Enhancement of domain-wall mobility at the angular momentum compensation temperature detected by NMR

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The angular momentum compensation temperature $T_A$ of ferrimagnets is attracting attention because of high-speed magnetic dynamics near $T_A$. We show that NMR measurement in ferrimagnets is capable of determining $T_A$ and domain-wall mobility. We perform a $^{57}$Fe-NMR study on the ferrimagnet Ho$_3$Fe$_5$O$_{12}$ with $T_A = 245$ K. The intensity of the NMR signal from nuclei inside domain walls shows a maximum at $T_A$. Conversely, the NMR signal intensity from nuclei inside domains does not show any anomalies at $T_A$. Because the domain-wall motion enhances NMR signal from nuclei inside domain walls, the NMR signal is enhanced at $T_A$, where the domain-wall mobility increases. The use of NMR in the multi-domain state is a powerful tool to estimate the domain-wall mobility as well as to determine $T_A$.

Recently, angular momentum compensation in ferrimagnets, where angular momenta on different sublattices cancel out each other, has attracted much attention aimed at the realization of high-speed magnetic dynamics in the field of spintronics [1,2]. The angular momentum compensation has long been discussed in ferrimagnetic materials and their shape [3]. However, ferrimagnets are incapable of determining $T_A$ and domain-wall mobility. We perform a $^{57}$Fe-NMR study on the ferrimagnet Ho$_3$Fe$_5$O$_{12}$ with $T_A = 245$ K. The intensity of the NMR signal from nuclei inside domain walls shows a maximum at $T_A$. Conversely, the NMR signal intensity from nuclei inside domains does not show any anomalies at $T_A$. Because the domain-wall motion enhances NMR signal from nuclei inside domain walls, the NMR signal is enhanced at $T_A$, where the domain-wall mobility increases. The use of NMR in the multi-domain state is a powerful tool to estimate the domain-wall mobility as well as to determine $T_A$.

The rare earth iron garnet system $R_3$Fe$_5$O$_{12}$ (RIG, where $R$ is a rare earth element) is also a ferrimagnet accompanied by $T_A$ [4,10]. However, RIG does not show any anomalies in FMR at $T_A$ because the angular momentum of $R^{3+}$ weakly couples with that of $\text{Fe}^{3+}$ and behaves almost as a free magnetic moment. As a result, the magnetic relaxation frequency of $R^{3+}$ magnetic moment is much higher than that of $\text{Fe}^{3+}$ magnetic moment and the exchange frequency of $R^{3+}$ and $\text{Fe}^{3+}$ [11,14]. In this case, the $R^{3+}$ magnetic moment follows the dynamics of the $\text{Fe}^{3+}$ magnetic moment. Hence, the $R^{3+}$ ions contribute to the magnetization but not to the angular momentum owing to heavy damping of the $R^{3+}$ site [14].

Although $T_A$ in RIG cannot be determined using FMR, it has been reported that the mobility of bubble domains formed in a substituted RIG system increases at a certain temperature, which is regarded as $T_A$ [12]. Recently, it has become possible to directly and exactly measure the net angular momentum regardless of the material and its shape by using the Barnett effect, in which magnetization is induced by mechanical rotation due to spin-rotation coupling [5,10]. Consequently, $T_A$ of Ho$_3$Fe$_5$O$_{12}$ (HoIG) was determined to be 245 K [3]. With the focus on magnetic dynamics at $T_A$, a microscopic method has been desired to explore the spin dynamics at $T_A$ regardless of materials and their shape.

Here, we propose an NMR method to explore the spin dynamics at $T_A$ and domain-wall mobility in HoIG. The rare earth iron garnet system Ho$_3$Fe$_5$O$_{12}$ (HoIG) is a ferrimagnet with $T_A = 245$ K. The intensity of the NMR signal from nuclei inside domain walls shows a maximum at $T_A$. Conversely, the NMR signal intensity from nuclei inside domains does not show any anomalies at $T_A$. Because the domain-wall motion enhances NMR signal from nuclei inside domain walls, the NMR signal is enhanced at $T_A$, where the domain-wall mobility increases. The use of NMR in the multi-domain state is a powerful tool to estimate the domain-wall mobility as well as to determine $T_A$.
In a magnetic ordered state such as in ferromagnets and ferrimagnets, the macroscopic magnetization of electrons enhances the NMR signal via hyperfine interactions. Particularly, the NMR signal from nuclei in domain walls is strongly enhanced owing to magnetic domain-wall motion, as shown in Fig. 1. An input radio frequency (RF) magnetic field $H_1$ used for NMR can move domain walls, so that magnetic moments in the walls rotate and generate a transverse component of a hyperfine field in synchronization with the RF field. As a result, $H_1$ is enhanced to become $\eta_{n}\hat{H}_1$, where $\eta_{n}$ is the enhancement factor for the input process. In the reverse process, the Larmor precession of nuclear spins causes domain-wall motion, because the electronic system feels an effective magnetic field $H_{\text{eff}}$ from the nuclear magnetization through the hyperfine interaction, which leads to oscillation of the bulk magnetization. Because the oscillation of the bulk magnetization induces a much stronger voltage in an NMR pickup coil than the precession of nuclear magnetic moment $m_n$, the output NMR signal is enhanced to be $\eta_{\text{out}}m_n$, where $\eta_{\text{out}}$ is the enhancement factor for the output process. This enhancement effect enables us to selectively observe the NMR signal from nuclei in domain walls, even though the volume fraction of domain walls is much smaller than that of domains.

In this Letter, we report the result of an NMR study of HoIG under a magnetic field of up to 1 T. For a multidomain state below 0.3 T, the temperature dependence of the NMR intensity shows a maximum at $T_A$. For a singledomain state above 0.5 T, the temperature dependence of the NMR intensity does not show any anomalies at $T_A$. These results indicate the enhancement of the domain-wall mobility at $T_A$. Extending a simple conventional model for describing $\eta_{\text{out}\, m_n}$, we formalize the modified enhancement factor $\eta_{\text{out}\, \text{eff}}$ taking the domain-wall mobility into account. This enhancement of the NMR intensity at $T_A$ enables us to estimate the domain-wall mobility as well as to determine $T_A$, even in a powder sample.

We synthesized HoIG by solid state reaction for this study. We ground the sample in a mortar to create a fine powder with a typical particle diameter of 5 $\mu$m. The NMR measurements of $^{57}$Fe nuclei were carried out using a standard phase-coherent pulsed spectrometer. The NMR signals were obtained using the spin-echo method, with a first pulse duration of $(1.0 \, \mu s)$ and a second pulse duration of $(2.0 \, \mu s)$. The spin-echo decay time $T_2$ was measured by varying the interval time $\tau$ between the first and second pulses. The value of $T_2$ is defined such that $\text{I}(2\tau) = \text{I}(0) \exp(-2\tau/T_2)$, where $\text{I}(2\tau)$ and $\text{I}(0)$ are the NMR intensity at $2\tau$ and $\tau = 0$, respectively. The nuclear spin-lattice relaxation time $T_1$ was measured using the inversion recovery method.

Figure 2(a) shows the temperature variation of NMR spectra of $^{57}$Fe at the $d$ site without external fields. Each NMR spectrum shows a single peak, and the peak shifts to higher frequencies with decreasing temperature. The NMR intensity shows the maximum at 245 K. The top panel of Fig. 2(b) shows integrated NMR intensities. Generally, the NMR intensities need to be calibrated when comparing them under different conditions. The NMR intensity $I$ is proportional to the voltage induced in an pickup NMR coil by the precession of the nuclear magnetization $m_n$. Thus, $I$ is proportional to $dm_n(t)/dt$. Because $m_n(t)$ rotates at the Larmor frequency $\nu$, $I$ is proportional to $\nu m_n$. The size of $m_n$ depends on the polarization of the nuclear spin derived from the Boltzmann distribution function. Thus $m_n$ is proportional to $\nu/T$, where $T$ is temperature. As a result, $I$ is proportional to $\nu^2/T$. Moreover, the NMR intensity measured by the spin-echo method depends on $T_2$. Therefore, we calibrated the NMR intensity by multiplying $T\nu^{-2} \exp(2\tau/T_2)$.

The calibrated NMR intensity continues to show maximum at 245 K, which coincides with $T_A$ determined by the Barnett effect, in which mechanical rotation induces magnetization $M_\Omega$ due to spin-rotation coupling. As shown in the bottom panel of Fig. 2(b), $M_\Omega$ becomes zero at two temperatures. The lower temperature coincides with $T_M$ determined by a conventional magnetization measurement. At $T_M$, spin-rotation coupling is effective, but $M_\Omega$ becomes zero due to disappearance of bulk magnetization. In contrast, the higher temperature can be assigned to $T_A$, where the bulk magnetization remains but the spin-rotation coupling is not effective due to disappearance of net angular momentum. There are no anomalies in the temperature dependence of $\nu$, $1/T_1$, and $1/T_2$ as shown in Fig. 2(c). These results indicate that the maximum of the NMR intensity can be attributed to an anomaly in the enhancement factor.

To perform NMR experiments for single-domain state, we characterized magnetic field dependence of HoIG as shown in Fig. 3. The top panel of Fig. 3 shows the NMR frequency in magnetic fields ranging from 0 to 1 T at 300 K. With the increase in magnetic field the resonance frequency decreases because the magnetic moment at the $d$ site aligns with the magnetic field above $T_M$ and the hyperfine coupling constant is negative. The line in the top panel of Fig. 3 shows a slope of $-57\gamma = -1.3757 \, \text{MHz/T}$. In the multi-domain state at low fields, the rate of decrease of the NMR frequency by applying external field is smaller than $-57\gamma$ until all the domain walls disappear. This is because the external field at nuclear positions is cancelled out by the demagnetizing field created by domain-wall displacement caused by the external magnetic field. In the single-domain state above 0.6 T, the NMR frequency decreases with the ratio of $57\gamma$ by the magnetic field.

The optimized RF input power is shown in the bottom panel of Fig. 3. At low magnetic fields, the RF input power is small due to the large $\eta_{in}$, suggesting that the NMR signal from the domain walls, which is more enhanced than that from domains, contributes mainly to the NMR intensity. The input power sharply increases in the region between 0.4 T and 0.5 T and saturates above 0.6 T. This result indicates that the domain structure changes from multi-domain to single-domain.
FIG. 2. The NMR results of $^{57}\text{Fe}$ at the $d$ site in Ho$_3$Fe$_5$O$_{12}$ at zero field. (a) Temperature dependence of the NMR spectra. (b) Temperature dependence of the signal intensity, magnetization induced by rotation $M_0$, namely, the Barnett effect, and magnetization in a magnetic field of 1000 Oe. The open and filled circles show the bare integrated signal intensity and calibrated intensity by multiplying $T\nu^{-2}\exp(2\tau/T_2)$, respectively. The blue crosses and blue line show $M_0$ and the guide of eye. The red line shows the magnetization. The black solid and dashed lines show the magnetization and angular momentum compensation temperatures of Ho$_3$Fe$_5$O$_{12}$, respectively. (c) Temperature dependence of resonance frequency (top), $1/T_1$, and $1/T_2$ (bottom).

Figure 3(a) shows the temperature dependence of the calibrated NMR intensity in various magnetic fields. In the multi-domain state at 0 T and 0.3 T, the NMR intensity shows a maximum at 245 K, and then decreases toward $T_M$. The temperature at which the NMR intensity shows a maximum at 0.3 T appears to slightly decrease. It is speculated that $T_A$ decreases under magnetic fields in RIG, because the expectation value of the angular momentum of $R^{3+}$ decreases above $T_M$ in a magnetic field due to the decrease of molecular field at the $R$ site.

Generally, in the ferro-(ferri-)magnetic state, the enhancement factor $\eta_{\text{out}}$ is proportional to the hyperfine field $H_n$; the hyperfine field $H_n$ is also proportional to the NMR frequency $\nu$. Therefore, the NMR intensity in the single-domain state above 0.5 T should be calibrated by multiplying $T\nu^{-3}\exp(2\tau/T_2)$. In the multi-domain state of this sample below 0.3 T; however, the enhancement factor does not depend on $\nu$ as shown later. The calibrated values of the NMR intensity at 0.5 T and 1.0 T are almost the same, except near $T_M$ and do not show any anomalies around $T_A$. The drop in intensity around $T_M$ is due to the decrease in signal enhancement, which is proportional to the magnetization. These results indicate that the maximum of the NMR intensity is attributed to the domain walls.

We discuss the enhancement of the NMR signal at $T_A$ via domain-wall motion using a simple conventional...
In this model, the domain-wall displacement $x$ is limited by a demagnetizing field $H_d(x)$. The maximum displacement $x_{\text{max}}$ is determined from a position in which $H_d(x_{\text{max}})$ is balanced with an oscillating effective field $H_{\text{eff}}$, which is created by the precession of the nuclear magnetic moment through the hyperfine interaction, $H_d(x_{\text{max}}) = H_{\text{eff}}$. Because the sample used for the NMR measurement in the present study is a powder, each particle in it is assumed to be spherical with radius $R$, as shown in Fig. 4(b). $H_d$ is expressed as $H_d(x_{\text{max}}) = 2\pi \mu \frac{m}{d} m$. The net electron magnetic moment $m$ is tilted by the effective field of $H_{\text{eff}} = \frac{\mu}{m} H_n$, where $H_n$ is the hyperfine field, and the tilt angle $\theta$ of $m$ can be described by $\theta = \pi x/d$, where $d$ is the domain-wall thickness. Then, the bulk magnetization induced by the nuclear magnetization is expressed as

$$\frac{R H_n m_n}{2 dm} = \eta_{\text{out}} m_n,$$

where $\eta_{\text{out}}$ is the enhancement factor of the NMR signal for the output process and is defined as $\eta_{\text{out}} = \frac{H_{\text{eff}}}{2 dm}$. This model assumes that the velocity of the domain-wall motion $v$ is fast enough to satisfy $v = \mu H_{\text{eff}} > 4x_{\text{max}} \nu$, where $\mu$ is the domain-wall mobility.

The general derivation of NMR enhancement induced by domain-wall motion does not include the mobility of the domain-wall. Herein, we consider that $v$ is not fast enough to follow the oscillating effective field, i.e., $v < 4x_{\text{max}} \nu$. In this case, the displacement $x$ is limited by $\mu$. Then, $x$ is expressed to be $\nu/4\nu$. The enhancement factor $\eta_{\text{out}}$ is modified such that

$$\eta_{\text{out}}' = \frac{\nu}{4x_{\text{max}} \nu} \eta_{\text{out}} = \frac{\pi}{4\nu} \mu.$$  

This formula indicates that $\eta_{\text{out}}'$ in the slow limit of domain-wall motion is proportional to $\mu$, and $\eta_{\text{out}}'$ in the fast limit of domain-wall motion is continually connected to the conventional $\eta_{\text{out}}$. It is noted that the $\eta_{\text{out}}'$ does not depend on the NMR frequency $\nu$.

The domain-wall mobility of HoIG has not been reported directly, but it can be estimated from the reported damping parameters [16, 20]. The domain-wall mobility of Gd$_3$Fe$_5$O$_{12}$ (GdIG) is 225 m sec$^{-1}$Oe$^{-1}$ at 298 K [19]. The magnitude of the damping is inversely proportional to the domain-wall mobility. Because the damping parameter of HoIG is 80 times as great as that of GdIG [20], the domain-wall mobility of HoIG at room temperature is estimated to be 2.8 m sec$^{-1}$Oe$^{-1}$. However, the domain-wall mobility required for motion $x_{\text{max}}$ is defined such that $4x_{\text{max}} \nu / H_{\text{eff}} = 2 \nu / \pi M$, which is estimated to be $4 \mu \text{sec}^{-1}$Oe$^{-1}$ with the following values, $4 \pi M \sim 500 \mu$, $R \sim 5 \mu$m, and $\nu \sim 50$ MHz. Thus, this evaluation indicates that, in HoIG, the displacement of domain walls induced by nuclear precession is limited by $\mu$. Therefore, in the multi-domain state in HoIG, we used the modified enhancement factor $\eta_{\text{out}}'$ in Eq. (2). We estimate the value of $\mu$ at $T_A$ to be $3.5 \mu$ sec$^{-1}$Oe$^{-1}$ using $\mu = 2.8 \mu$ sec$^{-1}$Oe$^{-1}$ at 300 K. When we assume $d$ to be 0.1-1.0 $\mu$m, $\eta_{\text{out}}'$ is estimated from $10^2$ to $10^3$, which is comparable to typical enhancement factors [17, 21]. Although the enhancement of $\mu$ is small at $T_A$, the NMR method is very sensitive to detecting such a small enhancement.

The existing methods to estimate $\mu$ require a single crystal or microfabricated samples [4, 15]. In comparison, using the present NMR method, we can estimate $\mu$ even in powder samples, because of the ability to selectively observe the NMR signal from the nuclei in the domain walls. Moreover, $T_A$ is detected to be the temperature at which the NMR intensity shows the maximum. In summary, NMR measurement in domain walls is a method to microscopically measure $\mu$ in ferrimagnetic samples, regardless of their conductivity and shape.

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