Nontrivial temperature dependence of ferromagnetic resonance frequency for spin reorientation transitions

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We find unusual temperature dependence of the ferromagnetic resonance (FMR) frequency f_R for the spin-reorientation (SR) transition, in which the easy axis changes depending on temperature, observed in the Nd permanent magnet, $\text{Nd}_2\text{Fe}_{14}\text{B}$: $f_R \sim 0$ below the SR transition temperature (T_R), drastic increase of f_R around T_R , and decrease from a peak at higher temperatures. It is nontrivial that the SR transition causes the unusual behavior of the FMR frequency in a wide temperature region. We show the mechanism of the temperature dependence by theoretical and computational analyses. We derive a general relation between f_R and magnetizations to help the understanding of the mechanism, and clarify that the fluctuation of the transverse magnetization is a key ingredient for the resonance in all temperature regions.

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Ferromagnetic resonance (FMR) measurement is one of the most important methods to study the dynamics of ferromagnetic materials. FMR frequency signals tell us the information about spin dynamics such as spin precession motion, damping factor, etc. However, how the FMR frequency behaves for spin-reorientation (SR) transitions is hardly understood. The neodymium (Nd) permanent magnet [1–3], $\text{Nd}_2\text{Fe}_{14}\text{B}$, which has strong coercivity, is an indispensable material in modern technologies, applied to information-storage devices, hybrid and electric vehicles, generators, etc. This magnet exhibits a temperature-induced SR transition. However, due to its strong magnetic anisotropy energy [4], it is difficult to observe FMR signals in such a hard magnet, and the spin dynamics on the SR transition in the magnet has not been clarified. The temperature dependence of the FMR frequency is an open question.

Time domain measurement (TDM) of spin dynamics has rapidly been developed for the coherent control of spin precession motion toward spintronics technologies. The change of the precession frequency accompanying a SR transition has been detected recently in a rare-earth orthoferrite (RFeO_3) by using a time-resolved magneto-optical effect microscope [5–7]. Very recently a TDM of spin dynamics has been performed to evaluate the interlayer exchange coupling between hard and soft Nd magnet layers, and the resonance frequency for a single thick layer of the hard Nd magnet was estimated to be $f = \omega/(2\pi) = 161$ GHz under an external field at 2 T at room temperature [8]. The SR transition in the Nd magnet is a new target for TDM. Precession motion associated with SR transitions becomes an important and attractive topic.

Motivated by the experimental situation, we study the dynamics of the SR transition for the Nd magnet in the

present Rapid Communication. We find that the FMR frequency exhibits nontrivial temperature dependence with a drastic change around T_R . We investigate the origin of the behavior and find a universal mechanism for systems with the SR transition.

In the present work we study an atomistic spin model for the Nd magnet [9–11]. Compared to micromagnetics modeling, i.e., the coarse-grained continuum model approach often used for studies of permanent magnets [12], the atomistic spin modeling has advantages for investigating microscopic magnetic properties and thermal fluctuation effects [13–16]. Indeed temperature dependences of magnetization, domain wall (DW) width, DW free energy, anisotropy effects of Nd and Fe, etc., have been clarified by using the atomistic spin model for the Nd magnet [see (1)] [9–11,17,18], in which the microscopic parameters were taken mainly from first-principles calculations. Here we focus on the temperature dependence of the FMR frequency at zero external field. Under a finite external field h_e , the resonance frequency shifts by $\gamma h_e/(2\pi)$.

The atomistic Hamiltonian for the Nd magnet is given by the form [9–11]

$$\mathcal{H} = - \sum_{i < j} 2J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j - \sum_i^{\text{Fe}} D_i (s_i^z)^2 + \sum_i^{\text{Nd}} \sum_{l,m} \Theta_{l,i} A_{l,i}^m \langle r^l \rangle_i \hat{O}_{l,i}^m. \quad (1)$$

Here J_{ij} denotes the Heisenberg exchange coupling between the i th and j th sites, and D_i is the magnetic anisotropy constant for Fe atoms. For Fe and B atoms, s_i is the magnetic moment at the i th site. For Nd atoms, s_i is the moment of the valence ($5d$ and $6s$) electrons and is coupled antiparallel to the

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FIG. 1. (a) Temperature dependences of m^2 , m_z^2 , and m_{xy}^2 for the Nd magnet model (1). (b) θ dependence of the per-site anisotropy energy of Eq. (2) for the Nd magnet model.

moment of the $4f$ electrons $\mathcal{J}_i$, and thus the total magnetic moment is given by $\mathbf{S}_i = \mathbf{s}_i + \mathcal{J}_i$, in which $\mathcal{J}_i = g_T \mathbf{J}_i \mu_B$ with the magnitude of the total angular momentum, $J = 9/2$, and Landé g factor, $g_T = 8/11$. For the Fe and B atoms, $\mathbf{S}_i = \mathbf{s}_i$ is defined. The Zeeman term for the model is given as $-\sum_i \mathbf{h}_e \cdot \mathbf{S}_i$.

The last term of Eq. (1) is the magnetic anisotropy energy for Nd atoms, where $\Theta_{l,i}$, $A_{l,i}^m$, $\langle r^l \rangle_i$, and $\hat{O}_{l,i}^m$ are the Stevens factor, the coefficient of the spherical harmonics of the crystalline electric field, the average of r^l over the radial wave function, and Stevens operator, respectively, at site i for Nd atoms. The summation for l runs $l = 2, 4, 6$ and diagonal operators ($m = 0$) give dominant contribution, and then the last term is given in the series of J_i^z as

$$\sum_i^{\text{Nd}} D_1^{\text{Nd}} (J_i^z)^2 + D_2^{\text{Nd}} (J_i^z)^4 + D_3^{\text{Nd}} (J_i^z)^6 + \text{const.} \quad (2)$$

Figure 1(a) depicts the temperature dependences of $m^2 \equiv \frac{1}{N^2} \langle \sum_{\alpha=x,y,z} S_i^\alpha \rangle^2$, $m_z^2 \equiv \frac{1}{N^2} \langle (\sum_i S_i^z)^2 \rangle$, and $m_{xy}^2 \equiv \frac{1}{N^2} \langle (\sum_i S_i^x)^2 + (\sum_i S_i^y)^2 \rangle$, where $\langle X \rangle$ is the thermal average of X for the system with $6 \times 6 \times 6$ unit cells with periodic boundary conditions [19]. The SR transition occurs at $T_R \sim 150$ K [9,10], close to the experimental estimation [20–22]. The critical temperature is $T_c \sim 850$ K, a little bit larger than the experimental estimation [20–22], which does not affect the discussion here. The spins align tilted from the c axis (z axis) for $T < T_R$, while they align parallel to the c axis for $T > T_R$. Because the minimum of the anisotropy energy (2) is realized at $\theta \simeq 0.2\pi$ [Fig. 1(b); see also Fig. S1 in the Supplemental Material (SM) [23]], the most preferable direction of the magnetization in the ground state is not the c axis (z direction), which is the easy axis at room temperature.

Now we study the temperature dependence of the FMR frequency at zero external field. The FMR spectrum $I(f)$ is calculated by the autocorrelation function of spins (power spectrum):

$$I^\alpha(f) \equiv \frac{1}{T} \int_{t_0}^{t_0+T} d\tau I^\alpha(\tau) e^{i2\pi f\tau}, \quad (3)$$

FIG. 2. Temperature dependences of f_R (blue diamonds) and f_{SW} (red circles) for the Nd magnet model.

where

$$I^\alpha(\tau) = \frac{1}{T} \int_{t_0}^{t_0+T} dt \langle \bar{S}^\alpha(t) \bar{S}^\alpha(t+\tau) \rangle. \quad (4)$$

Here $\bar{S}^\alpha = \frac{1}{N} \sum_i S_i^\alpha$ for $\alpha = x, y$, and z . For the spin dynamics, we adopt the stochastic Landau-Lifshitz-Gilbert (SLLG) equation [16,24]:

$$\begin{aligned} \frac{d}{dt} \mathbf{S}_i = & -\frac{\gamma}{1 + \alpha_i^2} \mathbf{S}_i \times (\mathbf{H}_i^{\text{eff}} + \boldsymbol{\xi}_i) \\ & - \frac{\alpha_i \gamma}{(1 + \alpha_i^2) S_i} \mathbf{S}_i \times [\mathbf{S}_i \times (\mathbf{H}_i^{\text{eff}} + \boldsymbol{\xi}_i)], \end{aligned} \quad (5)$$

where α_i is the damping factor at the i th site and γ is the gyromagnetic constant. Here $\mathbf{H}_i^{\text{eff}} = -\frac{\partial \mathcal{H}}{\partial \mathbf{S}_i}$ is the effective field applied at the i th site from the exchange interactions and the anisotropy terms, and $\boldsymbol{\xi}_i$ is a random noise introduced into each site and has the relation $\langle \xi_k^\mu(t) \rangle = 0$, $\langle \xi_k^\mu(t) \xi_l^\nu(s) \rangle = 2\tilde{D}_k \delta_{kl} \delta_{\mu\nu} \delta(t-s)$. Here ξ_i^μ is the μ ($=x, y$, or z) component of the white Gaussian noise. We assume $\alpha_i = 0.1$ [12]. The strength of the noise is given by the fluctuation-dissipation relation: $\tilde{D}_i = \frac{\alpha_i k_B T}{S_i \gamma}$, which guarantees the thermal equilibrium state in the steady state.

The temperature dependence of the FMR frequency f_R is plotted by blue diamonds in Fig. 2. The red circles are explained below. We find that f_R exhibits a drastic change around T_R and nonmonotonic temperature dependence. In particular, it is found that $f_R \sim 0$ for the magnetization tilted from the c axis and f_R drops sharply at the SR point. The spectrum of $\tilde{I}(f) \equiv \frac{I(f) + I^*(f)}{2}$ has a single peak (Fig. S2 in the SM [23]), and f_R was estimated by the median of the spectrum of $\tilde{I}(f)$ [25]. The estimated f_R at 400 K ($T/T_c \sim 0.5$) in the present simulation is 141 GHz under zero field and then $f_R = 197$ GHz under 2T field, which is the same order of the experimental value (161 GHz) at room temperature [8].

The sudden decrease of the resonance frequency is related to the SR transition. To capture the fundamental mechanism of the nonmonotonic temperature dependence of f_R , the Nd magnet model (1) is too complicated, and thus we introduce a

FIG. 3. For $D_1 = 1$ and $D_2 = 0$ in the minimal model (6), temperature dependences of m^2 , m_z^2 , and m_{xy}^2 are given in (a) and those of f_R (blue circles) and f_{SW} (red circles) are plotted in (b). For $D_1 = 1$ and $D_2 = -0.7$, the former and latter dependences are depicted in (c) and (d), respectively. $k_B = 1$ was set. The time step for Eq. (5) $dt = 0.005$ was used for the evaluation of f_R .

minimal model which exhibits the SR transition:

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - D_1 \sum_i (S_i^z)^2 - D_2 \sum_i (S_i^z)^4, \quad (6)$$

where $J = 1$ is the exchange constant for nearest-neighbor pairs, used as the unit of energy, D_1 and D_2 are primary and secondary uniaxial anisotropy constants, respectively, and S_i is a classical unit spin.

In the ground state (GS) all magnetic moments are aligned in the same direction and the per-site exchange energy is $E_{\text{ex}} = -\frac{z}{2}J$, where z is the coordination number. Thus the per-site GS energy is given by the minimum of $E_G = -\frac{z}{2}J - D_1(S^z)^2 - D_2(S^z)^4$. For $D_2 = 0$ the easy axis is given by $\theta_0 = 0$. However, for $D_2 < 0$, the magnetic moments are aligned in the direction of $\theta_0 = \cos^{-1}(\sqrt{-D_1/2D_2})$. Here we adopt $D_1 = 1$ and $D_2 = -0.7$ to have $\theta_0 \simeq 0.2\pi$, which shows a similar potential minimum as in the Nd magnet model (1) (see Fig. S3 in the SM [23]).

As a reference for the conventional case, we first check the temperature dependence of f_R for the simple ferromagnet with $D_1 = 1$ and $D_2 = 0$. The temperature dependences of m^2 , m_z^2 , and m_{xy}^2 ($k_B = 1$ is set) are depicted in Fig. 3(a), where $T_C \simeq 1.77J$, which is a little bit higher than that for the Heisenberg model ($1.44J$) [26]. The resonance frequency $f_R(T)$ is plotted by blue circles in Fig. 3(b) (see also Fig. S4 in the SM [23]), which shows a monotonic temperature dependence similar to that of m_z^2 .

Next, we study the case with the SR transition with $D_2 = -0.7$. We find a SR transition at $T_R \simeq 0.4$ and ferromagnetic

transition at $T_C \simeq 1.6$ in Fig. 3(c). In Fig. 3(d) $f_R(T)$ is shown (see also Fig. S5 in the SM [23]). We find $f_R \sim 0$ below T_R , f_R increases above T_R with rising temperature, and it exhibits a peak at an intermediate temperature and reduction at higher temperatures. This behavior is qualitatively similar to that found in the Nd magnet model. Thus, $f_R \sim 0$ at low temperatures is attributed to the SR transition.

Now we analyze the mechanism of $f_R \sim 0$ in the low-temperature phase. Below T_R the magnetization is tilted from the c axis, where the effective field applied to each site is given as

$$h_{i,\text{eff}} = -\frac{\partial H}{\partial S_i} = J \sum_j^{\text{NN}} S_j + h_{\text{eff}}^{\text{Aniso}} e_z, \quad (7)$$

where $h_{\text{eff}}^{\text{Aniso}} = 2D_1 S_i^z + 4D_2 (S_i^z)^3$ is the contribution from the anisotropy term. The field from the exchange interactions does not contribute to the precession motion because it is parallel to the spin alignment, and thus the frequency for the precession motion is given by

$$f = \gamma h_{\text{eff}}^{\text{Aniso}} / (2\pi). \quad (8)$$

For $D_2 = 0$, $f = \gamma h_{\text{eff}}^{\text{Aniso}} / (2\pi) = 2D_1 m_z / (2\pi) = 0.318$ [see Fig. 3(b)], which depends on the temperature proportionally to m_z . It is the conventional temperature dependence. On the other hand, for $D_2 \neq 0$, the situation is different. Considering the relations $\frac{dE_G}{d\theta}|_{\theta=\theta_0} = \frac{dE_G}{dS_z} \frac{dS_z}{d\theta}|_{\theta=\theta_0} = -h_{\text{eff}}^{\text{Aniso}} \sin \theta_0$, we notice a relation: $h_{\text{eff}}^{\text{Aniso}} = 0$ for $\theta = \theta_0$ because $\frac{dE_G}{d\theta}|_{\theta=\theta_0} = 0$ and $\sin \theta_0 \neq 0$. Thus, we have an important consequence:

$$f = \gamma h_{\text{eff}}^{\text{Aniso}} / (2\pi) = 0 \quad \text{for } \theta = \theta_0 \neq 0. \quad (9)$$

To deduce Eq. (9), nonzero θ_0 is essential. Here we adopted $-D_1 \sum_i (S_i^z)^2$ and $-D_2 \sum_i (S_i^z)^4$ anisotropy terms for the minimal model. Instead of this choice, for example, we can adopt $-D_2 \sum_i (S_i^z)^4$ and $-D_3 \sum_i (S_i^z)^6$, in which $D_2 > 0$ and $D_3 < 0$, without the D_1 term. This choice also can give nonzero θ_0 .

Finally we investigate the temperature dependence of f_R for $T > T_R$. For $T > T_R$, m_{xy}^2 does not appear and the D_2 term is not essential. Thus we consider the situation for $D_2 = 0$ with applying Stoner-Wohlfarth's single domain picture. The free energy for the uniform single domain with a magnetic anisotropy K under zero field is given as $F = -K(T) \cos^2 \theta$, where K is a function of temperature. Here $\cos \theta = \frac{M_z}{M}$. M and M_z are the total magnetization and its z component, respectively. It should be noted that this θ is not the same as θ_i defined for each spin as $S_i^z = S \cos \theta_i$. The internal field for each magnetic moment is

$$h_{\text{eff}} = -\frac{dF}{dM_z} = 2K(T) \frac{M_z}{M^2}. \quad (10)$$

Thus the resonance frequency is given as $f_{\text{SW}} = \gamma h_{\text{eff}} / (2\pi) = \gamma K \frac{M_z}{M^2} / \pi$.

To evaluate $K(T)$ in the microscopic models (1) and (6), we derive the following relation between $K(T)$ and the transverse-field susceptibility χ_{xx} . The free energy of the system under a transverse field is $F = -K \cos^2 \theta - H_x M \sin \theta$. Here $M_x = M \sin \theta_{\text{min}} = \frac{M^2}{2K} H_x$, where θ_{min} is the angle to realize the stable state. M_x is also expressed as $M_x = \chi_{xx} H_x$, where χ_{xx} is the susceptibility in the hard direction. From these, we have $K(T) = \frac{M^2}{2\chi_{xx}}$. The susceptibility χ_{xx}

FIG. 4. Temperature dependences of Tm_z (blue circles) and $1/m_{xy}^2$ (red diamonds) for the minimal model (6) with (a) $D_2 = 0$ and (b) $D_2 = -0.7$, and for (c) the Nd magnet model (1).

is evaluated by $\chi_{xx} = \frac{N^2 m_{xy}^2}{2k_B T}$. Then we find a relation $K(T) = \frac{k_B T M^2}{N^2 m_{xy}^2}$, from which we obtain the temperature dependence of the resonance frequency as

$$f_{\text{SW}} = \frac{1}{\pi} \gamma K \frac{M_z}{M^2} = \frac{1}{\pi} \gamma \frac{k_B T m_z}{N m_{xy}^2}. \quad (11)$$

We notice that this formula can be extended for lower temperatures below T_R . Because of $m_z \sim O(1)$ and $m_{xy}^2 \sim O(1)$ for $T < T_R$, $f_{\text{SW}} = \gamma k_B T \times O(1/N) \simeq 0$. It is also worth noting that for $T > T_R$, $m_{xy}^2 \sim O(1/N)$ and $f_{\text{SW}} \sim O(1)$. In Figs. 2, 3(b), and 3(d), f_{SW} are plotted by red circles. We find excellent agreements between f_{SW} and f_R in all cases including the temperature region $T < T_R$.

Figures 4(a), 4(b) and 4(c) depict the temperature dependences of Tm_z and $1/m_{xy}^2$ for model (6) with $D_2 = 0$, $D_2 \neq 0$,

and model (1), respectively. We find that the temperature dependence of Tm_z is qualitatively similar in all cases, while that of $1/m_{xy}^2$ is qualitatively different, and the behavior of m_{xy}^2 , i.e., the fluctuation of the transverse magnetization, is a very important ingredient for the FMR frequency.

To conclude, we showed unusual temperature dependence of the FMR frequency in a wide temperature region for the spin-reorientation transition in the Nd magnet, caused by the competition between magnetic anisotropy energies: $f_R \sim 0$ below T_R , f_R drastically changes around T_R , and it exhibits a peak and then decrement at higher temperatures. This is totally different from the dependence of conventional magnets with a single uniaxial anisotropy energy, in which a monotonic decrease of the FMR frequency is observed. We clarified the mechanism for $f_R \sim 0$ below T_R . It is worth noting that the state of the tilted spin alignment is stable, and the precession around the GS easy axis (parallel to the GS magnetic moments) does not occur. We also derived the formula (11) for the FMR frequency above T_R in connection to the temperature T , magnetization along the easy axis m_z , and the fluctuation of the magnetization along the hard axis (hard plane) m_{xy}^2 . We found that this formula is a good description for the overall temperature region, and m_{xy}^2 in the formula is important for the qualitative nature of the FMR frequency. This formula is generally applied to other materials with SR transitions.

This finding of the unusual temperature dependence of the FMR frequency will stimulate time-domain measurement studies on precession motions in SR transitions and spin-state switching such as ultrafast optical control of precession motion for $R\text{FeO}_3$ [5,7] and of spin state for epsilon- Fe_2O_3 [27] and microwave-assisted magnetic recording for the Co/Pd layer, etc. [28,29].

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