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ANALYSIS OF HIGH-FIELD MAGNETIZATION PROCESS IN $Sm_2Fe_{17}N_{3.0}$

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Abstract-The observed high-field magnetization curves of $Sm_2Fe_{17}N_{3.0}$ at 4.2 K and 296 K are well reproduced by the calculation using the Sm-Fe exchange field $2\mu_B H_{ex} = 320$ K and two crystalline electric field parameters $A_0^2 = -910$ K and $A_1^0 = 200$ K. The calculation shows that during the magnetization process along the hard axis at 4.2 K, the Sm moment rotates toward the direction antiparallel to **H** when $H < 110$ kOe and then returns to the field direction with further increase of the field. At 296 K, the Sm moment rotates toward the direction antiparallel to **H** monotonously with increasing field and finally becomes antiparallel to **H** when $H \geq H_A = 210$ kOe. The particular magnetization process of the Sm moment can be explained by the field-induced noncollinear coupling between the spin and orbital moments of the Sm ion.

I. INTRODUCTION

In recent years, the intrinsic magnetic properties of R_2Fe_{17} compounds have been improved considerably by introducing intersititial nitrogen atoms to form $R_2Fe_{17}N_x$ compounds. [1] The nitride $Sm_2Fe_{17}N_x$ exhibits strong uniaxial anisotropy at room temperature with $H_A = 140 \sim 210$ kOe, [2-4] which makes $Sm_2Fe_{17}N_x$ a very promising material for permanent magnet applications. The High-field magnetization curves at 4.2 K and 296 K were measured on the magnetically aligned powder $Sm_2Fe_{17}N_{3.0}$ samples by Kato et al.. [5] In this paper, we shall report the analysis of the magnetization process of the Fe and Sm sublattices in $Sm_2Fe_{17}N_{3.0}$ in terms of the exchange and crystalline electric field (CEF) model. It will be shown that the field-induced noncollinear coupling between the spin and orbital moments of the Sm ion causes a particular magnetization process of the Sm moment when the field is applied along the hard axis.

II. METHOD OF CALCULATION

$Sm_2Fe_{17}N_{3.0}$ crystallizes in the rhombohedral Th_2Zn_{17}-type structure, in which the Sm ions occupy the single 6c site. In the presence of an external magnetic field **H**, the Hamiltonian describing the 4f electrons of the Sm ion in $Sm_2Fe_{17}N_{3.0}$ is given by

$$\mathcal{H} = \lambda \mathbf{L} \cdot \mathbf{S} + \mathcal{H}_{CEF} + 2\mu_B \mathbf{S} \cdot \mathbf{H}_{ex} + \mu_B(\mathbf{L} + 2\mathbf{S}) \cdot \mathbf{H}, \quad (1)$$

where λ is the spin-orbit coupling constant, **L** and **S** are the total orbital and spin angular momenta, $\mathcal{H}_{CEF}$ is the CEF Hamiltonian, and $\mathbf{H}_{ex}$ is the Sm-Fe exchange field.

Since the sixth-order Stevens coefficient γ_J is zero for the ground-state $J = \frac{5}{2}$ multiplet of the Sm ion, the diagonal A_6^0 term in the CEF Hamiltonian, through mixing of the excited multiplets, has a smaller influence on the uniaxial anisotropy than the A_4^0 term. Neglecting the off-diagonal and sixth-order CEF terms, the CEF Hamiltonian of the Sm ion can be simply written as

$$\mathcal{H}_{CEF} = A_2^0 \sum_j \sqrt{\frac{4\pi}{5}} Y_2^0(\theta_j, \varphi_j) + A_4^0 \sum_j \sqrt{\frac{4\pi}{9}} Y_4^0(\theta_j, \varphi_j), \quad (2)$$

where $A_n^0 (n = 2, 4)$ are the CEF parameters in which the 4f radial expectation values $\langle r^n \rangle$ are involved, and the summation j is over all the 4f electrons.

The matrix elements of Eq. (1) are calculated by using the irreducible tensor operator technique. [6] Two first excited multiplets ($J = \frac{7}{2}$ and $J = \frac{9}{2}$) are taken into account in the calculation. For the spin-orbit coupling λ=410 K is used, corresponding to an energy separation of 1435 K between $J = \frac{5}{2}$ and $J = \frac{7}{2}$ multiplests of the trivalent Sm free ion. [7] The eigenvalues E_n and eigenfunctions $|n\rangle [n = 1, 2, \cdots, \sum_J (2J+1) = 24]$ are obtained by diagonalizing the 24×24 matrix of Eq. (1). The free energy for the $Sm_2Fe_{17}N_{3.0}$ system is given by

$$F(T, \mathbf{H}, \mathbf{H}_{ex}) = -2k_B T \ln Z + K_1 \sin^2 \theta_{Fe} - \mathbf{M}_{Fe} \cdot \mathbf{H}, \quad (3)$$

$$Z = \sum_n \exp(-E_n / k_B T), \quad (4)$$

where K_1 and M_{Fe} are the magnetic anisotropy constant and the magnetic moment of the Fe sublattice per formula unit, respectively. $\mathbf{H}_{ex}(T)$ is assumed to be proportional and antiparallel to $\mathbf{M}_{Fe}(T)$. For given temperature T and magnetic field **H**, the equilibrium direction of $\mathbf{M}_{Fe}$ can be determined by minimizing the free energy $F(T, \mathbf{H}, \mathbf{H}_{ex})$ with respect to the angle θ_{Fe}. The magnetic moment of the Sm ion is given by

$$\mathbf{M}_{Sm} = \mathbf{M}_{Sm}^L + \mathbf{M}_{Sm}^S, \quad (5)$$

where $\mathbf{M}_{Sm}^L$ and $\mathbf{M}_{Sm}^S$ represent the orbital and spin moments of the Sm ion, respectively, and can be calculated as

$$\mathbf{M}^{L}_{\mathrm{Sm}} = -\sum_{n} \mu_B \langle n|\mathbf{L}|n\rangle \frac{\exp(-E_n/k_BT)}{Z}, \tag{6}$$

$$\mathbf{M}^{S}_{\mathrm{Sm}} = -\sum_{n} \mu_B \langle n|2\mathbf{S}|n\rangle \frac{\exp(-E_n/k_BT)}{Z}. \tag{7}$$

The total magnetic moment of the system can be obtained by

$$\mathbf{M} = 2\mathbf{M}_{\mathrm{Sm}} + \mathbf{M}_{\mathrm{Fe}}. \tag{8}$$

In the calculation, the reduced temperature dependence of $K_1(T/T_c)/K_1(0)$ and $M_{\mathrm{Fe}}(T/T_c)/M_{\mathrm{Fe}}(0)$ for $Sm_2Fe_{17}N_{3.0}$ is assumed to be identical with those for Y_2Fe_{17}. [8] From the magnetization curves of $Y_2Fe_{17}N_x$ measured parallel and perpendicular to the alignment direction at 4.2 K, the anisotropy field was estimated to be about 40 kOe, which corresponds to $K_1(0) = -53$ K/f.u.. [9] The value for $M_{\mathrm{Fe}}(0)$ is taken to be $39.2\mu_B$/f.u..

III. RESULT AND DISCUSSION

Figure 1 shows the calculated and experimental magnetization curves at 4.2 K and 296 K for $Sm_2Fe_{17}N_{3.0}$. The dashed lines in Fig. 1 represent the calculation that the external field **H** makes an angle of 86° with the *c*-axis, which simulates a situation of incomplete *c*-axis alignment of powder particles. Following set of parameters: $A_2^0 = -910$ K, $A_4^0 = 200$ K, and $2\mu_B H_{\mathrm{ex}}(0) = 320$ K are

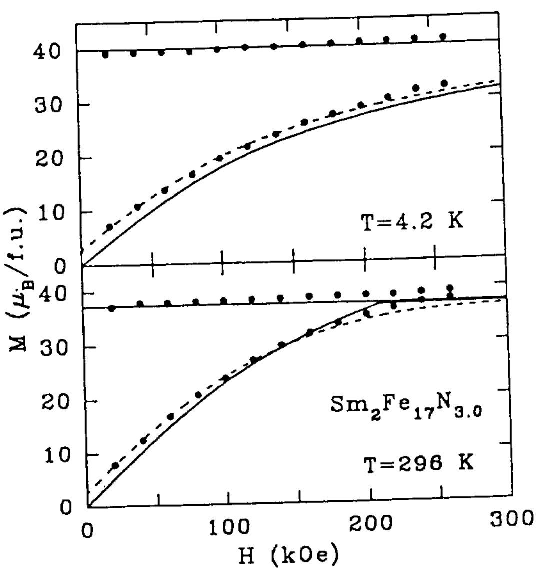

FIG. 1. The calculated and experimental magnetization curves at 4.2 K and 296 K for $Sm_2Fe_{17}N_{3.0}$. the solid and dashed lines represent the calculation with the applied field parallel and perpendicular to the *c* axis and making an angle of 86° with the *c* axis, respectively. Experimental data (full circles) are taken from Ref. 5.

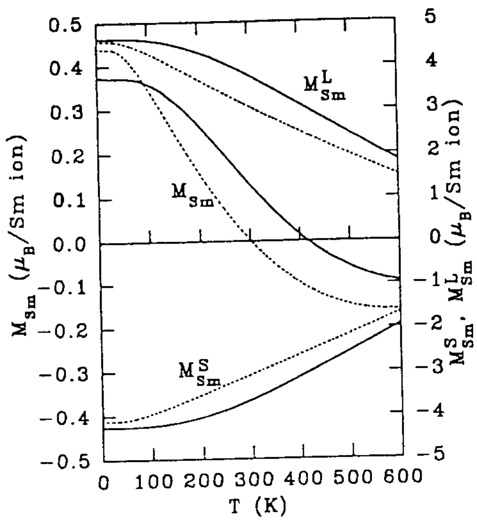

FIG. 2. The calculated M_{Sm}, M^{L}_{Sm} and M^{S}_{Sm} as a function of temperature. The dashed lines represent the calculation not including the CEF interaction.

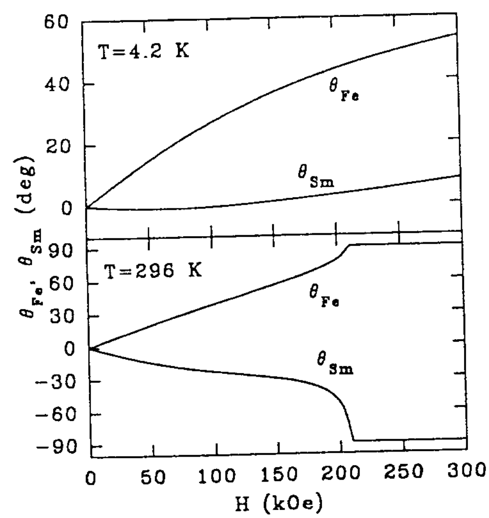

FIG. 3. The calculated field dependence of the angles θ_{Fe} and θ_{Sm} when the field is applied perpendicular to the *c* axis at 4.2 K and 296 K.

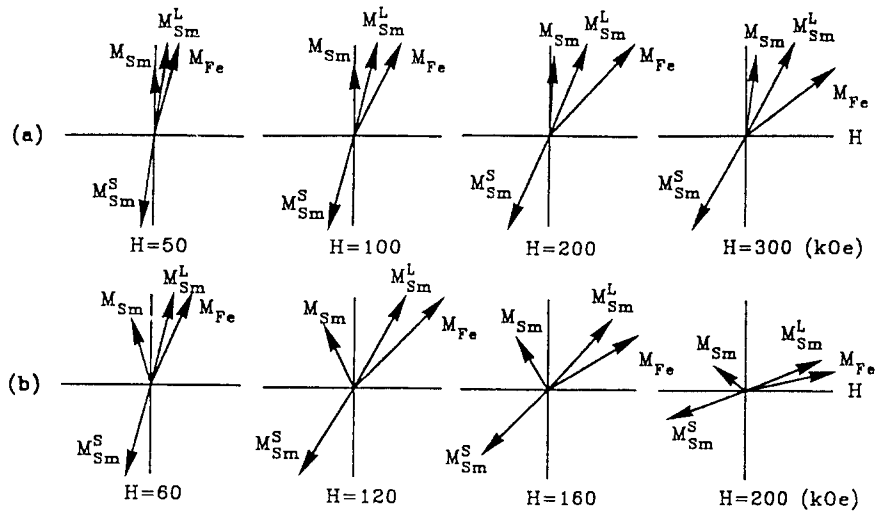

FIG. 4. The magnetic structures of $Sm_2Fe_{17}N_{3.0}$ at different field strengths at (a) T=4.2 K and (b) 296 K.

used in the calculation. It can be seen from Fig.1 that the present calculation reproduce well the experiment. The value of the anisotropy field H_A at 296 K is calculated to be 210 kOe for $Sm_2Fe_{17}N_{3.0}$, which may be compared with H_A = 214 kOe for $Sm_2Fe_{17}N_{2.94}$ determined by the singular point detection technique. [2] The anisotropy field H_A at 4.2 K is estimated to be more than 1300 kOe by the calculation.

The temperature dependence of the Sm magnetic moment and the magnetization process of the Sm and Fe sublattices in $Sm_2Fe_{17}N_{3.0}$ are analyzed by the calculation using the determined CEF and Sm-Fe exchange field parameters. Figure 2 shows the calculated temperature dependence of M_{Sm}, M_{Sm}^L, and M_{Sm}^S at zero magnetic field. The calculated values of M_{Sm} are $0.372\mu_B$ and $0.136\mu_B$ at 4.2 K and 296 K, respectively. The crossover temperature T_∞, at which the total Sm moment because negative, is found to be 418 K. The dashed lines in Fig. 2 represent the calculation that did not take the CEF interaction into account. In this case, the value of T_∞ is reduced to 301 K, indicating that the CEF interaction has a striking influence on the crossover temperature.

Figure 3 shows the calculated magnetic field dependence of the angles θ_{Fe} and θ_{Sm} for $Sm_2Fe_{17}N_{3.0}$ when the field is applied perpendicular to the *c* axis at 4.2 and 296 K. At 4.2 K, $\mathbf{M}_{Fe}$ rotates toward the **H** direction continuously, while $\mathbf{M}_{Sm}$ rotates toward the direction antiparallel to **H** with small θ_{Sm} value when $H < 110$ kOe and then returns to the field direction with further increase of the field. There exists a very pronounced noncollinearity between $\mathbf{M}_{Fe}$ and $\mathbf{M}_{Sm}$. The angle between $\mathbf{M}_{Fe}$ and $\mathbf{M}_{Sm}$ amount to as high as $\Delta\theta = \theta_{Fe} - \theta_{Sm} = 45.7°$ at 300 kOe. At 296 K, $\mathbf{M}_{Sm}$ rotates toward the direction antiparallel to **H** monotonously with increasing field and, finally, becomes antiparallel to **H** when $H \geq H_A = 210$ kOe. The particular magnetization process of $\mathbf{M}_{Sm}$ mentioned above is caused by the field-induced noncollinear coupling between $\mathbf{M}_{Sm}^L$ and $\mathbf{M}_{Sm}^S$. Because of the weak spin-orbit coupling of the Sm ion, a noncollinear coupling between $\mathbf{M}_{Sm}^L$ and $\mathbf{M}_{Sm}^S$ will occur under the strongly combined action of the CEF, the Sm-Fe exchange field and the external field. The maximum angle between $\mathbf{M}_{Sm}^L$ and $-\mathbf{M}_{Sm}^S$ reaches more than 2°. Figures 4(a) and 4(b) illustrate the variation of the magnetic structures of $Sm_2Fe_{17}N_{3.0}$ at 4.2 K and 296 K. At 4.2 K, due to the fact that $\mathbf{M}_{Sm}^L$ is always larger than M_{Sm}^S ($M_{Sm}^L - M_{Sm}^L \sim 0.4\mu_B$), the total Sm moment $\mathbf{M}_{Sm}$ is closer to $\mathbf{M}_{Sm}^L$ than $\mathbf{M}_{Sm}^S$, and is located in the region marked by the c axis and $\mathbf{H}$ when $H > 110$ kOe [see Fig. 4(a)]. At 296 K, however, the value difference between M^L_{Sm} and M^S_{Sm} is small ($|M^L_{\mathrm{Sm}} - M^S_{\mathrm{Sm}}| < 0.14\mu_B$), resulting in a fact that $\mathbf{M}_{\mathrm{Sm}}$ is located in the region marked by the c axis and $-\mathbf{H}$ [see Fig.4(b)]. In addition, the values of M^L_{Sm} and M^S_{Sm} decrease with increasing field. For instance, M^L_{Sm} and M^S_{Sm} vary from $3.625\mu_B$ and $3.512\mu_B$ at 80 kOe to $3.114\mu_B$ and $3.083\mu_B$ at 160 kOe. When H exceeds 178 kOe, M^S_{Sm} becomes larger than M^L_{Sm}, and $\mathbf{M}_{\mathrm{Sm}}$ finally becomes antiparellel to $\mathbf{H}$ when $H \geq H_A$.

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