Angular momentum compensation manipulation to room temperature of the ferrimagnet Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$ detected by the Barnett effect

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We demonstrate that the angular momentum compensation temperature $T_A$, at which the net angular momentum in the sample disappears, can be controlled in Ho$_3$Fe$_5$O$_{12}$ by partially substituting Dy for Ho. The $T_A$ can be detected using the Barnett effect, by which mechanical rotation magnetizes an object due to spin-rotation coupling. We found that $T_A$ increases with the Dy content and clarified that the $T_A$ of Ho$_{1.5}$Dy$_{1.5}$Fe$_5$O$_{12}$ coincides with room temperature. The Barnett effect enables us to explore materials applicable to magnetic devices utilizing the angular momentum compensation only by rotating the powder sample at room temperature.

Angular momentum compensation is a key characteristic in the field of spintronics, where attention is focused on the high-speed magnetic response at the angular momentum compensation temperature. N-type ferrimagnets have a magnetic compensation temperature $T_M$, at which magnetization disappears even in the ferrimagnetically ordered state. Furthermore, when $g$-factors of the magnetic moment belonging to different sublattices are different, the ferrimagnetic materials have an additional compensation, namely the angular momentum compensation temperature $T_A$, at which the net angular momentum $\langle J_{net} \rangle$ in the material disappears even in the magnetically ordered state. While $T_M$ is easily determined by magnetization measurements, $T_A$ cannot be determined by conventional magnetization measurements using a magnetic field. It was recently reported that the domain wall mobility was enhanced at $T_A$ in GdFeCo. However, this method requires microfabrication of materials and only applies to metals. Nevertheless, using the Barnett effect, a phenomenon by which a rotating object is magnetized by spin-rotation coupling, the angular momentum compensation can be determined.

The Barnett effect was discovered in 1915. The angular momentum of electrons in matter interacts with rotational motion through spin-rotation coupling, described as

$$\mathcal{H} = -J \cdot \Omega, \quad (1)$$

where $J$ is the angular momentum and $\Omega$ is the angular velocity. The angular momentum is then aligned with the direction of the rotation axis, which causes magnetization. With ferrimagnets, $J$ is represented as the sum of the angular momenta belonging to different sublattices ($J_{net}$). The Barnett effect does not apply when $\langle J_{net} \rangle = 0$, which is the definition of the angular momentum compensation.

In this letter, we demonstrate $T_A$ control for the insulating rare earth iron garnet Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$. $T_A$ is observed using the Barnett effect. To realize a high-speed magnetic device using the angular momentum compensation, $T_A$ should coincide with the temperature at which the device is operated. We found that $T_A$ coincides with room temperature ($20^\circ$C) at $x = 1.5$.

Powder samples of Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$ were prepared from Fe$_5$O$_7$ (4N), Ho$_2$O$_3$ (3N), and Dy$_2$O$_3$ (3N) via a solid-state reaction. Fe$_5$O$_7$, Ho$_2$O$_3$, and Dy$_2$O$_3$ powders were mixed in a molar ratio of 5 : 3 – $x$ : $x$ in an agate mortar. The mixed powder was pelletized and heated to 1200 °C – 1400 °C in the ambient atmosphere. To characterize the samples, the DC-magnetization measurements were performed by extraction magnetometry with a commercial magnetometer (PPMS, Quantum Design).

Figure 1(a) shows the temperature dependence of the magnetization of Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$. The magnetization of both the end materials, Ho$_2$Fe$_5$O$_{12}$ (HoIG) and Dy$_2$Fe$_5$O$_{12}$ (DyIG), vanishes at $T_M = 135$ and 218 K, respectively. For the substituted materials, the magnetization does not disappear completely due to the $T_M$ distribution; thus $T_M$ was defined as the temperature at which the magnetization shows a local minimum. Figure 1(b) shows the linear relation between $T_M$ and the Dy concentration, and a schematic of the magnetization at sublattices. In the high-temperature region above $T_M$, the net magnetization does not disappear completely due to the $T_M$.
FIG. 1. (a) Temperature dependence of Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$ magnetization in a magnetic field of 1,000 Oe. (b) The variation in magnetization compensation temperature $T_M$ with the Dy content $x$ in Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$. (c) Schematic of the apparatus for Barnett effect measurement using an air-driven rotor system.

since the magnetization at the Fe$^{3+}$ sublattice, $M_{Fe}$, is larger than that at the R$^{3+}$ sublattice, $M_R$. $M_R$ aligns parallel to the magnetic field. The magnitude of $M_R$ increases as the temperature decreases, and $M_R$ equals $M_{Fe}$ at $T_M$. The magnetization of these sublattices flips across $T_M$, and then $M_R$ aligns parallel to the magnetic field below $T_M$ (see Section I of Supplementary Material for more details).

We measured the Barnett effect to observe angular momentum compensation. Our apparatus for rotating samples uses an air-driven rotor system as shown in Fig. 1(c). The rotor system was placed in magnetic shields to exclude the geomagnetic field. The sample was rotated using compressed air with angular velocity $\Omega$ and magnetized to $M_\Omega$ by the rotation. We measured the stray field from $M_\Omega$ using a fluxgate magnetic sensor. To remove the residual background magnetic field, we measured the difference in the stray field $\Delta B = [B(+\Omega) - B(-\Omega)]/2$, where $B(+\Omega)$ and $B(-\Omega)$ represent the stray fields obtained at $+\Omega$ and $-\Omega$, re-

FIG. 2. (a) Temperature dependence of $M_\Omega$ of Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$ ($x = 0$ and 0.5). The solid lines are the guide for eyes. (b) Schematic illustration of angular momentum and magnetic moment. Relationship between angular momentum (dark red and blue arrows) and magnetic moment (light red and blue arrows) in Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$. (c) Relationship between angular momentum and the magnetization $M_\Omega$ induced by rotation at various temperature. The dark and light purple arrows shows net angular momentum and magnetization of Ho$_{3-x}$Dy$_x$Fe$_5$O$_{12}$, respectively.
According to the negative sign of the effective factor, the net angular momentum becomes negative as shown in Fig. 3(b), because Eq. (1) indicates that $\Omega$ rotates along the $+z$ direction. At $T_\Lambda$, since the Barnett effect does not apply due to the net zero angular momentum, $M_\Omega$ becomes zero. The range of temperatures between $T_\Lambda$ and $T_M$, $M_\Omega$ becomes positive. This indicates that $\langle J_{net} \rangle$ and $M_{net}$ are coupled antiparallel, the sign of the $g$ factor becomes positive: this is a gyromagnetic reversal state. At $T_M$, though $\langle J_{net} \rangle$ aligns with the rotation axis, $M_\Omega$ is zero due to the disappearance of $M_{net}$ at $T_M$. Below $T_M$, $M_\Omega$ becomes negative again due to the antiparallel coupling between $\langle J_{net} \rangle$ and $M_{net}$. The increase in $T_\Lambda$ with increasing $x$ indicates that $T_\Lambda$ coincides with room temperature (293 K) at appropriate Dy substitution.

To investigate the $\text{Ho}_3-\text{Dy}_x\text{Fe}_5\text{O}_{12}$ composition with $T_\Lambda = 293$ K, the rotational frequency dependence of $M_\Omega$ was measured for various $\text{Ho}_3-\text{Dy}_x\text{Fe}_5\text{O}_{12}$ compositions at 293 K as shown in Fig. 3(a). For all compositions, $M_\Omega$ is proportional to $\Omega$, indicating that the Barnett effect was correctly measured. In HoIG, $M_\Omega$ is negative and decreases as $\Omega$ increases because $\langle J_{net} \rangle$ and $M_{net}$ are coupled antiparallel above $T_\Lambda = 240$ K (see Fig. 2(a)). As the Dy content $x$ increases, the slope of $M_\Omega$ vs. $\Omega$ gradually increases, becoming almost 0 at $x = 1.5$, and becoming positive at $x \geq 2.0$. Figure 3(b) shows the initial susceptibility induced by rotation: $M_\Omega/2\pi\Omega$ at 293 K. $M_\Omega/2\pi\Omega$ increases with increasing Dy content $x$. From the linear fit to the data, $T_\Lambda$ is equal to 293 K at $x = 1.49 \pm 0.31$.

Figure 3(c) shows a phase diagram of $\text{Ho}_3-\text{Dy}_x\text{Fe}_5\text{O}_{12}$. Both $T_M$ and $T_\Lambda$ depend linearly on $x$. The black line is the linear fit to the $T_M$ data. The red line is obtained by calculating the temperature dependence of $\langle J_{R} \rangle$ with various Dy contents (see Sections II and III of Supplementary Material). The calculations are consistent with our experimental results.

Here, we consider the Dy substitution effect on $T_M$. In the general rare earth iron garnet (RIG) system, $T_M$ predominantly depends on $M_{R}$, which contains two factors: the size of the $R^{3+}$ magnetic moment $\mu = -gJ$, and $R^{3+}Fe^{3+}$ interaction (see Section I of Supplementary Material). Since the Néel temperature is almost irrelevant to the rare earth $R^{3+}$, the ferrimagnetic order in RIG is predominantly contributed by the $Fe^{3+}Fe^{3+}$ interaction. Thus, the temperature dependence of magnetization at the $Fe^{3+}$ sublattice can be regarded as almost the same in the RIG series. The $R^{3+}Fe^{3+}$ interaction contributes to the temperature dependence of the magnetization at $R^{3+}$ sublattice. The $R^{3+}R^{3+}$ interaction is much weaker than...
any other interactions in $R$IG and is negligible except in the very low temperature region. With HoIG and DyIG, since Ho$^{3+}$ and Dy$^{3+}$ have the same magnetic moment of $\mu = 10 \mu_{B}$, $T_M$ only depends on the $R^{3+}$Fe$^{3+}$ interaction. Figure 3 shows the calculated temperature dependence of sublattice magnetization (see Section II of Supplementary Material). Because of the stronger Dy$^{3+}$Fe$^{3+}$ interaction compared to Ho$^{3+}$Fe$^{3+}$, the magnetization at the Dy$^{3+}$ sublattice is larger than that at the Ho$^{3+}$ sublattice. Consequently, the $T_M$ of DyIG is higher than that of HoIG.

In contrast to $T_M$, $T_A$ depends on $\langle J_R \rangle$, which contains two factors: the size of the $R^{3+}$ angular momentum $J_R$, and the $R^{3+}$Fe$^{3+}$ interaction. The calculated temperature dependence of the angular momenta at the Ho$^{3+}$ and Dy$^{3+}$ sublattices is shown in Fig. 4. Although the angular momentum of Dy$^{3+}$ ($J_{Dy} = 15/2$) is smaller than that of Ho$^{3+}$ ($J_{Ho} = 8$), $\langle J_{Dy} \rangle$ is larger than $\langle J_{Ho} \rangle$ because the Dy$^{3+}$Fe$^{3+}$ interaction is stronger than the Ho$^{3+}$Fe$^{3+}$ interaction. Therefore, the $T_A$ of DyIG is larger than that of HoIG.

We now discuss the relative values of $T_M$ and $T_A$. Essentially, the difference between $T_M$ and $T_A$ originates in the difference in the $g$ factors of the ions belonging to different sublattices. Especially, when all the ions have the same $g$ factor, $T_A$ merges with $T_M$. When the difference in $g$ factors between Fe$^{3+}$ and $R^{3+}$ increases, so does the difference between $T_M$ and $T_A$. In the Ho$_{3-x}$Dy$_x$Fe$_3$O$_{12}$ system, the $g$ factors of Fe$^{3+}$, Ho$^{3+}$, and Dy$^{3+}$ are 2.5/4, and 4/3, respectively. Since the difference in the $g$ factors in DyIG is smaller than that in HoIG, the difference between $T_M$ and $T_A$ is expected to be smaller in DyIG than in HoIG. However, the experimental result shows that $T_A(x)$ increases at almost the same rate as $T_M(x)$ against temperature as shown in Fig. 3(c). This is due to competition between two effects of Dy substitution: $T_A$ increases due to the stronger Dy$^{3+}$Fe$^{3+}$ interaction compared to Ho$^{3+}$Fe$^{3+}$, and $T_A$ decreases relative to $T_M$ due to the larger $g$ factor of Dy$^{3+}$ compared to that of Ho$^{3+}$.

In summary, we controlled the angular momentum compensation temperature of Ho$_{3-x}$Dy$_x$Fe$_3$O$_{12}$ by Dy substitution, and found that $T_A$ coincides with room temperature at $x = 1.5$ in this system using our apparatus for measuring the Barnett effect. Using the Barnett effect, $T_A$ is easily obtained without microfabrication of the sample, regardless of the metal or insulator. The Barnett effect enables us to explore candidate materials for high-speed magnetic devices exploiting fast-magnetization reversal at angular momentum compensation.

SUPPLEMENTARY MATERIAL

See supplementary material for details of the magnetic structure of the samples and their calculated magnetization and angular momentum.

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