Ultrafast spin-damping effects in optically excited antiferromagnets

Christian Tzschaschel, Takuya Satoh, and Manfred Fiebig

1Department of Materials, ETH Zürich, 8093 Zurich, Switzerland
2Department of Physics, Tokyo Institute of Technology, Tokyo 152-8551, Japan

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Antiferromagnets are opening up new possibilities for high-speed spintronic applications as they potentially allow for unprecedented switching rates. A promising approach for utilising the intrinsically faster timescales of antiferromagnets is the ultrafast non-thermal control of magnetic order by laser-optically generated magnetic-field pulses. Because of the shortness of the magneto-optical interaction process, magnetic damping effects during the sub-picosecond laser-pulse excitation are usually not considered. Here, we point out that such damping can play a crucial role in antiferromagnets because of their unique interplay between exchange interaction and magnetic anisotropy. We show that due to spin damping, an ultrashort magnetic field pulse can impulsively generate a net magnetisation in an otherwise fully compensated antiferromagnet, which creates a new route for the optical manipulation of antiferromagnetism. We complement time-resolved measurements of the Faraday rotation by a theoretical model of spin-damping effects, thus providing a basis for a comprehensive description of optically excited antiferromagnets.

I. INTRODUCTION

Antiferromagnets (AFMs) are a promising material class for spintronic applications. The fully compensated magnetic order in AFMs makes them robust against external magnetic fields and at the same time leads to vanishing magnetic stray fields thus reducing cross-talk between neighboring antiferromagnetic domains. Furthermore, the intimate interaction between differently oriented magnetic sublattices in an AFM leads to spin dynamics in the THz-regime, which allows antiferromagnetic switching processes on picosecond timescales. This combination of properties is extremely appealing for antiferromagnetic spintronics and has the potential to revolutionise nowadays semiconductor-based information technologies. In fact, recently, the first electrically switched antiferromagnetic bit has been presented and employing synthetic AFMs in racetrack memories dramatically improved both their speed and data density. The efficient manipulation of AFMs relies on spin currents acting as effective magnetic fields that generate staggered (anti-) damping-like torques. While these can be conveniently generated from electrical currents, exploring the full potential of AFMs requires sub-picosecond access to the antiferromagnetic order.

Optical laser pulses naturally come to mind as those provide the required time-resolution to track the THz-spin dynamics. A prominent mechanism to induce coherent spin dynamics in AFMs is the inverse Faraday effect (IFE). Using circularly polarised laser pulses, this effect allows the non-thermal generation of effective magnetic field pulses in a material. Even in pure AFMs, these magnetic field pulses impulsively create an initial spin canting. As the IFE is not based on absorption, it provides the means to control the antiferromagnetic order without generating excess heat and thus enables energy-efficient, high-speed spin control. Despite the technological relevance of damping-like torques, ultrafast damping effects during optical excitations are typically neglected. While this provides a reasonable approximation for spin dynamics in ferromagnets, we question its
validity for antiferromagnets.

Here, we report a comprehensive theoretical and experimental analysis of optically induced spin dynamics in fully compensated AFMs. We present a complete phenomenological description of the IFE including spin damping during both the ultrafast excitation and the ensuing spin precession. By considering damping during the spin excitation, we predict an additional ultrafast damping-like torque that impulsively induces a net magnetisation in fully compensated antiferromagnets. This torque scales with the Gilbert-damping parameter $\alpha$, which we tune by varying the sample temperature. We experimentally verify the presence of that torque by scrutinizing the initial phase of an optically induced spin precession in antiferromagnetic hexagonal rare-earth manganites. In particular in regions of enhanced damping, the typically neglected damping-like torque of the IFE dominates the optically induced spin dynamics, thus opening up an unexpected, yet highly efficient energy-transfer channel for opto-spintronics applications.

A. Magnetic ordering in h-RMnO$_3$

The nine hexagonal rare-earth manganites h-RMnO$_3$ ($R =$ Sc, Y, In, Dy, Ho, Er, Tm, Yb, Lu) belong to the class of magneto-electric multiferroics\cite{29}. Taking h-YMnO$_3$ as its best-known representative, with decreasing temperature, the material undergoes a structural phase transition from $P6_3/mmc$ to $P6_3cm$ at $T_C \approx 1260$ K leading to the formation of six different structural domains characterised by a distortive order parameter $Q$\cite{30}. This structural phase transition is accompanied by the emergence of an improper ferroelectric polarisation $P(Q)$\cite{31}. Below the Neēl temperature $T_N \approx 75$ K, YMnO$_3$ orders antiferromagnetically\cite{32} and assumes the space group $P6_3cm'$. A projection of the magnetic order is shown in Fig. 1a. The magnetic moments order in a triangular lattice with the spins pointing along the three equivalent crystallographic $x$ axes\cite{33}. The inter-plane exchange interaction is approximately two orders of magnitude weaker than the intra-plane exchange interaction and can therefore be neglected\cite{34}.

In contrast to YMnO$_3$, the paramagnetic-antiferromagnetic phase transition at $T_N$ in HoMnO$_3$ leads to space group $P6_3c'm$\cite{32}. While the crystallographic structure is identical to the one shown in Fig. 1a, the magnetic moments are $90^\circ$ rotated, such that they point along equivalent crystallographic $y$ axes and the total toroidal moment in the unit cell vanishes. At $T_{SR} \approx 35$ K, HoMnO$_3$ undergoes a first-order spin-reorientation transition (SRT) from $P6_3c'm$ to $P6_3cm'$ with decreasing temperature and assumes the magnetic structure shown in Fig. 1a\cite{35}.
Both antiferromagnetic space groups exhibit three optically excitable magnon modes called X, Y and Z mode\textsuperscript{26} which induce an oscillating net magnetisation during the spin precession pointing along the x, y or z axes, respectively. In terms of symmetry, the X and Y modes correspond to doubly-degenerate E modes, while the Z mode is an $A_2$ mode\textsuperscript{36,37}. All three modes can be selectively excited by the inverse magneto-optical effects and probed individually by time-resolved measurements of either the Faraday effect (Z mode) or the Cotton-Mouton effect (X, Y mode)\textsuperscript{26}.

Note that within each of the six structural Q domain states, the spins can order in two $\ell$-domain states (with $\ell$ representing the antiferromagnetic order parameter) leading to a total of 12 different antiferromagnetic domain states. However, all these respond identically to the effective magnetic field pulse of the optical excitation and are hence indistinguishable\textsuperscript{26}. This allows us to work with an antiferromagnetic single domain state for the theoretical description.

Exemplarily, we focus the ensuing discussions onto the Z mode, which is illustrated in Fig. 1c. We performed time-resolved optical pump-probe measurements to study the coherent spin dynamics as a function of temperature. As opposed to the higher frequency X and Y modes, the low frequency Z mode is typically not accessible by Raman scattering or inelastic neutron scattering. Time-resolved measurements, however, cover the required frequency range of 10 GHz to 1 THz. This allows us to continuously trace the Z mode as a function of temperature including through the SRT and close to $T_N$.

From a fundamental point of view the Z-mode frequency is determined by the weak magneto-crystalline in-plane anisotropy\textsuperscript{38}, which is the driving force behind the SRT in HoMnO$_3$. Studying the Z mode might thus yield novel insights into the nature of the SRT. The Z mode is, however, also interesting for applications. A coherent large-angle Z-mode excitation can ultimately lead to a coherent precessional switching of the AFM order. Since only the weak in-plane anisotropy needs to be overcome, this facilitates the most energy efficient switching process\textsuperscript{6,38} with potential switching rates exceeding 100 GHz.

### B. Optical setup

It is crucial for polarisation sensitive optical experiments to avoid the pronounced crystallographic birefringence. This limits us to (0001)-cut samples, where the z axis is perpendicular to the surface. Our setup is schematically shown in Fig. 1b. The fundamental light source is a regeneratively amplified laser system providing 130 fs pulses at 1.55 eV. We use the output of an optical parametric amplifier to generate 0.95 eV circularly polarised pump pulses, which create an effective magnetic field in the sample via the IFE. By pumping the material far below the nearest absorption resonance at $\approx 1.6\text{ eV}$\textsuperscript{39}, we minimise parasitic thermal effects\textsuperscript{19}. We then probe the ensuing dynamics by measuring the time-resolved Faraday rotation $\Phi(t)$ of the linearly polarised probe pulse with a balanced detection scheme. The sample is mounted in a cryostat, which allows us to vary the temperature between $\approx 5\text{ K}$ and 300 K.

Figure 2a shows an exemplary measurement of the time-resolved Faraday rotation in YMnO$_3$. Two different regimes can be clearly distinguished: on the one hand we have a pronounced signal around 0 ps reflecting the direct interaction between pump and probe pulse. On the other hand there is a damped sinusoidal modulation of the Faraday rotation as the hallmark of the spin precession and relaxation after the excitation. The latter is a fitted according to
\[ \Phi(t) = A_0 + A_1 e^{-t/t_1} + A_2 e^{-t/\tau} \sin(\omega t + \phi_0), \]

where the \( A_0 + A_1 e^{-t/t_1} \) describes an exponentially decaying offset, while \( \tau, \omega \) and \( \phi_0 \) are the lifetime, frequency and initial phase of the optically induced spin precession, respectively. Figure 2b shows a zoom into the region of the temporal overlap together with an extrapolation of the oscillatory component of the fit function. As a consequence of the non-vanishing initial phase, the fit yields a finite Faraday rotation at 0 ps. Note that the initial phase is not accessible in spectroscopic, but only in time-resolved measurements. It contains valuable information about the excitation mechanism and has been used previously to distinguish between different excitation mechanisms of phonons and magnons.\(^{20,40–42}\) We will show in the following that the deviation of the Faraday rotation from the perfect sine-like time-dependence is consistent with the so far neglected effect of spin damping during the excitation.

II. THEORY

We describe the magnetic order with the following unit-cell Hamiltonian:

\[ \mathcal{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D_z \sum_i S^2_{i,z} + D_y \sum_i S^2_{i,y} + \Delta \sum_i S^2_{i,x} S_{i,y} \]  

Here, \( J > 0 \) is the antiferromagnetic intra-plane exchange interaction, \( D_z > 0 \) is the easy-plane anisotropy and \( \Delta > 0 \) is a weak fourth-order anisotropy, which is necessary to describe the SRT in HoMnO\(_3\). The sign of \( D_y \) determines the magnetic ground state as \( P6_3'c'm' (D_y < 0, \mathbf{S} \parallel \hat{y}) \) or \( P6_3'cm' (D_y > 0, \mathbf{S} \parallel \hat{x}) \). We neglect the weak inter-plane exchange interaction, which allows us to consider only the three spins in one lattice plane in the unit cell, i.e. \( i,j = \{1,2,3\} \). By defining the net magnetic moment \( \mathbf{M} = g\mu_B \sum_i \mathbf{S}_i \), we can rewrite the first term of the Hamiltonian \( \mathcal{H} \) as

\[ J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = \frac{3}{2} J \sum_i S^2_{i,z} + \frac{3}{2} \sum_i |S_i|^2. \]

Limiting our considerations to the \( Z \) mode, we set \( \mathbf{M} = M_z \hat{z} \). Furthermore, we use the fact that the \( Z \)-mode precession preserves the threefold rotational symmetry of the unit cell.\(^{22,26}\) Therefore, the three spins in one unit cell must have the same \( z \) component and \( M_z = 3 g\mu_B S_{i,z} \). Equation (3) thus yields

\[ J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = \frac{3}{2} J \sum_i S^2_{i,z} + \frac{3}{2} \sum_i |S_i|^2. \]
Hence, omitting the constant contribution $\propto |S_i|^2$, the Hamiltonian in Eq. (2) can be rewritten as $\mathcal{H} = \sum_i \mathcal{H}_i$ with
\[
\mathcal{H}_i = \left( \frac{3}{2} J + D_z \right) S_{i,z}^2 + D_y S_{i,y}^2 + \Delta \sum_i S_{i,x}^2 S_{i,y}^2 \tag{5}
\]

Thus, by taking the $Z$-mode symmetry into account, we could restrain $\mathbf{M}$ to be parallel to $\hat{z}$, which allows decoupling the individual magnetic sublattices. This enables us to describe the dynamics of the whole magnetic system by considering only one spin $S$. We will describe its dynamics by analytically solving the Landau-Lifshitz-Gilbert (LLG) equation, i.e.\[13]
\[
\mathbf{m} = -\gamma \mu_0 \mathbf{m} \times \mathbf{H} - \frac{\gamma \mu_0}{m_0} \alpha \mathbf{m} \times (\mathbf{m} \times \mathbf{H}) \tag{6}
\]
where $\mathbf{m} = g \mu_B \mathbf{S}$ denotes the magnetic moment per sublattice with magnitude $m_0 = g \mu_B |S|$\[14] and $\mu_0 \mathbf{H} = -\partial_m \mathcal{H}$ the effective magnetic field acting on one sublattice. $\gamma = g \mu_B h^{-1} \approx 2 \pi \cdot 28 \cdot 10^9 \text{rad s}^{-1} \text{T}^{-1}$ denotes the gyromagnetic ratio and the dimensionless quantity $\alpha$ the Gilbert damping parameter.

As highlighted by Figs. 2b and c, the optically induced spin dynamics can be separated into optical excitation and precessional relaxation. As different mechanisms are dominating in the different regimes, we will also theoretically describe them separately.

### A. Optical excitation

In this section, we will generalise the existing theory of magneto-optical spin excitations in undamped materials to be also applicable to damped spin systems. Prior to the excitation, we assume the system to be in the magnetic ground state. Thus, the effective magnetic field of the Hamiltonian in Eq. (2) is parallel to the sublattice magnetisation $\mathbf{m}$ and does not exert a torque. With the $P6_3/m\overline{3}$ symmetry of the ground state, we set $\mathbf{m}(t < 0) = m_0 \hat{x}$. Furthermore, as the optically induced magnetic field pulse of the IFE is short compared to the period of the spin precession, it can be modelled as $\mathbf{H}_{\text{IFE}}(t) = H \delta(t) \hat{z}$, where $H$ is the effective magnetic field strength, $\theta$ the laser pulse duration and $\delta(t)$ denotes the Dirac delta-function\[17]

The setting is sketched in Fig. 1b. The first term of Eq. (6) induces a field-like torque $\mathcal{T}_F = -\mu_0 \mathbf{m} \times \mathbf{H}$ leading to a spin canting by $\gamma \mu_0 H \theta$ within the easy-plane. Additionally, the second term gives rise to a damping-like torque $\mathcal{T}_D = -\alpha \frac{\mu_0}{m_0} \mathbf{m} \times (\mathbf{m} \times \mathbf{H})$, which induces a spin canting of $\alpha \gamma \mu_0 H \theta$ along the magnetic hard axis\[18]. This damping-like torque has not been considered so far for optical excitations.

For this configuration, we can analytically integrate the LLG equation from $t < -\infty$ to $t \geq 0$ and find
\[
\mathbf{m}(t \geq 0) = m_0 \begin{pmatrix}
1 \\
\gamma \mu_0 H \theta \\
\alpha \gamma \mu_0 H \theta
\end{pmatrix} \tag{7}
\]
with $\alpha \gamma \mu_0 H \theta, \gamma \mu_0 H \theta \ll 1$. The initial spin canting along the $z$ axis scales with the Gilbert damping parameter $\alpha$. It is thus typically much smaller than the initial canting along the $y$ axis. However, any spin canting along the magnetic hard axis ($z$) will be enhanced during the spin precession due to exchange enhancement\[17,18]. Since exchange enhancement is an effect intrinsic to antiferromagnets, but not present in ferromagnets, it marks a distinct difference between ferro- and antiferromagnets. Thus, while neglecting damping effects during the spin excitation is justifiable for ferromagnets, it might not be justified in antiferromagnets. In fact, we will show that despite $\alpha \ll 1$ the contribution of the damping-like torque to the total spin precession can even dominate over the field-like excitation.
B. Precessional relaxation

In order to understand how this new damping-like torque affects the spin precession, we will now linearise the LLG-equation and solve it with the initial conditions from Eq. (7).

We assume \( m_x \approx m_0 \) and \( m_y, m_z \ll m_x \). Combining the effective magnetic field created by the Hamiltonian in Eq. (5) with the LLG-equation (6) yields to the lowest order

\[
\dot{m} = -\frac{m_0}{2\hbar g \mu_B} \begin{pmatrix} 0 \\ J m_z + \alpha D m_y \\ -D m_y + \alpha J m_z \end{pmatrix} \tag{8}
\]

with \( J = \frac{3}{2} J + D_z + |D_y| \) and \( D = \Delta |S|^2 + |D_y| \) and hence

\[
\ddot{m}_z + \alpha \omega_0 \left( A + \frac{1}{A} \right) \dot{m}_z + \left( 1 + \alpha^2 \right) \omega_0^2 m_z = 0, \tag{9}
\]

where \( \hbar \omega_0 = 2|S| \sqrt{J D} \) is the "undamped" spin precession frequency and \( A = \sqrt{J D}^{-1} \) is the exchange enhancement factor\(^{217}\). Geometrically, \( A \) corresponds to the ratio of the maximum basal-plane spin canting relative to the maximum canting along the \( z \) axis during the spin precession. It thus parametrises the ellipticity of the spin precession’s trajectory\(^{20,15}\).

The solution to the LLG-equation is

\[
m_z(t) = z_0 e^{-t/\tau} \sin (\omega t + \phi_0). \tag{10}
\]

For \( \alpha \ll 1, A \gg 1 \) and initial conditions \( \phi_0 \), one finds the lifetime \( \tau \), frequency \( \omega \) and initial phase \( \phi_0 \) of the spin precession as

\[
\tau = \frac{2}{A \alpha \omega_0}, \tag{11}
\]

\[
\omega = \omega_0 \sqrt{1 - A^2 \alpha^2 / 4}, \tag{12}
\]

\[
\tan \phi_0 = A \alpha \sqrt{1 - A^2 \alpha^2 / 4} \frac{1}{1 - A^2 \alpha^2 / 2}. \tag{13}
\]

Combining Eqs. (11) and (12), we find

\[
\frac{A \alpha}{2} = \frac{1}{\sqrt{1 + \omega^2 \tau^2}}. \tag{14}
\]

Furthermore, we obtain \( A \alpha \) from the initial phase via Eq. (13). Differences between those results allow us to identify the presence of additional excitation mechanisms that are not based on the IFE. Thus, the initial phase provides new insights into the optically induced spin dynamics.

Independently, we obtain \( A \alpha \) from the initial phase \( \phi_0 \) via Eq. (13). For an optical spin excitation purely via the IFE, Eqs. (13) and (14) give the same value for \( A \alpha \). Any differences between those results allow us to identify the presence of additional excitation mechanisms that are not based on the IFE. Thus, the initial phase provides new insights into the optically induced spin dynamics, which we will elucidate later.

Note that \( A \alpha = 2 \) marks the case of a critically damped spin precession. For stronger damping, no spin oscillations can be observed. As \( A \gg 1 \) in all antiferromagnets, where the exchange interaction is the dominant energy scale, this justifies the initial assumption \( \alpha \ll 1 \). While this also implies that \( T_{DL} \ll T_{FL} \), the initial phase of the spin precession is determined by \( A \alpha \). Thus, owing to the exchange enhancement, the here predicted damping-like torque \( T_{DL} \) exerted by the IFE becomes experimentally accessible by evaluating initial phase of the spin precession.

III. RESULTS

The non-vanishing initial phase in Fig. 2 already points to the presence of an ultrafast damping-like torque \( T_{DL} \). Now, we show its relevance by performing temperature-dependent measurements of optically induced magnon dynamics. By varying the temperature, we tune the Gilbert-damping parameter \( \alpha \) and hence the
magnitude of $T_{DL}$. In particular, we trace the $Z$ mode through the spin reorientation in HoMnO$_3$ and study its evolution upon approaching the Néel temperature in YMnO$_3$. This allows us to experimentally verify Eqs. (11)–(13) in different scenarios and thus to showcase the importance of spin damping during ultrafast spin excitations.

**A. HoMnO$_3$: First-order spin-reorientation transition**

Equivalent to the exemplary measurement in Fig. 2 on YMnO$_3$, we performed time-resolved Faraday-rotation measurements of optically induced spin dynamics in HoMnO$_3$ as a function of temperature. The oscillatory component of the signal was fitted by Eq. (1), from which we extract the undamped frequency $\omega_0$, lifetime $\tau$ and initial phase $\phi_0$ as a function of temperature. The results are shown in Fig. 3a, b, and c, respectively.

The reduced in-plane anisotropy constant $D_y$ during the SRT causes a clear dip in the temperature dependence of $\omega_0$ around 33 K in Fig. 3a. The red curve shows a fit of the following minimal model to the data. The Mn ions carry a spin $S = \frac{5}{2}$. The temperature-dependent macrospin per sublattice $|S|(T)$ can be implicitly described as a paramagnet aligned by the exchange field of the neighbouring sublattices, i.e. $|S|(T) = SB_{S=2}|S|(T)T_NT^{-1}$, where $B_{S=2}$ is the Brillouin function. We modelled the SRT by introducing a linear dependence of the anisotropy parameter $D_y$ on temperature. The SRT temperature is defined as the point, where $D_y$ vanishes. Furthermore, as the SRT is a 1$^{st}$-order phase transition, it exhibits a hysteresis, which is accounted for by heuristically introducing a jump temperature $T_{jump} > T_{SR}$, such that $|D_y|(T) = \text{sign}(T - T_{jump})\kappa(T - T_{SR})$. The hysteretic behaviour is consistent with our observations, where, coming from low temperatures, the magnon frequency shows a discontinuity at a temperature higher than the SRT temperature (c.f. inset in Fig. 3). We find from the fit of $\omega_0$: $T_N = (69.9 \pm 0.8)$ K, $T_{SR} = (33.3 \pm 0.2)$ K. Taking the exchange interaction $J = 2.5$ meV from neutron scattering experiments yields furthermore $\Delta = (0.68 \pm 0.01)$ $\mu$eV and $\kappa = (0.16 \pm 0.01)$ $\mu$eV K$^{-1}$. The latter leads to an extrapolated in-plane anisotropy of $\approx 5.4$ $\mu$eV at 0 K, which is more than two orders of magnitude smaller than the out-of-plane anisotropy $D_z = 650$ $\mu$eV. Both weak anisotropy contributions are consistent with typical assumptions in inelastic neutron scattering experiments, where the in-
plane anisotropy is often neglected.\(^{23}\)

The lifetime of the spin precession, shown in Fig. 3b, follows approximately an exponential decay with an additional lifetime reduction between 35 K and 45 K. By combining Eqs. (13) and (14) we can calculate the temperature dependence of the initial phase \(\phi_0\) from the magnon frequency and lifetime and compare it to the directly measured value. The result of the calculation is shown in red in Fig. 3c together with the direct measurement of the initial phase in black. The agreement is striking around the SRT and towards higher temperatures. As the model only takes excitation by the IFE into account, we demonstrate here the importance of the additional damping-like torque \(T_{DL}\) that we considered in the theory section. Note that the initial phase changes from sine-like (\(\tan \phi_0 < 0\)) at low temperatures to cosine-like (\(\tan \phi_0 > 0\)) above \(\approx 30\) K. Therefore, magnon excitation via the damping-like torque of the IFE is even the dominant excitation mechanism above 30 K. Below 25 K, we find deviations from the model prediction, which can be attributed to the incipient ordering of the Ho(4b) moments\(^{31,32}\) and related changes in the magneto-crystalline out-of-plane anisotropy \(D_z\).

After presenting this consistent model for the low-energy spin dynamics, we will now focus on the damping. Combining the results of Figs. 3a and b, we can extract \(A\alpha\) using Eq. (14). The results are shown in Fig. 4a. Surprisingly, the low-temperature deviations are less pronounced, but we find a significant increase of \(A\alpha\) between 30 K and 45 K. Towards higher temperatures, \(A\alpha\) approaches the theoretical limit of \(\sqrt{2}\).

Since we previously determined \(\Delta\) as well as the temperature dependence of \(D_y\) and \(|S|\), we can calculate \(D\). Subsequently, using \(\hbar \omega_0 = 2|S|/DA\), we gain access to the exchange enhancement factor \(A\) (Fig. 4b). The minimum of \(D\) at the SRT leads to an enhancement of \(A\). We can now further determine the Gilbert damping parameter \(\alpha\) (Fig. 4c). Note that the increase of \(A\alpha\) around the SRT temperature can be mostly attributed to the enhancement of \(A\), while we find only a minor increase in the Gilbert damping \(\alpha\). Note further that the anomalous increase of \(\alpha\) is confined to small temperature range above \(T_{jump}\). Our measurements are consistent with previous non-linear optical imaging experiments on HoMnO\(_3\), which revealed the presence of a small-domain state after entering the reoriented phase\(^{33,34}\). Magnon scattering at the domain walls could be a possible explanation for the enhanced damping. Since the observed effect is only weakly pronounced, we conclude that the spin dynamics at any temperature are determined by the bulk-like response in the antiferromagnetic domains while the ratio of the domain walls on the probed volume is negligible even in the small-domain state.

Note that by combining Eqs. (7) and (5), the energy carried per spin after the excitation can be calculated as \(E(0^+) = (A^2 \alpha^2 + 1) \gamma^2 \mu_B^2 H^2 \theta^2 |S|^2 D_y\). The contribution proportional to \(A\alpha\) originates from the initial spin canting along the \(z\) axis. Thus, if \(A\alpha > 1\), then the so far neglected damping-like torque of the IFE becomes dominant.

Assuming that the Gilbert damping is microscopically related to scattering with bosonic quasiparticles, we can further evaluate the temperature dependence of \(\alpha\). The grey line in Fig. 4c is proportional to a Bose-Einstein distribution describing the population of a bosonic quasiparticle excitation with an energy of approximately 4.3 meV. This suggests that scattering with the crystal field excitations of the Ho 4\(f\) electrons (3.5 meV\(^{35}\)) yields a significant contribution to the spin damping.
FIG. 4: Interplay of Gilbert damping and ellipticity of the spin precession in HoMnO$_3$. 

a $A\alpha$ product extracted from Z-mode frequency and lifetime. 

b Temperature-dependent ellipticity $A$ of the spin precession. 

c Extracted Gilbert damping parameter $\alpha$.

B. YMnO$_3$: Second-order AFM-PM phase transition

We found that the Gilbert damping in HoMnO$_3$ is potentially related to scattering on crystal field excitations of the Ho 4$f$ electrons. To substantiate that, we therefore performed equivalent measurements to those on HoMnO$_3$ also on YMnO$_3$, where no 4$f$ electrons are present. The results are presented in Fig. 5. We fit the temperature dependence of $\omega_0$ with $\Delta = 0$ and a temperature independent $D_y$. $\omega_0$ thus directly reflects the temperature dependence of $|S|$. We find $T_N = (74.7 \pm 0.1)$ K in agreement with literature. Assuming $J = 2.45$ meV and $D_z = 0.48$ meV from neutron scattering, we extract $D_y = 0.86$ µeV and thus a temperature-independent value for the exchange enhancement $A = 69$.

As anticipated, the Z-mode lifetime in YMnO$_3$ is about a factor of 20 larger than in HoMnO$_3$ pointing towards scattering with crystal-field excitations of the Ho 4$f$ electrons as the dominant damping mechanism in HoMnO$_3$. The reduced damping allows us to study the spin dynamics of YMnO$_3$ also in the vicinity of the Néel temperature $T_N$. The lifetime decays approximately exponentially with increasing temperature, but drops drastically when approaching $T_N$. The lower damping is also reflected in a smaller initial phase. By again combining frequency and lifetime of the spin precession, we can calculate $A\alpha$ using Eq. (14). As we already
FIG. 6: Temperature dependence of Gilbert damping parameter in YMnO$_3$. Gilbert damping extracted from magnon lifetime in. Bold line: Bose-Einstein distribution assuming a temperature-independent quasi-particle energy of 2.6 eV. Dashed line: Bose-Einstein distribution assuming a softening of the quasi-particle energy proportional to the undamped magnon-frequency $\omega_0$.

quantified $A$ from the temperature-dependence of $\omega_0$, we thus obtain the Gilbert damping parameter $\alpha$. The results are shown in Fig. 6. Up to approximately 60 K, the Gilbert damping parameter stays below $10^{-3}$. It follows a Bose-Einstein distribution with a quasiparticle energy of $\approx 2.6$ meV (bold line), which is close to the energy of a hybrid spin-lattice mode. Assuming that this mode softens proportional to $\omega_0$, we can consistently fit the temperature dependence of $\alpha$ up to $T_N$.

When approaching the Néel temperature, however, the frequencies of all magnon modes approach zero as the ordered magnetic moment $|S|$ vanishes. Therefore, the thermal population of all modes diverges and magnon-magnon scattering contributes to the pronounced increase of $\alpha$ for $T > 65$ K.

Combining our results for $A$ and $\alpha$ with Eq. (13), we can calculate the temperature dependence of the initial phase. This is shown in Fig. 5c. While they closely follow the trend of the measured initial phase, confirming that the initial phase is indeed affected by the Gilbert damping, we can resolve a clear difference between model and measurement.

We propose to attribute this discrepancy to a secondary excitation mechanism, such as an optical spin transfer torque or optical orientation. Both processes are based on the residual absorption of the material at the pump photon energy, which can lead to an angular momentum transfer from the circularly polarised pump pulse to the material.

We heuristically include such a mechanism into our model by modifying the initial conditions in Eq. (7). We set:

$$m(0^+) = \begin{pmatrix} m_0 \\ \gamma \mu_0 H \theta m_0 \\ \alpha \gamma \mu_0 H \theta m_0 + \delta \end{pmatrix}, \quad (15)$$

This changes the initial phase to

$$\tan \phi_0 \approx A \left( \alpha + \frac{\delta}{\gamma \mu_0 H \theta m_0} \right), \quad (16)$$

Thus, as $\alpha$ is low in YMnO$_3$, a small absorption is sufficient to significantly affect the initial phase. Even though the pump photon energy of 0.95 eV is far away from the nearest absorption resonance at $\approx 1.6$ eV, the residual absorption cannot be neglected. For a Lorentzian resonance, the residual absorption scales proportional to the linewidth, which we assume to be proportional to $T$. Taking both contributions - the damping-like torque of the IFE and the absorption-based mechanism - into account, we achieve to obtain quantitative agreement between the measured and modeled initial phases (Fig. 5c). In the case of HoMnO$_3$, the absorption-based excitation was overshadowed by the strong damping-like torque of the IFE.
IV. SUMMARY

We presented a comprehensive theoretical and experimental study of antiferromagnetic spin dynamics, where we showed that the effects of spin damping during impulsive optical spin excitations cannot be neglected. The rich magnetic landscape of the hexagonal rare-earth manganites provides the necessary toolbox to study coherent spin excitations in various regimes. Based on a phenomenological theory, we develop a consistent picture involving changes of the magnetic order parameter, the magneto-crystalline anisotropy and the Gilbert-damping parameter $\alpha$. In particular by evaluating the initial phase, we show the presence of an ultrafast non-thermal damping-like torque during the spin excitation. This additional torque, which is induced by inevitable damping effects during the optical excitation, has not been considered so far. We show, however, that it can in fact provide the dominant contribution to the spin excitation in strongly damped scenarios. The high efficiency of the damping-like torque is based on the pronounced exchange enhancement in antiferromagnets. We emphasise though that any torque along the magnetic hard axis will profit from this enhancement. Exploiting this enhancement opens up new possibilities for the energy-efficient ultrafast control of antiferromagnetism paving the way towards antiferromagnetic spintronic applications.

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