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Specific heat and magnetic ordering of $NdNi_2B_2C$

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Abstract

In this work, the specific heat of $NdNi_2B_2C$ was computed with the three sets of crystal-field parameters proposed by previous authors. All curves of the heat capacity plotted with the calculated results exhibit sharp peaks around the magnetic transition temperature T_N as experimentally observed. To understand the mechanism of its magnetic ordering, we also calculated the magnetization of the material in low temperature region with the ground crystal-field (CF) level, the two lowest CF levels, and the full CF levels of $J = 9/2$ multiplet respectively for comparison. Using the two eigenstates of the ground CF level, we derived a formula for $\langle J_x \rangle$ with mean-field approach for theoretical analysis. Both our numeric and theoretical results suggest that the two lowest CF levels play dominant roles in the magnetic process of the material below T_N. It is also very interesting to notice that the ground CF level itself results in a larger T_N, but the inclusion of the first excited CF doublet in calculations instead hinders the magnetic ordering, leading to a weakly reduced transition temperature.

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Keywords: Crystal-field theory; Specific heat; Magnetic ordering

The discovery of rare-earth nickel boride carbides $RENi_2B_2C$ have attracted considerable interest in recent years, due to the co-existence of superconductivity and magnetism observed in low temperature region when the rare earth (RE) is either Dy, Ho, Er or Tm [1]. The crystal structure of the quaternary intermetallic borocarbide systems is body-centered tetragonal, similar to $ThCr_2Si_2$-type system, but with an additional carbon atom in each lanthanide layer [2]. In these materials, the localized 4f electrons are subject to strong crystal field interactions, and the rare-earth ions are coupled with each other through the valence electrons. It is such indirect interactions of the rare-earth ions, called Ruderman–Kittel–Kasuya–Yoshida (RKKY) exchange, that gives rise to magnetic ordering of the material in low temperature region.

The crystal field parameters (CFP's) of $RENi_2B_2C$ (RE = Ho, Er, Tm) have been determined by inelastic neutron scattering (INS), these parameters were then extrapolated to other borocarbides (RE = Pr, Nd, Sm, Tb, Dy, and Yb) [3]. However, the Raman scattering performed recently on $ErNi_2B_2C$ suggested a new CF level scheme for the ground state $J = 9/2$ multiplet [4], which is different from the set obtained previously by INS [3]. To investigate the crystal-field interaction in the light of its electronic structure, M. Divis et al. calculated the CFP's of $NdNi_2B_2C$ with the first principle technique based on density functional theory [5], and such obtained CFP's were then used to fit the Schottky specific heat, which is attributed to the splitting of the ground CF J-multiplet of the Nd^{3+} ion. Their calculated specific heat with the fitted parameters yielded qualitatively good agreement with experimental observation *above* $T_N = 4.8$ K.

To investigate the magnetic and thermodynamic properties of $NdNi_2B_2C$, we calculated its specific heat with the three sets of the CFP's proposed by Gasser and Divis et al. [3,5], and also

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Table 1
Three sets of the crystal-field parameters in Kelvin, obtained by U. Gasser and M. Divis et al., are used in our calculations

	$A_2^0\langle r^2\rangle$	$A_4^0\langle r^4\rangle$	$A_4^4\langle r^4\rangle$	$A_6^0\langle r^6\rangle$	$A_6^4\langle r^6\rangle$
Gasser	−238	57	−1669	−47	551
DFT	−32	0.9	131	19	−183
Fit	−18	65	250	50	−280

its magnetization in low temperature region with both theoretical and numeric approaches in present work.

$NdNi_2B_2C$ crystallizes in tetragonal structure, its magnetic moment is observed to be along a-axis below $T_N = 4.8$ K [6], so the single ion Hamiltonian of this system is given by

$$\mathcal{H} = \alpha_J A_2^0\langle r^2\rangle O_2^0 + \beta_J\left(A_4^0\langle r^4\rangle O_4^0 + A_4^4\langle r^4\rangle O_4^4\right) + \gamma_J\left(A_6^0\langle r^6\rangle O_6^0 + A_6^4\langle r^6\rangle O_6^4\right) - \mathcal{J}\langle J_x\rangle J_x + \frac{1}{2}\mathcal{J}\langle J_x\rangle^2 \quad (1)$$

in the mean-field approximation, where the first fives terms denotes the crystal field interactions, the sixth term represents the RKKY exchange coupling among the neighboring magnetic rare-earth ions, and the last one is a correction accounting for the duplicate count of the ions. *To describe the magnetic and thermodynamic properties of a rare-earth magnet below the magnetic transition temperature T_M, the RKKY exchange term must be considered in the calculations* [7–9]. Otherwise, the sharp peak at T_M in the curve of the specific heat cannot be reproduced theoretically [5,10–13]. The exchange coupling parameter $\mathcal{J}$ in this term can be determined by fitting T_M of the magnet in calculations.

$NdNi_2B_2C$ is antiferromagnetic, and has complicated magnetic structure below $T_N = 4.8$ K. However, its macroscopic properties, such as specific heat, susceptibility and overall magnetization in an external field, can be described rather nicely with a *ferromagnetic-like model* as Eq. (1) because the magnetic moments of the material align co-linearly below T_N. The validity of this approach has been justified and explained in detail in our previous article [14]. For an antiferromagnet, $\langle J_x\rangle$ appearing in the single-ion Hamiltonian represents the thermally averaged J_x of a neighboring magnetic ion, and has a different sign than that of the considered rare-earth ion. Since actually the corresponding quantity of the considered ion is evaluated in the computing process, thus a negative sign must be added when such calculated $\langle J_x\rangle$ is taken as the value of the neighboring ion in computations. Additionally, $\mathcal{J}$ should be negative for an antiferromagnetic system to have lower energy. Thus, if the calculated $\langle J_x\rangle$ is directly fed to the code, $\mathcal{J}$ becomes positive as seen below.

The three sets of crystal-field parameters mentioned above (see Table 1) were used to compute the specific heat of the compound, the calculated results are now depicted in Fig. 1 in comparison with the experimental data. With Gasser's CFP's, the RKKY coupling parameter was found to be 2.0 K, whereas with the two sets of CFP's of Divis, the coupling parameters were determined to be $\mathcal{J} = 1.22$ K and $\mathcal{J} = 1.25$ K respectively by fitting T_N. Just below the critical temperature, the three sets of CFP's all generate a sharp peak in the specific

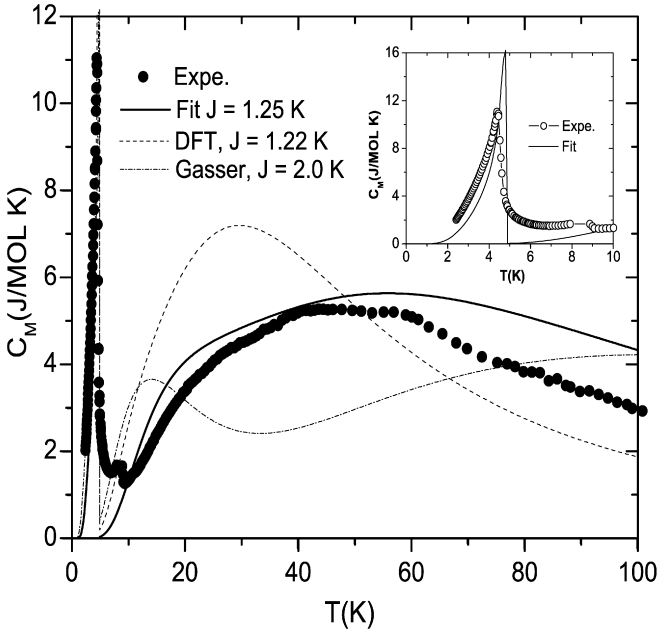

Fig. 1. The magnetic specific heat of $NdNi_2B_2C$ calculated with the three sets of CFP's and the exchange coupling constants in comparison with experimental results. The curve below 10 K for the fitted CFP's is also displayed in the inset.

heat curves, showing good agreement with the experimental results. However above T_N, the fitted CFP's by Divis and his co-workers based on DFT calculations and experimental results produce best agreement as found by them previously [5].

With the fitted CFP's, the ground CF level is found to be: $a|\pm\frac{3}{2}\rangle + b|\mp\frac{5}{2}\rangle$, where $a = 0.693$, $b = 0.721$; the first excited CF level is: $c|\pm\frac{9}{2}\rangle + d|\pm\frac{1}{2}\rangle + e|\mp\frac{7}{2}\rangle$, here $c = 0.901$, $d = -0.435$ and $e = -0.004$, which is 50.7 K above the lowest CF level; and other CF levels are well separated above them [5]. Therefore, it is reasonable to assume that the magnetism of the system below T_N is mainly governed by the ground CF doublet. To facilitate formulation, we denote $|\varphi_1\rangle = a|+\frac{3}{2}\rangle + b|-\frac{5}{2}\rangle$ and $|\varphi_2\rangle = a|-\frac{3}{2}\rangle + b|+\frac{5}{2}\rangle$ respectively. The system orders magnetically along a-axis below T_N, which we choose as x-axis, thus the RKKY exchange part of the Hamiltonian becomes

$$\mathcal{H}_{ex} = -\frac{1}{2}\mathcal{J}\langle J_x\rangle(J_+ + J_-), \quad (2)$$

where the corrective term has been neglected to simplify theoretical analysis. In the vector space spanned by these two states, the matrix of the Hamiltonian is

$$\mathcal{H} = \begin{pmatrix} \varepsilon_0 & -\sqrt{21}ab\mathcal{J}\langle J_x\rangle \\ -\sqrt{21}ab\mathcal{J}\langle J_x\rangle & \varepsilon_0 \end{pmatrix}, \quad (3)$$

and the ground CF doublet is split by the RKKY exchange interaction into two new states $|\psi_{1,2}\rangle = \frac{1}{\sqrt{2}}(|\varphi_1\rangle \pm |\varphi_2\rangle)$, which are shifted in energy by $\epsilon_{1,2} = \mp\sqrt{21}ab\mathcal{J}\langle J_x\rangle$ respectively. The thermal average of $J_\pm$ can be evaluated by

$$\langle J_\pm\rangle = \frac{1}{z}\big[\langle\psi_1|J_\pm|\psi_1\rangle\exp(-\epsilon_1/k_BT) + \langle\psi_2|J_\pm|\psi_2\rangle\exp(-\epsilon_2/k_BT)\big], \quad (4)$$