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Magnetic Orderings and Néel Temperature of $TbNi_2B_2C$ *

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We employ a two-ion model to calculate the spontaneous magnetization and susceptibility of $TbNi_2B_2C$ with the mean-field theory, and derive an analytic formula for the Néel temperature of the material with the perturbation theory. In both the cases, only the four lowest crystal-electric-field (CEF) states are taken into account as base functions. Our theoretical results are consistent with the experimental data, manifesting the strong effects of the low lying CEF states on the magnetic behaviors of the material and the suitability of the theoretical approach proposed here.

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Rare-earth nickel borocarbide RNi_2B_2C compounds have attracted considerable research interest in the last fifteen years due to the coexistence of superconductivity and magnetic ordering in low temperature regions.The [1−12] X-ray experiment revealed that RNi_2B_2C compounds all crystallize in the tetragonal structure of the $ThCr_2Si_2$ type, in which the R-C sheets and Ni_2B_2 layers stack alternatively along the c-axis.[13−15] Neutron scattering experiments found that the magnetic ordering in these systems arises from the rare earth ions, which bear localized $4f$ electrons and are coupled with each other through RKKY exchange interaction, but no ordered magnetic moment was detected on the Ni site.[16]

To study the anisotropic magnetic properties of these compounds quantitatively, Gasser *et al.*[17] performed inelastic neutron scattering experiments to determine the crystal-field parameters (CFPs) for the compounds with $R =$ Ho, Er and Tm, which were then utilized to estimate the CFPs of other members of the family with an extrapolation approach,[18] and to calculate the susceptibilities and magnetic specific heats of the Ho, Er and Tm compounds. Indeed, their calculated susceptibilities for these three materials were consistent with the measured ones above the magnetic transition temperatures.

In rear-earth antiferromagnets, the magnetic moments of the rare-earth ions still order antiferromagnetically well below T_N, even when a weak external magnetic field is exerted. Only when the external field is sufficiently strong can the magnetic moment orients in the opposite direction be rotated towards the field direction. Obviously when describing the magnetic structure of an antiferromagnet below T_N, the single-ion model usually used for ferromagnets is inadequate.[17] To solve the problem, we have proposed a two-ion model and used it to calculate the susceptibilities and magnetizations of $HoNi_2B_2C$,[19] $TbNi_2B_2C$ [20] and $DyFe_2Si_2$[21,22] successfully, and obtained good agreement with experimental results.[23−25]

In this model,[19−22] the A and B ions form two sublattices and they are all under CEF interaction. In addition, the A ion interacts with another A ion and a B ion through RKKY exchange interaction with coupling constants of λ_{11} and λ_{12}, the B ion with another B ion and the A ion with coupling constants of λ_{22} and λ_{21}, respectively. Thus, the Hamiltonians of the two magnetic ion are given by

$$\begin{aligned}\mathcal{H}^{(\mathrm{A})} &= \mathcal{H}_{CF} - \lambda_{11}\boldsymbol{J}\cdot\langle\boldsymbol{J}^{(\mathrm{A})}\rangle - \lambda_{12}\boldsymbol{J}\cdot\langle\boldsymbol{J}^{(\mathrm{B})}\rangle \\ &\quad - \mu_B g_J \boldsymbol{B}\cdot\boldsymbol{J}, \\ \mathcal{H}^{(\mathrm{B})} &= \mathcal{H}_{CF} - \lambda_{22}\boldsymbol{J}\cdot\langle\boldsymbol{J}^{(\mathrm{B})}\rangle - \lambda_{21}\boldsymbol{J}\cdot\langle\boldsymbol{J}^{(\mathrm{A})}\rangle \\ &\quad - \mu_B g_J \boldsymbol{B}\cdot\boldsymbol{J}. \end{aligned} \tag{1}$$

For simplicity, we have chosen $\lambda_{11} = \lambda_{22} = \lambda_1$ and $\lambda_{12} = \lambda_{21} = \lambda_2$, which have been found to be $\lambda_1 = 0.530\,\mathrm{K}$ and $\lambda_2 = -0.335\,\mathrm{K}$ for $TbNi_2B_2C$ by fitting its magnetization curves.[20]

For several decades, Bleaney's formula[26]

$$\chi = \chi_{CF}/(1 - \lambda\chi_{CF}), \tag{2}$$

has been used to calculate the susceptibility (denoted as χ) of a rare-earth compound from the susceptibility contributed by the pure CEF interaction (χ_{CF}) and the molecular-field constant (λ). The formula is also widely employed to estimate the molecular-field constant from the measured magnetic transition temperature or vice versa.

In a rare-earth magnet, if the second order crystal-field parameter B_2^0 is dominant but other CPFs are negligible, provided that the magnetic moments order in the z-direction, the magnetic transition temperature T_M can be estimated with the following formula

$$T_M = 2\mathcal{J}_0(g_J - 1)^2 \sum_{J_z} J_z^2 \exp\left(-3B_2^0 J_z^2/T_M\right)$$

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$$\times \left[\sum_{J_z} \exp\left(-\frac{3B_2^0 J_z^2}{T_M} \right) \right]^{-1}, \quad (3)$$

where $\mathcal{J}_0$ denotes the RKKY coupling constant.[27] Unfortunately, in most rare-earth magnets higher-order CFPs are also important and cannot be simply neglected, thus another formula

$$T_M = 2\mathcal{J}_0(g_J - 1)^2 \langle J_z^2(T_M)\rangle_{CF}, \quad (4)$$

should be used, where $\langle J_z^2(T_M)\rangle_{CF}$ is the thermally averaged value of J_z^2 evaluated at T_M with the pure CEF states only. The above formula was utilized by Noakes and Shenoy[27] to estimate the magnetic transition temperatures T_M's of rare-earth rhodium borides ($R\mathrm{Rh_4B_4}$). However, their calculated T_M's for $\mathrm{DyRh_4B_4}$ and $\mathrm{HoRh_4B_4}$ were both underestimated, in particular, their theoretical T_N value for antiferromagnet $\mathrm{TmRh_4B_4}$ was almost equal to zero.

To solve the above problem, Dunlap *et al.*[28] included all CEF terms in the Hamiltonian and performed numerical calculations to re-evaluate the T_M's of the rare-earth rhodium borides. However, their calculated T_M's of $\mathrm{DyRh_4B_4}$ and $\mathrm{HoRh_4B_4}$ were still both underestimated, and that of $\mathrm{TmRh_4B_4}$ remained almost unchanged.

Just seven years ago, Luong *et al.*[29] studied the Néel temperatures of $R\mathrm{Cu_2}$ with Eq. (4). To make use of the equation and to perform numeric calculations, the coordinate system has to be rotated so that the new z-axis is along the ordering direction for a considered compound.

To estimate the magnetic transition temperature T_M of a rare-earth magnet from its RKKY exchange constant or vice versa, we have developed an analytic approach based on mean-field and perturbation theories,[30–33] where the single-ion Hamiltonian was first used to derive analytic formulas for rare-earth ferromagnets, then extended to obtain formulas for rear-earth antiferromagnets. Now the two-ion model has been proposed for rare-earth antiferromagnets, it is really necessary to apply the model to a real antiferromagnet, for example, $\mathrm{TbNi_2B_2C}$ in the present work, to re-examine the problem and to test the theoretical approaches, that is, to derive formulae for the Néel temperature with the perturbation theory, and evaluate the magnetization numerically with the mean-field approach. We aim also to understand the mechanisms of the magnetic ordering and phase transition, i.e., to study how the CEF levels and RKKY coupling govern the magnetic process in the magnet at low temperatures. In contrast, our previous work only performed numerical calculations to study the magnetic properties of $\mathrm{TbNi_2B_2C}$, and to demonstrate the applicability of the two-sublattice model to antiferromagnets.[20]

As observed in experiments, $\mathrm{TbNi_2B_2C}$ shows no superconductivity at least down to 7 mK,[34]. The magnetic moments of Tb ions align along the a-axis below $T_N = 15$ K in an incommensurate antiferromagnetic structure.[34–36] Neutron diffraction measurement on the single crystal of $\mathrm{TbNi_2B_2C}$ suggested the development of a small ferromagnetic component below 8 K.[35] Further ac susceptibility and low field dc magnetization measurements also revealed two magnetic transitions, which correspond to the ordering of the Tb moments starting at $T_N = 15$ K and a spin reorientation at lower temperatures than $T_{NF} = 5.1$ K.[37]

The crystal-field parameters of $\mathrm{TbNi_2B_2C}$ have been found to be $A_2^0 = -15.4$ meV, $A_4^0 = 2.89$ meV, $A_4^4 = -84.4$ meV, $A_6^0 = -1.92$ meV and $A_6^4 = 22.6$ meV, respectively, by Gasser *et al.* with an extrapolation approach.[18] With these CFPs the three lowest crystal field levels are found to be

$$\begin{aligned} |\varphi_1^{(0)}\rangle &= a|4\rangle + b|0\rangle + c|-4\rangle, \\ |\varphi_{2,3}^{(0)}\rangle &= d|\pm 5\rangle + e|\pm 1\rangle + f|\mp 3\rangle, \\ |\varphi_4^{(0)}\rangle &= r|6\rangle + s|2\rangle + t|-2\rangle + u|-6\rangle, \end{aligned} \quad (5)$$

where $a = c = 0.44290$, $b = 0.77954$, $d = 0.28099$, $e = 0.76567$, $f = 0.57861$, $r = u = 0.10265$, $s = t = 0.69962$. In addition, the first excited CEF level $|\varphi_{2,3}^{(0)}\rangle$, a doublet as shown above, is located at 5.2859 K ($= \epsilon_2^{(0)}$) above $|\varphi_1^{(0)}\rangle$, and the second excited CEF level $|\varphi_4^{(0)}\rangle$, a singlet, situated at 10.8144 K ($= \epsilon_3^{(0)}$), respectively.

The ground CEF state $|\varphi_1^{(0)}\rangle$ is non-magnetic, so it cannot order magnetically itself below T_N in the absence of an external magnetic field. However, the two states of the first excited CEF levels $|\varphi_{2,3}^{(0)}\rangle$ are able to mix with $|\varphi_1^{(0)}\rangle$ through an RKKY exchange interaction to form new magnetic states and to give rise to magnetic ordering, since there exist non-zero matrix elements of $J_\pm$ between the two levels according to the established theory.[30–32,38,39] Moreover, though $|\varphi_4^{(0)}\rangle$ cannot mix with the ground state $|\varphi_1^{(0)}\rangle$ directly, it is able to mix with the two states $|\varphi_{2,3}^{(0)}\rangle$ of the first excited CEF level for the same reason, which bridges its indirect mixture with $|\varphi_1^{(0)}\rangle$ to form new magnetic states. Moreover, other CEF levels are firmly separated above. For instance, the third excited level $|\varphi_5^{(0)}\rangle = -0.707|4\rangle + 0.707|-4\rangle$ is situated at 144.939 K. Therefore, we believe that the magnetic properties of the material are mainly governed by the lowest four CEF states. In the vector space spanned by them as base functions, we have the matrices

$$\mathcal{J}_x = \begin{pmatrix} 0 & \alpha & \beta & 0 \\ \alpha & 0 & 0 & \gamma \\ \beta & 0 & 0 & \delta \\ 0 & \gamma & \delta & 0 \end{pmatrix},$$

$$\mathcal{H}_{EX}^{A,B} = -\mathcal{F}_{A,B} \begin{pmatrix} 0 & \alpha & \beta & 0 \\ \alpha & 0 & 0 & \gamma \\ \beta & 0 & 0 & \delta \\ 0 & \gamma & \delta & 0 \end{pmatrix}, \quad (6)$$

for the x component of the total angular momentum and the exchange Hamiltonians of the two rare-

earth ions, where $\alpha = 0.5(ad\sqrt{22} + be\sqrt{42} + cf\sqrt{30})$, $\beta = 0.5(cd\sqrt{22} + be\sqrt{42} + af\sqrt{30})$, $\gamma = rd\sqrt{3} + es\sqrt{10} + 3ft$, $\delta = du\sqrt{3} + et\sqrt{10} + 3fs$, and $\mathcal{F}_{A,B} = \lambda_1\langle J_{A,B}\rangle + \lambda_2\langle J_{B,A}\rangle + \mu_B g_J B$, respectively. Thus the total Hamiltonian matrices for the two ions are given by $\mathcal{H}_{A,B} = \mathcal{H}_{CF} + \mathcal{H}_{EX}^{A,B}$.

The two Hamiltonians are coupled together through $\langle J_A\rangle$ and $\langle J_B\rangle$, which are the functions of temperature and the external magnetic field. They are diagonalized separately to calculate $\langle J_A\rangle$ and $\langle J_B\rangle$ self-consistently in every computing loop. Such obtained spontaneous magnetization curves for the A and B ions are displayed in Fig. 1, and both are actually identical to those computed with the full CEF states of the lowest J-multiplet.[20] For comparison, we also calculated the magnetizations of the two ions with the three states of the two lowest CEF levels as depicted in the same figure, where the both curves just sustain up to $T = 10.6$ K, clearly suggesting that the spontaneous magnetic ordering of the material be governed by the three lowest CEF levels in the low temperature range.

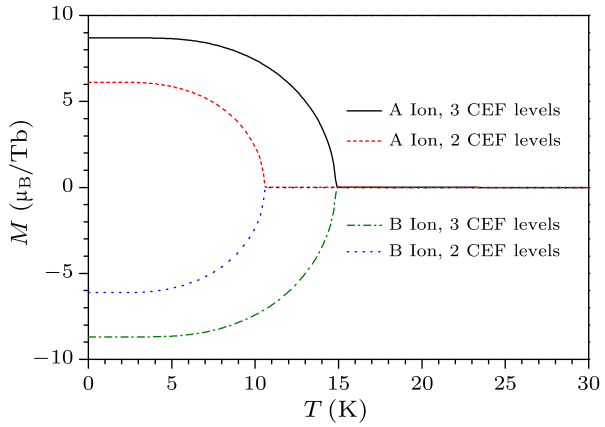


Fig. 1. The calculated magnetization curves of the A and B ions of $TbNi_2B_2C$ plotted as the functions of temperature with the four and three lowest CEF states in the absence of external magnetic field. Here the exchange constants $\lambda_1 = 0.530$ K and $\lambda_2 = -0.335$ K are used.

To further investigate, we also calculated the susceptibility data along the easy a-axis with the four states as base functions. Our theoretical susceptibility for the whole bulk sample is depicted in the main panel of Fig. 2. The susceptibilities of the two sublattices computed in the reduced vector space are also depicted in the inset of the figure. Surprisingly, these results, evaluated in the reduced vector space, are almost identical with those computed with the full CEF levels of the lowest J-mutiplet,[20] and the curve plotted in the main panel is consistent with the experimental one up to 160 K,[23] suggesting once again that the magnetic process of the magnet be dominantly governed by the lowest four CEF states. As already found before,[20] the susceptibility data of the two sublattices are significantly larger than those experimental data of the bulk sample well below T_N, however they are offset so that their sum values are considerably reduced as displayed in the main panel both theoretically and experimentally.

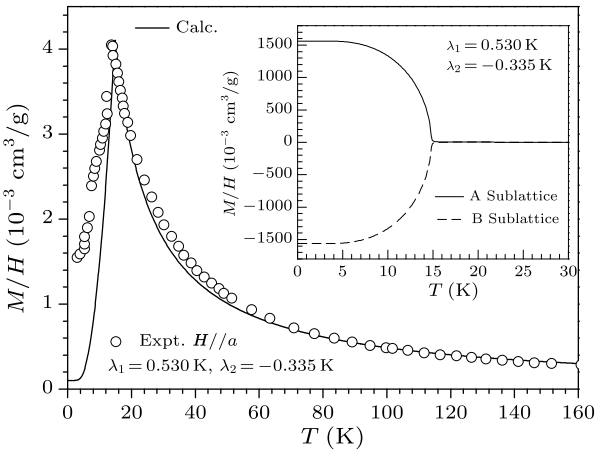


Fig. 2. The bulk susceptibility along the easy a-axis (in the main panel) and those of the A and B sublattices (in the inset) for $TbNi_2B_2C$ calculated with the lowest four CEF states as base functions. Here $\lambda_1 = 0.530$ K, $\lambda_2 = -0.335$ K are used.

The above analyses allow us to make further theoretical investigations into the four lowest CEF states. When no external magnetic field is applied and as T approaches T_N, the RKKY exchange interaction between the neighboring magnetic rare-earth ions in the material is considerably weak, however the CEF interaction still persists, therefore the exchange interaction can be treated as a weak perturbation. To the first order approximation, the new eigenstates of the four lowest states for the A ion are

$$
\begin{aligned}
|\psi_1^{(A)}\rangle &= |\varphi_1^{(0)}\rangle + \frac{\alpha\mathcal{F}_A}{\epsilon_2^{(0)}}|\varphi_2^{(0)}\rangle + \frac{\beta\mathcal{F}_A}{\epsilon_2^{(0)}}|\varphi_3^{(0)}\rangle,\\
|\psi_2^{(A)}\rangle &= |\varphi_2^{(0)}\rangle - \frac{\alpha\mathcal{F}_A}{\epsilon_2^{(0)}}|\varphi_1^{(0)}\rangle - \frac{\gamma\mathcal{F}_A}{\epsilon_2^{(0)} - \epsilon_3^{(0)}}|\varphi_4^{(0)}\rangle,\\
|\psi_3^{(A)}\rangle &= |\varphi_3^{(0)}\rangle - \frac{\beta\mathcal{F}_A}{\epsilon_2^{(0)}}|\varphi_1^{(0)}\rangle - \frac{\delta\mathcal{F}_A}{\epsilon_2^{(0)} - \epsilon_3^{(0)}}|\varphi_4^{(0)}\rangle,\\
|\psi_4^{(A)}\rangle &= |\varphi_4^{(0)}\rangle - \frac{\gamma\mathcal{F}_A}{\epsilon_3^{(0)} - \epsilon_2^{(0)}}|\varphi_2^{(0)}\rangle\\
&\quad - \frac{\delta\mathcal{F}_A}{\epsilon_3^{(0)} - \epsilon_2^{(0)}}|\varphi_3^{(0)}\rangle,
\end{aligned}
\tag{7}
$$

with eigenenergies

$$
\begin{aligned}
\epsilon_1 &= -\frac{(\alpha^2+\beta^2)\mathcal{F}_A^2}{\epsilon_2^{(0)}},\\
\epsilon_2 &= \epsilon_2^{(0)} + \frac{(\alpha\mathcal{F}_A)^2}{\epsilon_2^{(0)}} + \frac{(\gamma\mathcal{F}_A)^2}{\epsilon_2^{(0)} - \epsilon_3^{(0)}},\\
\epsilon_3 &= \epsilon_2^{(0)} + \frac{(\beta\mathcal{F}_A)^2}{\epsilon_2^{(0)}} + \frac{(\delta\mathcal{F}_A)^2}{\epsilon_2^{(0)} - \epsilon_3^{(0)}},\\
\epsilon_4 &= \epsilon_3^{(0)} + \frac{(\delta^2+\gamma^2)\mathcal{F}_A^2}{\epsilon_3^{(0)} - \epsilon_2^{(0)}},
\end{aligned}
\tag{8}
$$

to the second order approximation. For the B ion

we can obtain similar formulas. Evaluating the spontaneous magnetization with these new states and eigenenergies, and keeping in mind that when $T \to T_N$, $\langle J_A \rangle = -\langle J_B \rangle \to 0$ so that $\mathcal{F}_{A,B} \to 0$ as well, we then obtain an equation at the magnetic critical temperature

$$\begin{aligned}
&\frac{1 + 2\exp\left(-\frac{\epsilon_2^{(0)}}{k_B T_N}\right) + \exp\left(-\frac{\epsilon_3^{(0)}}{k_B T_N}\right)}{2(\lambda_1 - \lambda_2)} \\
&= \frac{\alpha^2 + \beta^2}{\epsilon_2^{(0)}} - \left(\frac{\alpha^2 + \beta^2}{\epsilon_2^{(0)}} + \frac{\gamma^2 + \delta^2}{\epsilon_2^{(0)} - \epsilon_3^{(0)}}\right) \\
&\cdot \exp\left(-\frac{\epsilon_2^{(0)}}{k_B T_N}\right) - \frac{\delta^2 + \gamma^2}{\epsilon_3^{(0)} - \epsilon_2^{(0)}} \\
&\cdot \exp\left(-\frac{\epsilon_3^{(0)}}{k_B T_N}\right).
\end{aligned}$$

Substituting $\lambda_1 = 0.530\,\mathrm{K}$ and $\lambda_2 = -0.335\,\mathrm{K}$, which were determined by fitting the measured susceptibility curves and T_N of the magnet,[20] and other parameters as given above, into this equation provides a solution of $T_N = 14.8\,\mathrm{K}$ for the Néel temperature, which is very close to 15 K, the value observed during experiments.

It is of interest to emphasize finally: (1) The ground CEF singlet of $TbNi_2B_2C$ is non-magnetic in the ordering direction, it cannot produce spontaneous magnetic ordering itself even when RKKY exchange is considered. (2) The first excited CEF level is able to mix directly with the ground CEF state through RKKY exchange interaction to form new magnetic states because there exist non-zero matrix elements of $J_\pm$ between the states of the two CEF levels to induce magnetic ordering. (3) The second excited CEF level, which is a singlet, can only mix with the ground CEF state *indirectly* through its direct mixture with the two states of the first excited CEF level to promote magnetic ordering. (4) The spontaneous magnetization is dominantly governed by these four lowest CEF states. (5) The spontaneous magnetization and susceptibility curves obtained with the four lowest CEF states as base functions are almost *identical* with those calculated with the full CEF states of the lowest J-multiplet, clearly indicating that our approximations, by neglecting other upper lying CEF levels, are very accurate. (6) The Néel temperature evaluated with the perturbation theory by using the four states as base functions is consistent with the experimental value, verifying the correctness of the theoretical approach and demonstrating the applicability of the two-ion model for rare-earth antiferromagnets.

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