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Antiferromagnetic spin dynamics in exchanged-coupled Fe/$GdFeO_3$ heterostructure*

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We investigate the ultrafast spin dynamics of an antiferromagnet in a ferromagnet/antiferromagnet heterostructure Fe/$GdFeO_3$ via an all-optical method. After laser irradiation, the terahertz spin precession is hard to be excited in a bare $GdFeO_3$ without spin reorientation phase but efficiently in Fe/$GdFeO_3$. Both quasi-ferromagnetic and impurity modes, as well as a phonon mode, are observed. We attribute it to the optical modification of interfacial exchange coupling between Fe and $GdFeO_3$. Moreover, the excitation efficiency of dynamics can be modified significantly via the pump laser influence. Our results elucidate that the interfacial exchange coupling is a feasible stimulation to efficiently excite terahertz spin dynamics in antiferromagnets. It will expand the exploration of terahertz spin dynamics for antiferromagnet-based opto-spintronic devices.

Keywords: ultrafast magnetization dynamics, antiferromagnetics, magnetic oxides, magnetization dynamics

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1. Introduction

In antiferromagnetic (AFM) materials, the neighboring magnetic moments point in opposite directions, resulting in null macroscopic magnetization and the absence of stray fields. These features make antiferromagnets invisible to common probes and quite robust over external magnetic perturbation.[1,2] In addition, due to strong AFM exchange interaction, antiferromagnets show typical terahertz (THz) resonance frequencies, which make them promising for future high-frequency spintronic applications and now attract significant attention.[3,4] However, the almost zero net magnetic moment makes it extremely challenging to effectively manipulate and probe the antiferromagnetic order.[2] Thus, the efficient detection of AFM spin dynamics with frequency up to the terahertz range has been interested and gazed widely.

The rare-earth orthoferrites $RFeO_3$ (R stands for a rare-earth element),[5] with canted spin and a weak net magnetic moment originating from antiferromagnetic exchange interaction and Dzyaloshinsky–Moriya interaction, are natural candidates for observing antiferromagnetic spin dynamics.[6] For typical orthoferrites, such as $HoFeO_3$ and $TmFeO_3$, the AFM spin dynamics has been widely investigated via the optical methods for its sensitivity and short stimulus.[7–11] However, some limitations are indeed non-negligible and to be addressed. Using the all-optical method, the spin dynamics of $RFeO_3$ can only be probed near the spin reorientation phase transition temperature region. In addition, the dynamic amplitude via the nonthermal stimulus dramatically decreases with temperature due to phonon scattering.[12,13] Moreover, there are also some rare-earth orthoferrites (R = Y, Eu, Gd, Tb) remained unexplored via all-optical technique due to their single magnetic phase. The $GdFeO_3$ (GFO) is a typical canted antiferromagnet without the spin reorientation phase transition, whose AFM spin dynamics triggered by optical method remains elusive. It has only been detected via second harmonic generation at extremely low temperature 1.5 K,[14] whereas the roomtemperature spin dynamics via optical excitation is to be explored. Recently, in the case of the interfacial exchange coupling with an Fe layer, the AFM spin dynamics in $ErFeO_3$ can be probed via an all-optical method away from the spin reorientation temperature range.[15] Inspired by this, it is with tremendous possibility of extending on GFO-type materials, where there is potential for greatly expanding the operating temperature and efficiency.

In this work, we investigate the room-temperature AFM spin dynamics of $GdFeO_3$ via an all-optical method in exchange-coupled Fe/$GdFeO_3$ (100) heterostructure. Both quasiferromagnetic and impurity modes, as well as phonon mode, are observed in low magnetic field in Fe/GFO while absent in pure GFO. Although the excitation efficiency of

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AFM dynamics is hardly affected by the magnetic moment arrangement (antiparallel or parallel) between interfacial layers, it can be modified by the fluence of pump laser presenting as the change of the AFM resonance amplitude, which results from optical modification of the interfacial exchange coupling interaction. Considering the great importance of the AFM resonance excitation for energy-efficient opto-spintronic devices, the efficient interaction stimulation via exchange coupling may drastically expand the advanced AFM dynamics detection of tremendous materials in near future.

2. Experimental methods

The GFO single crystal was grown using the floating zone method with crystallinity and crystallographic orientation confirmed by powder x-ray diffractometer and Laue x-ray diffraction as reported previously.[16] The sample was cut perpendicularly to the *a* axis and then polished, so a flat GFO (100) substrate was obtained. To fabricate the Fe/GFO (100) heterostructure, the GFO (100) substrate with thickness of 2 mm was put into the ultrahigh vacuum chamber ($\sim 5 \times 10^{-10}$ mbar) to warm up to about 500 K, which is far smaller than the growth temperature (1200 K) of the single crystal to remove the surface gas contaminants. Then, the GFO (100) substrate underwent natural cooling, and was kept at room temperature during the subsequent film growth. A 35-nm-thick Fe film was deposited by electron beam evaporator onto the GFO substrate, and with a 2.5-nm-thick Cu film as the capping layer for *ex situ* measurements. The deposition rate of Fe and Cu were kept at 0.15 nm/min and 0.10 nm/min, respectively. During the deposition, half the GFO substrate was shadowed for convenience to monitor the change of magnetic properties. Furthermore, the magnetic hysteresis loops of GFO (100) and Fe/GFO (100) heterostructure were determined by longitudinal magneto-optical Kerr effect (MOKE) at room temperature.

The AFM spin dynamics in Fe/GFO (100) was achieved using the all-optical magneto-optical Kerr effect (TRMOKE) technique at room temperature. A Ti:sapphire oscillator generates a train of laser pluses at repetition rate of 5.2 MHz, duration of 55 fs and with a wavelength of 780 nm, which was used as the pump laser with linearly polarization perpendicular to the projection direction. Moreover, the 780 nm femtosecond laser was doubled by a BBO crystal as the probe light, which was linearly polarized along the projection direction. Both the pump and probe beams were incident vertically onto the sample with spot diameters of about 10 μm and 5 μm, respectively. Here, the pump fluence varied from 0.5 to 4.0 mJ/cm^2 while the probe fluence was kept at 0.1 mJ/cm^2.

3. Results and discussion

The $GdFeO_3$(GFO) is a canted antiferromagnet possessing a weak net moment along the *c* axis below 641 K without spin reorientation.[17] Figure 1(a) shows the normalized longitudinal Kerr signals along the *c* axis for pure GFO (100) and Fe/GFO (100) heterostructure. The magnetic hysteresis loop of heterostructure presents two-step magnetization reversal process with a larger switching field reaching 160 Oe and a lower one about 50 Oe. It is noteworthy that the penetration depth of light with a wavelength of 632 nm is about 15 nm,[18] and consequently the MOKE can probe both the magnetic moment of Fe and the net magnetic moment of substrate simultaneously. Compared with the signal of pure substrate GFO (100), the coercivity of net moment of GFO almost keeps unchanged, revealing the influence of the net moment of Fe film and the treatments of substrate on the magnetic property of GFO is quite small.

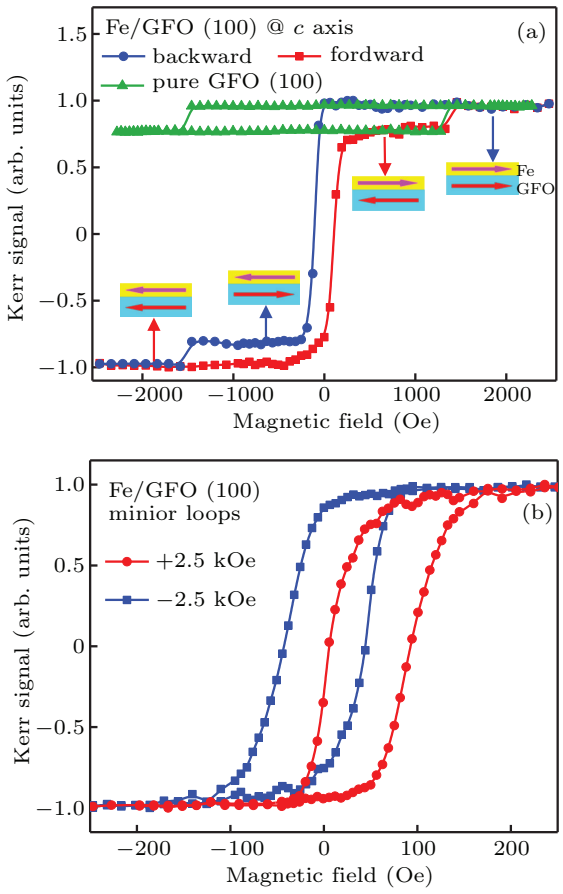


Fig. 1. (a) Normalized Kerr loops for the pure GFO (100) substrate and Fe/GFO (100) heterostructure with external magnetic field of 2.5 kOe applied along the *c* axis. The inset shows the parallel or anti-parallel spin configuration in the FM-AFM interface. (b) The minor loops along the *c* axis in Fe/GFO (100) heterostructure with maximum field of 250 Oe after the net moment of GFO fixed under an initial field of +2.5 kOe and −2.5 kOe, respectively.

Figure 1(b) presents the distinctly shifted minor loops known as exchange bias effect, with a maximum applied field of 250 Oe along the *c* axis in Fe/GFO (100) after the net moment of GFO fixed under an initial field of +2.5 and −2.5 kOe, respectively. When the GFO net moment fixed along the [001] axis under a positive field (+2.5 kOe), the minor loop exhibits a positive shift, and vice versa. That is to say, the net moment of GFO can reverse the Fe moment by inverting its own direction. Moreover, the positive (negative) shift

of minor hysteresis loop under positive (negative) initial magnetic field indicates the positive exchange bias, similar to the FM/AFM system.[19–21] Hence, the positive bias effect indicates the robust AFM interfacial exchange coupling between the net moment of GFO orthoferrite and the neighboring soft Fe layer, which is the keystone for further study of AFM spin dynamic via stimulation of Fe layer. In addition, the magnetization orientation of Fe layer and pinning layer (GFO layer) under various external magnetic fields is depicted schematically in Fig. 1(a), which provides the possibility to investigate the spin configuration related terahertz dynamics of GFO in heterostructure.

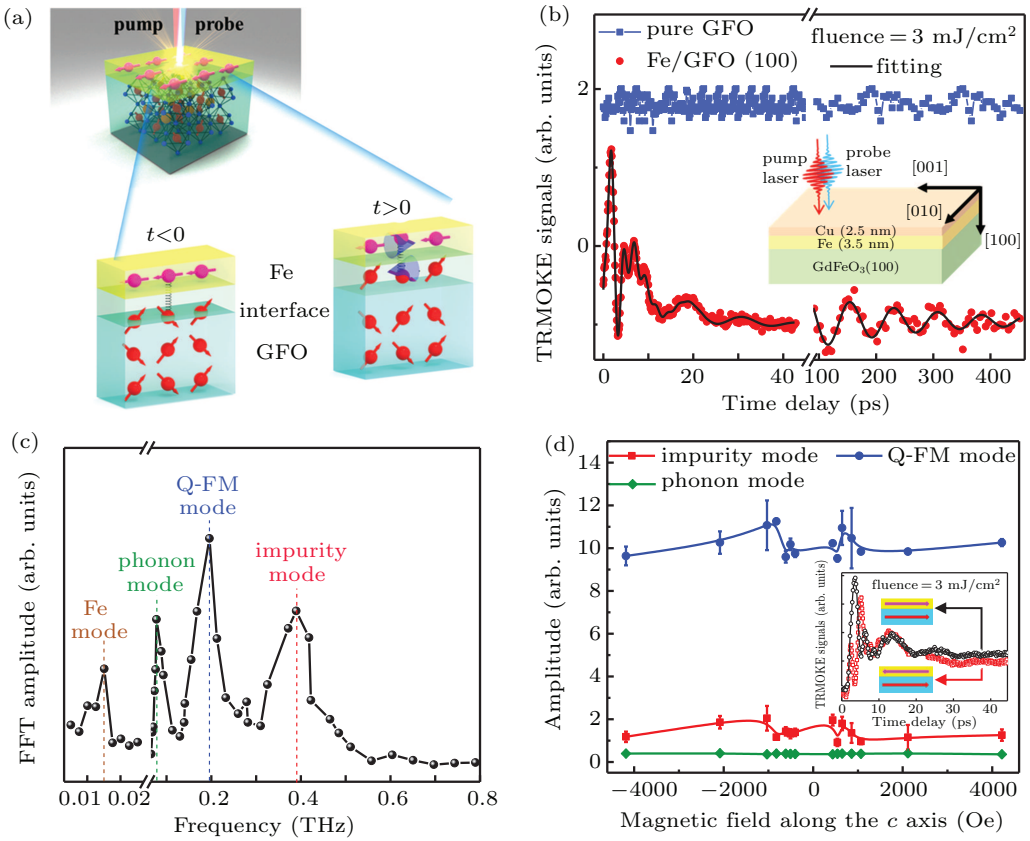


Fig. 2. (a) The schematic diagram of optical stimulus of AFM dynamics in Fe/GFO (100) heterostructure via optical modification of the interfacial exchange coupling at room temperature. (b) Room-temperature TRMOKE signals of Fe/GFO(100) heterostructure and pure GFO (100) substrate with a laser fluence of 3 mJ/cm^2 and a 2.5 kOe field applied along the c axis. The inset is the experimental configuration for TRMOKE measurement. (c) Frequency component obtained by the FFT method. (d) The magnetic field dependent resonance amplitude for impurity mode, phonon mode and Q-FM mode in Fe/GFO (100), respectively. The inset shows the typical TRMOKE signals for different magnetization configurations at the interface of Fe and GFO.

Figure 2(b) shows the typical dynamic signals of heterostructure and pure GFO substrate in a magnetic field of 2.5 kOe along the c axis of GFO at room temperature. As is expected, no obvious dynamic signal is observed in the pure GFO substrate because the all-optical method cannot effectively stimulate AFM resonance far away from the phase transition region.[9,12,22] Nevertheless, for Fe/GFO (100) heterostructure, a quite complex and fast response in 0–50 ps time range is observed and followed by a slow spin precession till 700 ps reasonably assigned to the precession of the Fe film. Naturally, the multiple high-frequency modes are obtained from the fast Fourier transform (FFT) power spectra with a low frequency of 8 GHz and three high resonance modes at 0.08 THz, 0.23 THz, and 0.38 THz presented in Fig. 2(c). In addition, the signal in the 0–50 ps time range is fitted by damped function with three frequency modes f_i via the following equation:

$$\Delta M(t) = \sum_{i=1,2,3} A_i \cos(2\pi f_i t + \phi_i)\, e^{t/\tau_i} + B e^{t/\tau_0} + C, \quad (1)$$

where A_i, φ_i and τ_i $(i = 1, 2, 3)$ represent oscillatory amplitude, initial phase of magnetic precession, and relaxation time, respectively. The last two terms are related to the background signal with the amplitude B and C, and recovery time constant τ_0. Similarly, the dynamic parameters of the Fe layer can be extracted by fitting the data[23] in a subsequent range but beyond the scope of our discussion here. Through the fitting curve in Fig. 2(b), we obtain three resonance modes at 0.078 THz, 0.23 THz, 0.39 THz, which are consistent with the FFT peaks. The two-sublattice model of the magnetic structure of the orthoferrites predicts the quasi-ferromagnetic (Q-FM) mode at 100–300 GHz.[24,25] Thus, we ascribe the 0.23 THz mode to photo-induced Q-FM mode of GFO, governed by the exchange interactions of Fe^{3+}–Fe^{3+} ions.[14] Moreover, the highest-frequency mode at 0.39 THz is assigned to the impurity mode because of its typical characteristic of possessing an initial phase almost π different from the Q-FM mode.[26,27] This mode stems from the occupation of the $^6A_1$

ground state of the Fe^{3+} ions in rare-earth positions.[6,26] In addition, we attribute the resonance of 0.08 THz to the phonon mode for its imperviousness to the magnetic field. The phonon mode derived from the lattice oscillation[14,28] was also reported in $ErFeO_3$ with the same frequency.[15] Therefore, the room-temperature AFM spin dynamics of GFO is successfully excited via the all-optical method in the exchange coupled Fe/GFO (100) system. As mentioned above, by adjusting the applied field, we can investigate the interfacial spin configuration related dynamics. Distinctly, the phases of the two signals are nearly 180° different for the parallel and antiparallel spin configurations (see the inset of Fig. 2(d)). From Fig. 2(d), the amplitude of each resonance mode is nearly invariant with the magnetic field, meaning that the excitation efficiency is hardly modulated by changing the in-plane magnetic moment configuration. It is quite sturdy for AFM order and intrinsic exchange interaction subject to the magnetic field. Furthermore, the relatively weak external field H cannot align the AFM spin configuration at the FM/AFM interface, with the internal exchange interaction remaining constant.

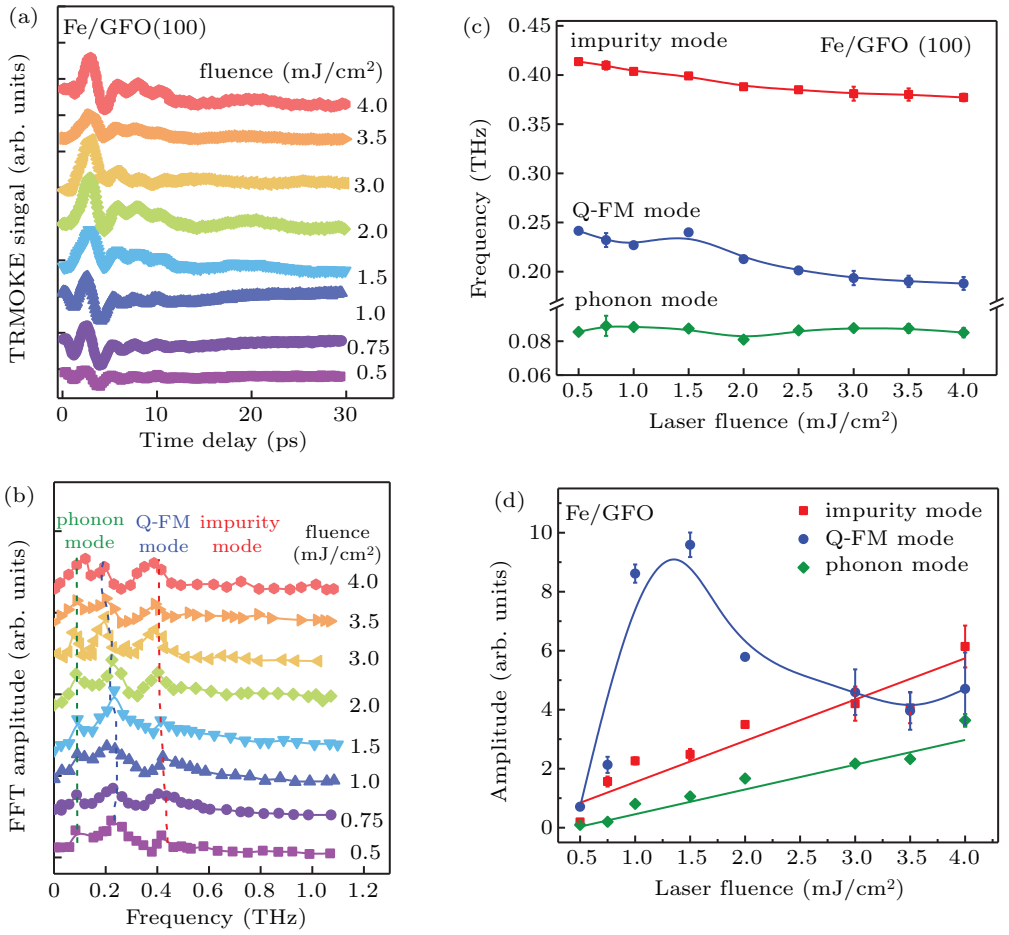


Fig. 3. The TRMOKE signals at different fluences of the pump laser (a) and their corresponding FFT spectra (b). The fluence of pump laser dependent precession frequency (c) and resonance amplitude (d) of impurity mode, phonon mode and Q-FM mode for Fe/GFO (100), respectively. The magnetic field (2.5 kOe) was applied along the c axis. The solid lines are of guide for the eyes.

To investigate the optical modification of excitation efficiency of AFM spin dynamics we adjust the pump laser fluence to excite the Fe/GFO (100) heterostructure with the typical signals shown in Figs. 3(a) and 3(b). Figure 3(c) reveals the resonance frequency of the phonon mode keeps constant, suggesting that the laser intensity cannot affect the lattice vibration rate. However, regarding the impurity mode and Q-FM mode, we observe a slight shift of the resonance frequency by varying laser fluence. According to the Sigma model,[29] the resonant frequency for Q-FM mode is determined by the uniaxial magnetic anisotropy, as well as the interfacial exchange coupling interaction for the interfacial AFM spins at room temperature in our system. Thus, the optical absorption of the system impacts the moment of the Fe layer, then modifies the interfacial exchange coupling with the GFO layer, and even the thermal effect, which are possibly responsible for the frequency shift of the Q-FM mode. Meanwhile, the impurity mode also comes from magnetic source, related to the Fe^{3+} ions in rare-earth positions. Thus, the shift of its frequency may have the same origins. Furthermore, the laser fluence can significantly modify the dynamic amplitude of all modes in heterostructure with different tendencies, which indicates the tunable excitation efficiency as shown in Fig. 3(d). Here, the direct interaction between laser and the GFO substrate can be ignored because of the low optical absorption of the orthoferrite,[30,31] namely, the pump laser can solely mod-

ulate the Fe layer. Consequently, we can conclude that optical modulation of AFM excitation amplitude is possible due to optical modification of interfacial exchange coupling interaction. The simple diagram of the excitation is depicted in Fig. 2(a). The laser induces the instantaneous nonequilibrium state of the Fe layer, then the interfacial exchange coupling is perturbed, as a result the precession of the net moment of GFO occurs.

The oscillation amplitude of the phonon mode exhibits nearly a linear relation with fluence, like that in $FeBO_3$[28] interpreted with an intense lattice vibration for intense optical absorption. Similarly, the excitation amplitude of impurity mode related to the Fe^{3+} ions in rare-earth is linearly increased with largened influence, which may result from the perturbated AFM state near the interface by coupling with more unbalanced Fe layer. The oscillation amplitude of Q-FM mode increases linearly with influence from 0.5 mJ/cm^2 to 1.5 mJ/cm^2, which can be ascribed to optical modification of the interfacial exchange interaction as well, according to early reports[6,31] Interestingly, continually intensifying the pump laser, the Q-FM resonance amplitude becomes weak. The impact of the high fluence on Fe film is comparatively large such as heat transport effect or the destruction of the magnetic anisotropy[32] resulting in the magnetic disorder or charge redistribution.[33–35] Therefore, weakening of the amplitude may come from the moment destruction or the charge redistribution by the intensive laser, leading to the saltation of the exchange coupling. However, the microscopic mechanism for optical modification of interfacial exchange coupling and the impact on magnetic order needs more experimental and theoretical studies. Besides, optically modifying the dynamics of the Fe layer may also need further research.

4. Conclusion

In summary, we have investigated the AFM spin dynamics of $GdFeO_3$ via an all-optical pump-probe method at room temperature by modification of interfacial exchange coupling in Fe/$GdFeO_3$ (100). Multimode AFM dynamic behavior of $GdFeO_3$ including quasiferromagnetic, impurity and phonon mode is observed in low magnetic field. In addition, the AFM dynamic properties are less affected by the relative moment arrangement (antiparallel or parallel) of Fe and $GdFeO_3$. The excitation efficiency of AFM resonance can be tuned tremendously by the pump influence via optical modification of the interfacial exchange coupling. The feasible triggering of multimode spin dynamics and easy tuning of excitation efficiency of orthoferrite via all-optical technique shed new lights on ultrafast magnetization manipulation of AFM for fast opto-spintronic devices.

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