Magnetic structure of $TmNi_2B_2C$

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Neutron-diffraction measurements have been carried out to determine the nature of the magnetic ordering in $TmNi_2B_2C$. $TmNi_2B_2C$ exhibits superconductivity below 11 K and magnetic ordering below 1.5 K. The magnetic structure is incommensurate and consists of ferromagnetic (110) planes of Tm moments aligned along the c axis with the magnitude of the moments modulated sinusoidally along the (110) direction, the modulation wave vector having a magnitude of 0.241 $Å^{-1}$. The magnetic structure as well as the modulation direction exhibited by $TmNi_2B_2C$ is different from that of other RNi_2B_2C (R=Gd–Er) compounds. The modulated state formed by the Tm moments allows the superconductivity to coexist with the magnetic ordering below T_N. [S0163-1829(96)00538-3]

RNi_2B_2C compounds, with R=magnetic rare-earth ion (Gd–Tm), have been studied very extensively[1–21] due to their interesting superconducting and/or magnetic properties at low temperatures. The compounds with R=Tm, Er, Ho, and Dy show superconductivity with relatively high T_c's (11 K for Tm and 6.5 K for Dy), which coexists with the magnetic ordering (1.5 K for Tm and 10.5 K for Dy) of the rare-earth moments. The compounds formed with Tb (Ref. 19) and Gd (Refs. 20 and 21) do not exhibit superconductivity, but do show magnetic ordering with T_N=15.5 and 20 K for Tb and Gd, respectively. These compounds form in the body-centered tetragonal structure[22] (space group $I4/mmm$) with alternate layers of R-C and Ni_2-B_2. Due to this layered structure, the conduction electrons in the Ni_2-B_2 plane may be partially shielded from the magnetic moments of the rare-earth ions giving rise to the possibility of coexistence for the superconductivity and the magnetic ordering.

Neutron-diffraction measurements have confirmed the nature of magnetic ordering in the Er,[8,9] Ho,[5–7] Dy,[14] and Tb (Ref. 23) compounds. $DyNi_2B_2C$ below T_N and $HoNi_2B_2C$ below 5 K show a simple commensurate antiferromagnetic (AFM) ordering where the ferromagnetic basal planes are coupled antiferromagnetically along the c axis. In $HoNi_2B_2C$, two additional incommensurate modulations of the magnetic order occur, one with a wave vector along c^* between 5 and 8 K and another along a^* between 5 and 6.5 K. The a modulation is also observed in the Er and Tb compounds. The wave vectors of the modulated magnetic structure along a^* have similar values in all three compounds (0.585, 0.553, and 0.552 for Ho, Er, and Tb, respectively). These values, in turn, are close to the wave-vector value of 0.6 obtained from a normal-state band-structure calculation of the $LuNi_2B_2C$ compound,[24] where a peak in $\chi(q)$ is observed along the a^* direction. In neutron inelastic-scattering measurements of the phonon dispersion in $LuNi_2B_2C$,[25] phonon anomalies associated with soft phonon modes were observed at wave vectors close to the incommensurate magnetic ordering wave vector along a^*. These modulations are taken as an indication of common Fermi-surface nesting features along the a^* direction in RNi_2B_2C compounds which may be responsible for the magnetic ordering of the rare-earth moments via the RKKY interaction.

$TmNi_2B_2C$ shows superconductivity below 11 K.[1] Specific-heat measurements show an anomaly which has been associated with the magnetic ordering at 1.5 K.[4] However, no magnetic structure determination has been reported. Detailed magnetization measurements[16] have shown that the magnetic anisotropy in this compound in the superconducting state, as well as the normal state, is different from that observed in other compounds [Ho,[3] Er,[18] Dy,[14,15] and Tb (Ref. 19)]. In all the latter compounds, the magnetization has larger values when the field is applied parallel to the ab plane, implying the magnetic moments are confined to the ab plane. Indeed, the magnetic structures determined from the neutron-diffraction measurements[5–9,14,23] show the magnetic moments in Ho, Er, Dy, and Tb compounds confined in the ab plane. In contrast, $TmNi_2B_2C$ may have a different orientation of the magnetic moments. In this paper, we report the magnetic structure of $TmNi_2B_2C$ obtained from neutron powder diffraction experiments. The compound shows magnetic ordering of the Tm moments below 1.5 K with an incommensurate magnetic structure which consists of ferromagnetically aligned Tm moments along the c axis in the (110) planes and a sinusoidal modulation of the magnitude of the moments along the (110) direction. The modulation wave vector has a magnitude of 0.241 $Å^{-1}$. $TmNi_2B_2C$ is the only compound in the RNi_2B_2C family of compounds to show a modulation in the (110) direction as well as the alignment of the magnetic moments along the c axis.

The samples were prepared by the standard arc-melting method and subsequent annealing. The isotope ^{11}B was used to prepare these samples to avoid the large absorption of neutrons by the naturally occurring ^{10}B. Neutron-diffraction experiments were carried out using the D1B multidetector at the ILL in Grenoble, using an incident wavelength of 2.524 Å. Since $TmNi_2B_2C$ has a magnetic ordering transition below 1.5 K, the experiment was first performed in a dilution

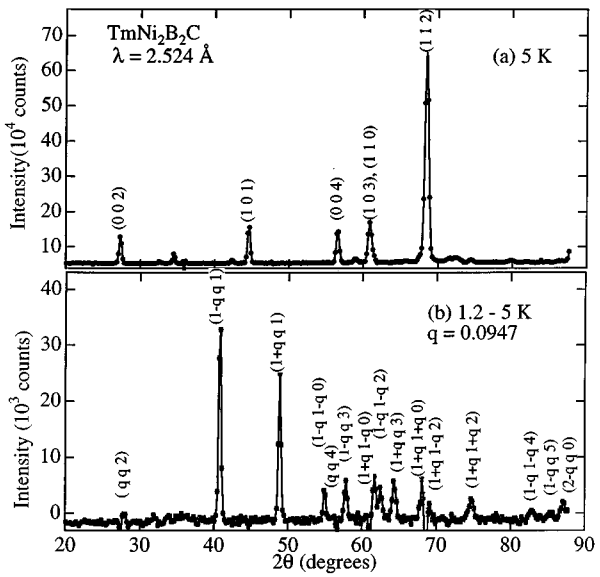

FIG. 1. Neutron-diffraction patterns from $TmNi_2B_2C$ at (a) 5 K for the nuclear peaks and (b) 1.2–5 K for the magnetic peaks.

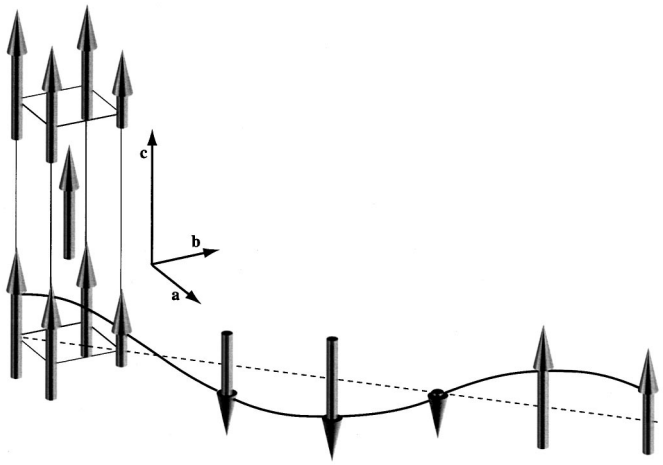

FIG. 2. Magnetic structure of $TmNi_2B_2C$ below 1.5 K. Tm moments are aligned ferromagnetically along the c axis in (110) planes and the magnitude of the moments are modulated sinusoidally along the (110) direction.

fridge at 50 mK and 2 K. The sample in the form of ingot was stuck directly on the copper sample holder to ensure temperature homogeneity. The sample was then ground to powder and the spectra were taken in an orange cryostat between 1.2 and 5 K. Both the spectra at 50 mK and 1.2 K were used for the data analysis.

At temperatures above 1.5 K, only nuclear reflections were observed, as shown in Fig. 1(a). The nuclear peaks may be indexed as $h+k+l=$even and were used to estimate the lattice parameters, $a=b=3.490\pm0.002$ Å, $c=10.628\pm0.004$ Å . The contribution from impurity phases was estimated to be less than 5%. As the temperature is lowered below 1.5 K, additional diffraction peaks were found to appear. These magnetic peaks, as shown in Fig. 1(b), can be indexed only if a modulation of the magnetic moments is taken along the (110) direction with the magnitude of the modulation wave vector as 0.241 $Å^{-1}$. The magnetic structure consists of ferromagnetic planes of Tm moments aligned along the c axis and modulated along the (110) direction as shown in Fig. 2. Since no resolved higher-order peaks (third or fifth) were observed, a simple sinusoidal modulation has been assumed in our model to calculate the magnetic intensities. The intensities of the magnetic peaks were calculated using the equation[26,27]

$$I_{\mathrm{calc}}=2\times\left(\frac{e^2\gamma}{2mc^2}\right)^2 jA(\theta_B)L(\theta_B)\left|\sum_n\left[\mathbf{m}_n-\mathbf{k}(\mathbf{k}\cdot\mathbf{m}_n)\right]f_n e^{-2\pi i(\mathbf{G}\cdot\mathbf{r}_n)}\right|^2,$$

where j, $A(\theta_B)$, $L(\theta_B)$, and f are the multiplicity, absorption, Lorentz, and magnetic form factors, respectively. **G** is a reciprocal-lattice vector defined by (h,k,l), **k** is the unit vector normal to the reflection plane, $\mathbf{m}_n$ is the magnetic moment vector, and $\mathbf{r}_n$ is the position of the nth magnetic moment. The factor 2 arises from the contribution of both the (110) and $(1\bar{1}0)$ domains. Table I lists the values of the calculated and observed intensities along with their indexing. Several models for the magnetic structure were considered while calculating the intensities of the observed magnetic peaks, for example, magnetic moments forming a spiral in the a-b plane modulating along the (110) direction from plane to plane, a conelike modulation along the (110) direction with the magnetic moments arranged on the surface of the cone at an angle to the c axis, etc. However, only one model given by

$$\mathbf{m}_n=\tfrac{1}{2}\cos(\mathbf{q}\cdot\mathbf{r}_n)|m_n|\hat{\mathbf{z}},$$

fits all the intensities of the magnetic peaks adequately where z is a unit vector along the c axis and **q** is the modulation wave vector. The factor, 1/2, arises from the assumption that the two domains are equally populated. The large error bars on the $(qq2)$, $(qq4)$, and $(1+q1+q0)$ peaks result from the subtraction of intense nuclear peaks which also occur at those positions. The form factor of free Tm^{3+} was used in our intensity calculations, even though the crystal-field effects in this compound, which are evident from the anisotropy of magnetic susceptibility,[16] may affect this value slightly. The magnetic moment of Tm^{3+} is estimated to be $3.74\mu_B\pm0.02$ and $4.8\mu_B\pm0.1$ at 1.2 K and 50 mK, respectively. The magnetic moment at 50 mK was estimated by scaling the magnitude of the moment at 1.2 K by the ratio of the intensities of the six lowest angle, clearly resolved, magnetic Bragg reflections. The ratio was constant within experimental accuracy for all six reflections indicating no change in the magnetic structure between the two temperatures. This

TABLE I. Comparison of the experimental (I_{obs}) and calculated (I_{calc}) intensities of the magnetic Bragg peaks at 1.2 K for $TmNi_2B_2C$ for neutron wavelength $\lambda = 2.524$ Å and $q = 0.0947$.

2θ	(hkl)	I_{obs}	I_{calc}
27.92	$(qq2)$	816 ± 234	710
40.79	$(1-qq1)$	$15\,724 \pm 243$	16 132
48.87	$(1+qq1)$	$11\,342 \pm 232$	10 700
54.95	$(1-q1-q0)$	2347 ± 237	2315
56.97	$(qq4)$	-619 ± 529	44
57.84	$(1-qq3)$	3458 ± 255	4231
61.68	$(1+q1-q0)$	4006 ± 297	4055
62.50	$(1-q1-q2)$	2899 ± 280	2871
64.34	$(1+qq3)$	3986 ± 283	3808
68.08	$(1+q1+q0)$	2913 ± 1500	— [a]
	$(1+q1-q2)$	—	—
74.70	$(1+q1+q2)$	3136 ± 358	2226
82.90	$(1-q1-q4)$	1521 ± 372	1078
85.38	$(1-qq5)$	1201 ± 406	1088
87.11	$(2-qq0)$	2596 ± 398	2245

[a]These two peaks were not included in the fit due to the large uncertainty from the subtraction of the nuclear background (see Fig. 1).

procedure was adopted since the ''background'' signal from the dilution fridge contained numerous Bragg reflections from Cu and Al components of the cryostat at higher angles which prevented an accurate refinement of both the nuclear and the magnetic data. It is also possible that the reduced value of the magnetic moment, when compared with the free-ion value ($7.3\mu_B$), is due to CEF effects in this compound.

The magnetic structure of $TmNi_2B_2C$ is quite different to that found for the other RNi_2B_2C (R=Er, Ho, Dy, and Tb) compounds. The latter compounds exhibit antiferromagnetic ordering with the magnetic moments confined to the ab plane. In $TmNi_2B_2C$, the moments are aligned along the c axis. Modulation of the magnetic ordering along a is observed for Er, Ho, and Tb compounds. However, the modulation in $TmNi_2B_2C$ is along the (110) direction. The Fermi-surface nesting features along the a^* direction,[24] considered responsible for the magnetic ordering of the rare-earth moments in RNi_2B_2C compounds, may be different in this compound. It is also possible that the nesting of the Fermi surface in this compound is considerably altered when the magnetic ordering takes place. A band-structure calculation for this compound will be particularly interesting to confirm whether such a Fermi surface alteration really occurs. However, it should be mentioned here that the band-structure calculations[24] did not include any matrix elements and apparent peaklike features are observed at other wave-vector values also. The phonon anomalies which occur along a^* may also be different from that in $LuNi_2B_2C$,[25] and hence an investigation of the phonon dispersion along the (110) direction in this compound would be useful in obtaining a complete picture of the role of the phonon modes in RNi_2B_2C compounds.

In conclusion, we have shown that in $TmNi_2B_2C$, the Tm moments order below 1.5 K with an incommensurate magnetic structure consisting of Tm moments, aligned ferromagnetically along the c axis in the (110) planes, and the magnitude of the moments modulated sinusoidally along the diagonal of the ab plane. It is possible that the modulation in the magnitude of the Tm moments allows the magnetic order to coexist with the superconducting state. $TmNi_2B_2C$ is the only compound in the family of RNi_2B_2C compounds where the modulation is along the (110) direction and magnetic moments aligned along the c axis. The Fermi surface nesting in this compound may be different compared to the other compounds in this series which needs further investigation.

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