Magnetic structure of $TbNi_2B_2C$

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Neutron-diffraction techniques have been used to study the magnetic structure of $TbNi_2B_2C$. The measurements, performed on single crystals of this compound, show that below approximately 15 K the moments order in an almost longitudinal spin wave with wave vector along **a***. The magnitude of the wave vector is close to that obtained in the Ho, Er, and Gd compounds. This observation provides additional evidence that there are common Fermi-surface nesting features along **a*** in the rare-earth nickel boride carbides which cause the magnetic ordering of the rare-earth moments via the Ruderman-Kittel-Kasuya-Yosida mechanism. Below approximately 8 K, the experimental results indicate the development of a small ferromagnetic component.

I. INTRODUCTION

The compounds of the RNi_2B_2C family (where R stands for a rare-earth element)[1–4] exhibit very interesting physical properties at low temperatures. The structure[3] of these compounds is body-centered-tetragonal (space group $I4/mmm$) and consists of R-C layers separated by Ni_2B_2 sheets. The compounds formed with the nonmagnetic rare-earth elements Lu and Y are superconductors[2–5] with relatively high superconductivity transition temperatures (16.6 and 15.6 K for the Lu and Y compounds, respectively). It is particularly interesting that the compounds formed with magnetic rare-earth elements such as Tm, Er, Ho, and Dy are also superconductors,[2–7] and that superconductivity coexists with magnetic order in these compounds. The compounds formed with Gd and Tb, on the other hand, order magnetically at low temperatures but they are not superconducting, at least down to approximately 2 K.[8,9]

The magnetic structure of the superconducting Ho, Er, and Dy compounds have been studied by neutron-diffraction techniques[10–13] using samples depleted in the heavily neutron absorbing B^{10} nuclei. The nonsuperconducting, down to 2 K, Gd compound has been recently studied by resonant magnetic x-ray scattering.[14] One of the most interesting results of these experiments is that in the magnetic state of Ho, Er, and Gd compounds an incommensurate modulation of the moments along **a*** is observed with wave vectors of 0.585, 0.553, and 0.553, respectively. This result suggests that there are common Fermi-surface nesting features along **a*** in these compounds which cause the magnetic ordering of the rare-earth moments via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism.

Electronic band-structure calculations[15–18] indicate that these compounds are three-dimensional metals with relatively high density of states at the Fermi level. These calculations,[15–18] as well as the recent observation[19] of soft phonons in $LuNi_2B_2C$, provided convincing evidence that the superconducting compounds of this family are strongly coupled conventional superconductors. More pertinent to an understanding of the magnetic ordering in these compounds is a recent calculation[20] (without matrix elements), of the generalized electronic susceptibility $\chi(q)$ of $LuNi_2B_2C$ based on the normal-state electronic band structure of this compound. It is encouraging that the calculated[20] $\chi(q)$ exhibits a peak along **a*** with a wave vector close to those observed in the Ho, Er, and Gd compounds.

In this paper we present the results obtained in a neutron-diffraction study of the magnetic ordering in the Tb compound which, like Gd, is not superconducting at least down to 2 K. Magnetization measurements[9] clearly show that this compound orders antiferromagnetically below approximately 15 K and that below approximately 8 K a weak ferromagnetic component may be developing.[9] Of particular interest in the present study, of course, is whether the magnetic structure of this compound at low temperatures includes an incommensurate modulation with wave vectors along **a*** and the determination of the magnitude of this wave vector.

II. EXPERIMENTAL DETAILS

Single-crystal specimens, depleted in the strongly neutron absorbing B^{10} nuclei, were grown at the Ames Laboratory by the high-temperature flux technique as described elsewhere.[21] Neutron-diffraction measurements showed that the as-grown platelets of the RNi_2B_2C compounds prepared at the Ames Laboratory were indeed single crystals of high quality (measured mosaic spread of less than 0.1°) with the c axis perpendicular to their flat surface. The $TbNi_2B_2C$ crystal used in the present experiment was a 3×2 mm^2 platelet of 0.5 mm thickness.

The neutron-diffraction experiments were performed using the HB1A triple-axis spectrometer at the High Flux Isotope Reactor of the Oak Ridge National Laboratory. This is a constant incident neutron energy (14.7 meV) spectrometer utilizing a double pyrolytic graphite monochromator and a pyrolytic graphite analyzer. A pyrolytic graphite filter was used to attenuate higher-order contaminations. Measurements, over the 2.3–300 K temperature range, were performed with the crystal oriented so that the a-c crystal plane coincides with the horizontal scattering plane.

The nuclear and magnetic structure factors evaluated from the single-crystal measured intensities are subject to large uncertainties because the latter quantities must be corrected for the effects of secondary extinction and absorption. The extinction problem is particularly severe in the present experiments because of the high perfection of the crystals (full

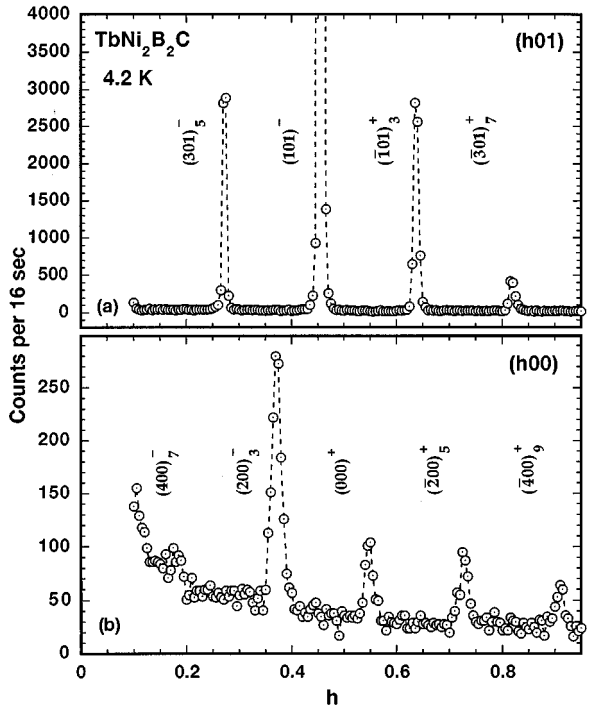

FIG. 1. Typical scans along the $[h01]$ and $[h00]$ symmetry directions. The satellites of a $(h0l)$ reflection are denoted by $(h0l)_n^{\pm}$ where the superscript $+$ (or $-$) means that the magnetic wave vector is added (or subtracted) from the reciprocal vector of the reflection, and the subscript n is the order of the satellite.

width at half maximum $\simeq 0.1°$) and the fact that as a result of magnetostriction, the mosaic of the crystals changes as the temperature is reduced below the magnetic transition temperature. The absorption correction is also subject to large uncertainties because of the irregular shape of the platelets and its extreme sensitivity to the exact amount of B^{10} contained in the depleted samples.

III. RESULTS AND DISCUSSION

At temperatures above approximately 14 K only nuclear reflections with $h+k+l=2n$ are observed as expected from the crystal structure of the compound. Therefore, at temperatures above approximately 15 K the magnetic moments in $TbNi_2B_2C$ are not ordered.

As the temperature is lowered below approximately 15 K additional diffraction peaks (see Fig. 1) start to develop in rows parallel to the reciprocal a axis (or the equivalent b axis of this tetragonal structure). These additional diffraction peaks can be indexed as first- and higher-order satellites of the allowed nuclear reflections $(h+k+l=2n)$. These observations imply that below approximately 15 K the magnetic moments are ordered in a modulated structure with wave vector parallel to the reciprocal a axis. The magnitude of the wave vector changes (see inset of Fig. 2) from 0.551 ± 0.001 at 15 K to 0.545 ± 0.001 at 2.3 K, the lowest temperature reached in the present experiment.

It can be seen (Fig. 1) that the intensities of the $(h00)$ satellites are considerably lower than those of the corresponding $(h0l)$ satellites. This implies that the direction of the moments is close to that of the **a*** axis. From the observed intensities the magnitude of the transverse component of the moment was estimated (see Sec. II) to be approximately 40 times smaller than the longitudinal component. Thus, below approximately 15 K the magnetic structure of $TbNi_2B_2C$ can be described as a nearly longitudinal spin wave with propagation wave vector parallel to the reciprocal a axis. This almost longitudinal spin wave is not purely sinusoidal but considerably squared as shown by the observation of relatively intense higher-order harmonics. Actually the estimated (see Sec. II) ratios of the moments of the third and fifth harmonics to that of the first harmonic are close to those expected (1/3 and 1/5, respectively) for a completely squared spin wave. Below 15 K the intensity of the observed satellites increases with decreasing temperature and approaches saturation below approximately 6 K (Fig. 2). No significant hysteresis effect on the observed intensities (Fig. 2) was found within the precision of the present experiment.

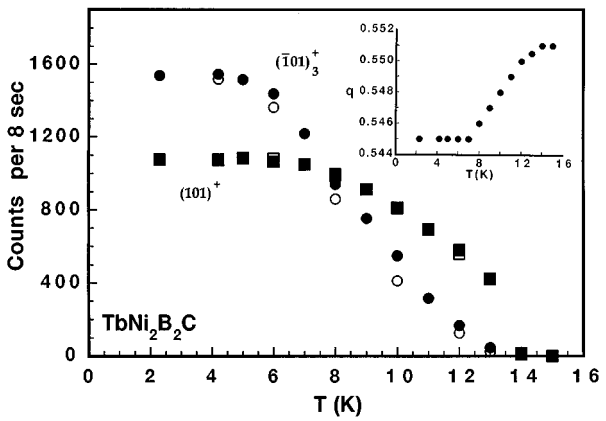

FIG. 2. Temperature dependence of the first- $(101)^+$, and third-$(\bar{1}01)_3^+$, order satellites. The data with open (filled) symbols were taken with decreasing (increasing) temperature.

To examine whether a ferromagnetic component develops below 8 K as indicated by low-field magnetization measurements[9] we recorded the temperature dependence of the intensities of the (006) and (105) reflections, which have the smallest intensities among all reflections that could be reached with the present experimental arrangement. The in-

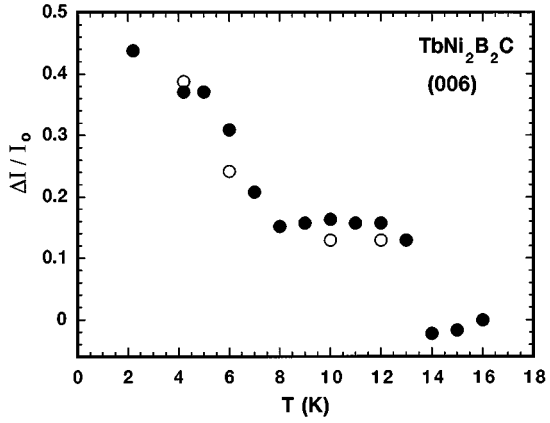

FIG. 3. Temperature dependence of the relative change $\Delta I=I-I_0$ of the (006) nuclear reflection. I_0 stands for the intensity at 16 K.

tensities of these nuclear reflections (Fig. 3) exhibit a small anomaly at the antiferromagnetic transition (a feature common to all nuclear reflections), remain constant down to approximately 8 K, and then increase with decreasing temperature. This result suggests the presence of a small ferromagnetic component below approximately 8 K in agreement with low-field magnetization results.[9] It is clear, however, that only polarized neutron experiments, presently in preparation, can provide a definitive proof of the existence of a ferromagnetic component below 8 K.

The magnitude of the wave vector of the modulated magnetic structure of $TbNi_2B_2C$ is very close to the values obtained for Ho, Er, and Gd (0.585, 0.553, 0.553, respectively); actually, the magnitude of the wave vector close to the transition (0.551) is almost identical to the magnitudes observed in Er and Gd. Therefore, the present results provide additional evidence that there are common Fermi-surface nesting features along **a*** in the RNi_2B_2C family that are responsible for the magnetic ordering of the rare-earth moments in these compounds via the RKKY mechanism. It is important to notice that the generalized electronic susceptibility $\chi(q)$ of $LuNi_2B_2C$ calculated by Rhee, Wang, and Harmon[20] using the normal-state electronic band structure of this compound, exhibits a pronounced peak along **a*** at a wave vector (0.6) close to the observed values of Ho, Er, Gd, and Tb compounds. Presumably, this nesting of the Fermi surface is not substantially altered by the opening of antiferromagnetic and, in the case of Ho and Er compounds, superconducting gaps at the Fermi level. Finally, it should be pointed out that the same Fermi-surface nesting feature may be responsible for the strong phonon anomalies observed[19] in the low-lying $[\xi 00]$ dispersion curves of $LuNi_2B_2C$. If this is the case, phonon softening and magnetic ordering are competing to decrease the energy of the system in the magnetic compounds of this family as a result of this nesting of the Fermi surface.

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