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Femtosecond single-shot imaging and control of a laser-induced first-order phase transition in HoFeO$_3$

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Abstract

Excitation of antiferromagnetic HoFeO$_3$ with a single 80 fs laser pulse triggers a first-order spin-reorientation phase transition. In the ultrafast kinetics of the transition one can distinguish the processes of impulsive excitation of spin precession, nucleation of the new domain and growth of the nuclei. The orientation of the spins in the nuclei is defined by the phase of the laser-induced coherent spin precession. The growth of the nuclei is further promoted by heating induced by the laser excitation. Hereby we demonstrate that in HoFeO$_3$ coherent control of the spin precession allows an effective control of the route of the heat-induced first-order magnetic phase transition. The theoretical description of the excitation of the spin precession by linearly-polarized ultrashort laser pulses is developed with the sigma model. The analysis showed high sensitivity of the excited dynamics to the initial spin orientations with respect to the crystallographic axes of the material.

Keywords: pump–probe, magneto-optics, imaging, domains, ultrafast, light-induced

(Some figures may appear in colour only in the online journal)
first-order spin-reorientation phase transition in HoFeO$_3$ and compare it with the results obtained earlier for (Sm, Pr)FeO$_3$ and DyFeO$_3$.

Here we investigate in detail ultrafast kinetics of the photo-induced first-order spin-reorientation phase transition in dielectric HoFeO$_3$. We show that a single linearly polarized laser pulse can launch the spin-reorientation and drive the collective spin rotation over 90 degrees. All-optical control of the magnetization in the photo-induced state can be achieved by varying the linear polarization of the pumping light and the sign of the magnetization in the ground state. We demonstrate that ultrafast time-resolved imaging of the photo-induced magnetization reveals stages of nucleation and consequent growth of the nuclei. These stages are typical for photo-induced magnetization and summarize the results.

In section 4 we demonstrate how the sign of the photo-induced magnetization component ($M_x$) can be controlled by the pump polarization and the direction of the net magnetic moment is pointing solely along the $z$-axis. This rotation results in an emergence of non-zero magnetization along the $z$-axis. The corresponding angular phase is denoted as the $\Gamma_1$ phase. At the temperature $T_5$ a first-order phase transition occurs during which the spins suddenly rotate toward the (010) plane retaining the angle $\theta$ with the $z$-axis. This rotation results in an emergence of non-zero magnetization along the $z$-axis. The corresponding angular phase is denoted as $\Gamma_{2\delta}$. A further increase of the temperature pulls the antiferromagnetic vector in the (010) plane towards the $x$-axis. This rotation is accomplished at $T_3 = 58$ K when the spins are oriented along the $x$-axis and the net magnetic moment is pointing solely along the $x$-direction. This $\Gamma_2$ phase persists up to the Neel temperature $T_N = 647$ K [17]. Figure 1(a) schematically shows the whole set of the transitions between the magnetic phases in the range of temperatures from $T_1$ to $T_3$.

3. Experimental set-up and magneto-optical characterization of the sample

For the magneto-optical study of the spin-reorientation phase transitions in HoFeO$_3$ we took a crystal cut perpendicularly to the $z$-crystallographic axis. The unfocused beam from an optical parametric amplifier at a central wavelength of 630 nm was used to probe the sample. The OPA was pumped by 80 fs laser pulses at the central wavelength of 800 nm from an amplified Ti:sapphire laser (Spectra Physics Spitfire). The repetition rate of the pulses was 1 kHz. A charge-coupled device (CCD) camera in combination with collective optics and two polarizers in the cross-Nicol configuration was used to visualize the magnetic structure by sensing the magneto-optical Faraday effect. The probing laser beam was nearly at normal incidence. Thus it is sensitive only to the out-of-plane magnetization component ($z$-component). It must be noted that in contrast to the measurements reported in [11], the sensitivity of this setup greatly depends on the orientation of the polarization of light with respect to the axis of the polarizer in front of the CCD camera. Below the temperature $T_2$ the interaction the magnetizations $M_1$ and $M_2$ of the two magnetic sublattices acquire a relative canting over an angle of about 0.5 degrees. Due to the canting the antiferromagnet acquires a non-zero net magnetic moment $M = M_1 + M_2$. The value and the direction of $M$ is given by the relative orientation of the antiferromagnetic vector $L = M_2 - M_1$ with respect to the crystallographic $y$-axis according to the relation:

$$M = \frac{H_D}{H_{ex}}e_y \times L,$$

where $H_D$ and $H_{ex}$ are the Dzyaloshinskii and the exchange fields, respectively; $e_y$ is a unit vector along the even axis of the crystal (the $y$-axis) [13, 14]. It is known that in a narrow temperature range from $T_1 = 39$ K to $T_3 = 58$ K the orthoferrite exhibits a number of magnetic phase transitions due to which the spins reorient from the $z$-axis to the $x$-axis [15–18]. Below $T_1 = 39$ K the spins are in the $\Gamma_2$ phase. In this phase $L$ is oriented along the $z$-axis and $M$ is oriented along the $x$-axis. In the temperature range from $T_1$ to $T_2 = 51$ K the antiferromagnetic vector gradually rotates in the (100) plane ultimately acquiring an angle $\theta$ with the $z$-axis nearly equal to 30$^\circ$. This angular phase is denoted as the $\Gamma_1$ phase. At the temperature $T_3$ a first-order phase transition occurs during which the spins suddenly rotate toward the (010) plane retaining the angle $\theta$ with the $z$-axis. This rotation results in an emergence of non-zero magnetization along the $z$-axis. The corresponding angular phase is denoted as $\Gamma_{2\delta}$. A further increase of the temperature pulls the antiferromagnetic vector in the (010) plane towards the $x$-axis. This rotation is accomplished at $T_3 = 58$ K when the spins are oriented along the $x$-axis and the net magnetic moment is pointing solely along the $z$-direction. This $\Gamma_2$ phase persists up to the Neel temperature $T_N = 647$ K [17]. Figure 1(a) schematically shows the whole set of the transitions between the magnetic phases in the range of temperatures from $T_1$ to $T_3$. 

2. Spontaneous spin-reorientation phase transitions in HoFeO$_3$

Holmium orthoferrite (HoFeO$_3$) crystallizes in an orthorhombic structure (point group is $D_{3d}^2$) [12]. Fe$^{3+}$ spins are coupled antiferromagnetically. Due to the Dzyaloshinskii–Moriya interaction the magnetizations $M_1$ and $M_2$ of the two magnetic sublattices acquire a relative canting over an angle of about 0.5 degrees. Due to the canting the antiferromagnet acquires a non-zero net magnetic moment $M = M_1 + M_2$. The value and the direction of $M$ is given by the relative orientation of the antiferromagnetic vector $L = M_2 - M_1$ with respect to the crystallographic $y$-axis according to the relation:
magnetic structure is characterized by the net magnetization oriented entirely along the $x$-axis. It cannot contribute to the Faraday effect and shows up as a homogeneous gray image (figure 1(b)). The emergence of the magnetization component along the $z$-axis results in a directly measurable Faraday rotation of the probe. This reveals an onset of the magnetic domain structure corresponding to the $\Gamma_{24}$ phase (figure 1(c)). A gradual increase of the temperature results in an enhancement of the magneto-optical contrast while maintaining the main features of the domain structure (figure 1(d)). Naturally, the bright and dark areas on the images were assigned to the magnetic domains in which $M_z$ points along and against the $z$-axis, respectively. Figure 1(e) summarizes the changes of the magneto-optical contrast as a function of temperature. It is seen that the contrast saturates at temperature nearly equal to 57 K. This value roughly agrees with the critical temperature $T_3$ [15].

In order to study the kinetics of the phase transition at an ultrafast timescale, we performed an all-optical pump–probe experiment. The sample was excited by a linearly polarized pump pulse with duration $\tau = 80$ fs. The central wavelength of the pump was 800 nm. The pump pulse had an incidence angle close to 20 degrees. The beam had a Gaussian spatial profile, being focused into a spot with the full width at half maximum $\sigma = 75 \mu$m. The thickness of the sample $d$ was equal to 70 $\mu$m. Similarly to [10], the repetition rate of the pump pulses was brought down to 2 Hz. The majority of the experiments was performed without external magnetic field.

4. Dependence of the photo-induced magnetic state on the polarization of the pump light and the antiferromagnetic vector

Figure 2(a) shows snapshots of the magneto-optical contrast in the low-temperature $\Gamma_{12}$ phase at 48 K recorded 550 ps after the pump excitation. This time delay is long enough to guarantee that the magnetization is in thermal equilibrium with the lattice [19]. It is seen from the images that a single linearly polarized pump pulse produces well pronounced changes of the magneto-optical contrast. The laser fluence used in the experiment was 100 mJ cm$^{-2}$. The corresponding absorption results in local heating of the lattice of about 10 K. Such a temperature increase is sufficient to trigger the spin-reorientation phase transition. Accordingly, the pump-induced changes of the
Faraday rotation were attributed to the emergence of magnetization oriented along the \( z \)-axis \( (M_z) \). This magnetization is naturally inherited by the high-temperature phases \( (\Gamma_4 \text{ and } \Gamma_2) \) and is not present in the low-temperature phases \( (\Gamma_3 \text{ and } \Gamma_1) \). Because of the continuous nature of the transition \( \Gamma_2 \rightarrow \Gamma_4 \), it is hard to unambiguously establish to which of these two phases the photo-induced magnetization belongs.

The photo-induced magnetization, shown in figure 2(a), demonstrates a strong dependence on the azimuthal angle \( \varphi \) which the polarization plane of the pump light makes with the \( x \)-axis (see figure 2(b)). To characterize changes in the photo-induced state quantitatively, images were digitized and averaged were taken over the areas where the magneto-optical contrast had been changed. The polarization dependence has a \( 180^\circ \) periodicity with maxima for the azimuthal angles equal to \( \pm 45 \) degrees. In order to study how the magnetic ground state affects the photo-induced magnetization, we applied a magnetic field \( H_x \) oriented along the \( x \)-axis. The field changes the sign of the net magnetization \( M_x \) and consequently the sign of the \( L_z \) projection of the antiferromagnetic vector in accordance with equation (1). Figure 2(c) demonstrates that the transient photo-induced magnetization is sensitive to the
magnetic ground state and shows well pronounced hysteretic behavior. This indicates that a change of the sign of the magnetization in the ground state affects the direction of the photo-induced magnetization. To confirm this hypothesis we pumped various areas of the sample without external magnetic field after heating above $T_3$ and subsequent cooling to the initial temperature. Figure 2(d) shows that pumping the sample at spatially different areas may lead to opposite results. The periodic areas in which white or black photo-induced domains emerge represent the magnetic domain pattern of the $\Gamma_{12}$ phase. Similarly to [11] the sign of the photo-induced magnetization depends on the initial orientation of the spins and the laser-induced effect change sign upon changing the magnetic domain. Our findings regarding the sign of the photo-induced magnetization can be summarized in a simple form:

$$\phi = \text{sign}(M') \cdot \text{sign}(\sin 2\varphi).$$

Recently, a similar behavior was reported for the Morin phase transition in DyFeO$_3$ [10]. In that case the degeneracy between two orientations of the photo-induced magnetization was lifted by light-induced excitation of coherent spin precession. To understand the mechanism of the control of the sign of the photo-induced magnetization in HoFeO$_3$, one has to perform time-resolved experiments.

5. Time-resolved dynamics of the photo-induced magnetization

Figure 3(a) demonstrates the dynamics of the photo-induced magnetization for two distinct orthogonal linear polarizations of the pump pulse. We discriminate three main features in the pump-induced dynamics of the magnetization: (i) pronounced time delay preceding the magnetization growth, (ii) coherent high-frequency oscillation, (iii) gradual growth of the magnetization (see figure 3(b)). The phase of the oscillation is sensitive to the polarization of the pumping light in the same extent as the sign of the magnetization growth. This observation is a strong indication that in HoFeO$_3$, similarly to other orthoferrites [9–11, 20], light-induced coherent spin precession can lift the degeneracy between the magnetic states with magnetizations '$\uparrow$' or '$\downarrow$', respectively.

Further we limit our analysis of magnetization dynamics to the cases of the pump polarizations corresponding to the angles $\varphi = \pm 45$ degrees. We extracted the value for the growth rate of the magneto-optical signal from those dynamic segments which demonstrate nearly linear growth. Figure 3(c) shows that for the growth rate, the polarization sensitive contribution to the magnetization dynamics persists even down to 28 K.
This is far below than the lower border of the $\Gamma_{12}$ phase. It implies that the laser excitation can drive not only a single transition from $\Gamma_{12} \rightarrow \Gamma_{24}$ but a cascade of phase transitions $\Gamma_{12} \rightarrow \Gamma_{12} \rightarrow \Gamma_{24} \rightarrow \Gamma_{4}$.

In the following sections we discuss all these features in detail.

5.1. High-frequency coherent quasi-ferromagnetic soft mode

The frequency of the photo-induced oscillation $f_{\text{FM}}$ shows a softening down to 60 GHz if the sample temperature approaches $T_{\text{c}}$, see figure 4(a). This temperature behavior in the vicinity of the phase transition from the $\Gamma_{12}$ to the $\Gamma_{24}$ phases is a hallmark of the so-called quasi-ferromagnetic mode of the spin oscillations in HoFeO$_3$ [15]. The oscillation is a precession of the net magnetization around the equilibrium orientation, so that time-varying components $L_x$, $M_y$ and $M_z$ emerge.

In order to understand the mechanism of the light-induced excitation of the quasi-ferromagnetic mode we employed a phenomenological free energy approach [14]. The lowest order term $\Delta \Phi$ which is allowed by the magnetic symmetry of the low-temperature phases ($\Gamma_2$ and $\Gamma_{24}$) which describes interaction of linearly polarized light, having electric field component in the (0 0 1) crystallographic plane, with spins reads [13]:

$$\Delta \Phi = \frac{1}{16 \pi} (\mathcal{A}_{xxy} L_x L_y + \mathcal{B}_{xxy} L_x M_z + \mathcal{B}_{xxy} L_y M_z) \left( \mathcal{E}_x \mathcal{E}_y + \text{c.c.} \right).$$

(2)

Here $\mathcal{A}_{xxy}$, $\mathcal{B}_{xxy}$ and $\mathcal{B}_{xxy}$ are phenomenological parameters. The second factor in equation (2) is convenient to rewrite as:

$$\frac{1}{16 \pi} (\mathcal{E}_x \mathcal{E}_y + \text{c.c.}) = I(t) \sin 2\varphi,$$

where $I(t) = \frac{1}{B} \left| \mathcal{E}_x \right|^2$. $\mathcal{E}_x(t)$ and $\mathcal{E}_y(t)$ are time-dependent $x$ and $y$ components of the electric field of the pump pulse, respectively. The light intensity $I_0$, measured experimentally, is given by $I_0 = \int_{-\infty}^{+\infty} I(t) dt$.

The sigma-model equations [14] are determined by variation of the Lagrangian. For one spin, in the units of the Planck constant $\hbar$ the Lagrangian reads:

$$\mathcal{L} = T + G - W(L, t)$$

$$T = \frac{1}{2} \gamma H_{\text{ex}} \left( \frac{dL_x}{dt} \right)^2$$

$$G = -\frac{1}{H_{\text{ex}}} \left( \mathbf{H}_{\text{eff}} \cdot \left( L_x \times \frac{dL_x}{dt} \right) \right).$$

(3)

The first two terms in equation (3) determine the inertial $T$ and the gyroscopic $G$ dynamics of an antiferromagnet. $\gamma$ is the gyromagnetic ratio. The effective magnetic field $\mathbf{H}_{\text{eff}}$ in orthoferrites is given by the sum of the static Dzyaloshinskii field $\mathbf{H}_D = \mathbf{H}_D \left[ e_y \times \mathbf{L} \right]$ and an effective optomagnetic field created by the light pulse. The optomagnetic field is defined as $\mathbf{h}_{\text{opt}} = -\frac{\partial \Delta \Phi}{\partial \mathbf{M}}$, so that in the present case the total effective magnetic field reads:

$$\mathbf{H}_{\text{eff}} = \mathbf{H}_D - (\mathcal{B}_{xxy} e_y + \mathcal{B}_{xxy} e_y) I(t) \sin 2\varphi.$$
As a result, we show that the magnetization dynamics in HoFeO\textsubscript{3} can be launched via the inertial mechanism by means of linearly polarized light in phases which are characterized by non-zero $x$ and $y$ components of $\mathbf{L}$. The efficiency of the excitation is controlled by the azimuthal angle $\phi$. The inertial mechanism explains the symmetry of the soft mode excitation in the $\Gamma_{12}$ phase, for which $L_{\chi} \neq 0$. In contrast, this result anticipates that linearly polarized light incident along the $z$-axis cannot trigger the magnetization dynamics in the $\Gamma_{2}$ phase, which is solely characterized by $L_{z} = 0$. Indeed, for temperatures below 38 K the slope of the amplitude dependence demonstrates a pronounced change (see figure 4(b)). This temperature nearly matches with $T_{1}$. Below $T_{1}$, despite a significant drop, the amplitude of the oscillations remains non-zero and nearly temperature-independent.

The excitation of the spin dynamics in the $\Gamma_{2}$ phase cannot be explained within the inertial mechanism. In order to describe the light-induced spin dynamics in this phase, we took into account the gyroscopic term in the Lagrangian, see equation (3). This term in the $\Gamma_{2}$ phase reads:

$$\Delta G = -\frac{1}{H_{ex}}(\mathcal{B}_{xy} L_{z} \frac{dL_{x}}{dt} - L_{x} \frac{dL_{z}}{dt})$$

$$+ \mathcal{B}_{xy} L_{y} \left( L_{z} \frac{dL_{y}}{dt} - L_{y} \frac{dL_{z}}{dt} \right) J(t) \sin 2\varphi. \quad (9)$$

Taking into account that $L_{z} \gg L_{x}, L_{y}$, which is valid for magnetization dynamics in the $\Gamma_{2}$ phase, equation (9) can be reduced to:

$$\Delta G \approx -\frac{1}{H_{ex}} \mathcal{B}_{xy} L_{z} \frac{dL_{x}}{dt} J(t) \sin 2\varphi. \quad (10)$$

The Lagrange–Euler equation accounting for the gyroscopic term in the $\Gamma_{2}$ phase can be written as a closed equation for $L_{z}$ only and reads:

$$\frac{1}{\gamma H_{ex}} \frac{d^{2} L_{x}}{dt^{2}} - \frac{\partial W}{\partial L_{x}} = -\frac{1}{\gamma H_{ex}} \mathcal{B}_{xy} \frac{df(t)}{dt} \sin 2\varphi. \quad (11)$$

The time-dependent optomagnetic effective field via the gyroscopic term leads to the initial deflection of the magnetization [23, 24]:

![Figure 5](image-url)

**Figure 5.** (a) Time-resolved traces of the magneto-optical signal for images of the photo-induced dynamics in HoFeO\textsubscript{3}. The plots are obtained by taking the difference between those measured for two azimuthal polarizations of the pumping light $\phi = +45$ and $\phi = -45$. The sample temperature is 32 K. The inset shows the simulated spatial profile of the pump pulse. The dashed lines are guides to the eye. (b) The phase diagram of the magnetization growth rate as a function of the pump-fluence and the temperature. The color code represents the measured value of the growth rate. (c) Comparison of the the magnetization growth rate dependencies on the fluence and the temperature.
Thus we have shown that the quasi-ferromagnetic mode can be excited by means of light in both $\Gamma_2$ and $\Gamma_{12}$ phases. It is clearly seen that different mechanisms result in different efficiencies of the excitation of the spin oscillations.

5.2. Thermally driven dynamics of the photo-induced magnetization

The magnetization dynamics, averaged over the photo-induced domain, for different temperatures is shown on figure 3(b). The growth rate increases approaching $T_2$ and shows a pronounced non-linear temperature behavior (figure 3(c)). Interestingly, no peculiarities in the growth rate are observed once the temperature passes $T_1$. This indicates that the factor which promotes the growth is most likely not sensitive to the magnetic order itself.

To clarify the origin of the magnetization growth, we compared the rate of the growth measured for various temperatures with the same value obtained for various fluences of the pump. The fluence dependence can be easily extracted from the digital images, if one accounts for the Gaussian distribution of the fluence in the spatial profile of the pumping light (see inset in figure 5(a)). One can see that the fluence dependence, shown on figure 5(a), demonstrates striking similarities with the temperature dependence (see figure 3(b)). A comparison of the fluence and temperature dependencies of the growth rate, shown on figure 5(b), demonstrates that they have the same trend. We plotted a color map (figure 5(c)) of the magnetization growth rate, measured in CCD counts per second. The color map shows that in the vicinity of the transition from the $\Gamma_{12}$ to the $\Gamma_{12}'$ phase the fluence and the temperature are nearly proportional with the proportionality $4 \text{ mJ cm}^{-2} \text{ K}$. This experiment demonstrates that the photo-induced growth of the magnetization in HoFeO$_3$ is solely driven by the pump-induced increase in the lattice temperature. Indeed, taking the specific heat $C = 10 J (K\text{-mol})^{-1}$ [16], mass density $\rho \approx 10 g \text{ cm}^{-3}$ [25], atomic mass $A = 269$, the irradiation of the sample with the pump pulse having energy $E = 4\mu J$ results in a heating $\Delta T$:

$$\Delta T = \frac{E \cdot A}{C \cdot \rho \cdot \pi (0.5\sigma)^2 \cdot d} \cdot e^{-nd} \approx 17 \text{ K}. \quad (13)$$

Note, that the estimate of the laser-induced heating given in [11], exceeds the one obtained here. This can be related to an error in the definition of spot size in the earlier works. Performing imaging, the errors are less probable. However, in both cases it was concluded that the temperature raise defines incoherent dynamics of the magnetization. This incoherent magnetization dynamics can be attributed to the temperature-driven growth of the nuclei of the high-temperature phases ($\Gamma_{24}$ or $\Gamma_2$) in the volume of the initial phase. The initial temperature defines the initial number of nuclei and their growth rate. This may explain the experimentally observed non-linear dependency of the magnetization growth rate as a function of temperature (see figures 3(c) and 5(b)).

Interestingly, the observed temperature dependence of the magnetization dynamics along with the fluence dependence are qualitatively similar to the recently observed ultrafast light-induced first-order metal–insulator transition in V$_2$O$_3$ [26]. Despite the differences in the microscopic nature of these transitions, the light-induced dynamics of the order parameter are qualitatively the same. This points to the universality of the ultrafast dynamics of the order parameter triggered at first-order phase transitions.

5.3. Polarization-independent delay in the growth of the photo-induced magnetization

The time delay $\Delta t$ which precedes the onset of the photo-induced magnetization is a conspicuous feature visible in the pump–probe time traces (see both figures 3(a) and 5(a)). It reaches values up to 20 ps for low fluences of the pumping light. Interestingly, while $\Delta t$ is a strong function of the pump fluence, it is nearly independent of the sample temperature (figure 6). Most likely this is an experimental artifact caused by a transient crystallographic birefringence induced by the pump in the medium. The birefringence can negatively affect the sensitivity of the polarimeter used in this work, making the actual dynamics invisible. In particular, here we cannot see an onset of the inertial motion of the spins as reported in
[11]. Only upon relaxation of the birefringence on the scale of 10 ps, the laser-induced spin dynamics can be distinguished in the measurements.

6. Conclusions

We have shown that an ultrashort laser pulse can trigger spin reorientation in HoFeO$_3$ over 90 degrees. The direction of the magnetization in the transient photo-induced state can be switched from ‘up’ to ‘down’ by varying the linear polarization of the pump pulse with respect to the crystallographic axes. The same effect can be achieved if one changes the sign of the magnetization in the initial phase. We show that the ultrafast time-resolved dynamics of the photo-induced magnetization reveals pronounced stages of nucleation and subsequent growth of the new phase. Comparing the dynamics of the magnetization for various temperatures and fluences of the pumping light, it is shown that the magnetization growth is an incoherent process driven by the laser heating. The sign of the magnetization in the growing domains is defined by the polarization of the pump and the magnetization of the initial state. Again we confirm that the mechanism of the control of the route of the phase transition is based on excitation of coherent spin precession, which predefines the sign of the magnetization in the nuclei. The striking difference in the amplitude of the coherent spin precession in the magnetic phases $\Gamma_2$ and $\Gamma_4$ is related to the efficiency of the excitation mechanisms acting independently in these phases: inertial and the gyroscopic, respectively.

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