

$$a_1 = \gamma H_z, \quad a_2 = -\gamma H_x, \quad \bar{H}_{x,y} \Delta t \equiv \int_{-\infty}^{+\infty} H_{x,y}(t)dt. \quad (5)$$

Thus, the amplitudes of the two components of the oscillations are determined by the pulse field components, $H_z$ and $H_x$. The oscillation of the vector $\vec{m}$ produces a modulation of the antisymmetric part of the permittivity tensor $\varepsilon_{ij}, \Delta\varepsilon_{ij} \propto \varepsilon_{ij\mu\nu}m_\mu \mu\nu$;
where $\epsilon_{ijk}$ is the absolute antisymmetric tensor. In linear approximation, the out-of-plane mode produces $\Delta_{z}^{\sigma} \propto m_{y}$, and the in-plane mode produces $\Delta_{x}^{\sigma} \propto m_{y}$.

For our measurements, a Ti domain inclined to the surface of the sample was chosen (see Fig. 1), and the values of $H_{k}$ and $H_{l}$ were approximately equal, which is in agreement with the observation that $a_{1} \sim a_{2}$. The measured frequency $f_{1} = 1.07$ THz is in good agreement with the out-of-plane mode of the antiferromagnetic spin oscillations. From the NiO out-of-plane anisotropy field, $\gamma H_{a1} \approx 23$ GHz [29], one obtains $f_{1} = 1.1$ THz. The 1.07 THz component has been observed in far-infrared antiferromagnetic resonance [27, 28] and Raman scattering [29]. Concerning the in-plane mode, the data for a small in-plane anisotropy field $\gamma H_{a2} \sim 1$ GHz are not well known [19], but spin oscillations with a frequency $f_{2} = 140$ GHz have recently been observed in NiO using Brillouin scattering [30]. Therefore, this strongly suggests that the observed oscillations in Fig. 2(a) within the wide time interval from 1 ps to tens of picoseconds after the pulse is turned off, are spin oscillations around their easy-axis, which are the usual spin-wave modes, triggered by the effective magnetic field $H$ generated via the IFE, with the time derivative of the field $dH/dt$ working as a torque acting on the vector $\vec{l}$.

The simultaneous observation of the magnetic response at both short times and long times, in processes (1) and (2), allows us to reach conclusions about the applicability of different approaches to describe the spin dynamics. The Landau–Lifshitz equation for ferromagnets (or, equivalently, the $\sigma$-model equation for AFMs) describes the dynamics of spin systems in terms of only the magnetization vector (or sublattice magnetizations, for AFMs). These equations are valid for quasi-equilibrium states, where these magnetizations are formed by the exchange interaction. This occurs (according to our observations) for times corresponding to process (2). On the other hand, our results show that the $\sigma$-model approach (as well as any common theories treating the dynamics of spin systems through the mean value of the magnetization) are not sufficient to describe shorter times $t \lesssim 0.5$ ps, namely, the behavior in regime (1). To stress this, it is enough to mention, that the amplitudes of the oscillation, Eq. (5), do not contain anisotropy fields, and it can be obtained by neglecting all relativistic interactions in the AFM, except for the Zeeman interaction of the spins with the pulsed magnetic field. In this approximation within the $\sigma$-model approach, the projection of the magnetization parallel to the field is conserved and cannot appear during the action of a short ($\Delta t \ll 1/\omega_{z}$) field pulse. For the $\sigma$-model, direct calculations show that the static and dynamic contributions to the magnetization, Eq. (2), compensate for each other in this short time interval [17]. On the other hand, the signal observed at these times is much higher than for process (2). Thus, to describe the initial stage (1) of the process, one needs to use a more detailed analysis involving the spin and orbital momenta of the solid, as well as the angular momentum of photons [25].

To conclude, the time-resolved magneto-optical response of antiferromagnetic NiO provides a direct measure of magnetization changes under the action of circularly polarized light. Remarkably, we found that even compensated antiferromagnetic NiO shows spin oscillations triggered non-thermally by a circularly polarized pulse, with the time derivative $dH/dt$ as the driving force acting on the antiferromagnetic vector $\vec{l}$. The 1.07 THz and 140 GHz components are in good agreement with experimentally reported frequencies of antiferromagnetic spin oscillations. The results of our experiment show the possibility of extending the potential of NiO as an antiferromagnetic constituent in spintronic devices, from static and thermally-limited spin-dynamical experiments into the promising range of all-optical magnetization control at THz frequencies.

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