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Magnetic and quadrupolar interactions in NdB_6

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Abstract

By analysis of the excitation spectrum of NdB_6, we extract the intersite magnetic interactions. The Néel temperature $T_N = 7.9$ K is obtained by using the value of the intersite interaction deduced from the fit, which compares favourably with the experimental value $T_N \simeq 8$ K. We take into consideration a ferroquadrupolar interaction g_3' in the analysis, and obtain $g_3' = 107$ mK, which agrees well with the experimental value of about 100 mK. The ferroquadrupolar interaction results in a gap in the excitation spectrum observed in the experiment.

1. Introduction

The rich variety of ordered states in cubic rare-earth hexaborides RB_6 has attracted much attention. In particular, CeB_6 is a typical material which shows a quadrupolar order. At low temperatures, CeB_6 orders in a $4 \times 4 \times 2$ structure [1]. This structure is often observed in other RB_6 compounds: with R = Pr [2], Dy [3], and Gd [4]. On the other hand, NdB_6 orders in the A-type (or type I) antiferromagnetic structure at $T_N \simeq 8$ K [5].

In this paper, we reanalyse the excitation spectrum of NdB_6 [6, 7] to obtain information on the interaction in NdB_6. The crystalline electric field ground state of Nd^{3+} ($J = 9/2$) is the $\Gamma_8^{(2)}$ quartet [8, 9]. The easy axis of the dipolar moment in the $\Gamma_8^{(2)}$ states is along the $\langle 111 \rangle$ direction (and equivalent ones) [10]. An isotropic dipolar interaction alone would align the moment $\boldsymbol{J}$ along the $\langle 111 \rangle$ direction in the ground state. Experimentally, however, $\boldsymbol{J}$ is along $\langle 001 \rangle$ in the ordered state [5]. We show that the quadrupolar interaction stabilizes the state with $\boldsymbol{J} \parallel \langle 001 \rangle$. The importance of the Γ_3-type ferroquadrupolar interaction in NdB_6 has been pointed out [9, 11].

2. Model

In this paper, we consider a model including the Γ_3-type quadrupolar interaction:

$$H = -\sum_{(i,j)} I_{ij} \boldsymbol{J}_i \cdot \boldsymbol{J}_j - \sum_{(i,j)} K_{ij} (O_{2i}^0 O_{2j}^0 + 3 O_{2i}^2 O_{2j}^2). \tag{1}$$

We study this model within the $\Gamma_8^{(2)}$ quartet of states. They are given by

$$|+\sigma_z\rangle = a_1|\pm 9/2\rangle + a_2|\pm 1/2\rangle + a_3|\mp 7/2\rangle, \tag{2}$$

$$|-\sigma_z\rangle = b_1|\pm 5/2\rangle + b_2|\mp 3/2\rangle, \tag{3}$$

where $\sigma_z = \uparrow$ or $\downarrow$ and the upper (lower) signs of $\pm$ and $\mp$ correspond to $\sigma_z = \uparrow(\downarrow)$. The coefficients in (2) and (3) depend on the Lea–Leask–Wolf (LLW) parameter x which characterizes a cubic crystalline field [12]. We define pseudospin operators $\boldsymbol{\tau}$ and $\boldsymbol{\sigma}$: $\tau^z|\pm\sigma_z\rangle = \pm|\pm\sigma_z\rangle$, $\sigma^z|\tau_z\uparrow\rangle = +|\tau_z\uparrow\rangle$, $\sigma^z|\tau_z\downarrow\rangle = -|\tau_z\downarrow\rangle$. The multipolar operators can be written in terms of $\boldsymbol{\tau}$ and $\boldsymbol{\sigma}$:

$$J^\alpha = \tfrac{1}{2}(\xi + \eta T^\alpha)\sigma^\alpha, \tag{4}$$

$$O_2^0 = 3J^{z2} - J(J+1) = -\tfrac{3}{2}(\xi - \eta - 9)\tau^z, \tag{5}$$

$$O_2^2 = J^{x2} - J^{y2} = \frac{\sqrt{3}}{2}(\xi - \eta - 9)\tau^x, \tag{6}$$

where $\alpha = x$, y, or z, $T^{x,y} = -\tau^z/2 \mp \sqrt{3}\tau^x/2$, $T^z = \tau^z$, $\xi = 5b_1^2/3 - b_2^2 + 4\sqrt{21}b_1b_2/3$, and $\eta = -10b_1^2/3 + 2b_2^2 + 4\sqrt{21}b_1b_2/3$. According to Raman spectroscopy [9], the LLW parameter is found to be $x = -0.82$ for NdB_6, and then $b_1 = 0.924$, $b_2 = -0.382$, $\xi = -0.883$, and $\eta = -4.71$. We use these values in the following analysis.

We consider the A-type ordered ground state (the ordering vector $\boldsymbol{Q}_A = (0, 0, 1/2)$ in units of $2\pi/a$, where a is the lattice constant) which was observed in a neutron diffraction experiment on NdB_6 [5]. Among the eigenstates of J^z (2) and (3), the ground state should be $|+\uparrow\rangle$ for the A sublattice and $|+\downarrow\rangle$ for the B sublattice, because the $\tau^z = +1$ states have larger $|J^z|$ than the $\tau^z = -1$ states and are energetically favourable. The ratio of transition probabilities is given by

$$\frac{|\langle +\downarrow|J^{x,y}|+\uparrow\rangle|^2}{|\langle -\downarrow|J^{x,y}|+\uparrow\rangle|^2} = \left(\frac{2\xi - \eta}{\sqrt{3}\eta}\right)^2 = 0.130, \tag{7}$$

and $|\langle -\uparrow|J^{x,y}|+\uparrow\rangle| = 0$. Experimentally, only one excitation mode was observed, which we interpret as coming from the excited state $|-\downarrow\rangle$ for the A sublattice with dominant transition probability. For the B sublattice, we consider only $|-\uparrow\rangle$ as an excited state. Possible mixing between excitation modes should result in a double-peaked spectrum for some wavenumbers even though the probability of another transition is negligible. In view of the absence of such a feature, we neglect the mode mixing in the following analysis.

We introduce a pseudospin which describes the ground state and the excited states mentioned above: $\rho^z|+\uparrow\rangle = +(1/2)|+\uparrow\rangle$, $\rho^z|-\downarrow\rangle = -(1/2)|-\downarrow\rangle$ for the A sublattice, and $\rho^z|+\downarrow\rangle = -(1/2)|+\downarrow\rangle$, $\rho^z|-\uparrow\rangle = +(1/2)|-\uparrow\rangle$ for the B sublattice. Then the model Hamiltonian takes the form of an anisotropic Heisenberg model in a magnetic field:

$$H = -2\sum_{(i,j)} J_{ij}[\rho_i^x\rho_j^x + \rho_i^y\rho_j^y + D\rho_i^z\rho_j^z] - 9(\xi - \eta - 9)^2\sum_{(i,j)} K_{ij}\mathrm{e}^{\mathrm{i}\boldsymbol{Q}_A\cdot\boldsymbol{R}_{ij}}\rho_i^z\rho_j^z - \sum_i h_i\rho^z, \tag{8}$$

where $J_{ij} = (3\eta^2/8)I_{ij}$, $D = 4\xi^2/(3\eta^2)$, $h_i = h_{\mathrm{eff}}$ ($h_i = -h_{\mathrm{eff}}$) for the A sublattice (B sublattice), and $h_{\mathrm{eff}} = (4\xi/(3\eta))J(\boldsymbol{Q}_A)$. The quadrupolar interaction K_{ij} affects the magnetic excitation. By using the Holstein–Primakoff transformation, we obtain the excitation spectrum:

$$\omega_q^2 = (\Delta - J(\boldsymbol{q}))(\Delta - J(\boldsymbol{q} + \boldsymbol{Q}_A)), \tag{9}$$

$$\Delta = DJ(\boldsymbol{Q}_A) + \frac{9}{2}(\xi - \eta - 9)^2 g_3' + h_{\mathrm{eff}} = \frac{4\xi(\xi + \eta)}{3\eta^2}J(\boldsymbol{Q}_A) + \frac{9}{2}(\xi - \eta - 9)^2 g_3', \tag{10}$$

where $g_3' = K(\boldsymbol{0})$.

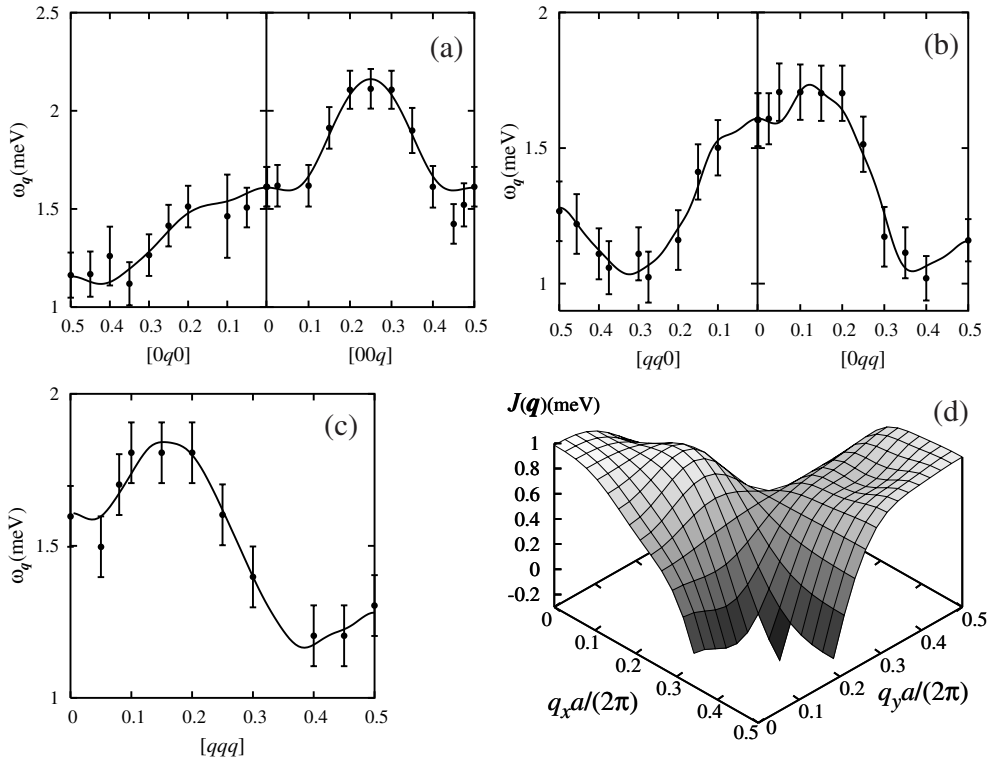

Figure 1. ((a)–(c)) The magnetic excitation spectrum of NdB_6 along symmetry directions. Solid circles are experimental data [6, 7], and the solid curve is obtained in the present work. (d) $J(\boldsymbol{q})$ in the $q_z = 0.5 \times 2\pi/a$ plane.

3. Result

The error bars of the experimental spectrum are rather large, and it does not seem sensible to take the centre of each error bar for the fit. Therefore we first draw a smooth curve which seems to be the best guess for the actual spectrum. Then we fit the smooth curve with ω_q given in (9) using the least-squares method. Obviously there is some ambiguity in guessing the curve to fit. However, a different choice of the curve hardly changes the overall feature of $J(\boldsymbol{q})$. The RKKY interaction has a long range in general, and we include a sufficient number (=34) of dipolar interactions to fit the excitation spectrum. We also include the quadrupolar interaction $g_3' = K(\boldsymbol{0})$. The result of the fitting is shown in figure 1. The experimental spectrum is well reproduced by the fit. In the present analysis, we also take into consideration the experimental data along the $\langle 111 \rangle$ direction [7] (see figure 1(c)), which were not published, and which we did not include in the previous study [13].

In the mean field theory, the Néel temperature of the model (1) is given by

$$T_{\mathrm{N}}^{\mathrm{MF}} = \frac{2}{3}\frac{\xi^2 + \eta^2}{\eta^2} J(\boldsymbol{Q}_{\mathrm{A}}). \tag{11}$$

The fit gives $J(\boldsymbol{Q}_{\mathrm{A}}) = 0.986$ meV and $T_{\mathrm{N}}^{\mathrm{MF}} = 7.9$ K. The magnitude of the quadrupolar interaction $g_3' = 107$ mK is obtained by our fit. In the mean field theory, the condition for

stabilizing the $\boldsymbol{J} \parallel \langle 001 \rangle$ state against $\boldsymbol{J} \parallel \langle 111 \rangle$ is given by

$$g_3' > g_{3\min}' = \frac{8}{27} \frac{\xi - 2\eta}{\eta(\xi - \eta - 9)^2} J(\boldsymbol{Q}_A), \tag{12}$$

and $g_{3\min}'$ is estimated to be 79 mK by using the fitted value of $J(\boldsymbol{Q}_A)$. Thus $g_3' > g_{3\min}'$ in our fit, and this is consistent with the experimental observation of the direction of $\boldsymbol{J}$. We note that the value $g_3' = 107$ mK agrees well with the experimental value of about 100 mK [9]. Figure 1(d) shows $J(\boldsymbol{q})$ in the $q_z = 0.5 \times 2\pi/a$ plane. We note that $J(\boldsymbol{q})$ has a maximum at $\boldsymbol{q} = \boldsymbol{Q}_A$, consistent with the A-type structure. The local maximum at $\boldsymbol{q} \simeq (0.15, 0.15, 1/2)$ is related to the softening of the excitation spectrum around $\boldsymbol{q} \simeq (0.3, 0.3, 0)$ (figure 1(b)) which is equivalent to $(0.3, 0.3, 1/2)$ in the A-type ordered state.

In summary, we have extracted the magnetic and quadrupolar interactions by fitting the excitation spectrum of NdB_6. The transition temperature T_N^{MF} and the quadrupolar interaction g_3' obtained from the fit agree well with experimental values. The ferroquadrupolar interaction is indispensable for aligning $\boldsymbol{J} \parallel \langle 001 \rangle$, and the value $g_3' = 107$ mK obtained is consistent with the observed direction of $\boldsymbol{J}$.

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