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Effect of laser pulse propagation on ultrafast magnetization dynamics in a birefringent medium

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Abstract

Light propagation effects can strongly influence the excitation and the detection of laser-induced magnetization dynamics. We investigated experimentally and analytically the effects of crystallographic linear birefringence on the excitation and detection of ultrafast magnetization dynamics in the rare-earth orthoferrites $(\text{Sm}_{0.5}\text{Pr}_{0.5})\text{FeO}_3$ and $(\text{Sm}_{0.55}\text{Tb}_{0.45})\text{FeO}_3$, which possess weak and strong linear birefringence, respectively. Our finding is that the effect of linear birefringence on the result of a magneto-optical pump-probe experiment strongly depends on the mechanism of excitation. When magnetization dynamics, probed by means of the Faraday effect, is excited via a rapid, heat-induced phase transition, the measured rotation of the probe pulse polarization is strongly suppressed due to the birefringence. This contrasts with the situation for magnetization dynamics induced by the ultrafast inverse Faraday effect, where the corresponding probe polarization rotation values were larger in the orthoferrite with strong linear birefringence. We show that this striking difference results from an interplay between the polarization transformations experienced by pump and probe pulses in the birefringent medium.

Keywords: laser-induced magnetization dynamics, ultrafast spin-reorientation transition, weak ferromagnets, crystallographic birefringence, ultrafast inverse Faraday effect

(Some figures may appear in colour only in the online journal)

1. Introduction

Efforts to find new, faster ways of manipulating the magnetic states of media have yielded spectacular and unexpected results over the past two decades [1–3]. Experiments showed that femtosecond laser pulses can trigger a variety of (sub)picosecond changes in magnetic materials, including ultrafast demagnetization [4–6]; coherent spin deflection followed by homogeneous precession [7] and spin waves [8, 9]; various phase transitions resulting in the emergence of magnetization [10–13] or its reorientation [14, 15]; emergence of non-equilibrium magnetic states [16]; complete magnetization reversal [17–24]; spin transport [6, 25, 26]; and even modification of exchange

interactions [27]. As these laser-induced events are 100 to 1000 times faster than the elementary operations in currently used magnetic recording devices, a study of ultrafast magnetism can facilitate new technologies for ever faster recording and processing of magnetically stored information.

Magneto-optical pump-probe techniques, where femtosecond laser pulses are used for both excitation and detection of the dynamics in media, are the primary tools for studying ultrafast magnetism. Here laser-induced magnetic changes are detected indirectly, via various magneto-optical phenomena. The most used measurement techniques employ the fact that the magnetization lifts the degeneracy between the right and left handed circularly polarized states of light, which results

in the Faraday effect in transmission and the magneto-optical Kerr effect in reflection. Such an indirect probe of spins naturally raises concerns about the interpretation of the results of the measurements. For example, it has long been a subject of intense discussion whether the relation between the magneto-optical Kerr rotation and the magnetization, known for thermal equilibrium, holds also on the subpicosecond time scale [28–30]. Another problem is how the results of magneto-optical pump-probe experiments are affected by the propagation of pump and probe pulses in media [31]. For instance, if crystallographic birefringence is not negligible, two circularly polarized waves are not eigen states of light in such a medium, meaning that both the effect of the polarized pump pulse on the spins and the effect of the spins on the probe pulse polarization will depend on the propagation distance in the medium. Various authors have so far addressed parts of this problem, for example related to effects of the birefringence on the probe polarization [32] or on the pump polarization [33] transformations in the process of excitation and detection of coherent spin precession. However, in order to interpret the result of a pump-probe experiment realistically, the effects of propagation of the pump and probe pulses have to be considered together. To the best of our knowledge, an analysis of the interplay of the effects of propagation of both pulses in birefringent media has not been done yet.

The striking example of experiments in which the interplay of the pump and probe propagation effects could play an important role is the pump-probe experiments on rare-earth (RE) orthoferrites. These strongly birefringent materials are model systems in ultrafast magnetism, in which several important pump-induced phenomena have been observed [7, 13–15]. Among these are some that are excited via strongly polarization dependent optomagnetic effects, as well as some that are based on ultrafast heating and thus mostly insensitive to the polarization of the pump. The latter distinction is of particular interest, since it is likely that birefringence will affect these two types of effects differently.

In this article we investigate experimentally and analytically the combined effects of crystallographic linear birefringence on the laser-induced excitation and detection of magnetization dynamics in mixed RE orthoferrites (RE1,RE2)FeO₃. Samples of two different compositions were chosen such that one possessed high and the other very low crystallographic birefringence. By performing magneto-optical pump-probe experiments and analytical calculations we demonstrate that the effect of linear birefringence on the measured amplitude of laser-induced magneto-optical signal depends strongly on the mechanism by which the dynamics is excited.

2. Samples

For our investigation we grew two types of RE orthoferrite single crystals, (Sm_{0.55}Tb_{0.45})FeO₃ and (Sm_{0.5}Pr_{0.5})FeO₃, using the floating zone technique [34]. These orthoferrites are dielectrics possessing weak ferromagnetism, i.e. both the magnetization **M** and the antiferromagnetic vector **L** are non-zero [35]. Also, they possess pronounced temperature dependence

of their magnetocrystalline anisotropy. As a result, the studied crystals show two second order spin reorientation (SR) phase transitions observed as a rotation of the magnetization from the *c* axis (Γ_4 phase) to the *a* axis (Γ_2 phase) via the angular Γ_{24} phase [35]. The phase transition temperatures, T_1 and T_2 , are defined by the rare-earth ions RE1 and RE2 and can be tuned up to room temperature and above, e.g. in the case of (Sm,Tb)FeO₃ [36, 37]. Optically, (RE1,RE2)FeO₃ are biaxial crystals and, therefore, their optical axes do not coincide with the crystallographic ones. Importantly, the strength of their crystallographic birefringence can be effectively tailored by changing the sample composition. In particular, in (Sm,Pr)FeO₃ the birefringence can effectively be tuned to zero by bringing the fraction of Pr-ions close to 0.5 [38, 39].

From each grown crystal, we cut a sample in the form of a thin plane-parallel plate (158 μm for (Sm,Tb)FeO₃ and 93 μm for (Sm,Pr)FeO₃). The faces of these plates were perpendicular to the crystallographic *c* axes, such that light propagating along the sample normal experienced a finite birefringence Δn_{ab} . In order to characterize the magnetic and magneto-optical properties of the samples, we measured the polarization rotation of light at the wavelength of 632.8 nm, propagating along the *c* axis. The measurements were done as a function of temperature and in an external magnetic field applied along the *c* axis. With no birefringence, the polarization rotation β of the light passing through the crystal would be fully caused by the Faraday effect and therefore directly proportional to the magnetization along the *c* axis M_c and the sample thickness. However, since orthoferrites are birefringent and the *c* axis is not their optical axis, the absolute value of the polarization rotation angle can be severely reduced. Inside these samples, the polarization will typically vary periodically between linear and elliptical states under the influence of crystallographic linear birefringence, as illustrated in figure 1(a) [40]. Moreover, for some thicknesses the polarization rotation might even vanish completely (see figure 1(a) panel 3).

The results of the measurements are plotted in figures 1(b) and (c). Two important conclusions can be drawn from these data. First, both samples possess a spin reorientation region in which the measured polarization rotation β as a function of temperature grows from zero, corresponding to the magnetization lying in the sample plane, to its maximum value β_{max} , indicating that the magnetization has aligned along the *c* axis of the sample. In (Sm,Tb)FeO₃ (figure 1(b)) and (Sm,Pr)FeO₃ (figure 1(c)) we find the spin reorientation between $T_1 = 210$ K and $T_2 = 265$ K and between $T_1 = 98$ K and $T_2 = 130$ K, respectively. Secondly, the absolute value of the polarization rotation in the high temperature region $T > T_2$ differs in the studied samples by an order of magnitude. Generally, for orthoferrite samples with a thicknesses of the order of 100 μm one would expect to measure at most a few degrees of polarization rotation [41]. Our data for (Sm,Tb)FeO₃ are in line with this expectation. However, the absolute value of the polarization rotation angle β_{max} in (Sm,Pr)FeO₃ is much larger, confirming that, in agreement with [38] and [39], the birefringence Δn_{ab} is strongly reduced.

3. Spin dynamics experiment

To excite and detect ultrafast magnetization dynamics we used a conventional magneto-optical pump-probe technique [1]. Pump and probe laser pulses had a central wavelength of 800 nm and a pulse length of 100 fs. The energy of the pump pulse was 9 μ J and the probe pulse was about 50 times weaker. In our experiments the circularly polarized pump pulses were incident under a small angle with respect to the sample normal. The probe pulses were at normal incidence, i.e. propagating along the c axis. They had their linear polarizations aligned along one of the crystallographic axes a or b such that the rotation of the light polarization, originating from the Faraday effect, would be proportional to the c component of the magnetization M_c [32]. On the sample, the pump formed a spot of approximately 100 μ m (full width at half maximum), while the probed area was somewhat smaller. A DC magnetic field of 1.3 kG was applied at an angle of 14° from the a axis in the direction of the c axis.

It has been demonstrated that magnetization dynamics in RE orthoferrites with a spin reorientation phase transition can be excited via two distinct mechanisms [7, 14]. In the first one a laser pulse is used to rapidly heat the material across the phase transition region, which causes a change of magnetic anisotropy on the picosecond timescale. It results in precession of the magnetization around the new quasi-equilibrium position and, upon damping of the precession, reorientation [14]. The second mechanism is based on the ultrafast inverse Faraday effect (IFE), which quasi-instantaneously brings the magnetic system out-of-equilibrium, thus triggering precession of the magnetization around its initial direction [7]. The dynamics induced by these two mechanisms can be distinguished by their dependencies on the pump polarization and the direction of the applied magnetic field [42, 43]. Therefore, we have measured the pump-induced dynamics of the probe polarization rotation β for four sets of parameters: left-handed σ^- and right-handed σ^+ circularly polarized pump pulses as well as positive H^+ and negative H^- applied magnetic fields (see appendix A). The contributions of the heat-induced and the IFE-induced magnetization dynamics are distinguished by calculating combinations $\beta^H = \frac{1}{4}\beta(H^+\sigma^+) + \frac{1}{4}\beta(H^+\sigma^-) - \frac{1}{4}\beta(H^-\sigma^+) - \frac{1}{4}\beta(H^-\sigma^-)$ and $\beta^{\text{IFE}} = \frac{1}{4}\beta(H^+\sigma^+) - \frac{1}{4}\beta(H^+\sigma^-) + \frac{1}{4}\beta(H^-\sigma^+) - \frac{1}{4}\beta(H^-\sigma^-)$, respectively.

4. Experimental results

The experimentally measured dynamics of the probe polarization is plotted in figure 2. The graphs in figure 2(a) show the pump-polarization-independent, magnetic-field-dependent pump-probe traces for both samples. In the data one can observe decaying oscillations as well as a change in quasi-equilibrium value, i.e. after several tens of picoseconds the signal settles at a value different from the starting one. The oscillation frequencies and their temperature dependence are in line with what one would expect for the quasi-ferromagnetic (qFMR) mode of spin precession in RE orthoferrites. The settling of

Figure 1. (a) Change of the polarization of light under the influence of both the Faraday effect and birefringence upon propagation through a non-excited medium (the static case). The graph shows how, with propagation distance z along the c axis of the orthoferrite crystal, the major axis of the polarization ellipse periodically rotates away from and towards its initial position along the crystal axis (dotted line). A period of a few tens of μ m is typical for 800 nm light in orthoferrites with moderate birefringence. The upper panels 1–4 contain schematic drawings of the polarization ellipse at four different propagation distances. ((b) and (c)) Polarization rotation in a 632.8 nm laser beam as a function of temperature in (Sm,Tb)FeO₃ and the (Sm,Pr)FeO₃, respectively. The laser beam was propagating along the c axes of the samples and was initially linearly polarized along one of the other crystal axes. The grey areas mark the spin reorientation regions. A DC magnetic field was applied along the c axis. Insets in ((b) and (c)) show magnetic field dependences of the polarization rotation measured in the high temperature T_4 phases. The lines are guides to the eye.

the signal at the new level originates from the magnetization reorientation. Both these processes are triggered by the ultrafast laser-induced heating across the temperature of the phase transition [14].

Figure 2. Laser-induced rotation of the probe polarization in $(\text{Sm,Tb})\text{FeO}_3$ (left) and $(\text{Sm,Pr})\text{FeO}_3$ (right) at different temperatures. We distinguish between dynamics excited by the pump-polarization-independent, magnetic-field-dependent effect of heating across a phase transition boundary (a) and the pump-polarization-dependent, magnetic-field-independent ultrafast inverse Faraday effect (b).

The graphs in figure 2(b) show the pump-polarization-dependent, magnetic-field-independent data. Here we see a strong peak at the point of pump-probe overlap ($t = 0$), followed by oscillations superimposed on the exponentially decaying process. The peak is due to a change in the magneto-optical rather than magnetic properties of the materials [43]. The oscillations, on the other hand, correspond to the qFMR mode of spin precession and are induced by the ultrafast IFE [7]. Note that in $(\text{Sm,Tb})\text{FeO}_3$, at low temperature, the ultrafast IFE excites precession at two frequencies. Identification of the origin of the second mode of oscillation, with frequency around 230 GHz, requires further investigation and is beyond the scope of this paper.

To compare the results of the pump-probe experiments in $(\text{Sm,Tb})\text{FeO}_3$ and the low-birefringent $(\text{Sm,Pr})\text{FeO}_3$, we examined the amplitudes of the oscillations shown in figures 2(a) and (b). The values of the amplitudes are extracted by fitting the data at positive time delays with the following function:

$$\beta(t) = Ae^{-t/\gamma} \cos(2\pi ft - \phi) + Be^{-t/\tau} + C. \quad (1)$$

The four fit parameters A , f , ϕ , and γ represent, respectively, the amplitude, frequency, initial phase and damping constant

Figure 3. (a) Frequencies of the magnetization precession excited via laser-induced heating and IFE as a function of the relative temperature $T_{\text{rel}} = T - 0.5(T_1 + T_2)$ in the two studied samples. ((b) and (c)) The amplitudes of the oscillations in the measured polarization rotation originating from qFMR spin precession in $(\text{Sm,Tb})\text{FeO}_3$ (filled dots) and $(\text{Sm,Pr})\text{FeO}_3$ (open dots) induced by heating (b) and the ultrafast IFE (c) as a function of the relative temperature. Also shown (squares) are the amplitudes of the 230 GHz oscillations observed in $(\text{Sm,Tb})\text{FeO}_3$ at low temperatures. The lines are guides to the eye.

of the oscillations. The term $Be^{-t/\tau}$ is introduced to describe the exponential relaxation process. We note that the characteristic time of the laser-induced SR transition is a few ps [14, 43] and it can therefore be treated as instantaneous on the time scale of interest. Therefore, the spin-reorientation is described in equation (1) by the constant C . As discussed above, in a few pump-probe traces in $(\text{Sm,Tb})\text{FeO}_3$ below $T = 80$ K, we also observed oscillations at a second frequency. For those cases the fit function was modified to include a second oscillation term. The oscillation frequencies are plotted as a function of temperature in figure 3(a). The qFMR frequencies approach zero near the SR boundaries, thus displaying typical behaviour.

Figures 3(b) and (c) show the temperature dependences of the oscillation amplitudes obtained from the fit. For heat-induced magnetization dynamics (figure 3(b)) we observe

much larger amplitudes of the oscillation in (Sm,Pr)FeO₃ than in (Sm,Tb)FeO₃. A part of this difference can be explained by differences in the onset temperatures and widths of the SR transitions. First, the specific heat at $T_1 = 210$ K in (Sm,Tb)FeO₃ is approximately twice as large as at $T_1 = 98$ K in (Sm,Pr)FeO₃ [44, 45]. Thus, when comparing amplitudes of the heat-induced precession in these two cases one should scale up the data for (Sm,Tb)FeO₃ by a factor of two. Moreover, in (Sm,Tb)FeO₃ as compared to (Sm,Pr)FeO₃, the spin reorientation region $T_2 - T_1$ is twice broader. It gives another factor of two. After taking these corrections into account, the amplitudes of the oscillations in the low-birefringent orthoferrite (Sm,Pr)FeO₃ are still about a factor of two higher.

This behaviour strongly contrasts with the results of the magnetization dynamics induced by the ultrafast IFE as shown in figure 3(c). Surprisingly, here the amplitudes of the oscillations measured in low-birefringent (Sm,Pr)FeO₃ are clearly smaller than in (Sm,Tb)FeO₃. This result is very counterintuitive. It cannot be explained by the effect of linear birefringence on the probe pulse, as this would simply cause a decrease in the measured polarization rotation, affecting heat-induced and IFE-induced magnetization dynamics equally. However, it can also not be explained by the effect of birefringence on the pump pulse, because that effect suppresses the probe pulse polarization rotation for IFE-induced magnetization dynamics more in samples with higher birefringence. We observe the opposite and, therefore, a more thorough analysis of the excitation and detection of spin dynamics in these two orthoferrites is required.

5. Calculations

To obtain deeper insight into this problem, we first calculated for various values of Δn_{ab} the distorting effect of linear birefringence on the pump pulse. Using this result, we then determined the magneto-optical Faraday rotation experienced by the probe pulse in the excited sample. To find the effect on the probe we also took into account linear birefringence. We consider a 100 μm -thick orthoferrite c sample, instantaneously excited at $t = 0$ via ultrafast laser-induced heating or the ultrafast IFE and chose the material to have a spin reorientation transition between 100 and 130 K. For the parameters of the excitation we chose values close to the ones in our experiment: a circularly polarized laser pulse with energy $E_p = 9 \mu\text{J}$ and central wavelength $\lambda_0 = 800$ nm is incident along the sample normal (c axis) and forms on the sample a spot with diameter $d = 100 \mu\text{m}$. Using these parameters, a calculation of the maximum temperature increase in the center of the spot yields $\Delta T_{\text{max}} = 11$ K (see appendix B).

Consequently, for each value of the linear birefringence Δn_{ab} our calculations involved two steps: an analytical calculation of the c -component of the amplitude of the spin precession δM_c as a function of the travel distance of light z in the sample and a calculation of the probe polarization rotation at the corresponding distance. Note that here we calculate the c component, because in the experiment we only detect changes in the magnetization along the c axis (the probe propagation

direction). The amplitude of the heat-induced precession is defined by the change of the equilibrium orientation of the spins due to spin reorientation phase transition. As a model for this spin reorientation we used the phenomenological description developed in [46]. It relates the angle θ , between the c axis and the equilibrium magnetization direction, to the temperature T as:

$$\sin^2(\theta) = \frac{T_2 - T}{T_2 - T_1}, \quad T_1 \leq T \leq T_2. \quad (2)$$

Considering the case when the initial temperature $T_0 = T_1$, the amplitude of the c -component of the heat-induced precession then becomes:

$$\delta M_c^H(z) = M \cos(\theta) = M \sqrt{\frac{\Delta T_{\text{max}} e^{-\alpha z}}{T_2 - T_1}}, \quad (3)$$

where M stands for the saturation magnetization.

For the IFE-induced dynamics we study the case $T_0 + \Delta T_{\text{max}} \leq T_1$. This allows us to treat the excitation of the sample via heating and the ultrafast IFE independently. Also, it simplifies our analysis, as we do not have to take into account the Faraday effect for the pump pulse. Considering that the strength of the ultrafast IFE is proportional to the pulse fluence and the fraction of circularly polarized light [7] as well as taking into account the effects of linear birefringence and absorption experienced by the pump pulse, the amplitude of the c -component of the IFE-induced precession will have the form:

$$\delta M_c^{\text{IFE}}(z) = R \cos\left(\frac{2\pi \Delta n_{ab} z}{\lambda_0}\right) e^{-\alpha z}. \quad (4)$$

Here R is the component of the magnetization along the c axis at $z = 0$ after a quarter period of IFE-induced precession. For the calculations we chose $R = \frac{M}{10}$, which is comparable with earlier reported values [7].

In the second step we calculated the probe polarization as a function of z considering both the z -dependence of the precession amplitudes and the influence of the crystallographic birefringence on the probe polarization. To that end we approximated $\delta M_c(z)$ by a staircase function (1000 pieces) and calculated the rotation of the probe pulse polarization for each piece separately using the transformation derived in [40]. For the specific Faraday rotation we took a value of 2170 deg cm⁻¹ [41].

Figure 4 shows the results of our calculations. In (a) and (d) we plotted the precession amplitudes $\delta M_c(z)$ of the heat-induced and IFE-induced magnetization dynamics for the case of a linear birefringence $\Delta n_{ab} = 0.02$. Note that the precession amplitude decreases monotonically with z for magnetization dynamics excited via ultrafast heating, whereas it varies periodically when the excitation mechanism is the ultrafast IFE. This periodic behaviour is due to the effect of birefringence on the polarization of the pump pulse, causing it to alternate between left-handed and right-handed circular polarization. If circular dichroism is not strong, the heat-induced effect is not sensitive to such changes in the pump polarization. For the given case of $\Delta n_{ab} = 0.02$ we also plotted how the major axis of the polarization ellipse of the probe pulse changes upon

Figure 4. Calculations for magnetization dynamics induced by heating and the ultrafast inverse Faraday effect. (a) and (d) show the calculated amplitudes of magnetization precession $\delta M_c(z)$ and (b) and (e) the corresponding rotation of the probe polarization with propagation in an orthoferrite sample with $\Delta n_{ab} = 0.02$ for heat and IFE-induced magnetization precession, respectively. (c) and (f) show, as a function of linear birefringence, the largest absolute polarization rotation that one could measure from samples not thicker than $100 \mu\text{m}$.

propagation through the sample (figures 4(b) and (e)). As in the static case depicted in figure 1(a), excitation through either mechanism causes it to periodically rotate away from and towards the c axis. However, the case of IFE-induced dynamics differs significantly from both the static case and the heat-induced dynamics, because on average the absolute value of the polarization rotation still increases upon propagation.

Finally, the lowest graphs show the largest absolute rotation that the polarization experiences in the first $100 \mu\text{m}$ of propagation for the cases of heat-induced (c) and IFE-induced (f) magnetization dynamics. One can see that generally a lower birefringence leads to a larger polarization rotation of the probe pulse for heat-induced magnetization dynamics. This is not the case for magnetization dynamics caused by the ultrafast IFE, given the birefringence is not very low ($\Delta n_{ab} > 0.003$ in figure 4(f)). This is a remarkable result that supports our experimental observation that birefringence affects the detection of heat-induced and IFE-induced magnetization dynamics in pronouncedly different ways.

6. Discussion

The calculation results shown in figure 4 allow us to understand why the two excitation methods affect the measured polarization rotation of the probe pulse so differently. With

propagation, the probe polarization alternates between a positive and a negative rotation away from the crystal axis. In the case of heat-induced magnetization dynamics, the Faraday effect will rotate the probe polarization further away from the crystal axis in the first half of the alternation period, but back towards the crystal axis in the second half (see figure 4(b)). Therefore the maximum polarization rotation experienced by the probe, depends on the length of the period. For a longer period (smaller birefringence), one generally measures a larger polarization rotation. On the other hand, in the case of excitation via the ultrafast IFE the direction and the value of the c -component of magnetization M_c alternate as well due to the alternations of the pump polarization. In case of a one-color experiment, this period is equal to the alternation period of the probe polarization. Therefore, the probe encounters a magnetization pointing towards its propagation direction in one half of its alternation period and against its propagation direction in the other half. This first of all causes the Faraday effect to always rotate the probe polarization further away from the crystal axis and, consequently, the amplitude of its periodic rotation increases with propagation distance (see figure 4(e)). A higher birefringence thus does not limit the maximum polarization rotation of a pulse probing magnetization dynamics induced by the ultrafast IFE (as shown in figure 4(f)).

The second consequence of the alternating nature of the IFE-induced magnetization dynamics is that it can shift the phase of the alternation cycle of the probe. The size of this phase shift depends on the input polarizations of the pump and probe pulses. In our experiment the choice of these parameters did likely result in a shift of almost $\frac{\pi}{2}$, as can be seen when comparing the graphs in figures 4(b) and (e). This allows us to make an important conclusion: by adjusting the input polarizations of the pump and probe pulses it is always possible to optimize the detection of IFE-induced magnetization dynamics independently of the sample thickness.

Additionally one can see in figure 4(f), and in one instance also in figure 4(c), that for certain values of the birefringence the largest absolute polarization rotation of the probe makes small jumps upwards. This is related to the fact that the absolute polarization rotation of the probe reaches maxima at discrete points in the sample. In case of the IFE, where the amplitude of periodic rotation of the probe polarization increases with propagation distance, one finds a slightly higher largest absolute polarization rotation each time the point where its maximum is reached coincides with the surface where the probe exits the sample.

Our analysis above reveals that the effect of linear birefringence on the polarization rotation of a probe pulse can indeed vary, depending on the excitation mechanism. It thus explains our experimental results. Note though that one can only find this effect when the pump and probe pulses are similarly affected by linear birefringence, as in the case when they have the same wavelength. The latter leads to a very important remark on the interpretation of two-color pump-probe experiments, e.g. the ones for DyFeO_3 reported by Iida *et al* [33]. In those situations the limitation discussed above is not working and by changing the wavelength of the pump pulses, one changes not only the efficiency of the excitation, but also the value of the linear birefringence experienced by the pump pulses and thus the amount of suppression of the rotation of probe polarization. This should be carefully taken into account when studying the spectral dependences of the laser-induced magnetization dynamics.

7. Conclusions

To conclude, we have shown both experimentally and analytically that in transmission pump-probe experiments linear birefringence strongly affects the outcome of the measurements of laser-induced ultrafast magnetization dynamics. For heat-induced magnetization dynamics one typically measures lower values of probe polarization rotation in samples with higher birefringence. However, due to the interplay between the effects of birefringence on the pump and probe laser pulses, the IFE-induced dynamics in high birefringence samples can produce similar values of probe polarization rotation as the IFE-induced dynamics in low birefringence samples. This is a consequence of the fact that birefringence distorts the polarizations of the same-color pump and probe pulses in a similar manner. As a result, transient magnetization changes its sign through the sample with the same period as the

polarization of the probe pulses changes from linear to elliptical. It can be seen as some analogy to the phase matching realized between laser-induced transient magnetization and probe pulses. Importantly, for such a phenomenon to be realized, similar birefringence values for pump and probe pulses are required. Furthermore, one can influence this interplay between pump and probe pulses by changing their input polarizations, making it possible to optimize the detection of the IFE-induced magnetization dynamics in orthoferrite samples of any thickness.

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Appendix A. Raw data

Figure A1 shows the raw data for each sample at three different temperatures: the first one is well below the SRT, the second is approximately in the middle of the SRT range, and the third is at the upper border of the SRT range (near T_2). We included each combination of left/right handed pump polarization and positive/negative applied magnetic field. One can see that far below the SRT the signal depends only on the helicity of the pump polarization, while within the SRT range also the direction of the applied magnetic field plays an important role. Close to T_2 both effects begin to vanish.

Appendix B. Calculation of sample heating

One way to excite ultrafast magnetization dynamics is by heating the sample with an ultrashort laser pulse. This will then trigger magnetization precession on a picosecond timescale if the temperature rise induces a spin reorientation phase transition. After the excitation, the temperature T inside the sample depends on the initial temperature T_0 and on the distance z from the surface where the light entered:

$$T(z) = T_0 + \Delta T_{\max} e^{-\alpha z}. \quad (\text{B.1})$$

$\Delta T_{\max}$ is the temperature in the middle of the pump spot at the surface where the pump pulse enters the sample. It can be calculated from the pump pulse fluence U and the fraction $\mathcal{T}$ of light remaining after reflection, as

Figure A1. Raw data for three different temperatures: the first is well below the SRT, the second in the middle of the SRT range and the third is at the upper border of the SRT range, near T_2 .

$$\Delta T_{\max} = \frac{\alpha N_A V T U}{4C}, \quad (\text{B.2})$$

with N_A being the Avogadro constant and

$$U = \frac{4 \ln(2) E_p}{\pi d^2}, \quad (\text{B.3})$$

$$\mathcal{T} = 1 - \left(\frac{n-1}{n+1} \right)^2, \quad (\text{B.4})$$

where $E_p = 9 \mu\text{J}$ is the pump pulse energy, and $d = 100 \mu\text{m}$ is the pump spot diameter. Using realistic parameters for the absorption coefficient $\alpha = 240 \text{ cm}^{-1}$ [47], the molar heat capacity $C = 50 \text{ J K}^{-1} \text{ mol}^{-1}$ [44, 45], the unit cell volume $V = 236 \text{ \AA}^3$ [48], and the refractive index $n = 2.35$ [47], $\Delta T_{\max}$ is then calculated as 11 K.

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