Magnetic and Thermal Properties of SmRh$_2$Zn$_{20}$ Single Crystal

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The magnetization, magnetic susceptibility, and specific heat of the single crystalline sample SmRh$_2$Zn$_{20}$ were measured. The valence of Sm ions in SmRh$_2$Zn$_{20}$ was found to be trivalent. No evidence of valence fluctuations was detected. SmRh$_2$Zn$_{20}$ is an antiferromagnet with $T_N = 2.46$ K. The observed magnetic phase transition temperature in the $C(T, H)$ curves showed that $T_N$ splits into two in the external field $H$ along the [001] and [101] directions. On the other hand, $T_N$ in $H$ along the [111] direction did not split, decreasing to 2.20 K at $H = 7$ T. At 2 K, the magnetization $M_{111}$ in $H$ along the [111] direction increased linearly with increasing field, while $M_{001}$ and $M_{101}$ deviated upward slightly from the linear dependence. We analyzed the observed magnetic and thermal properties of SmRh$_2$Zn$_{20}$ taking into account the crystalline-electric-field effect, the Zeeman energy, and the exchange interaction. The theoretical calculation well reproduced the experimental $\chi(T)$, $M(H)$, $C(T, H)$ and $T_N(H)$, suggesting that the energy scheme of Sm$^{3+}$ is composed of the ground state $\Gamma_7$ and the excited state $\Gamma_8$ with an energy gap of 10.8 K. The sublattice magnetic moments are expected to be along the $\langle 111 \rangle$ direction below $T_N$ at $H = 0$ T. Variations of the magnetic structures induced by the external magnetic fields in a narrow temperature region around $T_N$ are inferred on the basis of theoretical calculations.

1. Introduction

Many Sm-based compounds have been investigated for more than three decades because they show the fundamental physical properties of condensed matters such as valence fluctuation and intermediate valence.\textsuperscript{1,2) Recently, $RT_rX_{20}$-type compounds ($R =$ rare earth, $Tr =$ transition metal, $X =$ Al, Zn, Cd) have attracted much attention because of their various physical properties.\textsuperscript{3–8) They crystallize in the cubic structure, and the rare-earth atoms are in the cubic symmetric sites. Sakai and Nakatsuji, Higashinaka et al., and Yamada et al. presented the interesting experimental results\textsuperscript{9–11) indicating that Sm$Tr_2Al_{20}$ ($Tr =$ Ti, V, Cr, Ta) are an-

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tiferromagnets with strong valence fluctuation, which brought about characteristic behaviors such as a large electronic specific-heat coefficient $C/T$, a weak temperature dependence of magnetic susceptibility, a $-\ln T$-dependent resistivity, and a field-insensitive phase transition. The peculiar valence fluctuation is ascribed to the strong $c$-$f$ hybridization. It is contradictory, however, to the fact that the ground state of Sm ions in all Sm$Tr_2$Al$_{20}$ is said to be a quartet $\Gamma_8$ of trivalent Sm. Kuwai et al. measured the thermoelectric power $S$ of Sm$Tr_2$Al$_{20}$ ($Tr = Ti, V, Cr$) and found the large values of $\Delta S/T$ at temperatures above and near the Néel temperature $T_N$. The large values of $S$ correspond approximately to the large values of $C/T$ since $S$ is proportional to $C$ when both originate from the density of states of the conduction electrons at the Fermi energy.

Sm$Tr_2$Zn$_{20}$ ($Tr = Fe, Co, Ru$) and Sm$Tr_2$Cd$_{20}$ ($Tr = Ni, Pd$) have been investigated by Yazici et al. and Jia et al. Sm$Tr_2$Zn$_{20}$ ($Tr = Fe, Ru$) and SmNi$_2$Cd$_{20}$ exhibit ferromagnetic order, whereas SmPd$_2$Cd$_{20}$ is an antiferromagnet and SmCo$_2$Zn$_{20}$ is nonmagnetic down to 110 mK. The valence of Sm ions in these series is close to trivalent. The ground state of Sm$^{3+}$ due to the crystalline-electric-field (CEF) effect is quartet $\Gamma_8$ for SmRu$_2$Zn$_{20}$ and SmPd$_2$Cd$_{20}$. SmRu$_2$Zn$_{20}$ shows an anomalous magnetic anisotropy of magnetization below the Curie temperature $T_C$, which contradicts the anisotropy predicted from the $\Gamma_8$ ground state. Isikawa et al. suggested the possibility of the octupole-octupole interaction as a mechanism to explain the anomalous magnetic anisotropy. Yazici et al. suggested that SmRu$_2$Zn$_{20}$ is a rare compound of Sm-based heavy-fermion ferromagnet based on the Sommerfeld–Wilson and Kadowaki–Woods ratios.

A few experimental data revealed that SmIr$_2$Zn$_{20}$ and SmRh$_2$Zn$_{20}$ are antiferromagnets with $T_N$ at 1.3 and 2.4 K, respectively. The former has an additional $T_N$ at 1.2 K. The ground state of Sm ions is the $\Gamma_7$ doublet for both compounds, which are rare examples among the Sm$Tr_2$X$_{20}$-type compounds. In SmRh$_2$Zn$_{20}$, the field-induced new phases and the field-induced first-order transition were observed. However, this first-order transition was sample-sensitive.

In this paper, we report the magnetic susceptibility $\chi(T)$, magnetization $M(H)$, and specific heat $C(T, H)$ of SmRh$_2$Zn$_{20}$ to elucidate the fundamental physical properties of the sample. SmRh$_2$Zn$_{20}$ is an antiferromagnet with a Néel temperature $T_N = 2.46$ K. The observed peak in the $C(T)$ curve at $T_N$ is split into two by the external field $H$, depending on the field direction. A phase diagram of $T_N$ vs $H$ is given. The experimental results of $\chi(T)$, $M(H)$, $C(T, H)$, and the field-direction dependence of $T_N$ are well reproduced by the theoretical calculations based on the CEF effect, Zeeman effect, and exchange interaction. The variations
of the magnetic structures in $H$ are discussed on the basis of theoretical calculations.

2. Experimental Procedure

Single crystals of $\text{SmRh}_2\text{Zn}_{20}$ and the reference sample $\text{YRh}_2\text{Zn}_{20}$ were grown by the Zn-self-flux method, which was the same as that described previously.\textsuperscript{21,22} The crystal structure of the cubic $\text{CeCr}_2\text{Al}_{20}$ type was confirmed from the X-ray powder diffraction pattern. There was no trace of impurity phases. The lattice parameters $a$ of $\text{SmRh}_2\text{Zn}_{20}$ and $\text{YRh}_2\text{Zn}_{20}$ were obtained to be 14.226 and 14.200 Å, respectively, which agree with those in the literature.\textsuperscript{23,24} The crystal axis was determined from Laue pictures. The samples were shaped using a spark-cutting machine, and the weights of the samples $\text{SmRh}_2\text{Zn}_{20}$ and $\text{YRh}_2\text{Zn}_{20}$ are 4.46 and 7.67 mg, respectively, which were used for all the measurements. We recognized a sample dependence of the physical properties in $\text{SmRh}_2\text{Zn}_{20}$; thus, we present here the data of the sample, the specific heat $C(T)$ of which shows the sharpest peak at $T_N$ and the highest $T_N$.

The magnetization $M$ and the magnetic susceptibility $\chi$ were measured at temperatures down to 2.0 K using a magnetic property measurement system (MPMS, Quantum Design Inc.). The specific heat was measured at temperatures down to 0.5 K using a physical property measurement system (PPMS, Quantum Design Inc.).

3. Experimental Results

Figure 1 shows the temperature dependence of the magnetic susceptibility of $\text{SmRh}_2\text{Zn}_{20}$ in the field 1 T along the [001] direction. The value of $\chi$ at 300 K suggests that the Sm ions in $\text{SmRh}_2\text{Zn}_{20}$ are in the trivalent state, not in the valence-mixing state.\textsuperscript{1} The magnetism of the trivalent Sm compound is generally expressed by the sum of two components: the Curie term that originated from $J = 5/2$ and the Van Vleck term that originated from the mixing of $J = 5/2$ with $7/2$, that is, $\chi^{3+}(T) = C/T + \chi_{\text{VV}}$, where $C$ is the Curie constant of $J = 5/2$. In addition, the magnetic susceptibility is affected by the exchange interaction as follows: $1/\chi = 1/\chi^{3+} - n$. The solid line in Fig. 1 shows the calculated $\chi(T)$ curve using the two parameters, $\chi_{\text{VV}} = 0.97\times10^{-3}$ emu/mol and $n = -47$ mol/emu. The experimental susceptibility is in good agreement with the calculated one. The inset in Fig. 1 shows the reciprocal susceptibility $1/\chi$ and corrected reciprocal susceptibility $1/(\chi(T) - \chi_{\text{VV}})$ at temperatures below 30 K in the same field as in the main figure. The corrected susceptibility shows a linear dependence of temperature representing the usual Curie–Weiss law. The thick solid line in the inset is the calculated line using $n$. At low temperatures between 3 and 30 K, the experimental data is also in good agreement with the calculated one. The good agreement at temperatures between 3
and 300 K indicates that the CEF effect is small. The paramagnetic Curie temperature $\theta_p$ was deduced to be $-5$ K from the data in Fig. 1, which is related theoretically to the parameter $n$ as $\theta_p = nC$.

![Figure 1](image1.png)

**Fig. 1.** (Color online) Magnetic susceptibility of SmRh$_2$Zn$_{20}$ in the field 1 T along the [001] direction. The solid line is a calculated curve. Inset: reciprocal magnetic susceptibilities $1/\chi$ (closed red circles) and $1/(\chi - \chi_{VV})$ (blue open circles) in low-temperature region. The thick solid line in the inset is a calculated curve. See text for details.

Figure 2 shows the $\chi(T)$ curve in the field 1 T along the [001] direction at low temperatures. The magnetic phase transition $T_N$ is observed at 2.46 K. The temperature dependences of $\chi(T)$ along the [101] and [111] directions were also measured (not shown here) and they were almost the same as that along the [001] direction at temperatures down to 2 K, implying that $\chi(T)$ does not show any field-direction dependence even at temperatures below $T_N$. However, the absence of the field-direction dependence of $\chi(T)$ is limited at temperatures in the vicinity of $T_N$. The inset in Fig. 2 shows the field dependences of magnetization at 2 K in the fields along the [001], [101], and [111] directions. The magnetization along the [111] direction shows an almost linear dependence against the field, but the magnetizations along the [001] and [101] directions gradually deviate upward slightly from the linear dependence at around 2 T. This result does not indicate that the easy direction of magnetization in fields is parallel to the [001] or [101] direction.

Figure 3 shows the temperature dependences of the specific heat $C$ of SmRh$_2$Zn$_{20}$ and YRh$_2$Zn$_{20}$. The sharp peak at $T_N$ is observed at 2.46 K, which is the same temperature as that determined in the $\chi(T)$ curve. The magnetic component $C_{mag}$ of the specific heat is evaluated
as $C_{\text{SmRh}_2\text{Zn}_{20}} - C_{\text{YRh}_2\text{Zn}_{20}}$ and is shown by the black open circles in this figure. The entropy $S$ is numerically calculated by integrating $C/T$ on $T$ and is shown in the inset in Fig. 3. The entropy at $T_N$ is approximately equal to $R \ln 2$, indicating that the ground state of Sm$^{3+}$ is doublet $\Gamma_7$. Moreover, the rapid increase in entropy above $T_N$ with increasing temperature suggests that the excited state $\Gamma_8$ is close to the ground state. The thick solid line indicated by $C_{\text{Schottky}}$ in Fig. 3 shows the calculated $C(T)$ curve of SmRh$_2$Zn$_{20}$, where the energy scheme was assumed to be composed of the doublet ground state and the quartet excited state with a gap $\Delta = 10.8$ K. The solid line indicated by $S_{\text{Schottky}}$ in the inset is the calculated entropy $S(T)$ curve. The calculated two lines are in good agreement with the experimental ones above $T_N$. This narrow energy gap indicates that the CEF effect is weak, which is consistent with the consideration of the temperature dependence of $\chi(T)$ above $T_N$. $\chi(T)$ was mostly understood on the basis of the Curie–Weiss law of Sm$^{3+}$ without the CEF effect.

Figures 4(a)–4(c) show the temperature dependences of the specific heat of SmRh$_2$Zn$_{20}$ in magnetic fields along the [001], [101], and [111] directions. Interestingly, in the fields along the [001] and [101] directions, $T_N$ splits into two and the width of splitting increases with increasing field. The upper $T_N$ does not change with increasing field, whereas the lower one decreases with increasing field. In the field along the [111] direction, $T_N$ gradually decreases without splitting. Figure 4(d) shows the field-direction dependence of $T_N$. The splitting widths of $T_N$ at 7 T are 0.26 K in $H // [001]$ and 0.14 K in $H // [101]$. These of $T_N$ changes appear in the narrow temperature range between 2.16 and 2.46 K.

![Figure 2](image-url)
Fig. 3. (Color online) Temperature dependences of the specific heat $C$ of SmRh$_2$Zn$_{20}$ (red closed circles) and YRh$_2$Zn$_{20}$ (blue open squares). $C_{\text{mag}}$ denotes the magnetic part of $C$ (black open circles) evaluated as $C_{\text{SmRh}_2Zn_{20}} - C_{\text{YRh}_2Zn_{20}}$. The thick solid line denoted by $C_{\text{Schottky}}$ is a calculated curve of the Schottky-type specific heat. Inset: temperature dependence of the entropy of SmRh$_2$Zn$_{20}$. The solid line denoted by $S_{\text{Schottky}}$ is a calculated curve. See text for details.

4. Analysis and Discussion

4.1 Hamiltonian

We discuss the small magnetic anisotropy observed in the magnetization curves at 2 K (inset in Fig. 2), the field-direction dependence of the specific heat $C(T)$, and the complex behaviors of $T_N$ in fields (Fig. 4). Generally, as a consequence of the characteristics of $\Gamma_7$ and $\Gamma_8$ of Sm$^{3+}$ in the cubic symmetry, the $\Gamma_7$ state is magnetically isotropic, whereas the $\Gamma_8$ state is anisotropic in fields. Thus, it is expected that this magnetic anisotropy will be caused by the mixing of the $\Gamma_8$ state with the ground state $\Gamma_7$ through the external magnetic field and the exchange field. We analyze this magnetic anisotropy in the frame of the molecular-field approximation. For simplicity, a simple antiferromagnetic structure with two sublattices (A and B) is assumed, that is, the up- and down-magnetic moments are fixed in the A and B sublattices, respectively. We define the following Hamiltonian to analyze the magnetic and thermal properties of Sm$^{3+}$ in SmRh$_2$Zn$_{20}$,

$$\mathcal{H}_i = A_4(O_4^i + 5O_4^i) + g\mu_BJ_iH_{\text{ext}} + g\mu_BJ_iH_{\text{mol}}(i),$$

where $i$ denotes the sublattices (A and B). The first term in this equation is the CEF energy, where the six-order term in the CEF energy is excluded because the quantum number $J$ is 5/2. The mixing effect with the upper multiplet of $J = 7/2$ will be taken into account by adding $\chi^{VVV}$ to $\chi$ calculated using Eq. (1). The second term in Eq. (1) is the Zeeman energy due to the external magnetic field $H_{\text{ext}}$, and the third term is the exchange energy expressed in

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Fig. 4. (Color online) Temperature dependences of the specific heat $C$ of $\text{SmRh}_2\text{Zn}_{20}$ in magnetic fields along the [001] (a), [101] (b), and [111] (c) directions. (d) Magnetic field dependences of $T_N$ in the fields along the [001] (filled and open diamonds), [101] (filled and open squares), and [111] (filled circles) directions. See text for details.

the molecular-field approximation. The molecular field $H_{\text{mol}}(i)$ at the $i$ sublattice is expressed using the average magnetic moment in the other sublattice $j$ as $n_{BA}(g_J\mu_B)\langle J_j \rangle$, where $\langle \cdots \rangle$ is the thermal average and $n_{BA}$ is the exchange coupling parameter between the atoms in the A and B sublattices. The other parameters used in Eq. (1) are conventionally defined.\textsuperscript{25,26}

The average magnetization at each sublattice is calculated as

$$M_i = -g_J\mu_B\langle J_i \rangle = -g_J\mu_B \frac{\text{Tr}\, J_i\, \exp(-\beta H_i)}{\text{Tr}\, \exp(-\beta H_i)},$$

(2)

where $\beta = 1/k_B T$. The average magnetization $M$ per atom is $M = (M_A + M_B)/2$. Note that the physical quantities of $J_i$, $H_{\text{mol}}$, and $H_{\text{ext}}$ are three-dimensional vectors. Thus, Eq. (2) is a set of six equations, which represent the thermal averages of the $x$-, $y$-, and $z$-components of $M_A$ and $M_B$. Equation (2) is solved by the iteration method. By using both values of $\langle J_A \rangle$ and $\langle J_B \rangle$, which have been obtained using Eq. (2), the specific heat $C$ per mole is evaluated as

$$C = \left(\frac{N_A}{2}\right) \frac{\partial}{\partial T} \left(\langle H_A \rangle + \langle H_B \rangle - \frac{1}{2}\langle H_{\text{exch}} \rangle\right),$$

(3)

where $N_A$ is Avogadro’s number, and the third term in parentheses is used to correct the double counting of the exchange energy between the Sm atoms at A and B sublattices. $H_{\text{exch}}$
in this equation is the same as the third term in Eq. (1) and is expressed as \( n_{BA}(g_J\mu_B)^2J_AJ_B \).

We have only two fitting parameters, i.e., \( A_4 \) and \( n_{BA} \), when we numerically calculate \( M(T, H) \) and \( C(T, H) \) using the above Hamiltonians, where \( H \) is the magnitude of \( H_{\text{ext}} \). \( A_4 \) is proportional to \( \Delta \) as \( A_4 = \Delta/360 \) in the unit [K], and \( n_{BA} \) is physically the same as \( n \) [mol/emu] through the relationship of \( n_{BA} = nN_A \). As already mentioned in Sect. 3, the energy scheme of \( \text{Sm}^{3+} \) is composed of the doublet ground state \( \Gamma_7 \) and the quartet excited state \( \Gamma_8 \) with \( \Delta = 10.8 \) K. Thus, \( A_4 \) is set to be 0.030 [K]. The value of \( n_{BA} \) was accurately determined to be \(-1.45 k_B/(g_J \mu_B)^2\) by adjusting the calculated \( T_N \) to the experimental \( T_N \). Consequently, the sublattice magnetic moments \( M_A \) and \( M_B \) are evaluated as a result of the iteration process of Eq. (2) using the two parameters \( A_4 \) and \( n_{BA} \). This means that the magnetic alignments of each sublattice moment in \( H \) are inferred. First, we present the calculated results of \( \chi(T) \), \( M(H) \), \( C(T, H) \), and \( T_N(H) \) in comparison with the respective experimental data in the next subsection. After that, we discuss the magnetic alignments of sublattice magnetic moments in \( H \).

We have to note one important result concluded from this CEF calculation. At temperatures below \( T_N \), the magnetic moments at the sublattices are along the \( \langle 111 \rangle \) direction at \( H = 0 \), namely, there exist four magnetic domains below \( T_N \). For simplicity, we have calculated the \( \chi(T) \), \( M(H) \), and \( C(T) \) curves in fields for a single domain, in which the up- and down-magnetic moments are along the [111] and [\bar{1} \bar{1} \bar{1}] directions, respectively.

4.2 Calculation and comparison with the experimental data

Figure 5 shows the calculated temperature dependences of the magnetic susceptibility \( \chi \) of \( \text{SmRh}_2\text{Zn}_{20} \) in the field 1 T along the [001], [101], and [111] directions, where \( \chi \) has been shifted by the amount of \( \chi_{\text{VV}} \) because the mixing effect with the multiplet of \( J = 7/2 \) is not taken into account in Eq. (2). In the temperature range between 2 K and \( T_N \), the magnetic anisotropy is very small, which is consistent with the experimental results. Below 2 K, however, the calculated \( \chi \) shows a magnetic anisotropy, although we cannot compare it with the experimental data, which is limited above 2 K.

The inset in Fig. 5 shows the calculated magnetization curves at 2 K in the fields along the [001], [101], and [111] directions. \( M_{111}(H) \) in \( H // [111] \) increases linearly with increasing field without any anomalies such as spin fops, which is in good agreement with the experimental behavior. On the other hand, the \( M_{101}(H) \) curve deviates slightly from the \( M_{111}(H) \) curve around 3 T, and the \( M_{001}(H) \) curve deviates further from the \( M_{111}(H) \) curve. The characteristic behaviors of the three calculated \( M(H) \) curves at 2 K are in good agreement with
Fig. 5. (Color online) Temperature dependences of the calculated magnetic susceptibility of SmRh$_2$Zn$_{20}$ in the field 1 T along the [001], [101], and [111] directions. Inset: calculated magnetization curves at 2 K along the three directions. See text for details.

Fig. 6. (Color online) Temperature dependences of the calculated specific heat of SmRh$_2$Zn$_{20}$ in the fields along the [001] (a), [101] (b), and [111] (c) directions. (d) Calculated field dependences of $T_N$. See text for details.

Figures 6(a)–6(c) show the calculated specific heat $C$ in the fields of 0, 2, 4, 6, and 7 T along the [001], [101], and [111] directions. In the case of $H // [001]$ and [101], $T_N$ splits into two, and the width of the splitting increases with increasing field. In $H // [111]$, $T_N$ does not
split but moderately decreases with increasing $H$. In Fig. 6(d), the external-field dependences of $T_N$ are plotted. The splitting widths of $T_N$ at 7 T are 0.7 K in $H // [001]$, 0.3 K in $H // [101]$, and zero in $H // [111]$. These characteristic behaviors are also in good agreement with the experimental ones. However, note that the height of the calculated specific heat at $T_N$ at 0 T is one-third compared with that of the experimental specific heat at $T_N$, and the second-peak height separated by $H$ is also lowered compared with the experimental one. This is caused by the fact that the numerical calculation is based on the molecular-field approximation. The magnetic fluctuation $\langle (J_A - \langle J_A \rangle) (J_B - \langle J_B \rangle) \rangle$, which is more important in the vicinity of $T_N$ than at temperatures far from $T_N$, cannot be taken into account in this approximation. In fact, at 2.0 and 2.8 K, which are far below and far above $T_N$, respectively, the values of the calculated $C$ are approximately equal to those of the experimental $C$.

It is concluded that the theoretical calculations have successfully reproduced the macroscopic experimental data, $\chi(T)$, $M(T, H)$, $C(T, H)$, and $T_N(H)$, by using only two parameters, $A_4$ and $n_{BA}$.

### 4.3 Alignment of sublattice magnetic moments

In this subsection, we present the microscopic magnetic structures of SmRh$_2$Zn$_{20}$ in the vicinity of $T_N$, which have been deduced from the model calculations. Here, we discuss the single magnetic domain.

Figures 7(a)–7(c) show the temperature dependences of the calculated $x$-, $y$-, and $z$-components of the respective sublattice magnetic moments of SmRh$_2$Zn$_{20}$ in the field 4 T. The subscripts 1 and 2 denote the up- and down-sublattice moments, respectively. In $H$ along the [001] direction, as seen in Fig. 7(a), the lower $T_N$ is found to be the temperature at which the antiferromagnetic alignment along the [001] direction ($z$-component) disappears, and the upper $T_N$ is the temperature at which the antiferromagnetic alignment of both the $x$- and $y$-components disappears. In $H$ along the [101] direction [Fig. 7(b)], the lower $T_N$ is the temperature at which the antiferromagnetic alignment along the [101] direction disappears, and the upper $T_N$ is the temperature at which the antiferromagnetic alignment of the $y$-component disappears. In $H$ along the [111] direction [Fig. 7(c)], the magnetizations $M_A$ and $M_B$ are parallel to the [111] direction, and the antiferromagnetic alignment disappears at $T_N$. Thus, the magnetic susceptibility along the [111] direction can be considered a parallel magnetic susceptibility. In the cases of $H$ along the [001] and [101] directions, the antiferromagnetic component along the $H$ directions disappears at temperatures between the lower and upper $T_N$’s. Thus, the magnetic susceptibility along these directions can be considered a perpen-
dicular magnetic susceptibility. The lower and upper $T_N$’s as well as the $T_N$ along the [111] direction are second-order phase-transition temperatures.

Figures 8(a)–8(c) show the field dependences of the calculated $x$-, $y$-, and $z$-components of the respective sublattice magnetic moments of SmRh$_2$Zn$_{20}$ at 2.3 K. In the cases of $H$ along the [001] and [101] directions, as seen in Figs. 8(a) and 8(b), the antiferromagnetic alignments along the $H$ directions disappear near 4 and 5 T, respectively. At these points, any anomalous discontinuity such as spin flopping is not theoretically concluded. On the other hand, the antiferromagnetic alignments perpendicular to the $H$ directions are robust against $H$ as seen in the figures. In $H$ along the [111] direction [Fig. 8(c)], the magnetic moments $M_A$ and $M_B$ are parallel to the $H$ direction below 6 T. Thus, the magnetization along this direction is due to the parallel susceptibility, as mentioned previously.

The susceptibility $\chi_{111}$ in $H$ along the [111] direction has been considered the parallel susceptibility $\chi_{//}$. However, as seen in Fig. 5, the calculated $\chi_{111}(T)$ does not approach zero at 0 K. This behavior is contrary to the common behaviors of usual antiferromagnets. We briefly comment on the discrepancy between the calculated $\chi_{//}(T)$ and the $\chi_{//}(T)$ for usual antiferromagnets, although the experimental $\chi(T)$ is limited above 2 K. In the case of usual antiferromagnets, the sublattice moments $M_A$ and $M_B$ are saturated at 0 K, and they do not change even if $H$ is applied; thus, $\chi_{//}$ is equal to zero. However, when $\Delta \neq 0$, the sublattice moments $M_A$ and $M_B$ are not saturated at 0 K, that is, the moment $M_A$ increases and $M_B$ decreases with the application of the external field via a Van Vleck-type mechanism between $\Gamma_7$ and $\Gamma_8$ states in Sm$^{3+}$; thus, $\chi_{//}$ is not equal to zero.

![Fig. 7.](image-url) (Color online) Temperature dependences of the calculated $x$-, $y$-, and $z$-components of respective sublattice magnetic moments of SmRh$_2$Zn$_{20}$ in the field 4 T along the [001] (a), [101] (b), and [111] (c) directions. The subscripts 1 and 2 denote the up- and down-sublattice moments, respectively.
The microscopic magnetic structures deduced from the theoretical calculations have provided an insight into SmRh$_2$Zn$_{20}$. To confirm the magnetic structures of SmRh$_2$Zn$_{20}$, neutron scattering experiments should be performed.

4.4 Conclusions

We prepared single crystals of SmRh$_2$Zn$_{20}$ and measured $\chi(T)$, $M(T,H)$, and $C(T,H)$. Sm ions in SmRh$_2$Zn$_{20}$ are in the trivalent state, not in the valence fluctuating state. SmRh$_2$Zn$_{20}$ is an antiferromagnet with $T_N = 2.46$ K. $T_N$ splits into two in $H // [001]$ and [101]. The upper $T_N$ does not change in fields up to the maximum $H$ of 7 T, whereas the lower $T_N$ decreases. In $H // [111]$, $T_N$ does not split and decreases to 2.20 K at 7 T. At 2 K, the magnetization $M_{111}$ along the [111] direction increases linearly with increasing field, whereas $M_{001}$ and $M_{101}$ deviate upward slightly from the linear dependence of $M_{111}$. We analyzed these magnetic and thermal properties of SmRh$_2$Zn$_{20}$ taking into account the CEF effect, Zeeman energy, and exchange interaction. The theoretical calculations have successfully reproduced the experimental $\chi(T)$, $M(H)$, $C(T,H)$, and $T_N(H)$. The energy scheme of Sm$^{3+}$ obtained is composed of the ground state $\Gamma_7$ and the excited state $\Gamma_8$ with the energy gap of 10.8 K. The sublattice magnetic moments are along the $\langle 111 \rangle$ direction below $T_N$ at $H = 0$ T. The magnetic structures in magnetic fields in the temperature region between the split $T_N$’s could be inferred on the basis of the theoretical calculations.

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27) In the notation of Lea et al.,26) $A_4$ is equal to $Wx/60$. 