Magnetization dynamics in dysprosium orthoferrites via inverse Faraday effect

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The ultrafast non-thermal control of magnetization has recently become feasible in canted antiferromagnets through photomagnetic instantaneous pulses [A.V. Kimel et al., Nature 435, 655 (2005)]. In this experiment circularly polarized femtosecond laser pulses set up a strong magnetic field along the wave vector of the radiation through the inverse Faraday effect, thereby exciting non-thermally the spin dynamics of dysprosium orthoferrites. A theoretical study is performed by using a model for orthoferrites based on a general form of free energy whose parameters are extracted from experimental measurements. The magnetization dynamics is described by solving coupled sublattice Landau-Lifshitz-Gilbert equations whose damping term is associated with the scattering rate due to magnon-magnon interaction. Due to the inverse Faraday effect and the non-thermal excitation, the effect of the laser is simulated by magnetic field Gaussian pulses with temporal width of the order of hundred femtoseconds. When the field is along the z-axis, a single resonance mode of the magnetization is excited. The amplitude of the magnetization and out-of-phase behavior of the oscillations for fields in z and -z directions are in good agreement with the cited experiment. The analysis of the effect of the temperature shows that magnon-magnon scattering mechanism affects the decay of the oscillations on the picosecond scale. Finally, when the field pulse is along the x-axis, another mode is excited, as observed in experiments. In this case the comparison between theoretical and experimental results shows some discrepancies whose origin is related to the role played by anisotropies in orthoferrites.

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I. INTRODUCTION

In the last years the need of enhancing the speed of modern spin-electronic and magneto-optic devices, and of further developing the magnetic storage technology, has stimulated many studies aimed at achieving a fundamental understanding of the mechanisms of magnetization dynamics and switching. In addition to ultrafast magnetic field pulses which require complex devices for their generation, the spin dynamics has been induced by ultrafast optical laser pulses. Recent experiments have shown that significant demagnetization of magnetic compounds can be measured on the scale of a few hundred femtoseconds. Typically the light is absorbed by the material, giving rise to a rapid increase of temperature responsible for the change of magnetization and spin reorientation. The question concerning the exact speed of the initial sub-picosecond magnetization breakdown is still subject of debate and partly relates to the question of how to interpret magneto-optical experiments. Furthermore, the cooling time could limit the repetition frequency whose value is fundamental for actual applications.

Recently, non-thermal ultrafast optical control of magnetization has been achieved in canted antiferromagnet samples of dysprosium orthoferrites by using circularly polarized femtosecond pulses. Via the inverse Faraday effect, the light excitation acts on the spins of the system as a magnetic field pulse directed along the wave vector of the radiation and proportional to its intensity. The inverse Faraday effect does not rely on absorption and has its fingerprint in the fact that the helicity of the pump controls the sign of the photo-induced magnetization. This effect plays a role also in the femtosecond photomagnetic switching of spins in ferromagnetic garnet films.

Since the manipulation of spins by means of circularly polarized laser pulses represents an advance in the field of ultrafast magnetization dynamics, we analyze thoroughly the experimental work by Kimel et al. and discuss its peculiarities. In this experiment the difference between the Faraday rotations induced by right- and left-handed polarized pulses has been studied in the temperature range between 20 K and 175 K. A characteristic spin-wave mode, called quasi-antiferro mode, is excited by the light pulse along the z-direction. When the sample is heated, the frequency of the mode oscillations increases and the amplitude decreases from the low-temperature maximum value of the order of $M_S/16$, where $M_S$ is the saturation magnetization. We point out that there is no theoretical explanation concerning the magnitude and the temperature behavior of the oscillations. Furthermore, some aspects of the cited experiment deserve attention. Actually, in the so called $\Gamma_4$ phase, stable at temperatures higher than 50 K, the oscillations have temperature-dependent frequencies in full agreement with those measured by Raman experiments.

On the other hand, at temperatures below 50 K, the frequency of the photoinduced magnetization stays constant in contrast with the results of the Raman spectra and other measurements which signal the discontinuous transition to another phase, called $\Gamma_1$. Therefore, the experimental data obtained by laser pulses reflect the excitation of resonance modes characteristic of the $\Gamma_4$ phase even at very low temperatures. Finally, the field pulse is also directed along the x-axis: another spin-wave mode, called
quasi-ferro mode, is excited and is characterized by an extraordinarily, not well understood, small amplitude.

In order to obtain a deeper understanding of the nonthermal control of magnetization, in this paper we perform simulations related to the experiment by Kimel et al.\textsuperscript{12} We have studied the magnetization dynamics employing a model for orthoferrites that was previously proposed for the analysis of resonance and high-frequency susceptibility.\textsuperscript{12} The parameters of the free energy, such as the symmetric and antisymmetric exchange, and the anisotropy constants, are determined by using the experimental Raman spectra of Ref.\textsuperscript{17}. The dynamical behavior is described by solving two coupled sublattice nonlinear Landau-Lifshitz-Gilbert equations through a fifth-order Runge-Kutta algorithm. The damping term in the dynamical equations is related to magnon-magnon interaction and its temperature behavior is provided by a calculation of the scattering rate in orthoferrites.\textsuperscript{12} Exploiting the inverse Faraday effect, we have analyzed the effect of Gaussian magnetic field pulses whose time width is of the order of hundred femtoseconds. Since, in the regime considered in the experiments, the effective magnetic fields are not large if compared with exchange fields, the solution of the linearized system represents a reasonable approximation to the numerical results. Therefore we have studied the dynamics within the linear solution after the excitation by a pulse shaped as a delta function, since the magnetic field pulse takes place on a time scale shorter than the period of the resonance modes.

One result of this work is that, in the $\Gamma_4$ phase, the quasi-antiferro mode of the magnetization is excited by a field pulse along the $z$-axis and the oscillations have amplitudes in agreement with experimental results. Moreover, the oscillations induced by pulses directed along the $z$ and $-z$ axis show the characteristic out-of-phase behavior. We point out that, even for the dynamics, the ratio between the antisymmetric and symmetric exchange energies is important. Furthermore, we stress that in the canted antiferromagnets, such as rare-earth orthoferrites, the largest amplitudes of the oscillation are not obtained for the ferromagnetic sum vector of the sublattice magnetizations but for the antiferromagnetic difference vector. The behavior of the magnetization has been analyzed in the temperature range between 20 K and 175 K. The damping process based on magnon-magnon scattering describes the decay of the oscillations with results consistent with the experiment.

The case of the field pulse directed along the $x$-axis has been analyzed in the $\Gamma_1$ phase. The magnetization along the $x$-axis oscillates with the frequency of the quasi-ferro mode as found in the experiment. However, the calculated and the experimental amplitudes of the oscillation are different if the magnetic field pulse along the $x$-axis has the same intensity of that along the $z$-axis. Therefore, in the comparison with experimental data, we discuss the role of anisotropies between $z$ and $x$ directions as a source of discrepancy between theory and experiment.

Finally, we consider the actual stable phase in equilibrium at low temperatures, the $\Gamma_1$ phase, and its resonance modes. We point out that the difference in energy between the $\Gamma_1$ and $\Gamma_4$ can be very small, so that even a small laser-heating effect could be responsible for the stabilization of the $\Gamma_4$ phase at very low temperatures on a picosecond time scale.

The outline of this paper is as follows. In the next section we discuss the numerical approach for the system and the analytic solution of the linearized equations for excitation by a delta function shaped magnetic field pulse. Section III provides the numerical and analytical results: in the first and second subsection the excitation due to the pulses along the $z$-axis and $x$-axis, respectively, is analyzed when the system at equilibrium is in the $\Gamma_4$ phase. In the final subsection the effect on the dynamics of a $\Gamma_1$ phase stable at low temperatures is considered. Section IV provides a summary.

\section{II. Free Energy and Dynamical Equations}

Rare-earth orthoferrites are represented by the formula $ReFeO_3$, where $Re$ stands for rare-earth. They have a perovskite-type structure with slight deformation from cubic to orthorombic. In many of them the spins of iron ions are antiferromagnetically aligned through a strong super-exchange interaction with a Néel transition temperature of about 700K. Moreover, promoted by the orthorombic deformation, an antisymmetric exchange interaction acts between iron spins, resulting in spin canted magnetism with a feeble saturation moment. In the case of dysprosium orthoferrites, the temperature dependence of the ferromagnetic moment is characterized by a steep rise around 50 K in coincidence with the stabilization of the $\Gamma_4$ phase.\textsuperscript{12} Actually these compounds are also known for their spin reorientation properties: continuous rotational-type (ferromagnetism present in the low-temperature phase) and, only for dysprosium, abrupt-type from $\Gamma_1$ (antiferromagnetic) to $\Gamma_4$ with increasing temperature.

We use the free energy and the dynamical equations proposed in a previous work and focus on the behavior of the magnetization in the $\Gamma_4$ phase.\textsuperscript{18} The static and dynamical behavior is studied within the two-sublattice model for iron spins that takes into account the two active resonance modes. This represents an excellent approximation since the remaining resonance modes are almost inactive, and their interaction with active modes is negligible. In Fig.1 we show a schematic representation of equilibrium positions of two-sublattice magnetization vectors for dysprosium orthoferrites.\textsuperscript{18}

The normalized free energy $V = F/M_0$, with $M_0$ the modulus of the sublattice magnetization, is composed of a part $V_{exc}$ due to exchange interactions and a part $V_{ani}$ due to the anisotropy:

$$V = V_{exc} + V_{ani}. \quad (1)$$
The anisotropy constants $A$ by the equation change energy $Oe$. $g$ is of the order of 20. $v$ is of the order of 1. $M_H$ is of the order of $10^5$ cm$^{-1}$ $E$ is quite large compared to the other terms. $\beta_0 = 0.022$. $M_S = |\vec{M}_1 + \vec{M}_2| = 2M_0 \sin(\beta_0) \sim 0.022 M_0$ is two orders of magnitude less than $M_0$. The equilibrium position of $\vec{M}_1$ and $\vec{M}_2$ represents a stationary solution of the nonlinear Landau-Lifshitz-Gilbert equations

$$\frac{1}{\gamma} \frac{d\vec{R}_1}{dt} = -\vec{R}_1 \wedge (\vec{H}(t) - \vec{\nabla}_1 V) + \alpha \vec{R}_1 \wedge \frac{d\vec{R}_1}{dt}$$  \hspace{1cm} (5)$$

$$\frac{1}{\gamma} \frac{d\vec{R}_2}{dt} = -\vec{R}_2 \wedge (\vec{H}(t) - \vec{\nabla}_2 V) + \alpha \vec{R}_2 \wedge \frac{d\vec{R}_2}{dt},$$  \hspace{1cm} (6)

where $\gamma = 17.6$ MHz/Oe is the gyroscopic ratio, $\vec{\nabla}_1$ and $\vec{\nabla}_2$ are gradients with respect to $\vec{R}_1$ and $\vec{R}_2$, respectively, and $V$ is the energy in Eq. (1). Clearly the dynamical equations satisfy the following constraints: $X_1^2 + Y_1^2 + Z_1^2 = 1$ and $X_2^2 + Y_2^2 + Z_2^2 = 1$.

The quantity $\vec{H}(t)$ in Eqs. (5,6) is the time-dependent magnetic field simulating the effect of laser pulses due to the inverse Faraday effect. In the experiment by Kimel et al., the magnetic field pulse is of the order of fractions of Tesla with time width of hundred femtoseconds. In the numerical simulations we consider a pulse directed along the propagation direction of the light with a Gaussian shape

$$\vec{H}(t) = \hat{k} \frac{F_0}{\sqrt{\pi \tau_p}} \exp\left[-(t/\tau_p)^2\right],$$  \hspace{1cm} (7)

where $\hat{k}$ defines the direction of the light wave-vector and $\tau_p$ indicates approximately the duration of the pulse. $\alpha$ is the Gilbert constant. Actually, $\alpha$ takes into account the damping of the oscillations due to the magnon-magnon scattering and to the interaction of magnons with dysprosium spins and phonons. We notice that the scattering via dysprosium spins should be larger at very low temperatures where dysprosium ions tend to order. Moreover, the phonon-magnon scattering should be effective only on the nanosecond time scale. It is the magnon-magnon interaction that provides the prominent source of scattering on the picosecond time scale.

### A. Solution of linearized system

In this subsection we consider the solution determined by linearizing Eqs. (5,6) and study the excitation of this linear system due to a magnetic field pulse shaped as a delta function. This is reasonable since, in the regime considered in the experiments, the effective magnetic fields are not large if compared with exchange fields, and the temporal width of the pulse is much smaller than the periods of the resonance modes.

In order to take into account small deviations from the equilibrium, the standard approach is to consider two separate coordinate systems, $(S_1,T_1,Y_1)$ and $(S_2,T_2,Y_2)$, which describe the dynamics of $\vec{M}_1$ and $\vec{M}_2$, respectively.
The frequencies is important also to determine the value of the phase: from 150 GHz at 50 K to 450 GHz at about 220 K. Therefore this pulse will excite only the quasi-antiferro mode. The field along the z-axis, starting from the equilibrium position at $t = 0^-$, we find at $t = 0^+$

$$
\Delta X_1 (0^+) = -\Delta X_2 (0^+) = \cos (\beta_0) \cos (\gamma F_0) - 1,
\Delta Y_1 (0^+) = -\Delta Y_2 (0^+) = \cos (\beta_0) \sin (\gamma F_0),
\Delta Z_1 (0^+) = -\Delta Z_2 (0^+) = 0.
$$

Thus the system will be excited in the quasi-ferro mode. As the experimental field magnitude is lower than the exchange fields, we study the subsequent dynamical evolution within the linear approximation. Within a relaxation time approximation, we include the damping effect describing the decay towards the equilibrium position.

From the solution of the linear system in the case of a pulse along the z-axis, we get the evolution of the quasi-antiferro mode. The only ferromagnetic component, defined in Eq. (19), is

$$
\Delta M_z = e^{-2/\tau} \left[ \frac{\cos (\beta_0)}{2} \sin (2\beta_0) \sin^2 \left( \frac{\gamma F_0}{2} \right) \cos (\omega_{AFM} t) + 2 e^{-2/\tau} R \cos^2 (\beta_0) \sin (\gamma F_0) \sin (\omega_{AFM} t) + 2 e^{-2/\tau} \sin \beta_0 \cos^2 (\beta_0) [\cos (\gamma F_0) - 1] \right]
$$

when the pulse is instantaneous, it provides an initial condition to the dynamics described by the linearized equations of motions. We have analyzed two cases: field along z- and x-axis, since these are prominent for the experiment that we want to discuss.

For the field along the z-axis, starting from the equilibrium position at $t = 0^-$, we find at $t = 0^+$

$$
\Delta X_1 (0^+) = -\Delta X_2 (0^+) = \cos (\beta_0) \cos (\gamma F_0) - 1
\Delta Y_1 (0^+) = -\Delta Y_2 (0^+) = \cos (\beta_0) \sin (\gamma F_0),
\Delta Z_1 (0^+) = -\Delta Z_2 (0^+) = 0.
$$

Therefore this pulse will excite only the quasi-antiferro mode.

The field along the x-axis yields at $t = 0^+$

$$
\Delta X_1 (0^+) = -\Delta X_2 (0^+) = 0
\Delta Y_1 (0^+) = -\Delta Y_2 (0^+) = -\sin (\beta_0) \sin (\gamma F_0),
\Delta Z_1 (0^+) = -\Delta Z_2 (0^+) = \sin (\beta_0) [\cos (\gamma F_0) - 1].
$$

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\Delta Y_1 (0^+) = -\Delta Y_2 (0^+) = \cos (\beta_0) \sin (\gamma F_0),
\Delta Z_1 (0^+) = -\Delta Z_2 (0^+) = 0.
$$

Therefore this pulse will excite only the quasi-antiferro mode.

The field along the x-axis yields at $t = 0^+$

$$
\Delta X_1 (0^+) = -\Delta X_2 (0^+) = 0
\Delta Y_1 (0^+) = -\Delta Y_2 (0^+) = -\sin (\beta_0) \sin (\gamma F_0),
\Delta Z_1 (0^+) = -\Delta Z_2 (0^+) = \sin (\beta_0) [\cos (\gamma F_0) - 1].
$$

Thus the system will be excited in the quasi-ferro mode.

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$$

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Therefore this pulse will excite only the quasi-antiferro mode.

The field along the x-axis yields at $t = 0^+$

$$
\Delta X_1 (0^+) = -\Delta X_2 (0^+) = 0
\Delta Y_1 (0^+) = -\Delta Y_2 (0^+) = -\sin (\beta_0) \sin (\gamma F_0),
\Delta Z_1 (0^+) = -\Delta Z_2 (0^+) = \sin (\beta_0) [\cos (\gamma F_0) - 1].
$$

Thus the system will be excited in the quasi-ferro mode.
We notice in Eq. (22) that the even terms in the magnetic field are at least two orders of magnitude less than the odd term.

Along the other two directions, there are the antiferromagnetic component along x-axis (with respect to the equilibrium), \( \Delta A_X = M_0 \Delta \tilde{A}_X \), with \( \Delta \tilde{A}_X \) defined as

\[ \Delta \tilde{A}_X = \Delta X_1(t) - \Delta X_2(t) = X_1(t) - X_2(t) - (X_1^{eq} - X_2^{eq}), \]  

and the antiferromagnetic component along y-axis (with respect to the equilibrium), \( \Delta A_Y = M_0 \Delta \tilde{A}_Y \), with \( \Delta \tilde{A}_Y \)

\[ \Delta \tilde{A}_Y = \Delta Y_1(t) - \Delta Y_2(t) = Y_1(t) - Y_2(t) - (Y_1^{eq} - Y_2^{eq}). \]  

Using again the linearized form of Eqs. (22, 25, 26) are determined by the mode frequency \( \omega_{AFM} \), the angular \( \beta_0 \), and the quantity \( R \) which depends on the model parameters in the form

\[ R = \sqrt{ \frac{2A_{xx} + D^2/2E}{2E} }. \]  

At intermediate temperatures \( (A_{xx} \) very small) one gets

\[ R \approx D/2E. \]

When the pulse is directed along the x-direction, the quasi-ferro mode is excited. The components of the magnetization different from zero are those along x- and y-axis. In particular we consider the magnetization along x-axis (with respect to equilibrium), \( \Delta M_X = M_0 \Delta \tilde{M}_X \), where \( \Delta \tilde{M}_X \) is defined as

\[ \Delta \tilde{M}_X = \Delta X_1(t) + \Delta X_2(t). \]  

From the linearized dynamical equations this quantity is calculated as

\[ \Delta \tilde{M}_X = e^{-t/\tau} \sqrt{ \frac{4E}{A_{xx} - A_{zz}} } \sin^2(\beta_0) \sin(\gamma_F t) \sin(\omega_{FM} t). \]  

III. NUMERICAL AND ANALYTICAL RESULTS

Using the procedure outlined in the previous section, all the parameters appearing in the dynamical equations \( \{5, 6, 13\} \) can be determined. These nonlinear equations are numerically integrated through a fifth-order Runge-Kutta algorithm. The experimental laser pulses along the z-axis are estimated to be equivalent to magnetic fields with an amplitude of 0.3 T and a full-width at half-maximum of about 200 fs. Therefore, we consider Gaussian fields of the form given in Eq. (1), with \( \tau_\rho = 200 \) fs and \( Amp(H) \) typically 3000 Oe. This value of the effective field is small when compared to the exchange fields, so that effects due to the nonlinearity of the equations are negligible. Since the time width of the pulse is small on the scale of the mode periods, the results are not strongly dependent on \( \tau_\rho \). The linear solution with a delta pulse represents a good approximation, providing the right orders of magnitude but does not reproduce exactly the numerical results. Therefore, in all the following figures we plot the results obtained by the numerical integration.

In the first and second subsections we discuss the effects of a Gaussian pulse along z and x directions, respectively, when the system at equilibrium is in the \( \Gamma_4 \) phase. In the final one we briefly analyze the resonance modes and magnetization dynamics if the \( \Gamma_4 \) phase is stable at low temperatures.

A. Pulse along z-direction

As illustrated in Ref. 12, pulses along the z and -z directions excite the quasi-antiferro mode. Using the numerical procedure discussed in the preceding section, we have calculated the oscillating behavior of the ferromagnetic vector along z focusing on \( \Delta M_Z \) at \( T = 95 K \). As shown in Fig.2(a), the magnetization has a sine-like behavior with weak damping on a picosecond scale. In addition, opposite fields give rise to out-of-phase oscillations in agreement with the experimental data. We notice that after 50 picoseconds the amplitude is about half of the initial one, a value that is compatible with results reported in Fig.1 of Ref.12. This suggests that the damping term associated to magnon-magnon scattering provides a reasonable description of the reduction of the amplitude as a function of temperature.

Comparing the experimental data shown in Fig.1 of Ref.12 with the theoretical results shown in Fig.2(a) of this paper, we point out similarities and differences. Indeed, on the scale of the initial pulse width, there is a strong enhancement of the experimental Faraday rotation, probably due to the interference between pump and probe pulses. After this transient the oscillations induced by pulses with opposite helicities show similar amplitudes in time. In contrast with theoretical results, the equilibrium point of the Faraday rotations shows a decay and its behavior is different for left- and right-handed polarized pulses. Only later the oscillations tend toward a common equilibrium point that is different, however, from that before the pulse excitation. If the shift of the equilibrium positions is caused by a change of the anisotropy constants due to an intrinsic photomagnetic effect, then the amplitude of the oscillations should be...
different for the two helicities, as reported in a recent experiment. Since this is not the case in dysprosium orthoferrites, the change in the orientation equilibrium is more probably associated with a small but unavoidable laser-heating effect. Actually, within the model proposed in this paper, a shift of the equilibrium configuration could in principle be caused by the magnetic pulse during the excitation. This shift is negligible, however, since the amplitude of the effective magnetic field is much smaller than the antisymmetrical exchange field \( D \). Only by enhancing the amplitude of the pulse to values comparable to \( D \), the shift becomes important, and, upon further increase, even a precessional switching can occur. These amplitudes imply very high light intensities that are probably above the damage threshold of the samples.

In Fig. 2(b) we focus on the related quantity \( d(\Delta M_Z) \), defined as

\[
d(\Delta M_Z) = \Delta M_Z(H+) - \Delta M_Z(H-),
\]

with \( H+ \) and \( H- \) indicating positive and negative amplitudes, respectively. As reported in the experimental work, this quantity is less dependent on initial effects. Just after the transient induced by the pulse, this quantity has an amplitude of the order of \( M_S/16 \), in close agreement with the calculated data reported in Fig. 2(b). It is worthwhile understanding the order of magnitude of this amplitude by exploiting the result of the previous section for the delta pulse. From Eq. (22) we get for short times \( (t \ll \tau_2) \)

\[
\Delta M_Z \approx 2M_0R \sin(\gamma F_0) \sin(\omega_{AFM} t),
\]

with \( R \) given in Eq. (27). The amplitude is determined by the term \( R \) depending on the model parameters and by \( \sin(\gamma F_0) \) related to the pulse intensity. Using the experimental data, the impulse \( F_0 \) is estimated to be of the order of 3000 \( \times \) 400 Oe-fs. This implies that \( \gamma F_0 \) is of the same order as the angle \( \beta_0 \) responsible for the canted antiferromagnetism: \( \sin(\gamma F_0) \approx \gamma F_0 \approx 2\beta_0 \). Thus, we have

\[
\Delta M_Z/M_S \approx 2R \sin(\omega_{AFM} t).
\]

If we consider \( R \approx D/2E \), we obtain the order of magnitude \( 16d(\Delta M_Z)/M_S \approx 16 \times 2 \times 0.02 = 0.64 \) not so far from the experiment. Hence the ratio \( D/E \) is fundamental not only for the equilibrium configuration, but also for the magnetization dynamics in the canted antiferromagnet.

Within the theoretical approach, the dynamics of the antiferromagnetic vectors can be easily obtained, but these quantities have not been measured in the experimental work. Due to the fact that the equilibrium position corresponds to a maximum of \( A_X = M_0(X_1 - X_2) \), the temporal evolution of \( \Delta A_X = M_0\Delta A_X \), with \( \Delta A_X \) defined in Eq. (28), is characterized by a very small amplitude. From Eq. (29) one can deduce that the amplitude of \( \Delta A_X \) should be at least an order of magnitude smaller than that of \( \Delta M_Z \).

In Fig. 3 we show the results of the numerical calculation for the quantity \( \Delta A_Y = M_0\Delta A_Y \), where \( \Delta A_Y \) is defined in Eq. (24). We point out that now the amplitude is an order of magnitude larger than that of \( \Delta M_Z \) and the response is more sensitive to the pulse. Actually this is due to the fact that the dynamics of the components along the \( y \)-axis is strongly influenced by the highest energy scale \( E \). From Eq. (26) we obtain a rough estimate for short times \( (t \ll \tau_2) \)

\[
\Delta A_Y \approx 2M_0 \sin(\gamma F_0) \cos(\omega_{AFM} t).
\]

Therefore the amplitude is \( 2\sin(\gamma F_0)M_0 \approx 2\beta_0 M_0 \sim 0.022 M_0 = M_S \). Finally, we notice that, after 50 picoseconds, the damping acts in the same way as for the ferromagnetic vector along the \( z \)-direction, causing a reduction of about half of the initial amplitude.

In order to make contact to the experimental measurements, it is interesting to analyze the behavior of the
magnetization dynamics at different temperatures. As reported in Fig. 3 of the paper by Kimel et al. upon heating the sample, the frequency of the oscillations increases and the amplitude decreases. The oscillations have temperature-dependent frequencies that are very close to those measured by Raman experiments at temperatures higher than 50 K. In Fig. 4 of this paper the calculated $d(\Delta M_z)/M_0$ is shown. We find agreement with experimental decay of amplitudes and behavior of the frequency upon increasing the temperature for times after the transient. Within the theoretical approach, the temperature dependence of the anisotropy constants $A_{xx}$ and $A_{zz}$ affects the frequency of the modes, but is not so important in the change of the amplitude after the laser transient. The decrease of the magnetization seen at a fixed time (of the order of tens of picoseconds) for different temperatures is dominated by the increase of the damping constant $\alpha$ in temperature. Due to the agreement with experiment, the estimate of the damping constant derived on the basis of the magnon-magnon scattering is reliable.

The results of Fig. 4 show the magnetization normalized by the sublattice magnetization modulus $M_0$. We point out that, in principle, $M_0$ can vary as function of temperature. However, the inclusion of the temperature dependence of $M_0 = M_z/(2\sin(\beta_0))$ is not easy, since only the magnetization $M_z$ is typically experimentally available. In any case, even if the angle $\beta_0$ is assumed fixed, the change of $M_z$ is small in the considered temperature range.

Before closing this section, we focus on the dependence of the magnetic response on the amplitude of the pulse.

As inferred by Eq. (31), for the effective magnetic pulses used in the experiments, the ferromagnetic vector is proportional to $F_0$, i.e., to the intensity of the light pulse used in the inverse Faraday effect. In Fig. 5 we plot the numerical results for the amplitude of the magnetization at $t=40$ ps, i.e., after about 10 periods as in the experimental data. We find the expected linear behavior as a function of the amplitude of the pulse. The value obtained at 5 T for $T = 60$ K is not far from unity. Thus, the saturation of the magnetization along $z$ is reached for this high intensity, in agreement with the extrapolation of the experimental data in the inset of Fig. 2 of Ref. 12.

Summarizing, in this subsection we have focused on the magnetization dynamics in the $\Gamma_4$ phase when the field pulse is directed along the $z$-axis. We have analyzed the amplitude and the decay of the oscillations as a function of temperature finding agreement with experimental data for times longer than the initial transient. Actually, on the time scale of the initial pulse width, discrepancies with experiments appear. These could be due to a small but unavoidable laser-heating effect that is not included in the present theoretical approach.

B. Pulse along x-direction: role of anisotropy

In this subsection we analyze the effects of the excitation due to the pulse along the x-axis.

The fields along the x-direction excite another mode. As shown in Eq. (29), $\Delta M_x$ oscillates with the frequency of the quasi-ferro mode in agreement with experiment.
On the other hand, the magnitude of the magnetic response along x is of the same order as that obtained along the z-axis with a pulse of equal amplitude. Even if the torque exerted by the field along x is tiny, the subsequent temporal evolution, in particular, that of the variables $T_1$ and $T_2$, is able to give a non-negligible amplitude to the response along x. This is in discrepancy with the experimental results which show that the Faraday rotation along x is at least an order of magnitude smaller than that along the z-axis.2 Also, as shown in Fig.3 of Ref.12, it does not show any temperature dependence following the behavior of the frequency of the quasi-ferro mode, in striking contrast with the case of a pulse along the z-axis.

In order to properly compare the experimental and theoretical results, we should take into account several effects in orthoferrites.20 First of all, the experimental estimate of the effective magnetic field pulse due to the inverse Faraday effect is available only when the pulse is along the z-axis. Moreover, it is well known that in rare-earth orthoferrites optical birefringence is not negligible.27,28 Therefore the experimental Faraday rotations along the z- and x-axis can be different, even if the oscillations of the magnetizations are of the same order. Since the Faraday rotation along the x direction is very small in comparison with that along the z-axis, part of the effect could be also associated with the anisotropy of the magneto-optical susceptibility. Indeed, the effective magnetic field generated via the inverse Faraday effect can be different for the two orthogonal orientations. Finally, there is another source of anisotropy: the field along the z-axis can show a renormalization of the coupling to the system that is different from that along the x-axis.

In order to elucidate this last point, we have included in our model the anisotropy induced by the dysprosium ions. As a result, a modification of the coupling of the magnetic field to the iron ions takes place. According to a simple scheme proposed long ago by Zvezdin and Matveev,29 the coupling to the field along the z-direction is reduced by the factor $1 + \eta_z \simeq 0.74$ (see in Fig.2(b) the effect of this reduction). Instead, the reduction $1 + \eta_x$ of the coupling along the x-axis is temperature dependent and can be extracted by the measurements of magnetic properties.29 With temperature decreasing toward 50 K, these coupling factors can become a non-negligible source of anisotropy. As shown in Fig. 6, the difference in the amplitudes for the response along the z- and x-axis can become relevant at those temperatures. However, as shown in the inset of Fig. 6, the ratio between the amplitudes along x and z axis increases with temperature up to an inversion point. Therefore the source of the anisotropy associated only to dysprosium ions seems not to be effective for the interpretation of the experimental data.

### C. Stable phase at low temperatures

In this last subsection we focus on the $\Gamma_1$ phase that is stable at low temperatures.

The measurements reported in Ref.12 show the maximum value of the amplitude of the magnetization between 20 and 50 K. Moreover, the frequency of the photoinduced magnetization is constant in this temperature
The condition $V_1 < V_4$ implies that

$$A_{yy}^{(2)} > \frac{D^2}{16E} + \frac{A_{xx}}{8}. \quad (37)$$

Taking into account the values of the exchange fields given in the previous section and the anisotropy constant $A_{xx}(\Gamma_1)$, we derive $A_{yy}^{(2)} > 60$ Oe. Therefore the value of the constant $A_{yy}^{(2)}$ is consistent with the expansion of the free energy being only a fraction of the anisotropy energies $A_{xx}$ and $A_{zz}$.

If the parameter $A_{yy}^{(2)}$ is of the order of hundreds of Oersted, the phases $\Gamma_4$ and $\Gamma_1$ are close in energy. Due to a small light absorption, the laser pulse could affect the stability of the system by favoring the $\Gamma_4$ phase on a short time scale. Therefore the femtosecond pulse could induce a reorientational phase transition from $\Gamma_1$ to $\Gamma_4$ state in analogy with the antiferromagnetic-to-ferromagnetic phase transition induced by heating with a laser in FeRh films. Finally, we point out that a static magnetic field gives rise to a spin reorientational transition in orthoferrites. Hence the role of the magnetic field pulse induced via the inverse Faraday effect could be investigated in relation to the perturbation of the phase stability. This is left for future investigations.

IV. SUMMARY

Stimulated by recent experimental results showing ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses in dysprosium orthoferrites, a theoretical study of magnetization dynamics has been presented in this paper. We have employed a general form of free energy suitable for dysprosium orthoferrites whose parameters are derived from experimental measurements. We have solved coupled sublattice Landau-Lifshitz-Gilbert equations whose damping parameter is determined by considering the scattering rate due to magnon-magnon interaction. Due to the inverse Faraday effect, the magnetic fields perturbing the equilibrium configuration can be modeled as Gaussian pulses with amplitude proportional to the intensity of the light pulse and time width of the order of hundred femtoseconds. The non-linear dynamical equations have been integrated through an optimized Runge-Kutta algorithm and an analytical solution of the linearized system has been discussed in the case when the magnetic field pulse is assumed to have the shape of a delta function. This solution provides the right orders of magnitude allowing to interpret the experimental results in simple terms.

We have found that the quasi-antiferro mode is excited by the pulse along the $z$-axis and the oscillations of the magnetization have amplitudes compatible with experiment. Magnetic fields in opposite directions give rise to out-of-phase oscillations showing a behavior in agreement with experimental results for times longer than the initial transient. We have stressed that the magnetization dynamics is not only strongly influenced by the amplitude of the magnetic field pulse, but also by the parameters determining the free energy, in particular the ratio between the antisymmetric and symmetric exchange energies. The temperature dependence of the magnetization dynamics has been discussed showing that the proposed damping mechanism based on magnon-magnon scattering can be effective on the picosecond scale. When the field pulse is along the $x$-axis, the quasi-ferro mode is excited, but there are some discrepancies in the comparison between theory and data. We point out that the response along the $x$-axis can be strongly influenced in orthoferrites by several effects such as the optical birefringence and the anisotropy of the magneto-optical susceptibility. Finally, the behavior of the magnetization has been analyzed in the low-temperature range where, due to an unavoidable heating effect, the laser pulse could perturb the stability between $\Gamma_1$ and $\Gamma_4$ state.

We notice that the model proposed in this work has neglected dipolar contributions because they are orders of magnitude smaller than exchange and anisotropy terms. Moreover, due to the fact that the effective magnetic field obtained through the inverse Faraday effect shows spatial variations negligible on the microscopic scale, only the spin-wave modes at zero wave-vector are excited. Since the static magnetization changes slowly in the investigated temperature range, the presence of spin-waves with
wave-vectors different from zero should not provide siz-
able contributions to the dynamic behavior. Therefore,
the macrospin approximation employed in this paper can
be considered reliable.

Finally, we point out that the approach employed for
dysprosium orthoferrites can be also generalized to de-
scribe the magnetization dynamics of other rare-earth
orthoferrites, at least in the $\Gamma_4$ phase. The anisotropy
constants are the only quantities strongly dependent on
the rare-earth ion, but these do not play a major role in
affecting statics and dynamics. On the other hand, the
most important values of the exchange fields are of the
same order in several rare-earth orthoferrites. Clearly
the approach proposed in this paper is suitable for mag-
netic dielectrics and not for metallic itinerant magnets.

However, up to now, due to the unavoidable light absorp-
tion, it has been impossible to ascertain the role played
by the inverse Faraday effect on the magnetization dy-
namics of itinerant magnets.

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