Research Letter

Molecular Field Calculation of Magnetization on NdRh₂Ge₂ Single Crystal

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Calculation of magnetization of the ternary single crystal compound NdRh₂Ge₂ has been carried out by using the wave-like molecular field model to explain the complex magnetic behavior. The field-induced magnetic structures having the propagation vectors, $\mathbf{Q}_2 = (0, 0, 39/40)$, $\mathbf{Q}_3 = (0, 0, 35/40)$, $\mathbf{Q}_4 = (0, 0, 31/40)$, and $\mathbf{Q}_5 = (0, 0, 0/40)$ (= the field-induced ferromagnetic phase) were proposed. Calculation on the basis of these structures and the antiferromagnetic phase with $\mathbf{Q}_1 = (0, 0, 1)$ well reproduces the experimental magnetization processes and H - T magnetic phase diagram.

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1. INTRODUCTION

Ternary rare earth compounds RM₂X₂ (R = rare earth, M = Rh, Ru, X = Si or Ge) crystallize in the tetragonal ThCr₂Si₂-type structure (I4/mmm) [1, 2]. In the numerous series, the RRh₂Ge₂ is worth studying because of the great diversity of its magnetic properties [3]. Recently, we reported very interesting magnetic behavior of NdRh₂Ge₂ single crystal [4] as follows.

- (i) The compound shows an antiferromagnetic behavior with neel temperature of 50 K.
- (ii) In the temperature dependence of magnetic susceptibility, there is another anomaly at 37 K which indicates a magnetic transition. A magnetic transition is also suggested at 20 K from magnetization measurements.
- (iii) There is a strong uniaxial magnetic anisotropy with the easy c -axis which leads to an Ising-like behavior in the compound.
- (iv) At a low temperature, a four-step metamagnetic process appears along the easy c -axis (see Figure 1).
- (v) The H - T magnetic phase diagram, where there are five magnetic phases, was constructed (see Figure 2).

In the present study, in order to explain this complex magnetic behavior, calculations of the magnetization and an analysis of moment arrangements at various temperatures and under various magnetic fields have been carried out with a wave-like molecular field model [5].

2. WAVE-LIKE MOLECULAR FIELD MODEL

The s-f interaction may be an important interaction in metallic rare earth compounds. Considering this interaction of i th atom as the effective Hamiltonian in terms of the molecular field $\mathbf{H}_m(i)$,

$$\mathbf{H}(i) = -g_J\mu_B\mathbf{J}_i\mathbf{H}_m(i), \quad (1)$$

where $\mathbf{J}_i$ is a good quantum number.

The molecular field acting on an atom on the i th c -plane is given by introducing molecular field coefficient $\lambda(q)$ depended on Fourier q component as

$$\mathbf{H}_m(i) = \sum_q \lambda(q) \langle \mathbf{J}_q \rangle \cos(\pi q i + \varphi_q), \quad (2)$$

where $\mathbf{J}_q$ is the Fourier q component of $\mathbf{J}_i$. These equations are to be solved self-consistently. The details have been reported by Iwata [5].

FIGURE 1: Experimental magnetization versus applied field for NdRh_2Ge_2 at $T = 4.2$ K (bold line) by Himori et al. [4] and calculated magnetization (thin line).

3. RESULTS AND DISCUSSION

3.1. Assumption of the propagation vectors at each magnetic phase

For the sake of calculating the magnetic processes by the wave-like molecular field model, knowledge of magnetic structures is required. Only antiferromagnetic structure has been reported on the NdRh_2Ge_2 compound; it is a simple structure having the propagation vector $\mathbf{Q}_1 = (0, 0, 1)$ and magnetic moments along the c -axis [6]. On the basis of the facts mentioned below, we can, now, propose magnetic structures for the field-induced magnetic phases.

On most of compounds which show an Ising-like multistep metamagnetic process along the c -axis and have the simple antiferromagnetic structure with $\mathbf{Q}_1$, the field-induced phases have propagation vectors $\mathbf{Q}_z = (0, 0, q_z)$ ($z = 1, 2, \dots$), for example, on PrCo_2Si_2 [7], NdCo_2Si_2 [8], and so on. Thus, we assume that the field-induced magnetic phases of NdRh_2Ge_2 compound have $\mathbf{Q}_z$. Wave numbers q_z 's are supposed from the magnetization process as shown in Figure 1. There are five magnetic phases in the metamagnetic process though it is hardly seen in the figure. The magnetization in each phase is $0\mu_B$, $0.06\mu_B$, $0.32\mu_B$, $0.58\mu_B$, and $2.59\mu_B$ ($=$ saturation value M_s), corresponding to $(0/40)M_s$, $(1/40)M_s$, $(5/40)M_s$, $(9/40)M_s$, and $(40/40)M_s$, respectively. This large common denominator suggests a long period structure. Metamagnetic transitions should be responsible for a spin-flip due to the strong uniaxial magnetic anisotropy. The induced $(1/40)M_s$ -phase requires 40 unit cells. Then, we can propose that the field-induced phases have the propagation vectors, $\mathbf{Q}_2 = (0, 0, 39/40)$, $\mathbf{Q}_3 = (0, 0, 35/40)$, $\mathbf{Q}_4 = (0, 0, 31/40)$, $\mathbf{Q}_5 = (0, 0, 0/40)$ ($=$ the field-induced ferromagnetic phase). Of course, the antiferromagnetic phase has $\mathbf{Q}_1 = (0, 0, 1)$.

3.2. Determination of the wave-dependent molecular field coefficients

Molecular-field coefficients $\lambda(\mathbf{Q}_z)$ are estimated by finding the best fit of the calculated values with the experimental data of the magnetic susceptibility, specific heat, and the

FIGURE 2: Molecular field coefficients $\lambda(\mathbf{Q}_z)$ versus propagation vector $(0, 0, q_z)$.

(a) $T = 4.2$ K $\mathbf{Q}_1$ -structure ($q_z = 1$)

(b) $T = 33$ K $\mathbf{Q}_2$ -structure ($q_z = 39/40$)

○ Paramagnetic moment

(c) $T = 40$ K $\mathbf{Q}_3$ -structure ($q_z = 31/40$)

FIGURE 3: (a) Moment arrangements and wave-like molecular fields (solid curves) at 4 K, (b) at 33 K, and (c) at 40 K. The open circles represent paramagnetic Nd.

magnetization process. The values of $\lambda(\mathbf{Q}_z)$ obtained in this study are plotted in Figure 2. The similar figure of this characteristic curve has been seen in PrCo_2Si_2 and NdCo_2Si_2 .

3.3. Moment arrangements in nonexternal magnetic field

The calculated Nd magnetic moment arrangements at various temperatures in nonexternal magnetic field together with the wave-like molecular fields (H_m) are illustrated in Figures 3(a)–3(c). The $\mathbf{Q}_1$ -structure ($=$ phase I) is expressed by only one function with $q_z = 1$ since the magnetic structure is AFI-type [6], whereas the $\mathbf{Q}_2$ -, $\mathbf{Q}_3$ -structure ($=$ phase II, phase III) are expressed by a sum of forty

FIGURE 4: (a) Moment arrangements along the c -axis and total $H_m + H$ (solid curves) at 4 K and under $H = 0$ T, (b) $H = 5.6$ T, (c) $H = 9.6$ T, (d) $H = 10.6$ T, and (e) $H = 13.6$ T applied along the c -axis.

one harmonic functions with $q_z = 0/40, 1/40, \dots, 40/40$ since the magnetic phases are intermediate phases below the induced ferromagnetic phase. Obviously, these moment arrangements have been corresponded with the wave-like molecular fields. It is very interesting results that some paramagnetic Nd ions appear in the Q_2 - and Q_3 -structure. It is caused by the $H_m (= 0)$ around the moments and it is seen in TbRu_2Ge_2 [9].

3.4. Moment arrangements under various applied field

Figures 4(a)–4(e) illustrate the calculated Nd magnetic moment arrangements and the behaviors of the total field ($H + H_m$) at 4.2 K. It is seen that the moments flips together with the change of a total field's sign. The magnitudes of the moments are smaller than $gJ\mu_B$ for all phases because of the CEF effect. It is noticed that no paramagnetic Nd ions appear in this case.

3.5. Magnetization process and magnetic phase diagram

The calculated magnetization process at 4.2 K under applied fields is shown in Figure 1. The magnetizations are $0\mu_B/\text{f.u.}$, $0.068\mu_B/\text{f.u.}$, $0.32\mu_B/\text{f.u.}$, $0.58\mu_B/\text{f.u.}$, and $2.59\mu_B/\text{f.u.}$ at 0 T, 1.4 T, 8.9 T, 10.1 T, and 13.7 T, respectively. These values are in good agreement with the experimental values of $0\mu_B/\text{f.u.}$, $0.06\mu_B/\text{f.u.}$, $0.32\mu_B/\text{f.u.}$, $0.58\mu_B/\text{f.u.}$, and $2.59\mu_B/\text{f.u.}$ It is clear

FIGURE 5: Magnetic phase diagram along the c -axis. The solid curves and open circles with dash line represent the calculation critical fields and the experimental ones (by Himori et al. [4]), respectively.

that the magnetization curve is reproduced by the calculated values fairly well at 4.2 K.

The calculated H - T diagram together with the experimental points obtained from the magnetization measurements are illustrated in Figure 5. It is shown that the calculations reproduce the main feature of the experimental H - T phase diagram.

4. SUMMARY

The interesting magnetic behavior on the NdRh_2Ge_2 single crystal had been reported; successive magnetic phase transitions occur at 20 K, 37 K, and 50 K ($= T_N$). At low temperatures, a four-step metamagnetic process appears. We try to explain this complex magnetic behavior by the wave-like molecular field model. For the sake of calculation, the field-induced magnetic structures having the propagation vectors, $Q_2 = (0, 0, 39/40)$, $Q_3 = (0, 0, 35/40)$, $Q_4 = (0, 0, 31/40)$, and $Q_5 = (0, 0, 0/40)$ (= the field-induced ferromagnetic phase) were proposed. On the basis of these structures and the antiferromagnetic structure reported, calculation of magnetization for various temperatures and fields has been performed. The calculation well reproduces main features of the experimental magnetization processes and H - T magnetic phase diagram. So, we believe that the magnetic structures proposed for the field-induced phases are right. To confirm the magnetic structures proposed, neutron diffraction study is needed.

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