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Optical excitation of antiferromagnetic resonance in $TmFeO_3$

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Femtosecond laser pulses are observed to excite antiferromagnetic resonances in $TmFeO_3$ via both thermal and nonthermal mechanisms. These mechanisms dominate in two different temperature ranges. The analysis shows that thermally and nonthermally triggered spin oscillations have different frequencies. The experimental data reveal that femtosecond laser excitation results in frequency changes of the antiferromagnetic resonance within 1 ps, demonstrating the feasibility of ultrafast frequency modulation.

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Ultrafast optical control of the magnetic state of a medium is presently a subject of intense research.[1–12] Experiments using femtosecond high-intensity laser pulses have revealed many fascinating laser-induced magnetic phenomena such as ultrafast demagnetization within 200 fs,[1–4,12] or modification of the magnetic anisotropy within 1 ps.[7] All these observations are of importance for the development of novel concepts for high-speed magnetic recording and information processing. However, the physics of these phenomena is still poorly understood. This is mainly because the excitation with femtosecond laser pulses puts a medium in a highly nonequilibrium state, where a conventional description of magnetic phenomena in terms of thermodynamics is no longer valid. Therefore in addition to the potential applications, ultrafast magnetic phenomena are a subject of fundamental importance in the physics of magnetism.

One of the recently observed intriguing phenomena is optical excitation of magnetic resonance, when subpicosecond laser pulses trigger homogeneous spin precessions.[5,6,9,11] The combination of optically excited magnetic resonance and optical detection of the spin precession provides a unique local probe of dynamical magnetic properties of a medium. With such a technique one is able to detect spin precession frequencies up to 10 THz even in the case of large damping. However, the application of this technique would require better understanding of the mechanisms responsible for optical excitation of magnetic resonance.

In this Rapid Communication, we report the study of the optically excited antiferromagnetic resonance in $TmFeO_3$. This compound offers a unique situation such that antiferromagnetic resonance is excited via thermal and nonthermal mechanisms simultaneously. This allowed us to use one type of oscillation as a reference for the other. Such a comparative analysis reveals a relation between efficiencies of the thermal and nonthermal mechanisms and shows that these mechanisms trigger spin oscillations of different frequencies. The experiment clearly shows that excitation of a medium with an ultrashort laser pulse can change the frequency of the antiferromagnetic resonance within 1 ps.

Thulium orthoferrite $TmFeO_3$ crystallizes in an orthorhombically distorted perovskite-type structure with four molecular units per unit cell and space-group symmetry *Pbnm*. The iron ions Fe^{3+} form two magnetic sublattices that are antiferromagnetically coupled. The equilibrium orientation of the spins in this material is given by the minimum of the thermodynamical potential Φ[13]:

$$\Phi = J(\mathbf{S}_1\mathbf{S}_2) + \mathbf{D}\cdot[\mathbf{S}_1 \times \mathbf{S}_2] + K_x(S_{1x}^2 + S_{2x}^2) + K_z(S_{1z}^2 + S_{2z}^2) + K_4(S_{1x}^4 + S_{1y}^4 + S_{1z}^4 + S_{2x}^4 + S_{2y}^4 + S_{2z}^4) + \mathbf{H}\cdot(\mathbf{S}_1 + \mathbf{S}_2), \quad (1)$$

where $\mathbf{S}_1$ and $\mathbf{S}_2$ are the vectors that characterize the spins of the iron ions in the two magnetic sublattices, J is the nearest-neighbor isotropic exchange interaction constant; D is the Dzialoshinsky-Moriya antisymmetric exchange constant; K_x, K_z, K_4 are constants of the magnetocrystalline anisotropy, and $\mathbf{H}$ is the magnetic field. The exchange J favors an antiferromagnetic configuration of the Fe^{3+} spins, whereas the antisymmetric exchange interaction D results in a slight canting of the spins from the antiparallel orientation over an angle $\beta \approx 0.5°$, so that the system acquires a weak ferromagnetic moment.

It follows from Eq. (1) that the equilibrium spin structure of a medium can be perturbed via a modification of any of the magnetic parameters,[14] such as the exchange interaction or the magnetic anisotropy, or due to an impact of an external stimulus such as a magnetic field. Both J and D are believed to be temperature independent, whereas the magnetocrystalline anisotropy of $TmFeO_3$ is characterized by a strong temperature dependence.[15] Below $T=80$ K, $K_x > K_z$ and the magnetic anisotropy favors alignment of the antiferromagnetic spins along the z axis, with a weak magnetic moment along the x axis [the magnetic structure is $\Gamma_2(G_zF_x)$]. Above $T=91$ K, $K_x < K_z$ and the spins are aligned along the x axis with a weak magnetic moment along the z axis. In this case the magnetic structure is $\Gamma_4(G_xF_z)$. In the range between 80 and 91 K the easy axis of the magnetic anisotropy continuously rotates in the xz plane, while keeping the weak ferromagnetic moment in the same plane. Microscopically, such a spin reorientation is due to a temperature-driven redistribution of the electrons in Tm ions, which leads to a renormalization of the Tm-Fe interaction and thus results in the change of the magnetic anisotropy. An ultrashort laser excitation of a solid leads to the creation of a large number of nonequilibrium phonons. These phonons may cause a redistribution of electrons in Tm ions on a time scale given by the electron-phonon interaction,[16] modify the anisotropy, perturb the equilibrium of the Fe spins, and thus excite magnetic precession.

On the other hand, a high-intensity laser pulse can also affect antiferromagnetic Fe spins directly in a *nonthermal*

way.[9] Via the inverse Faraday effect, light can induce an effective magnetic field **H**,[17,18] which can be phenomenologically described by

$$\mathbf{H}(0) = \frac{\chi}{16\pi}[\mathbf{E}(\omega) \times \mathbf{E}^*(\omega)], \tag{2}$$

where $\mathbf{E}(\omega)$ and $\mathbf{E}^*(\omega)$ are the electric field of the light and its complex conjugate, respectively; χ characterizes the magneto-optical susceptibility being a real scalar for nondissipative isotropic media. In the case of isotropic media, the maximum value of $\mathbf{H}(0)$ is reached at circularly polarized laser excitation. A spectral analysis shows that the magneto-optical susceptibility of $TmFeO_3$ is mainly defined by charge-transfer transitions in FeO complexes and hardly affected by transitions in Tm ions.[19] Thus it is expected that via the inverse Faraday effect, an ultrashort laser pulse will directly act on Fe spins in a way similar to an external magnetic field, but will not affect the Tm ions.

The equation of motion for the antiferromagnetic spins $\mathbf{S}_1$ and $\mathbf{S}_2$ shows that ultrafast optical excitation triggers two resonance modes with energies $h\omega_{FM}=[24JS(K_x-K_z)S]^{1/2}$ and $h\omega_{AFM}=[24JS(6DS\tan\beta+K_xS)]^{1/2}$.[13,15,20,21] The first "quasiferromagnetic" mode softens in the spin reorientation region, while the second quasiantiferromagnetic mode is usually characterized by a weaker temperature dependence.

In our experiment, the time-resolved measurements with subpicosecond temporal resolution were performed in a pump and probe configuration at a photon energy of 1.59 eV using amplified 200-fs pulses from a Ti:sapphire laser at a repetition rate of 1 kHz.[9] The pump beam was circularly polarized, while the probe had linear polarization. The intensity ratio between the pump and probe pulses was about 100. Both beams were focused on the sample to a spot diameter of about 200 μm for the pump and somewhat smaller for the probe beam. The pump fluence on the sample was around 30 mJ/cm^2. We have studied $TmFeO_3$ samples of 60-μm-thick plane plates cut perpendicular to the z axis of the crystal. Optical properties and electronic structure of the studied $TmFeO_3$ have been analyzed elsewhere.[22] The measurements were done in a cold finger cryostat where the temperature could be stabilized in the range of 5–300 K with a precision of better than 0.5 K.

To monitor the magnetization precession we used the magneto-optical Faraday rotation $\alpha_F=\chi(\mathbf{M}\cdot\mathbf{k})$, where $\mathbf{M}$ is the magnetization and $\mathbf{k}$ is the wave vector of light.[19] The static measurements of the Faraday rotation as a function of temperature are summarized in Fig. 1 and show the temperature behavior of the z component of the magnetization M_z that is clearly present above 90 K and vanishes below 80 K.

Figure 2 shows the temporal evolution of the Faraday rotation for two circularly polarized pumps of opposite helicities at temperatures between 7 and 85 K. At every temperature we observe rapid changes of the signal during the overlap of the pump and probe pulses.[23] The instantaneous changes of the Faraday rotation are followed by oscillations. At low temperature circularly polarized pump pulses of opposite helicities trigger oscillations of opposite phase. Starting from 75 K the oscillations induced by right- and left-handed helicities are in phase, having, however, different amplitudes. Above 85 K no oscillations are observed.

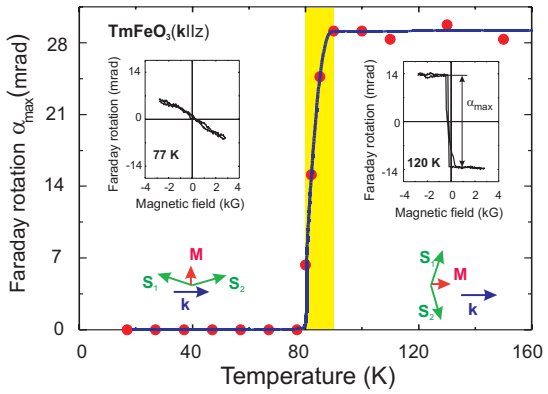

FIG. 1. (Color online) Amplitude of the hysteresis loop α_{max} as a function of sample temperature. The spin reorientation region is marked by the colored strip. Below 80 K no hysteresis is observed and the amplitude α_{max} is zero. The insets show field dependencies of the magneto-optical Faraday effect in the z cut $TmFeO_3$ at 77 and 120 K.

To separate thermal and nonthermal mechanisms of the optical excitation of antiferromagnetic oscillations, one should realize that thermal effects are insensitive to the helicity of the pump light, while the nonthermal excitation with right- σ^+ and left-handed σ^- circularly polarized laser pulses should trigger spin waves of opposite phase. Therefore the thermal δM_{th} and nonthermal δM_{nonth} laser-induced effects on the magnetization can simply be obtained as the sum and difference of the experimental curves, respectively $[\delta M_{th} \propto \alpha_F(\sigma^-)+\alpha_F(\sigma^+)$ and $\delta M_{nonth} \propto \alpha_F(\sigma^-)-\alpha_F(\sigma^+)]$.

The thus derived thermal and nonthermal effects are plotted in Fig. 3. One can see that the thermal excitation of

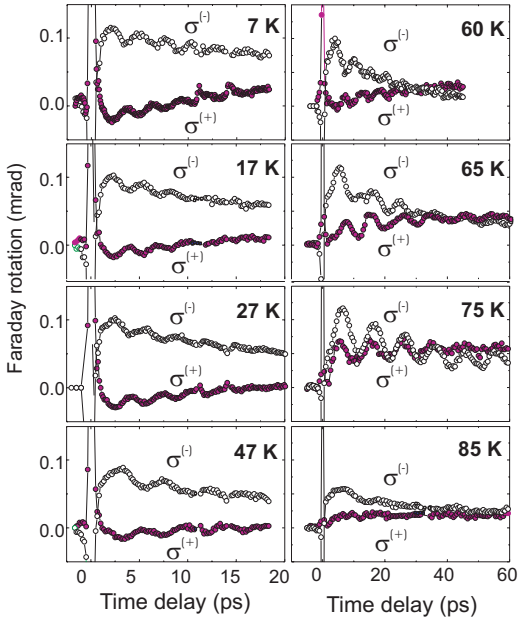

FIG. 2. (Color online) Magnetic excitations by circularly polarized pump pulses in $TmFeO_3$ probed by the magneto-optical Faraday effect.

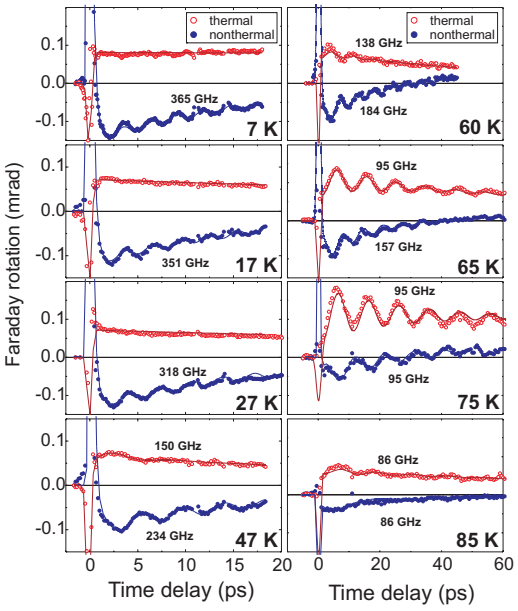

FIG. 3. (Color online) The sum and difference of the experimental curves obtained for right-handed σ^+ and left-handed σ^- circularly polarized pumps, characterizing the thermal and nonthermal optomagnetic excitation, respectively.

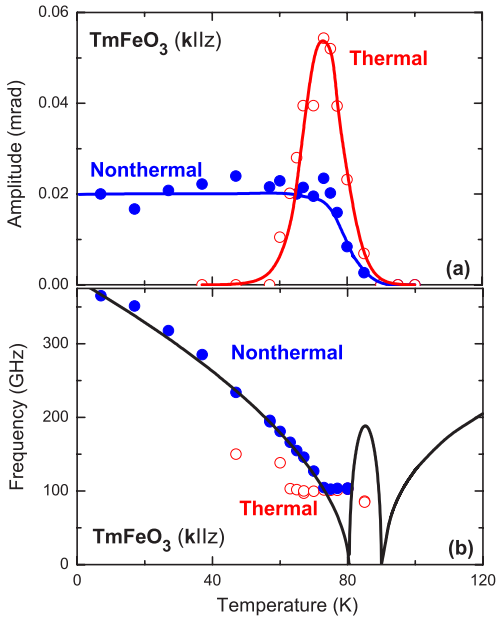

FIG. 4. (Color online) (a) Amplitudes of the thermally (circles) and nonthermally (dots) excited antiferromagnetic resonance as a function of temperature in the z-cut $TmFeO_3$; (b) frequency of the thermally and nonthermally excited spin oscillation as a function of temperature. Solid line is the best fit to the earlier experimental data (Refs. 15, 21, 24, and 25).

antiferromagnetic resonance appears to be effective only in a relatively narrow temperature range from 47 K up to 85 K. The frequency of the thermally excited spin precession decreases from 150 GHz at 47 K down to 86 GHz at 85 K. In contrast, the nonthermal mechanism is observed in a broad range of temperatures. At 7 K the nonthermal optical excitation results in a spin precession with a frequency of 365 GHz, which gradually decreases down to 95 GHz at 75 K. It is remarkable that in the range from 47 to 70 K the frequency of the thermally excited antiferromagnetic resonance is smaller than the frequency of the nonthermally excited resonance. This observation clearly demonstrates the fundamental difference between thermal and nonthermal mechanisms of the optomagnetic excitation.

Figure 4(a) shows the temperature behavior of the amplitude of the spin oscillations triggered via thermal and nonthermal mechanisms in $TmFeO_3$. The thermal effect, which is only observed in the range between 47 and 85 K, reaches a maximum value around 75 K. In contrast, the nonthermal excitation of the antiferromagnetic resonance is radically different. In particular, its efficiency does not vary from 7 K up to 75 K, above which it starts to decrease until it vanishes above 85 K.

The drastic difference between the temperature dependences of the thermal and nonthermal effects is related to their fundamentally distinct origins. Thermal excitation results in an increase of the lattice temperature of the sample from T to $T+\delta T$ and this leads to a reorientation of the weak ferromagnetic moment in the xz plane. Such perturbation triggers spin oscillations in the same plane that corresponds to the quasiferromagnetic mode. This spin precession can be seen in the experiment as oscillations of the Faraday rotation. For an excitation at temperature T, the amplitude of the magneto-optical oscillations is proportional to $M_z(T)-M_z(T+\delta T)$. Therefore the thermal excitation of the antiferromagnetic resonance in $TmFeO_3$ is inefficient at very low temperatures and should have a maximum slightly below the onset of the spin reorientation.

The phenomenology of the nonthermal inverse Faraday effect states [see Eq. (2)] that light pulse propagating along the z axis acts on the spins as a magnetic-field pulse directed along the same axis. In the low-temperature magnetic phase, where the antiferromagnetic spins are oriented along the z axis and a weak ferromagnetic moment is along the x axis, such magnetic field triggers oscillations in the xz plane. However, in contrast to the thermal effect, the efficiency of the nonthermal optomagnetic excitation at a temperature T is proportional to the z component of the weak ferromagnetic moment at this temperature $M_z(T)$. For the high-temperature magnetic structure of $TmFeO_3$, where the antiferromagnetic spins are aligned along the x axis and a weak ferromagnetic moment is directed along the z axis, the same kind of laser exitation would deviate the spins out of the xz plane and thus trigger the quasiantiferromagnetic mode with a frequency of about 1 THz. The latter, however, has not been detected in our experiment.

Figure 4(b) summarizes the temperature behavior of the precession frequencies triggered via thermal and nonthermal laser excitation. One can see that the precession frequencies triggered via the nonthermal mechanism are in excellent agreement with the temperature behavior of the quasiferromagnetic resonance mode in $TmFeO_3$.[13,15,20,21,24,25] The frequency of this mode gradually decreases and approaches 95 GHz in the vicinity of the spin reorientation at 70 K. Further increase of the temperature does not result in fre-

quency changes, while a strong temperature dependence is expected. This can be attributed to the fact that intense laser excitation induces a strong effective magnetic field via the inverse Faraday effect and can affect the magnetic anisotropy.[10] Both of these effects may seriously influence the magnetic structure of the medium and thus result in drastic changes of the frequencies of antiferromagnetic resonance, especially in the vicinity of the region of the spin reorientation.[15]

In contrast to the nonthermal effect, the thermal laser excitation most effectively triggers spin oscillations at a frequency of 95 GHz, while excitation of other frequencies is quite ineffective. This can simply be understood, since for the thermal excitation of the antiferromagnetic resonance one has to bring the sample into the regime of spin reorientation, where the frequency of the quasiferromagnetic mode is low. Similarly to the nonthermal effect, the thermal excitation does not trigger frequencies lower than 86 GHz, which again can be attributed to laser-induced renormalization of the frequencies of the resonances.

Indeed, Figs. 3 and 4 clearly show that laser excitation results in an ultrafast change of the frequency of the antiferromagnetic resonance. In particular, it is seen from the data obtained at 65 K that thermally and nonthermally triggered spin oscillations have frequencies of 95 and 157 GHz, respectively. Due to the relatively low repetition rate of the pump pulses, the steady-state heating of the sample is small (less than 5 K) and uniform. However, the initial temperature profile created by a single pump pulse is not uniform over the sample thickness due to optical absorption. The thermally triggered oscillations are observed in the sample region, heated by the laser pulse above spin reorientation temperature $T=80$ K. Nonthermally triggered spin oscillations dominate in the sample regions with lower level of laser excitation. Therefore the different frequencies of the thermally and nonthermally triggered spin oscillations give evidence to the fact that optical excitation is able to change the frequency of the antiferromagnetic resonance on a subpicosecond time scale. This laser-induced phenomenon demonstrates the feasibility of ultrafast frequency modulation.

To conclude, we have shown that femtosecond laser pulses excite antiferromagnetic resonances in $TmFeO_3$. This compound offers a unique situation in that the resonance is excited via both thermal and nonthermal mechanisms simultaneously. A comparative analysis reveals a relation between efficiencies of the thermal and nonthermal mechanisms and shows that these mechanisms trigger spin oscillations of different frequencies. Laser excitation is able to change the frequency of the antiferromagnetic resonance on a subpicosecond time scale. This phenomenon opens interesting insights into laser excitation of spin resonance and demonstrates the feasibility of ultrafast frequency modulation.

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