Estimation of angular momentum compensation temperature in GdFe film by magnetic Compton scattering

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The angular momentum compensation temperature, \( T_A \), in transition metal rare earth (TM-RE) ferrimagnetic materials is a crucial property for utilizing antiferromagnetic spin dynamics, which is much faster than its ferromagnetic counterpart. However, reports on the estimation of \( T_A \) in ferrimagnets are limited. In this study, we measured the temperature dependence of the spin magnetization and orbital magnetization of GdFe amorphous perpendicular magnetization film by using magnetic Compton scattering. Then, we estimated \( T_A \) experimentally.

**Keywords**: magnetic Compton scattering, TM-RE ferrimagnets, angular momentum compensation temperature

Antiferromagnets are a promising candidate for the core elements of future spintronic devices because they exhibit ultra-fast spin dynamics and a low magnetic susceptibility to magnetic fields. \(^{1-4}\) These desirable properties originate from the antiferromagnetic ordering, in which the magnetic moments are compensated for on an atomic scale. \(^{5}\) However, generally, antiferromagnets are difficult to manipulate and probe due to the small net magnetization and magnetic immunity. Hence, recently, transition metal and rare earth (TM-RE) ferrimagnetic materials, in which two inequivalent magnetic sublattices are antiferromagnetically coupled, have been attracting attention. \(^{6-11}\) This is because these materials have nonzero magnetization, which enables us to manipulate and probe them. These ferrimagnets exhibit two compensation temperatures due to the different Landé g-factors of the TM and RE elements. One is the magnetization compensation temperature, \( T_M \), at which the net magnetic moment vanishes. The other is the angular momentum compensation temperature, \( T_A \), at which the net angular momentum vanishes. Ultra-fast domain wall (DW) motions were demonstrated at \( T_A \) in TM-RE ferrimagnets in both field-driven and current-driven cases. \(^{12-17}\) These results suggest that the ferrimagnets exhibit antiferromagnetic spin dynamics at \( T_A \). This is because the time evolution of the state of a magnet is governed by the commutation relation of the angular momentum, not of the magnetic moment. Therefore, the estimation of \( T_A \) is a crucial step toward utilizing antiferromagnetic spin dynamics, but reports on this have been limited.

Recently, Imai et al. \(^{18}\) reported an angular momentum compensation temperature in a ferrimagnet insulator HoFe\(_2\)O\(_4\), by using the Barnett effect measurement technique. This measurement is simple, but there is no element specific information, which is important to understanding ferrimagnetic properties. Here, as a pioneering new experimental method, we focus on magnetic Compton scattering. Compton scattering photons reflect the momentum density distribution of the target electron. Experimentally, by irradiating a magnetic substance with circular polarized X-rays, the spin magnetization of magnetically active electrons can be measured. If the system is composed of independent particles described by a single wave function, \( \psi_\sigma(r) \) ( \( \sigma = \) majority spin or minority spin), within the limit of the impulse approximation, we can write the momentum density \( n_\sigma(p) \) by summing over all occupied single electron states of the system \( X_\sigma(p) \) as follows.

\[
 n_\sigma(p) = \sum_{i}^{occ} |X_\sigma(p)|^2. \tag{1}
\]

\[
 X_\sigma(p) = \left( \frac{1}{2\pi\hbar} \right) \int \psi_\sigma(r) \exp(-i\vec{p}\cdot\vec{r}) d\vec{r}. \tag{2}
\]

Here, \( p = (p_x, p_y, p_z) \) is the momentum of an electron in a solid, and \( p_z \) is the momentum of an electron in the material along the X-ray scattering vector, \( \mathbf{K} \). The scattering vector is defined as

\[
 \mathbf{K} = \mathbf{K}_2 - \mathbf{K}_1. \tag{3}
\]

Here, \( \mathbf{K}_2 \) and \( \mathbf{K}_1 \) denote the wave vectors of incident and Compton scattering X-rays, respectively. A magnetic

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Compton profile, (MCP), \( J_{\text{mag}}(p_z) \), is given by the following equation.

\[
J_{\text{mag}}(p_z) = \int (\mathbf{n}_{\text{mag}}(\mathbf{p}) - \mathbf{n}_{\text{min}}(\mathbf{p})) \, dp_x \, dp_y.
\]  
(4)

Here, \( \mathbf{n}_{\text{mag}}(\mathbf{p}) \) (\( \mathbf{n}_{\text{min}}(\mathbf{p}) \)) denotes the momentum density with the majority (minority) spin. Then, the spin magnetic moment, \( \mu_s \), is determined from the \( J_{\text{mag}}(p_z) \) as \(^{19)-20}\)

\[
\mu_s = \int J_{\text{mag}}(p_z) \, dp_z.
\]  
(5)

Total magnetization \( M_t \), which is measured by a superconducting quantum interference device magnetometer, is expressed as the following.

\[
M_t = M_s + M_l.
\]  
(6)

Here, \( M_s \) denotes the spin magnetization, which is obtained from a magnetic Compton scattering measurement, and \( M_l \) denotes the orbital magnetization. Therefore, it is possible to estimate individual \( M_t \), \( M_s \), and \( M_l \).

In this paper, we focus on a ferrimagnetic GdFe alloy in which Gd and Fe are antiferromagnetically coupled. First, the magnetization compensation temperature \( T_M \) is obtained from the temperature dependence of \( M_t \). Next, the magnitudes of the spin magnetizations of Gd and Fe (\( M_{s,Gd} \) and \( M_{s,Fe} \)) in the ferrimagnetic GdFe alloy are measured using magnetic Compton scattering, and the spin magnetization compensation temperature \( T_s \), at which \( M_{s,Gd} \) is equal to \( M_{s,Fe} \), is determined. Then, assuming that the \( M_{l,Gd} \) of Gd is zero, the orbital magnetization \( M_{l,Fe} \) of Fe is obtained from the difference between \( M_t \) and \( M_s \). Then, the \( g \)-factor of Gd and Fe is determined, and finally, the \( T_A \) of GdFe alloy is estimated.

GdFe films with a thickness of 1 \( \mu \text{m} \) were grown by DC sputtering using Gd and Fe targets on Al film substrates with a thickness of 6 \( \mu \text{m} \) as shown in Fig. 1(a). The GdFe was covered with a 60-nm-thick SiN cap layer. The composition of the GdFe film was determined to be Gd\(_{22.5}\)Fe\(_{77.5}\) by an electron probe micro analyzer. X-ray diffraction of the film did not show any peak, suggesting that the film was amorphous. To increase the intensity of the MCP signal, 64 films were deposited layer-on-layer.

MCP measurements were performed at the BL08W beamline at SPring-8, Japan. BL08W provides circularly polarized high energy X-rays from a wiggler device \(^{21)-22}\). The energy of the incident X-rays was 182.6 keV \(^{23}\). The sample was set in a vacuum chamber to suppress the scattering of the X-rays by air. MCP was measured in magnetic field switching mode with a scattering angle of 178 degrees, in which the incident X-rays were parallel to the applied magnetic field. Superconducting magnets can apply magnetic fields ranging from -0.5 T to 0.5 T perpendicular to the film plane. MCP measurements were performed at 289, 250, 225, 200, 175, 150, 100, 50, and 10 K. The magnetization curves of the \( M_t \) were measured by a superconducting quantum interference device (SQUID) magnetometer. The MCP of a polycrystalline Fe plate with a thickness of 0.1 mm was measured at room temperature as a standard measurement.

Figures 1(b) and 1(c) show the magnetization curves and temperature dependence of the saturation magnetization for the remanent state of the Gd\(_{22.5}\)Fe\(_{77.5}\) film, respectively. The temperature dependence of the total magnetization \( M_t \) can be written as

\[
M_t(T) = \left[ M_{t,Fe}(T) \right] - \left[ M_{t,Gd}(T) \right] = M_{t,Fe}(0) \left( 1 - \frac{T}{T_c} \right)^{\gamma_{Fe}} - M_{t,Gd}(0) \left( 1 - \frac{T}{T_c} \right)^{\gamma_{Gd}}.
\]  
(7)

Here, \( M_{t,Fe}(0) \) and \( M_{t,Gd}(0) \) are the total magnetization of Fe and Gd at 0 K, respectively. \( \gamma_{Fe} \) and \( \gamma_{Gd} \) are the critical exponents of Fe and Gd, respectively, and \( T_c \) is the Curie temperature \(^{14), 24}\). We assume \( M_{t,Fe}(0) = 1130 \text{ emu/cm}^3 \) \(^{25}\), \( \beta_{Fe} = 0.50 \) \(^{26}\). Moreover, we determined \( T_c = 520 \text{ K} \) by measuring the temperature dependence of the magnetization \(^{14}\). It was estimated that \( M_{t,Gd}(0) = 1161 \pm 30 \text{ emu/cm}^3 \) and \( \beta_{Gd} = 0.60 \pm 0.005 \), leading to \( T_M = 131 \text{ K} \) with Eq. (7).

Fig. 1. (a) Structure of thin film sample used in experiment. For magnetic Compton scattering experiment, films were stacked in 64 layers to amplify signal intensity. (b) Magnetization curves at 289, 150, and 10 K. (c) Temperature dependence of saturation magnetization. Temperature at which saturation magnetization becomes zero on this curve is defined as magnetization compensation temperature \( T_M \), and \( T_M = 131 \text{ K} \) for this sample.
Figures 2(a)–2(c) show MCP as a function of the electron momentum ($p_e$) in atomic units (au) at 288, 150, and 10 K. The magnetic field ($H = 0.5$ T) was applied perpendicular to the film plane. The measurement points are indicated by the circles. The measured MCP for GdFe alloy can be reproduced by a linear combination of MCPs from 4f orbitals of Gd and those from 3d orbitals of Fe. This is because the electron orbitals of Gd from 1s to 5p and those of Fe from 1s to 3p are filled, and they are magnetically inactive and thus do not contribute to MCP. Furthermore, the contribution from the Gd 5d orbitals is negligibly small. Therefore, fitting free atom profiles to a high momentum line shape has been exploited to separate 4f contributions from 3d contributions in 4f rare earth–3d transition metal alloys. The purple solid lines in Figs. 2(a)–2(c) are the best fit with the least square method based on the above assumption, and the solid blue lines and the solid red lines indicate the MCPs of Gd and Fe, respectively. The 4f contribution of Gd obtained from the literature was used for the fitting. Above $T_M$, we observed that the MCP of Fe was oriented positive and the MCP of Gd was oriented negative as shown in Figs. 2(a) and 2(b) because $M_{\text{Fe}}(T) > M_{\text{Gd}}(T)$. In contrast, below $T_M$, we observed that the MCP of Fe was oriented negative and the MCP of Gd was oriented positive as shown in Fig. 2(c) because $M_{\text{Gd}}(T) > M_{\text{Fe}}(T)$. $T_M$ was 131 K for this sample.

Figure 2(d) shows the temperature dependences of the spin magnetization of Fe, $M_{\text{Fe}}(T)$, and that of Gd, $M_{\text{Gd}}(T)$, calculated from the MCPs for Fe and Gd obtained by the analysis described in a previous paragraph. The smaller spin magnetizations at 100 and 150 K indicate that the magnetizations in the GdFe film were not magnetically saturated because of the small external magnetic field ($\leq 0.5$ T). Note that the coercive field drastically increased around $T_M$. Figure 2(e) shows the temperature dependence of the total spin magnetic moments, which were calculated from $M_s(T) = |M_{\text{Fe}}(T)| - |M_{\text{Gd}}(T)|$ [Eq. (7)]. Here, we assume that the spin magnetizations also follow Eq. (7). Then, the fitting indicated that $M_{\text{Fe}}(0) = 1104 \pm 40$ emu/cm$^3$ and $T_S = 207 \pm 5$ K as shown in Fig. 2(e). Here, the measurement points at 100 K and 150 K are excluded from the fitting. As we have determined the total magnetization $M_t$ and the spin magnetization $M_s$, $M_l$ can be obtained from Eq. (6). Figure 3 shows the temperature dependences of $M_t$, $M_l$, and $M_s$. Since the orbital magnetization of Gd, $M_{\text{Gd}}(T)$ is zero, the difference between $M_t$ and $M_s$ is considered to correspond to the orbital magnetization of Fe, $M_{\text{Fe}}$. By assuming that $M_{\text{Fe}}$ has the same temperature dependence as for $M_{\text{Fe}}(T)$, $M_{\text{Fe}}(0)$ is estimated to be $25 \pm 2$ emu/cm$^3$. Therefore, the ratio of the orbital magnetization to the spin magnetization in Fe is $2.2 \pm 0.3$ %, which is consistent with a previous study.

As the g-factor is given by

$$g_{\text{Fe}} = \frac{2(|M_{\text{Fe}}(0)| + M_{\text{Fe}}(0))}{M_{\text{Fe}}(0)},$$

the g-factor of Fe in the sample is estimated to be $2.04 \pm 0.01$ by using determined values.

Fig. 2. (a) Magnetic Compton profiles (MCP) at 288 K, (b) 150 K, and (c) 10 K. Measurement points are indicated by circles. Solid purple lines are best fits with least square method. Red diamonds and blue triangles are deduced contributions from Fe and Gd, respectively. (d) Temperature dependences of spin magnetizations of Fe and Gd. (e) Temperature dependence of spin magnetization of GdFe. Blue dashed line represents fitting result with Eq. (7). Temperature at which spin magnetization is zero on this line is defined as spin magnetization compensation temperature $T_S$, and $T_S = 207 \pm 5$ K for this sample.
Fig. 3. Temperature dependences of orbital magnetization $M_l$ (diamonds), spin magnetization $M_s$ (green line), and total magnetization $M_t$ (blue line) of GdFe. $M_t$ was obtained by magnetization measurements using SQUID magnetometer, and $M_s$ was determined by magnetic Compton scattering measurements.

By applying the relationship between $T_A$, $T_M$, and $T_C$ reported in [20], the following equation is obtained for the present system,

$$T_A = \left( \frac{\beta_{Gd}}{\beta_{Fe}} \right)_{Fe-Fe} T_M + \left( 1 - \left( \frac{\beta_{Gd}}{\beta_{Fe}} \right)_{Fe-Fe} \right) T_C$$

Inserting determined values leads to $T_A = 202 \pm 2$ K, which is lower than $T_S = 207 \pm 5$ K. The small difference between $T_S$ and $T_A$ originate from $M_{S,Fe}(T) \gg M_{L,Fe}(T)$, $M_{S,Fe}(T) \sim M_{S,Fe}(T)$ and $M_{Gd}(T) = M_{Gd}(T)$ and it is confirmed that $T_M < T_A < T_S$ in this GdFe system.

In summary, we observed the magnetization compensation temperature and the spin magnetization compensation temperature in ferrimagnetic GdFe alloy film with perpendicular magnetic anisotropy using a SQUID magnetometer and magnetic Compton scattering. The spin magnetization and orbital magnetization of each element constituting the ferrimagnetic GdFe alloy were independently determined. Furthermore, we succeeded in estimating the angular momentum compensation temperature of the ferrimagnetic GdFe alloy film. Our results show that magnetic Compton scattering is a unique method for investigating the element specific spin magnetization and angular momentum compensation temperature in ferrimagnets.

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References

1) T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich: Nat. Nanotechnol., 11, 231 (2016).
2) V. Baltz, A. Manchon, M. Tsui, T. Moriyama, T. Ono, and Y. Tserkovnyak: Rev. Mod. Phys., 90, 015005 (2018).
3) J. Železny, P. Wadley, K. Obelnik, A. Hoffmann, and H. Ohno: Nat. Phys., 14, 220 (2018).
4) M. B. Jungfleisch, W. Zhang, and A. Hoffmann: Phys. Lett. A, 382, 865 (2018).
5) O. Gomonay, V. Baltz, A. Brataas, and Y. Tserkovnyak: Nat. Phys., 14, 213 (2018).
6) C. D. Stanciu, A. V. Kimel, F. Hansteen, A. Tsukamoto, A. Itoh, A. Kirilyuk, and T. Rasing: Phys. Rev. B, 73, 220402(R) (2006).
7) M. Binder, A. Weber, O. Mosecz, G. Wetzelsdorff, M. Izquierdo, J. R. Dahn, T. D. Hatchard, J.-U. Thiele, C. H. Baek, and M. R. Scheinfein: Phys. Rev. B, 74, 134404 (2006).
8) C. D. Stanciu, A. Tsukamoto, A. V. Kimel, F. Hansteen, A. Kirilyuk, A. Itoh, and T. Rasing: Phys. Rev. Lett., 99, 217204 (2007).
9) A. Kirilyuk, A. V. Kimel, and T. Rasing: Rev. Mod. Phys., 82, 2731 (2010).
10) A. Mekonnen, M. Cernier, A. V. Kimel, A. Kirilyuk, A. Hrabec, L. Ranno, and T. Rasing: Phys. Rev. Lett., 107, 117202 (2011).
11) A. Kirilyuk, A. V. Kimel, and T. Rasing: Rep. Prog. Phys., 76, 026501 (2013).
12) K.-J. Kim, S. K. Kim, Y. Hirata, S.-H. Oh, T. Tono, D.-H. Kim, T. Okuno, W. S. Kim, S. Kim, G. Go, Y. Tserkovnyak, A. Tsukamoto, T. Moriyama, K.-J. Lee, and T. Ono: Nat. Mater., 16, 1187 (2017).
13) S. A. Siddiqui, J. Han, J. T. Finley, C. A. Ross, and L. Liu: Phys. Rev. Lett., 121, 057701 (2018).
14) L. Caretta, M. Mann, F. Bittner, K. Ueda, R. Pfau, C. M. Günther, P. Hessing, A. Churikova, C. Klose, M. Schneider, D. Engel, C. Marcus, D. Bono, K. Bugansch, and G. S. D. Beach: Nat. Nanotechnol., 13 (2018).
15) K. Aoshima, R. Ebisawa, N. Funabashi, K. Kuga, and M. Machida: Jpn. J. Appl. Phys., 57, 09FT03 (2018).
16) D.-H. Kim, T. Okuno, S. K. Kim, S.-H. Oh, T. Nishimura, Y. Hirata, Y. Futakawa, H. Yoshikawa, A. Tsukamoto, Y. Tserkovnyak, Y. Shiota, T. Moriyama, K.-J. Kim, K.-J. Lee, and T. Ono: Phys. Rev. Lett., 122, 127203 (2019).
17) T. Okuno, S. K. Kim, T. Moriyama, D.-H. Kim, H. Mizuno, T. Ikebuchi, Y. Hirata, H. Yoshikawa, A. Tsukamoto, K.-J. Kim, Y. Shiota, K.-J. Lee, and T. Ono: Appl. Phys. Express, 12, 093001 (2019).
18) M. Imai, Y. Ogata, H. Chudo, M. Ono, K. Hariri, M. Matsuo, Y. Ohnouma, S. Maekawa, and E. Saitoh: Appl. Phys. Lett., 113, 052402 (2018).
19) N. Sakai, M. Ito, H. Kawata, I. Iwama, M. Ando, N. Shiotani, F. Itoh, Y. Sakurai, and S. Nanao: Nanot. Instrum. Methods A, 303, 488 (1991).
20) N. Sakai: J. Appl. Cryst., 29, 81 (1996).
21) Y. Kikutani, Y. Kubo, A. Koizumi, N. Sakai, L. B. Abuha, and K. B. Sharma: J. Phys. Soc. Jpn., 72, 72599 (2003).
22) M. Itou, A. Koizumi, and Y. Sakurai: Appl. Phys. Lett., 102, 082403 (2013).
23) A. Agui, A. Harako, A. Shibayama, K. Haisi, N. Tsui, X. Liu, G. Ma, and T. Sakurai: Journal of Magn. and Magn. Mater., 484 (2019).
24) Y. Hirata, D.-H. Kim, T. Okuno, T. Nishimura, D.-Y. Kim, Y. Futakawa, H. Yoshikawa, A. Tsukamoto, K.-J. Kim, S.-B. Choe, and T. Ono: Phys. Rev. B, 97, 220403(R) (2018).
25) R. Hasegawa and R. Ray: *Phys. Rev. B*, **20**, 1 (1979).
26) S. Blindell: Magnetism in Condensed Matter (2001).
27) H. Sakurai, M. Ota, X. Liu, A. Morisako, Y. Sakurai, M. Itou, T. Nagao, and A. Koizumi: *J. Appl. Phys.*, **102**, 013902 (2007).
28) F. Biggs, L. B. Mendelsohn, and J. B. Mann: *Atom. Data Nucl. Data*, **16** (1975).
29) S. Mangin, C. Bellouard, S. Andrieu, F. Montaigne, P. Ohresser, N. B. Brookes, and B. Barbara: *Phys. Rev. B*, **70**, 014401 (2004).

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