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Magnetic properties of DyB$_2$C, HoB$_2$C, and ErB$_2$C

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Magnetic properties of layered borocarbides RB$_2$C (R= Dy, Ho, and Er) have been studied by the magnetization and specific heat measurements at 1.8 – 300 K under the field up to 5 T. ErB$_2$C has a basic two-sublattice antiferromagnetic order below $T_N$ = 16.3 K. HoB$_2$C and DyB$_2$C show the three-dimensional magnetic order below $T_C$ = 7.0 K and 8.5 K, respectively. However, they have still large magnetic fluctuations below $T_C$. The magnetic ordering temperatures of RB$_2$C (R= Dy, Ho, and Er) do not follow the de Gennes relation, where the ordering temperatures of HoB$_2$C and DyB$_2$C are suppressed. The suppressions of ordering temperature and the unconventional fluctuating ground states of HoB$_2$C and DyB$_2$C originate from the spin frustration effects. The instability of the unconventional magnetic phase associated with the frustration is significant in this series; the fluctuating ground state results from the complex spin–spin interactions or spin–quadrupole interactions.

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I. INTRODUCTION

Layered metal borocarbides are of interest due to their low temperature electronic and magnetic properties. In recent years, it was discovered that DyB$_2$C$_2$ and HoB$_2$C$_2$ have an antiferroquadrupolar ordering at $T_Q$ = 24.7 K (Refs. 1, 2) and 5.0 K, respectively. YB$_2$C-type structure described below is found for R = Sc, Y and Tb–Lu. 4–6 The YB$_2$C-type structure consists of alternating layers of rare earth metals and covalently bonded boron–carbon (B–C) sheets as shown in Fig. 1. The metal sheets are made of fused squares and triangles. The arrangement of the rare earth ions in RB$_2$C is topologically equivalent to the Shastry-Sutherland lattice. 7 The B–C sheets are formed by fused four- and seven-membered rings. Metal cations are situated below and above the centers of the seven-membered B–C rings. It was originally reported that successive B–C layers were rotated by 90° giving a tetragonal $P_{4}2/mbc$ structure. 4–6 However, a recent powder x-ray diffraction study of HoB$_2$C (Ref. 8) and a powder neutron diffraction study of ErB$_2$C have shown the possibility of a different structure model, where the B–C layers are stacked directly above each other leading to an orthorhombic $Pbam$ structure. The geometry of this B–C network can be derived from the tetragonal rare earth tetraboride RB$_4$ arrangement. A boron network of RB$_4$ consists of fused four- and seven-membered rings in the (001) plane. By removing the two out-of-plane boron atoms at the apices of the B$_8$ octahedral cluster, i.e., those that convert B$_4$ squares into B$_8$ octahedra, we obtain a hypothetical RB$_3$ stratified-structure. Substitution of the carbon atoms for the two of four boron atoms in the four-membered rings then gives the two-dimensional B$_2$C network system.

Recently, the low temperature neutron powder diffraction of DyB$_2$C, HoB$_2$C, and ErB$_2$C was investigated. 8 ErB$_2$C has two-sublattice antiferromagnetic order below 16.3 K. DyB$_2$C and HoB$_2$C show an unusual scattering in low wave number ($Q$) region. In HoB$_2$C, three peaks appear in low-$Q$ range

FIG. 1. Crystal structures of RB$_2$C (R= Dy, Ho, and Er) in a [001] projection.
between 8 and 16 K. Sharp peaks appear at 15.7 K and at 10.5 K, and a broad, asymmetric peak occur at 8 K. Those peaks are not typical spin wave or other magnetic superstructures, but can be explained by the formation of a Warren–type magnetic random layer (MRL) lattice. A conventional magnetic transition occurs at 7 K, although the unconventional correlations indicated by the low-\(Q\) peaks are maintained below this transition. The low temperature behavior of DyB\(_2\)C is very similar to that of HoB\(_2\)C. The origins of the unconventional properties are still unclear. Other physical properties of R\(_B\)\(_2\)C have not been reported. In this paper, we report the magnetic behaviors of DyB\(_2\)C, HoB\(_2\)C, and ErB\(_2\)C studied by magnetization and specific heat measurements. The magnetic anisotropy of HoB\(_2\)C was examined using a single crystal.

II. EXPERIMENT

Polycrystalline samples of R\(_B\)\(_2\)C (R=Dy, Ho, and Er) were prepared by an arc-melting method. Mixtures of powders of the starting materials, which were elemental rare earth metals (99.9% pure in powder), amorphous boron (99% pure, 1 micron powder size) and graphite (99% pure, 1–2 micron powder size), were compacted in stainless steel dies without the use of binders or lubricants. The 1 g pellets were arc-melted on a water-cooled copper hearth in an argon atmosphere. To obtain homogeneous samples, each melted die was turned over and remelted several times. The x-ray diffraction data were obtained on powdered samples with Cu-K\(_\alpha\) radiation, and confirm that the samples were single phase. A single crystal of HoB\(_2\)C was prepared by the Czochralski method in an argon atmosphere using a tetra-arc furnace.

The magnetizations of R\(_B\)\(_2\)C were measured using a SQUID magnetometer (MPMS, Quantum Design Co.) in the temperature range of 1.8–300 K and under magnetic fields up to 5 T. The magnetic susceptibility measurements were performed after zero-field-cooled (ZFC) and after field-cooled (FC) conditions. For the single crystal of HoB\(_2\)C, the magnetization measurements, where the magnetic field \(H\) is applied parallel (\(H\parallel c\)) and perpendicular (\(H \perp c\)) to the \(c\)-axis, were carried out between 1.8 K and 300 K, and at several temperatures under magnetic fields up to 5 T. Specific heat measurements were carried out by a heat-relaxation method down to 1.9 K (using PPMS, Quantum Design Co.) and by a homemade light-irradiation AC method. The absolute value of the specific heat was obtained by the heat-relaxation method. The relative accuracy of the AC measurement is exceedingly high compared with that of the adiabatic or the heat-relaxation measurement, although the absolute value of specific heat cannot be estimated.

III. RESULTS AND DISCUSSION

A. ErB\(_2\)C

The temperature dependence of the magnetic susceptibility \(\chi\) for a powder sample of ErB\(_2\)C measured in FC and ZFC conditions is shown in Fig. 2(a). The \(\chi\) increases with decreasing temperature, and then passes through a peak at 16.7 K, and succeedingly it shows a minimum at 5.0 K. The peak of \(\chi\) is attributed to the antiferromagnetic transition. The coincidence of the \(\chi\) measured in FC and ZFC conditions suggests that no spontaneous magnetization appears in ErB\(_2\)C below \(T_N\). This is consistent with the recent powder neutron diffraction study of ErB\(_2\)C, which found a collinear antiferromagnetic arrangement of the moments along the \(c\) axis.\(^6\) However, the origin of the minimum of \(\chi\) at 5.0 K is not clear.

The inverse molar susceptibility \(\chi^{-1}\) [inset in Fig. 2(a)] shows Curie–Weiss behavior above 130 K. The effective magnetic moment (\(\mu_{\text{eff}}\)) is estimated at 9.28 \(\mu_B\). Since this value is close to that expected for a free Er\(^{3+}\) ion value, 9.59 \(\mu_B\), the 4\(f\) electrons in ErB\(_2\)C are well localized. The Weiss temperature is \(\theta_w = 38.6\) K. A nonlinear temperature dependence of \(\chi^{-1}\) at low temperatures just above \(T_N\) may be due to crystal field effects.

The magnetization curves of the powder sample of ErB\(_2\)C (Fig. 3) were measured at 1.8 K, 5.0 K, 10.0 K, and 15.0 K under fields up to 5 T. The magnetization curve at 1.8 K shows a metamagnetic transition at the critical field of \(H_{\text{C}} = 1.5\) T. At higher temperatures, the transitions become smoother. The one-stepped field induced transition indicates that the ErB\(_2\)C has a basic two-sublattice antiferromagnetic structure, and a strong Ising-type anisotropy. The saturation magnetization of 7.4 \(\mu_B/\text{Er}\) is less than a free Er\(^{3+}\) ion value \(gJ = 9 \mu_B/\text{Er}\), where \(g\) is the Landé \(g\) factor and \(J\) is the total momentum of the \(f\) electron configuration.

The specific heat of the polycrystalline sample of ErB\(_2\)C shows a \(\lambda\)-like anomaly at \(T_N = 16.3\) K [Fig. 2(b)]. In addi-
tion, a small anomaly in the specific heat data of ErB$_2$C is visible at 3.3 K, which corresponds to the minimum of the susceptibility. This may be due to a reorientation of the magnetic moment configuration. The temperature dependence of the magnetic specific heat divided by temperature $C_{\text{mag}}/T$ measured by a heat-relaxation method is shown in Fig. 2.

Subtracting the phonon component from the total specific heat, we obtain the temperature dependence of the magnetic specific heat. The phonon contribution was obtained from a measurement of the isostructural and nonmagnetic compound LuB$_2$C. We obtained the temperature dependence of the total entropy $S$ by numerically integrating the data of $C_{\text{mag}}/T$ vs $T$. Since our measurements are made above 1.9 K, only $\Delta S = S(T) - S(1.9 \text{ K})$ can be obtained in this study. Therefore, we tentatively plot $S$ putting $S(1.9 \text{ K}) = 0$. The total entropy of approximately $R \ln 2$ is released around $T_N = 16.3 \text{ K}$ showing that Er here has a Kramers doublet ground state. The magnetic specific heat of ErB$_2$C shows a Schottky anomaly around 40 K, which is due to the crystalline electric field (CEF) splitting of the $J = 15/2$ multiplet of Er$^{3+}$.

B. HoB$_2$C

Figure 4(a) shows the magnetic susceptibility $M/H$ for a single crystal of HoB$_2$C. The susceptibilities measured in the ZFC condition show a maximum at 7.0 K, while that in the FC condition shows weak increase below this temperature. The divergence of the FC and ZFC data may be ascribed to the appearance of weak spontaneous magnetization in HoB$_2$C. However, magnetization anomalies are not evident in the 8 –16 K region, in which the unusual low-$Q$ peaks are observed by the neutron diffraction.\textsuperscript{9}

$\chi^{-1}$ of HoB$_2$C [Fig. 4(b)] follows the Curie–Weiss law above 40 K. The effective paramagnetic moments and the Weiss temperatures are estimated at $\mu_{\text{eff}}^j = 10.64 \mu_B$ and $\theta_p^j = -0.58 \text{ K}$. For $H \perp c$, and $\mu_{\text{eff}}^j = 10.05 \mu_B$ and $\theta_p^j = -4.75 \text{ K}$. For $H \parallel c$, respectively. The effective magnetic moment is close to the calculated value of 10.58 $\mu_B$ for a free Ho$^{3+}$ ion. The negative values of $\theta_p$ indicate the existence of antiferromagnetic interactions. However, the small absolute values of $\theta_p$, which are much less than the value of the magnetic ordering temperature, suggest the presence of competing interactions. The competition may originate from geometric spin frustration and may cause the unconventional properties observed in the neutron diffraction measurement of HoB$_2$C.\textsuperscript{9}

Figure 5 shows the magnetization curves of the single crystal of HoB$_2$C at 1.8 K under a magnetic field up to 5 T, applied parallel and perpendicular to the c-axis. Below 1.5 T, the magnetization for $H \parallel c$ was larger than the magnetization for $H \perp c$. The anisotropic behaviors of the susceptibilities and the magnetization curves in the magnetic ordered phase indicate that the ordered magnetic moments favor to point within the (001) plane.

The spontaneous magnetization extrapolated to the zero field is estimated to be 4.7 $\mu_B$/Ho for $H \parallel c$ and 4.4 $\mu_B$/Ho for $H \perp c$. These values are less than half of the theoretical
value of $gJ = 10 \mu_B$/Ho. The residual magnetizations for $H \perp c$ and $H \parallel c$ are 0.28 $\mu_B$/Ho and 0.07 $\mu_B$/Ho, respectively. The small values of the spontaneous magnetizations and the residual magnetizations suggest that HoB$_2$C is not an ordinary ferromagnet. The magnetic ordered state below 7.0 K is a canted antiferromagnetic one with a weak ferromagnetic component.

The specific heat of HoB$_2$C is shown in Fig. 6(a). A broad peak was observed at $T_C = 7.0$ K, showing that HoB$_2$C has still large magnetic fluctuations in spite of the formation of three-dimensional magnetic order below $T_C$, since the shape of the peak is rather broader than a $\lambda$-type one usually observed in an ordinary magnet. This is consistent with the fact that the ordered magnetic moment in the conventional magnetic ordered phase is only 2.9 $\mu_B$/Ho, which is about only a 29% of 10 $\mu_B$/Ho is expected from $J = 8$. However, no anomalies in the specific heat or the susceptibilities were observed between 8 and 16 K. Moreover, the magnetic specific heat shows a tail in high temperature region, and the magnetic entropy is still released even far above $T_C = 7.0$ K, as shown in Fig. 6(b).

In order to check for the existence of anomalies in the susceptibility and the specific heat between 8 and 16 K more precisely, AC measurements of the susceptibility and the specific heat was performed. Anomalies of the AC susceptibilities [Fig. 7(a)] and the AC specific heat [Fig. 7(b)] were not observed between 8 and 16 K by these measurements. The noncritical behavior of the specific heat between 8 and 16 K implies that the transition is not long-range order (LRO). In contrast, the temperature dependences of the integrated intensity of (001)$_m$ and (002)$_m$ neutron reflections [Fig. 7(c)] show the critical behavior between 8 and 16 K, respectively. Furthermore, the reflections widths of the (001)$_m$ and (002)$_m$ (the subscript $m$ refers to the Warren-type MRL lattice) between 8 and 16 K indicate that the correlation length in the $c$ direction is $>1000$ Å. The result of the neutron diffraction study is contradictory to the result of the specific heat and the magnetization study. It is evident that the magnetic transition between 8 and 16 K are unconventional. Presumably, these transitions are invisible in the specific heat and the susceptibility since the entropy changes are small, and the anomalies may be broadened due to the nature of the MRL lattice.

C. DyB$_2$C

Figure 8 shows the magnetic susceptibility $M/H$ for a powder sample of DyB$_2$C; these behaviors are similar to that of HoB$_2$C. The susceptibility measured in the ZFC condition
shows a maximum at 9.2 K, while that in the FC condition shows weak increase below this temperature. It is possible that the divergence of the FC and ZFC data below 15 K illustrates the appearance of spontaneous magnetization of DyB$_2$C. However, no anomaly of the susceptibility around 22 K was observed.

$H/M$ of DyB$_2$C (inset in Fig. 8) follows the Curie–Weiss law above 40 K, and the effective paramagnetic moment and Weiss temperature of DyB$_2$C are $\mu_{\text{eff}} = 10.63 \mu_B$ (which is identical to the value of a free Dy$^{3+}$ ion) and $\theta_W = 0.1$ K, respectively.

The magnetization curve of the powder sample of DyB$_2$C (Fig. 9) at 1.8 K under the field up to 5 T shows a ferromagnetic behavior. However, the spontaneous magnetization of 5.1 $\mu_B$/Dy is only half of the theoretical value of $gJ = 10$ $\mu_B$/Dy. It seems likely that at least one more metamagnetic transition occurs in high field regions above 5 T. The small values of the spontaneous magnetizations and the residual magnetizations suggest that DyB$_2$C is not an ordinary ferromagnet.

The specific heat of DyB$_2$C is shown in Fig. 10(a). A broad peak was observed at $T_C = 8.5$ K and a very small hump around 20.5 K. The broad peak corresponds to the magnetic transition. DyB$_2$C has still large magnetic fluctuations in spite of the formation of three-dimensional magnetic order below $T_C = 8.5$ K. This is consistent with the ordered magnetic moment being only 3.3 $\mu_B$/Dy, whereas

$10 \mu_B$/Dy is expected from $J = 15/2$. Moreover, the magnetic specific heat shows a tail in high temperature region, and the magnetic entropy is still released even far above $T_C = 8.5$ K, as shown in Fig. 10(b). The very small anomaly at 20.5 K coincides with the appearance of the sharp Warren (002)$_m$ peak observed by neutron diffraction measurements for DyB$_2$C.

D. Magnetic ordering temperature

The magnetic properties of RB$_2$C ($R =$ Dy, Ho, and Er) are summarized in Table I. The effective paramagnetic moments are close to those expected for a free $R^{3+}$ ion values. These results indicate that the 4$f$ electrons in the rare earth ions are well localized and the 4$f$ levels locate far below the Fermi level ($E_F$). Hence, the influence of the 4$f$-band hybridization with the conduction band at $E_F$ is most likely very small. We believe that the density of states at $E_F$ of RB$_2$C ($R =$ Dy, Ho, and Er) does not so much change with rare earth element. Moreover, the neutron diffraction study of RB$_2$C ($R =$ Dy, Ho, and Er) shows that the lattice constants and the interatomic distances between the rare earth ions change only 2% or less with the rare earth element. Therefore, it is reasonable to assume that the magnetic coupling between the localized 4$f$ moments in RB$_2$C is the Ruderman-Kittel-Kasuya-Yosida (RKKY) type one via conduction electron polarization, and the RKKY exchange coefficient remain unchanged for all rare earth elements in this series. In the RKKY model, magnetic transition temperatures
TABLE I. Magnetic properties of $\text{R}_2\text{B}_2\text{C}$ ($\text{R}=\text{Dy}, \text{Ho}, \text{and Er}$). The magnetic transition temperatures $T_C$ are determined by the temperature at the maximum of the specific heat. The Weiss temperature $\theta_W$ and the effective paramagnetic moment $\mu_{\text{eff}}$ are estimated from the Curie–Weiss fitting. $g[J(J+1)]^{1/2}$ is the effective paramagnetic moment of the free trivalent rare earth ion, where $g$ is the Landé $g$ factor and $J$ is the total momentum of the $f$ electron configuration. $G$ is the de Gennes factor $G=(g-1)^2J(J+1)$.

<table>
<thead>
<tr>
<th>Compound</th>
<th>$T_C$ (K)</th>
<th>$\theta_W$ (K)</th>
<th>$\mu_{\text{eff}}$ ((\mu_\text{B}))</th>
<th>$g[J(J+1)]^{1/2}$</th>
<th>$G$</th>
</tr>
</thead>
<tbody>
<tr>
<td>DyB$_2$C</td>
<td>8.5</td>
<td>0.1</td>
<td>10.63</td>
<td>10.63</td>
<td>7.08</td>
</tr>
<tr>
<td>HoB$_2$C ((\perp c))</td>
<td>7.0</td>
<td>-0.58</td>
<td>10.64</td>
<td>10.58</td>
<td>4.5</td>
</tr>
<tr>
<td>HoB$_2$C ((\parallel c))</td>
<td></td>
<td>-4.75</td>
<td>10.05</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ErB$_2$C</td>
<td>16.3$^a$</td>
<td>38.6</td>
<td>9.28</td>
<td>9.59</td>
<td>2.55</td>
</tr>
</tbody>
</table>

$^a$Neel temperature $T_N$.

are approximately proportional to the de Gennes factor $G = (g-1)^2J(J+1)$.$^{10}$ In $\text{R}_2\text{B}_2\text{C}$ ($\text{R}=\text{Dy}, \text{Ho}, \text{and Er}$), however, the magnetic ordering temperatures do not follow the de Gennes relation. The disagree with the de Gennes scaling suggest that not only the RKKY interaction but also various mechanisms have to be considered when describing the magnetic coupling in $\text{R}_2\text{B}_2\text{C}$ ($\text{R}=\text{Dy}, \text{Ho}, \text{and Er}$).

The magnetic susceptibilities and magnetization curves of $\text{HoB}_2\text{C}$ suggest that the magnetic system of $\text{HoB}_2\text{C}$ is anisotropic-Heisenberg-type. The magnetic system of $\text{DyB}_2\text{C}$ is probably the same as that of $\text{HoB}_2\text{C}$. This is in contrast to the Ising-type behavior in $\text{ErB}_2\text{C}$, where the magnetic moment lies parallel to the $c$-axis.$^9$ The magnetic anisotropies in $\text{R}_2\text{B}_2\text{C}$ are due to CEF effects. It is natural that the increased degrees of freedom in the Heisenberg system lower the ordering temperature, because fluctuations are usually enhanced when the order parameter has continuous degrees of freedom. Therefore, the ordering temperatures of $\text{HoB}_2\text{C}$ and $\text{DyB}_2\text{C}$ are lower than those estimated from the de Gennes factor on the basis of $\text{ErB}_2\text{C}$.

The CEF has also considerable influence on the magnetic ordering temperature. Taking the CEF term added to the exchange Hamiltonian into account, the discrepancies between observed ordering temperatures and those predicted by the de Gennes relation was discussed.$^{11}$ The results of the inverse susceptibilities and the magnetic specific heat measurements of $\text{R}_2\text{B}_2\text{C}$ ($\text{R}=\text{Dy}, \text{Ho}, \text{and Er}$) indicate that the CEF splitting of the low-lying levels of $\text{ErB}_2\text{C}$ are larger than those of $\text{HoB}_2\text{C}$ and $\text{DyB}_2\text{C}$. Thus, the magnetic ordering temperature of $\text{ErB}_2\text{C}$ may be affected by CEF more strongly than those of $\text{HoB}_2\text{C}$ and $\text{DyB}_2\text{C}$.

Magnetic ordering temperatures of the $\text{R}_2\text{B}_2\text{C}$ analogs also do not follow the de Gennes relation. The magnetic ordering temperatures $T_N$ or $T_C$ of $\text{TbB}_2\text{C}_2$, $\text{DyB}_2\text{C}_2$, $\text{HoB}_2\text{C}_2$, and $\text{ErB}_2\text{C}_2$ are 21.7 K, 15.3 K, 5.8 K, and 15.9 K, respectively.$^{1-3,12,13}$ The magnetic ordering temperatures of $R=\text{Tb}$, $\text{Dy}$, and $\text{Ho}$ are suppressed by the competition between the quadrupole interaction and the magnetic interaction. It seems likely that the suppression of the ordering temperatures of $\text{HoB}_2\text{C}$ and $\text{DyB}_2\text{C}$ is also related to the quadrupole or/and higher multipole interaction of the 4$f$ electrons.

E. Spin frustration in the Shastry-Sutherland lattice

In this section, we discuss the magnetic order in view of the geometric arrangement of the rare earth metals. The 2D-layer of the rare earth metals in $\text{R}_2\text{B}_2\text{C}$ [Fig. 11(a)] is topologically equivalent to the Shastry-Sutherland model (SSM) [Fig. 11(b)], which is a 2D-model.$^7$ For the diagonal interaction $J_1$ and the square lattice interaction $J_2$, the Hamiltonian is written as

$$\mathcal{H} = J_1 \sum_{\text{diagonals}} S_i \cdot S_j + J_2 \sum_{\text{square}} S_i \cdot S_j.$$ (1)

This becomes a magnetically frustrated system if $J_1$ and $J_2$ are antiferromagnetic. In the Ising limit of SSM, the ground state for $J_1/J_2<2$ is the Néel LRO state (\((++-+)\) structure) which is known to be the ground state of $\mathcal{H}$ for $J_1 = 0$, and that for $J_1/J_2>2$ is the “paramagnetic” dimer state, in which spins at the ends of a dimer bond must be antiparallel.

In the classical Heisenberg system of SSM, the phase diagram was exactly determined for Eq. (1) as a function of $J_1/J_2$. The phase diagram of the Heisenberg system of SSM differs greatly from that of the Ising system. For $J_1/J_2<1$, the Néel LRO state ((++-+) structure) is the ground state, whereas for $J_1/J_2>1$, the ground state is the helical LRO state with the incommensurate wave vector \((\pi, q)\) or \((q, \pi)\). With decreasing the spin $S$, the system falls into a quantum spin liquid state, where the dimer forms a spin singlet in the case of $J_1/J_2<0.7$ for $S=1/2$.\(^{14}\)

I. $\text{ErB}_2\text{C}$: Ising-type system

The magnetic susceptibilities and magnetization curves of $\text{ErB}_2\text{C}$ suggest that the magnetic system of $\text{ErB}_2\text{C}$ is an Ising-type one. The neutron diffraction studies show that the magnetic structure of $\text{ErB}_2\text{C}$ is a collinear (++-+) antiferromagnet as shown in Fig. 12, where the magnetic mo-
The Shastry-Sutherland theory does not include the magnetic fluctuations that appear in HoB$_2$C and DyB$_2$C. In the present case, additional interactions competing with $J_1$ and $J_2$ are necessary to explain the instability of the magnetic phase. The possible candidate is the diagonal coupling of the square lattices [J$_3$ in Fig. 11(a)]. For $J_2>0$ and $J_3>0$, the Shastry-Sutherland lattice is expected to be further frustrated. Thus the long-distance interaction $J_3$ seem to play an important role in the spin frustration, because the magnetic coupling between the localized 4$f$ moments in the RB$_2$C is RKKY type. Furthermore, a quadrupole or/and higher multipole interaction of the 4$f$ electrons competing with the magnetic interactions may be also related to the frustration of the magnetic fluctuations. A possible origin of the formation of the MRL lattice, which is characterized by the short in-plane magnetic correlation, is due to the formation of helical domains of SSM.

IV. CONCLUSION

We have studied the magnetic properties of RB$_2$C ($R$ = Dy, Ho, and Er) by the magnetization and specific heat measurements. ErB$_2$C has a basic two-sublattice antiferromagnetic structure, and a strong Ising-type magnetic anisotropy. HoB$_2$C and DyB$_2$C show large magnetic fluctuations below $T_C$ as well as the three-dimensional magnetic order. In RB$_2$C ($R$ = Dy, Ho, and Er) both $\theta_1$ and the magnetic ordering temperatures do not follow the de Gennes relation, as HoB$_2$C and DyB$_2$C show a suppression of ordering temperature. The instability of the unconventional magnetic phase associated with the frustration is significant in this series; the fluctuating ground state results from the complex spin–spin interactions or spin–quadrupole interactions. For HoB$_2$C, no anomalies of the magnetizations and the specific heat are observed in the temperature region, in which the unusual low-$Q$ peaks are observed by the neutron diffraction measurements. For DyB$_2$C, the magnetization anomalies are not evident, but the specific heat shows an anomaly in that region. The noncritical behavior of the specific heat in the temperature region implies the transitions are not LRO. The interesting magnetic properties of HoB$_2$C and DyB$_2$C may originate from the frustrated nature of the magnetic lattice containing triangular interactions between rare earth atoms. The specific ratio of the $J_1$, $J_2$, and $J_3$ interaction gives rise to complex magnetic structures, one of which may bring about the formation of the MRL lattice. At present, however, we have insufficient information about the order parameter in these temperature regions. NMR and inelastic neutron scattering measurements will be performed to determined the spin dynamics of those unconventional materials.
12 K. Kaneko, H. Onodera, H. Yamauchi, K. Ohoyama, A. Tobo, and
13 K. Ohoyama, K. Indoh, A. Tobo, H. Onodera, and Y. Yamaguchi,