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Non-thermal optical excitation of terahertz-spin precession in a magneto-optical insulator

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We demonstrate non-thermal ultrafast laser excitation of spin precession with THz frequency in Gd-Bi-substituted iron garnet via the inverse Faraday effect. The modulation of THz precession by about 60 GHz below the compensation temperature of magnetic moment was observed. The THz frequency precession was caused by the exchange resonance between the Gd and Fe sublattices; we attributed the low-frequency modulation to dielectric resonance mode with a magnetic contribution. We demonstrate the possibility of polarization-sensitive control of spin precession under THz generation by laser pulses, helping to develop high-speed magneto-optical devices. © *2016 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4940241]

Understanding ultrafast spin dynamics on a femtosecond time scale promises to enable technologies based on the quantum-level interplay of nonlinear optics and magnetism. Ultrashort laser pulses can be used to investigate spin dynamics in magnetic materials on the time scale of the exchange interaction, which is much faster than spin-orbit interactions.[1] One of the most important factors that affects the viability of potential applications is the speed of magnetic triggering at high frequencies (HFs), making it important to understand the mechanisms of high-frequency magnetization dynamics in magnetic materials. The mechanism of exchange interaction plays an important role in all-optical switching of magnetization in multisublattice metallic alloys. In GdFeCo, demagnetization from the interaction between the Gd and Fe sublattices has been observed over a few picoseconds,[2] but this mechanism is thermal. Non-thermal excitation of ultrafast magnetization dynamics by laser pulses is based on photomagnetism[3] and the inverse Faraday effect (IFE).[4] Recently, these effects under polarized light excitation of the magnetization states have been demonstrated in transparent dielectrics, avoiding thermal side effects.

Generally, in magnetic systems with two and more magnetic sublattices, along with ferromagnetic resonance, there exists another type of collective precession of magnetic moments: the exchange resonance, predicted by Kaplan and Kittel.[5] At an exchange resonance during the full period of the magnetization precession, the angle between sublattices is not zero, and the resonant frequency is displaced to the far infrared (FIR) spectral range.[5] The optical excitation of exchange resonance between Fe sublattices in LuBi-iron garnets at 400–700 GHz has been observed.[6] The influence of the rare-earth (RE) sublattice is important, but this work did not account for this. Recently, at room temperature in Gd-Yb-Bi-iron garnet, we demonstrated unusual excitation of magnetization precession with a frequency of about 420 GHz using the IFE.[7] Based on our analysis and our preliminary experimental results, we expect excitation of magnetization precession up to the THz frequency range as the difference in magnetization increases between the RE and transition-metal (TM) sublattices. None have yet explored how the interaction between the RE and TM ions with a strong difference in magnetization between these sublattices affects the processes of ultrafast laser-induced magnetization dynamics in optically transparent magnetic dielectrics.

The THz frequency regime corresponds to the far infrared wavelength spectrum. This regime is important in various fundamental phenomena related to collective oscillations of atoms in solids, plasmonic oscillations,[8] and spin waves.[9] Along with FIR light, visible and near IR light (NIR) can couple with magnetic switching with unprecedented speed, allowing for ultrafast, efficient control of the spin system without causing heating.

In this letter, we study an RE-TM magneto-optical insulator, a promising model material for many topics in magneto-optics, magneto-plasmonics, and non-thermal femtomagnetic research. We studied non-thermal light-induced magnetization dynamics in RE-TM garnet via the IFE by using the time-resolved pump-probe method. We demonstrated all-optical excitation of the collective exchange resonance between rare-earth and transition metal sublattices at THz frequencies by using a single femtosecond NIR pulse. The experimental results are in good agreement with modeling using the Kaplan–Kittel resonance. We demonstrate THz magnetization precession at 12–60 K. At these temperatures, the octahedral and tetrahedral Fe sublattices are essentially completely ordered and magnetization changes because of ordering of the dodecahedral Gd sublattice. We also observed that the HF exchange resonance mode in the THz range was modulated by an unexpected low-frequency (LF) mode at about 60 GHz below the compensation point of the magnetic moment.

Experiments were performed on 200 μm thick single-crystal $(GdBi)_3Fe_5O_{12}$, grown by liquid-phase epitaxy, with a (111) plane orientation. At room temperature, the

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saturation magnetization, $4\pi M_S$, was 300 G; the compensation temperature of magnetic moment, T_M, was 220 K; and the Curie temperature, T_C, was 538 K. To investigate the ultrafast magnetization dynamics of the garnet with femtosecond laser pulse excitation, we performed time-resolved measurements in a transmission geometry by using a magneto-optical pump-probe method.

Pump pulses generated by a Ti:Sapphire laser with an amplifier and optical parametric oscillator (Spectra-Physics) at a repetition rate of 500 Hz, wavelength of 1400 nm, and duration of ~100 fs were directed with an angle of incidence of about 10° from the sample normal parallel to the [111] crystallographic axis, while probe pulses with a wavelength of 800 nm were directed incident along the sample normal. The pump beam was focused on a spot diameter of about 200 μm, giving a fluence of about 40 mJ/cm^2. The linearly polarized probe beam had a spot diameter about twice as small.

The ratio of probe intensity to pump intensity was about 1:100. The delay time Δt between the pump and probe pulses could be adjusted to up to 2 ns (see inset of Fig. 1). The helicity of circular polarization of the pump light was tuned with a Berek compensator. A magnetic field H of 2 kOe was applied parallel to the sample plane. We measured the perpendicular component of the magnetization, determined by the Faraday rotation angle, θ_F, of the probe pulses as a function of the delay time, Δt.

Figure 1 shows the time-resolved Faraday rotation as a function of delay time at $T = 12$ K under excitation by circular polarization of laser pulses via the IFE. Such materials with Bi ions have giant Faraday rotation, as previously demonstrated at room temperature.[7] Thus, the IFE in our material should be strong.[4] The behavior of the magnetization precession, as shown in Fig. 1, has the more complex character with low-frequency modulation. Fitting the dependence of time-resolved Faraday rotation with two damping sinusoidal functions, we obtained two frequencies: the LF mode at $f_{LF} = 62$ GHz and the high-frequency mode at $f_{HF} = 1.47$ THz, an extremely high frequency.

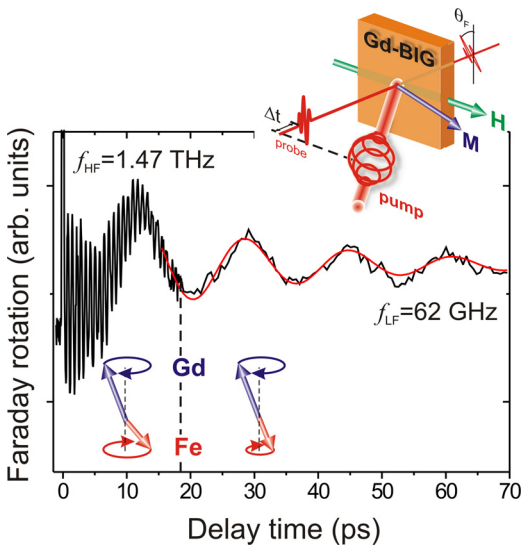

FIG. 1. Time-resolved Faraday rotation covers the high-frequency mode at 1.47 THz, modulated by the low-frequency mode at 62 GHz, measured at 12 K. The inset illustrates geometry of the experiment.

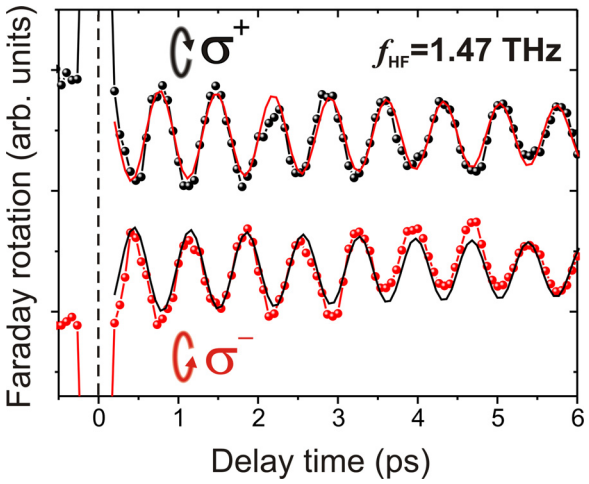

FIG. 2. Short initial time scale of magnetization dynamics, showing opposite helicity of the circular polarization of the pump pulse at $T = 12$ K. The solid lines are the damping sinusoidal functions.

At the initial time scale after the pump pulse, both the HF and LF modes were excited via the IFE. The HF and LF modes relaxed over about 20 ps and over about 100 ps, respectively. The polarization-sensitive HF mode that appeared for the left and the right helicity of the circular polarization of the pump pulses is the fingerprint of the non-thermal mechanism of magnetization precession (see Fig. 2). These precessions have opposite phases, which is characteristic of excitation via the IFE. The LF mode shows similar pump polarization behavior, with the opposite phase of the magnetization precession as the HF mode, which means its excitation has a non-thermal origin. This IFE excitation with a non-thermal mechanism is similar to our previous finding in two rare-earth-sublattice Gd-Yb-BIG crystals.[7,10]

The frequency of the HF mode was independent of the external magnetic field up to 2 kOe. To verify the HF mode, we measured the temperature-dependent magnetization saturation M_s by using classical magnetometry, as shown in Fig. 3. In general, RE-iron garnets can be treated as two-sublattice ferrimagnets: one made up of Fe ions occupying

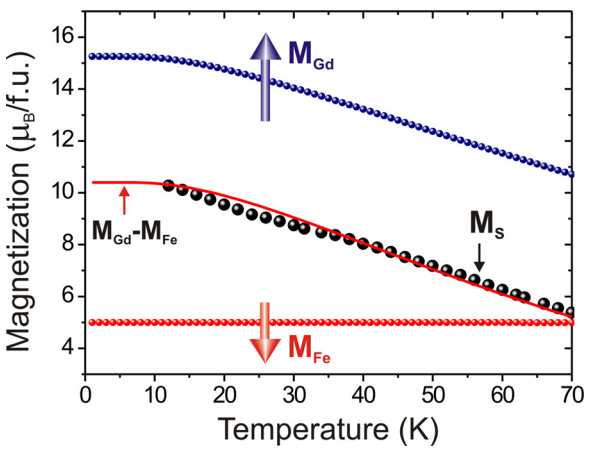

FIG. 3. Temperature dependence of the measured magnetization saturation M_s, theoretical calculation of $M_{Gd} - M_{Fe}$, and each magnetization of the Fe and Gd sublattices. We obtained the magnetizations by using molecular field theory with the following coefficients between respective sublattices: $\lambda_{aa} = 65$, $\lambda_{ad} = 95$, $\lambda_{dd} = -30.4$, $\lambda_{dc} = 6$, and $\lambda_{ac} = -2.9$ (mol cm^{-3}).

tetrahedral (d) and octahedral (a) sites, and the other of rare-earth ions occupying dodecahedral (c) sites. In our case, the magnetization saturation is given by $M_d - M_a - M_c$ or $M_{Fe} - M_{Gd}$. We obtained the temperature dependence of the Gd and Fe sublattice magnetizations by using molecular field theory with a molecular field coefficient for the three-sublattice system,[11] as shown in Fig. 3.

The Gd ions exhibit antiferromagnetic exchange coupling to the Fe ions; the Gd ions tend to align their spins against the net moment of the Fe ions. Because this coupling is relatively weak, the Gd ions order significantly only at low temperatures (<60 K), where the Gd sublattice is much more strongly magnetized than the Fe sublattice. As the temperature decreases, the frequency of the HF mode tends to increase with increases in M_s, which suggests an excitation of exchange Kittel–Kaplan resonance.[5] In our garnet, this resonance exists because of the exchange interaction between RE and TM sublattices. The magnetic moments of these sublattices are noncolinear during the whole precession, as shown in the inset of Fig. 1. Below 60 K, the Fe ions are very strongly coupled and the excitation between them is difficult in contrast to the previous work.[6] Thus, in garnets, the Gd–Fe coupling is relatively weak, and the Gd–Gd coupling is even weaker and is neglected here. Magnetization precession in our garnets occurs in a molecular field close to about 500 kOe.[11]

Figure 4(a) shows the time-resolved Faraday rotation at various temperatures. Over this whole temperature range, the HF mode with LF modulation appeared. By fitting with two frequencies,[12] we determined the temperature dependence of both modes' frequencies. As the total magnetization increased, the HF mode increased in frequency within the THz range via the excitation of exchange resonance (see Fig. 4(a)). The temperature dependence of the HF mode is direct evidence of this resonance. We can calculate the temperature dependence of the HF mode by using the Kaplan–Kittel formula[5] for the exchange resonance between the Fe and Gd sublattices

$$f_{ex}(T) = -\frac{\gamma \lambda_{Fe-Gd}}{2\pi}\left(M_{Fe}(T) - M_{Gd}(T)\right), \qquad (1)$$

where λ_{Fe-Gd} is the exchange parameter between the Fe and Gd sublattices, γ is the gyromagnetic ratio, and M_{Fe} and M_{Gd} are the magnetizations of the Fe and Gd sublattices, respectively. The gyromagnetic ratio is the same for these sublattices ($\gamma = \gamma_{Gd} = \gamma_{Fe}$) because, for Gd ions $L = 0$. The solid line in Fig. 4(b) was calculated using Eq. (1). The experimental data and theoretical calculations agree well.

To understand the qualitative nature of the LF mode, we measured f_{LF} as functions of temperature and external magnetic field. Figure 5(a) shows how the frequency depended on magnetic field H in the range of 0.9–2 kOe. Below 0.9 kOe, coherent magnetization precession did not appear because magnetic domain structures were present. The LF mode with $f_{LF} \sim 60$ GHz can be attributed to the spin-wave mode excitation by assuming a very high effective magnetic field of 20–30 kOe, which is one order higher than the magnitude of our external field. Thus, the LF mode cannot be explained by the excitation of the spin-wave mode, which was observed in a similar garnet at room temperature.[10] In our garnet, the frequency of the LF mode was slightly increased under applied magnetic field by a nonlinear manner. At a magnetic field above 2 kOe, the frequency was stabilized; however, by extrapolating this dependence at $H = 0$, the frequency does not reach zero. This dependence of the LF mode on magnetic field suggests the possibility of excitation of the dielectric resonance with a magnetic contribution. In yttrium iron garnet, because of the low magnetic and dielectric losses, dielectric resonance was observed at ~55 GHz using a U-band spectrometer at magnetic field 2 kOe.[13] Figure 5(b) shows that, as temperature increases, the frequency of the LF mode increases in the range of 60–70 GHz. This behavior may be related to the approach to the compensation point of the angular momentum, where the effective gyromagnetic ratio is enhanced.

In conclusion, we demonstrated non-thermal all-optical excitation of the collective exchange resonance between Gd and Fe sublattices in the THz frequency range via a single femtosecond NIR pulse. Our garnet exhibited exchange resonance with an extremely high frequency compared with those of RE-TM metallic alloys.[14,15] This resonance suggests the presence of an effective molecular field between the Gd and Fe sublattices.

Our results show that the NIR pulse induced phenomena that are usually observed in the FIR range at energy of a few meV. In general, this energy corresponds to atomic interactions in condensed matter. Through this, we show that time-resolved pump-probe techniques can provide valuable information about ultrafast exchange interactions in which

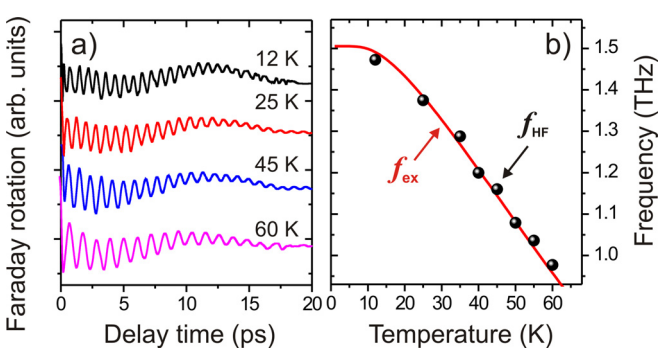

FIG. 4. Time-resolved Faraday rotation curves at various temperatures (a). Temperature dependence of the high-frequency mode f_{HF} (b). The solid line $f_{ex}(T)$ was calculated with Eq. (1).

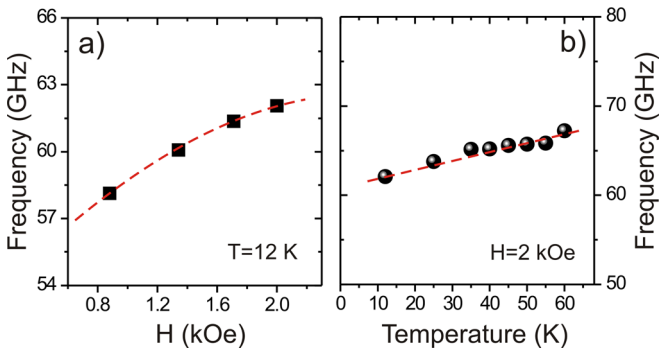

FIG. 5. Dependence of the low-frequency mode on the external magnetic field H at 12 K (a). Temperature dependence of f_{LF} at a magnetic field of 2 kOe (b). The dashed lines are guides for the eye.

energy transfers between the crystal lattice and spins in dielectric materials. These scientific problems are still open and require further investigation. Additionally, we observed an unexpected mode at low temperatures with a frequency of 60–70 GHz at an external magnetic field of 2 kOe. This mode could not be explained by spin-wave excitation. Instead, we attributed it to dielectric resonance with some magnetic contribution.

This work also contributes to the development of high-speed magnetic devices because, for example, garnets are widely used in magneto-optical modulators and microwave high-frequency filters. Our results will help in the development of materials with controlled magnetic and optical properties that allow the use of optical femtosecond pulses for a variety of magneto-optical devices. Recently, all-optical control by arbitrary light polarization state to possibility of magnetic writing has been demonstrated.[16] By understanding optical excitation of THz-level spin oscillations and local spin manipulation using polarized femtosecond pulses in an optical insulator, this may lead to memory based on both optical and magnetic mechanisms, enabling high-speed recording.

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