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Excitation of Electric-Field-induced Spin Wave in the Strained Garnet Ferrite Thin Films Using Sub-Picosecond Pulsed Wave

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Abstract

Garnet-type ferrite thin films of $Sm_3Fe_5O_{12}$ and $Lu_3Fe_5O_{12}$ have been grown on (0 0 1)-oriented $Gd_3Ga_5O_{12}$ substrates by a pulsed laser deposition. An electric-fields-induced spin wave can be excited in the strained $Sm_3Fe_5O_{12}$ films at room temperature using sub-picosecond pulsed wave. On the other hand, it is not observed in the strained $Lu_3Fe_5O_{12}$ films, due to the anisotropic crystal elongation based on the epitaxial strain.

1. Introduction and Experimental

Conduction-electron spin-currents in magnetic semiconductors and metals have a critical problem; they disappear within a very short range, typically hundreds of nanometers [1]. A spin-wave spin-current in magnetic insulator, on the other hand, persists several centimeters [2]. The main reason why the conductive spin-currents decay is thermal diffusion. Because there are no conductive electrons in magnetic insulators, spin-wave spin-currents, which are the collective motion of spins coupled by exchange interaction, can be propagated over a relatively long distances. In addition, spin pumping technique enables to convert the spin-wave spin-currents into conduction-electron spin-currents [3]. Since the success of the transmission of spin-currents across 1 mm using spin-pumping technique, much attention has been paid to spin-wave spin-currents in magnetic insulators [4].

Among the all of the types of magnons, such as electron spin resonance, Kaplan-Kittel exchange mode, ligand field mode [5], there are few types of magnons which can be excited by electromagnetic wave at room temperature (RT) without the application of the external magnetic field. These types of magnons are ferromagnetic resonance, ferrimagnetic resonance, anti-ferromagnetic resonance and electromagnons. Among them, we have focused on the electromagnons. Electromagnons are the newly discovered magnons which can be excited by the electric field component of terahertz (THz) wave [6]. The excitation strength of electromagnons is much greater than the conventional magnons existing in microwave region. Therefore, electromagnon spin-currents will enable the much longer and stronger signal transmission by using a sub-picosecond pulsed wave which involves the THz wave.

Herein, the electromagnons can be excited only in multiferroic materials, which exhibit the both ferroelectric and ferromagnetic properties simultaneously. However, there is no material which has the both multiferroicity and transparency in a wide THz regime at room temperature. Therefore, we firstly challenged to create the new material to realize the THz spintronics at RT. Above all, we have focused on an epitaxial strain. An epitaxial strain can change paraelectric into ferroelectric [7]. That is, the epitaxial strain can change paraelectric magnet into multiferroics.

In this study, garnet ferrites of $Sm_3Fe_5O_{12}$ (SmIG) and $Lu_3Fe_5O_{12}$ (LuIG) were selected as the target materials. SmIG and LuIG are ferrimagnetic and paraelectric at RT, and transparent in a wide THz regime. Moreover, the magnetization damping of SmIG and LuIG is very small: $\alpha \approx 6.7 \times 10^{-5}$, where α is the Gilbert damping coefficient [8]. Hence, SmIG and LuIG are the promising candidates for spintronics such as electromagnon spin-currents conductor.

The ultrathin films of SmIG (a = 12.530 Å) and LuIG (a = 12.277 Å) deposited on (0 0 1)-oriented $Gd_3Ga_5O_{12}$ (GGG: a = 12.383 Å) substrates using a pulsed laser deposition (PLD). The lattice mismatch causes the compressive strain to SmIG and the expansive strain to LuIG, which can be confirmed by reciprocal space mapping (RSM).

2. Results and Discussions

In the XRD (2θ-ω) patterns of SmIG and LuIG films on GGG (0 0 4) and (0 0 8) reflection peaks were observed for all of the films, suggesting that the SmIG and LuIG films were grown with a (0 0 1) orientation normal to the substrate surface. The rocking curves of SmIG and LuIG are 0.150 and 0.155 degrees, respectively, which suggests that the crystallinity of the thin films is as good as the previous report [9]. The RSM of the (-4 0 8) plane for all films indicates that SmIG and LuIG thin films were succeeded to grow epitaxially. According to the calculation of the lattice constants for out-of-plane and in-plane direction, the strain rates ($(c-a)/c$) of SmIG and LuIG films are 7.62 and 2.31%, respectively. The epitaxial strain can be induced in case where the thickness is thinner than the critical thickness. The critical thickness of SmIG and LuIG are 660 and 1250 Å, respectively, calculated by the equation shown in [10]. The thicknesses of the both films are 100 Å. Therefore, all of the films are successfully strained.

Figure 1(a)(b)(c) shows the THz spectroscopy of all films with an in-plane magnetization. The complex permittivity of the film was extracted using the way described in [11]. External magnetic fields of different strength were applied parallel to the film surface using neodymium magnets so as to confirm the magnetization dependence of the absorption peaks.

Two strong absorption peaks which are dependent to magnetization strength were observed in SmIG whereas there were no peaks in LuIG. In the bulk state of SmIG, the

lowest IR-active phonon exists in 10.3 meV. Hence, there are no characteristic peaks in the range shown in figure 1 in the case of bulk state at RT. When to assume that SmIG were changed into ferroelectric by applying the epitaxial strain, there are three possible origins of the absorption peaks: electromagnons, rigid layer mode [12] and soft mode [13]. However, the electromagnons only are dependent to magnetization strength. In addition, there are three distinctive properties of electromagnons: the magnetization dependence, two strong absorption peaks and the crystal azimuth dependence [14]. Figure 1(d)(e)(f) shows the THz spectroscopy of all films with an in-plane magnetization, rotating 90 degrees of the polarization of THz wave from the configuration in figure 1. The absorption peaks disappeared in figure 2, attributing the crystal azimuth. In-plane magnetization induces the anisotropy to in-plane direction. That is, they are considered to be derived from electromagnons because these absorption peaks matched all of the aforementioned three properties. The higher energy peak is due to the Brillouin Zone-edge magnon mode, and the lower energy peak is due to the harmonic magnon mode [14].

The reason why the electromagnon is not observed in the strained LuIG film is the difference of the expansive direction due to the epitaxial strain. The displacive ferroelectric represented by $BaTiO_3$ has the spontaneous polarization along the expansion direction [15]. The strained

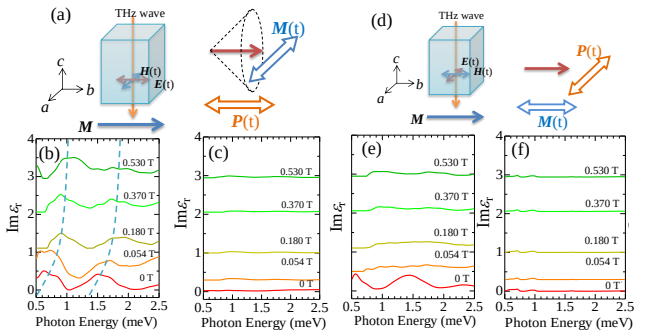

Fig. 1 (a) Configuration of spin precession induced by THz wave. THz spectroscopy of the strained SmIG film (b) and LuIG (c) in the configuration shown in (a). (d) Configuration which is rotating 90 degrees of the polarization of THz wave from the configuration (a). THz spectroscopy of the strained SmIG film (e) and LuIG (f) in the configuration shown in (d).

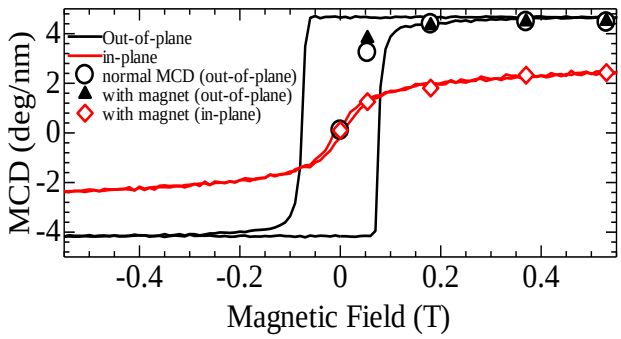

Fig. 2 Magnetic field dependence of MCD spectra for the strained SmIG film at room temperature. The excited photon energy is around 2.88-2.99 eV.

SmIG film uniaxially expands to out-of-plane direction, so that it has the spontaneous polarization along *c* axis. In contrast, the strained LuIG film biaxially expands to in-plane direction without anisotropic expansion. As a consequence, it does not have the spontaneous polarization. This result would not be unrelated that the magnetostriction constant of SmIG at 300 K is higher than any other rare-earth ferrite garnet ($\lambda_{100} = 21$, $\lambda_{111} = -8.5$) [8].

Figure 2 shows the magnetic property of the strained SmIG thin film measured by a magnetic circular dichroism (MCD). The external magnetic field dependence of MCD at 2.88-2.99 eV, corresponding to charge transfer energy: $O^{2-}(2p) \rightarrow Fe^{3þ}$ (3d), shows the hysteresis loops. The out-of-plane magnetization is saturated around 0.2 T whereas the in-plane magnetization is not saturated even at 0.5 T. That is, the easy axis flips to perpendicular direction, attributing to the shape magnetic anisotropy of ultrathin films [10]. The saturation magnetization is matched to the previous research (1.35 A/m ≈ 0.17 T) [8]. Next, we verified the validity of the magnetization measurement of THz spectroscopy using neodymium magnets. The MCD values using neodymium magnets are almost same with that of the electromagnet which is a guaranteed product as genuine MCD apparatus. Thus, the magnetization measurement of THz spectroscopy is guaranteed to have been succeeded.

3. Conclusions

As we design, the electromagnons can be excited using the strained SmIG thin film and sub-picosecond pulsed wave. The properties of two absorption peaks are matched to the properties of electromagnons. These results indicate that the SmIG thin film is the first candidate for electromagnon spin-current conductor.

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