Anisotropic magnetic properties of $TbNi_2B_2C$ single crystals

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Magnetic and transport properties of single crystals of $TbNi_2B_2C$ have been investigated by ac susceptibility, dc magnetization, specific heat, and resistance measurements. The compound shows highly anisotropic magnetic properties which come about as a result of the Tb magnetic moments lying predominantly in the ab plane. The ac susceptibility and low-field dc magnetization measurements indicate the presence of two magnetic transitions; one due to the antiferromagnetic (AFM) ordering of the Tb moments around 15 K, and another at 5 K, which we believe is due to a spin reorientation of the Tb moments. The resistivity (ρ_{ab}) measurements show a sharp decrease in the resistance at 15 K but contain no observable features corresponding to the lower-temperature transition. The compound does not show superconductivity above 300 mK. Specific heat at low temperatures shows a large λ-type anomaly at 13.8 K in addition to the anomalies associated with the AFM ordering of the Tb moments at 15 K (T_{AF}) and the spin reorientation at 5 K. Magnetization (M) and $\rho_{ab}(T)$ measurements indicate a decrease in T_{AF} as a function of applied field (H) for $H\|ab$, whereas for $H\|c$, T_{AF} is independent of field. The M-H isotherms at 2 K show that for $H\|ab$ the compound goes through a series of temperature-dependent metamagnetic states before finally saturating into a ferromagnetic state for $H>21$ kOe. For $H\|c$ the compound shows a linear M-H behavior expected for a normal AFM compound. The $\rho_{ab}(H)$ measurements show anomalies due to the metamagnetic transitions and a large negative magnetoresistance above 21 kOe for $H\|ab$.

INTRODUCTION

The discovery of the RNi_2B_2C (R=rare earth) family of magnetic superconductors has opened up the possibility of studying the interplay between superconductivity and long-range magnetic order.[1,2] These compounds form for most of the R ions[2] and show superconductivity for magnetic (Tm, Er, Ho, and Dy; T_c=10.8, 10.5, 8.5, and 6.5 K, respectively)[2–5] as well as nonmagnetic rare-earth ions (Y and Lu; T_c=16 and 15.5 K, respectively).[2] The layered structure[6] of these compounds (tetragonal, space group $I4/mmm$), which resembles that of the high-temperature copper-oxide superconductors, consists of R-C layers well separated from the conducting Ni_2-B_2 layers. However, the band-structure calculations[7] indicate that these compounds are closer to conventional superconductors than to the high-T_c oxides. As a result of the layered structure, the conduction electrons are partially isolated from the magnetic R moments, giving rise to the possibility that these materials may exhibit both magnetic ordering and superconductivity. Neutron-diffraction and specific-heat measurements have confirmed the coexistence of magnetic ordering of the R ion moments (R=Tm, Er, Ho, and Dy) with the superconducting state.[8–14] The materials containing Tm, Er, and Ho order magnetically (T_N=1.5, 5.9, and 6 K, respectively) after entering the superconducting state, while the Dy compound becomes superconducting (T_c=6.5 K) in the magnetically ordered state (T_N=10.5 K). Evidence that a degree of interaction between the magnetic moments of the R ions and the conduction electrons, present in these materials, comes from the fact that the compounds with nonmagnetic rare-earth ions (Y and Lu) have the highest T_c, while for the magnetic R ions, T_c decreases from Tm through to Dy. Detailed magnetization measurements have been reported for RNi_2B_2C compounds with R=Tm, Er, and Ho,[15–17] which demonstrate the anisotropic properties of these compounds. In the case of Ho and Er compounds, the anisotropy results in a Curie-Weiss susceptibility for $H\|ab$ and a much weaker, temperature-dependent susceptibility for $H\|c$ at low temperatures. For $TmNi_2B_2C$, anisotropy exists between 2 and 300 K, but with a larger value of susceptibility for $H\|c$ than for $H\|ab$. For $ErNi_2B_2C$, a change in the sign of anisotropy is observed, with $\chi\|ab<\chi\|c$ for $T>150$ K. In all the cases, crystal-field effects are cited as an explanation for the observed anisotropy.

The slow decrease in T_c observed moving across the RNi_2B_2C series of compounds from Tm to Dy (10.5 to 6.5 K) suggests that $TbNi_2B_2C$, the next compound in the series, may be expected to become superconducting at finite temperatures. The magnetic behavior exhibited by this class of materials makes it likely that $TbNi_2B_2C$, which orders magnetically around 15 K,[2] will also exhibit interesting anisotropic magnetic behavior. These facts, together with the availability of good quality single crystals of this material have motivated us to study this compound. Our earlier investigations of $DyNi_2B_2C$ (Ref. 3) have shown that phase purity plays an important role in the observation of superconductivity in a magnetically ordered state. In a careful search for a superconducting phase in $TbNi_2B_2C$ we have investigated various nonstoichiometric and stoichiometric polycrystalline materials[18] as well as stoichiometric single crystals. The results of our resistance measurements show no evidence for superconductivity above 300 mK in any of the materials tested. However, as expected, the $TbNi_2B_2C$ single crystals do display interesting anisotropic magnetic properties. Here we report on detailed resistance, ac susceptibility, dc magnetization, specific heat, and magnetoresistance measurements on $TbNi_2B_2C$ single crystals which highlight the magnetic properties of this material. Magnetization measurements show that $TbNi_2B_2C$ exhibits a second magnetic transition at 5 K in addition to the magnetic transition due to the magnetic ordering of the Tb moments at 15 K. A comparison of the magnetization values at these two transitions suggests the possibility of a spin reorientation of the Tb moments at 5 K with an enhanced magnetic component. It is possible that this spin reorientation could be one of the reasons for the absence of superconductivity in $TbNi_2B_2C$.

EXPERIMENTAL DETAILS

Polycrystalline samples of $TbNi_2B_2C$ were prepared by the standard arc-melting method under argon gas on a water-cooled copper hearth. The samples were wrapped in Ta foil, sealed in evacuated quartz tubes, and annealed at 1050 °C for 24 h. Single crystals of $TbNi_2B_2C$ were grown from these annealed buttons by the flux method using an equal amount of Ni_2B as flux.[19] Several sizeable, good quality single-crystal platelets were separated from the flux. The crystals were characterized by x-ray Laue photographs as having their *c* axis pointing out of the plane of the platelet. Single crystals weighing between 4 and 6 mg were used for the measurements. A standard four-probe dc method was used to measure the resistance in the *ab* plane of the crystals. A Heliox cryostat (Oxford Instruments) was used for the zero-field resistance measurements down to 300 mK. The magnetoresistance measurements were carried out at temperatures of between 2 and 25 K in magnetic fields up to 80 kOe, with the field applied parallel and perpendicular to the plane of the crystal. A mutual inductance method was employed to measure the ac susceptibility using a measuring frequency of 113 Hz and an ac excitation field of 2 Oe. Heat-capacity data were collected using a relaxation method from 2 to above 20 K. The dc magnetization was measured from 2 to 300 K in magnetic fields of up to 120 kOe using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design) and a vibrating sample magnetometer (Oxford Instruments).

RESULTS AND DISCUSSION

The ac susceptibility and low-field dc susceptibility of a $TbNi_2B_2C$ single crystal are shown in Figs. 1 and 2. Both the figures clearly indicate the presence of two magnetic transitions, one at 15 K and another around 5 K. The transition at 15 K (T_{AF}) corresponds to the magnetic ordering of the Tb moments. This has been confirmed independently via neutron-diffraction measurements, which show that the compound orders antiferromagnetically below 15 K.[20] The low-temperature transition is present in all the $TbNi_2B_2C$ crystals and is independent of frequency in ac susceptibility measurements between 10 Hz and 3 kHz. As we shall see shortly this transition is field dependent and is not observed for higher fields (>5 kOe). It is possible that this transition is caused by

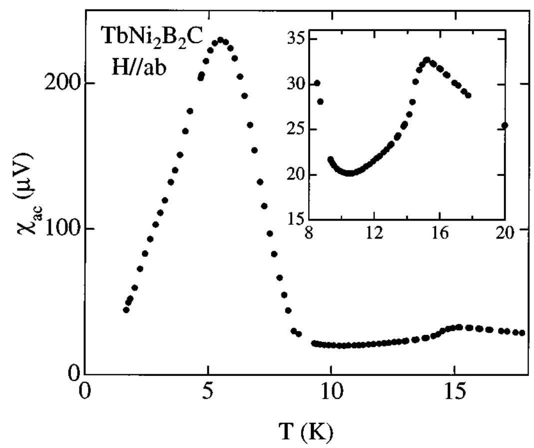

FIG. 1. Temperature variation of the real part of the ac susceptibility for a $TbNi_2B_2C$ crystal with the ac field applied parallel to the *ab* plane of the crystal. The transition at 5 K corresponds to the spin reorientation of the Tb moments. The inset shows the expanded version of the graph to highlight the magnetic ordering of the Tb moments at 15 K.

a reorientation of the Tb moments in the magnetically ordered state. Figure 3 shows the results of the zero-field resistance measurements along the *ab* plane. The compound shows metallic behavior down to the ordering temperature, where a kink characteristic of a magnetic ordering occurs. Below T_{AF}, the resistance continues to decrease with decreasing temperature flattening off below 5 K. The measurements were extended down to 300 mK (inset of Fig. 3) to search for any superconducting phases which may have been present in the sample. The resistance remains constant down

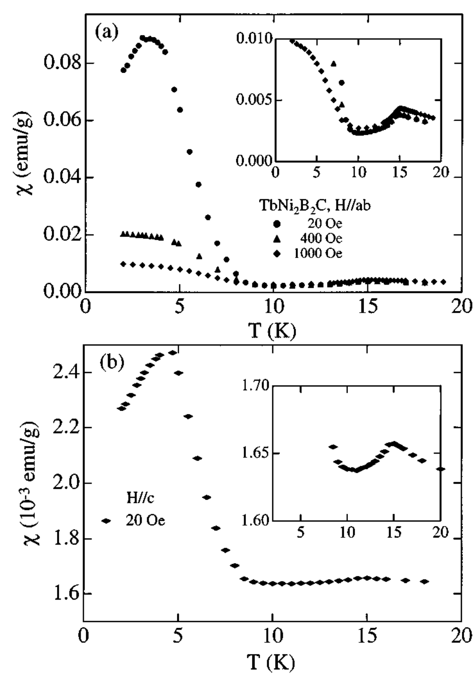

FIG. 2. Dc magnetic susceptibility versus temperature for a crystal of $TbNi_2B_2C$ with the magnetic field applied (a) parallel to the *ab* plane and (b) parallel to the *c* axis. The insets show the expanded version of the graphs to highlight the magnetic ordering of the Tb moments at 15 K.

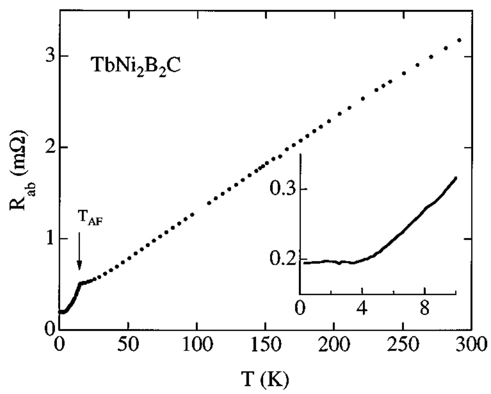

FIG. 3. Electrical resistance in the *ab* plane versus temperature for a $TbNi_2B_2C$ crystal. The kink at 15 K corresponds to the magnetic ordering of the Tb moments. The inset shows the extended resistance measurements down to 300 mK.

to the lowest temperatures, reflecting the residual resistance of the material. No feature indicating the presence of a superconducting phase was detected in our measurements.

Specific-heat measurements were made on a single crystal of $TbNi_2B_2C$, weighing 5.6 mg. The lattice contribution was subtracted from the total specific heat using a Debye temperature of 300 K, obtained from the data for the nonmagnetic YNi_2B_2C compound in Ref. 13 after correction for the relative masses of Tb and Y ions. The resulting magnetic contribution (C_m) shown in Fig. 4, contains three distinct anomalies delineating three magnetically ordered regions. The first evidence of magnetic ordering comes from a jump in $C_m(T)$ at T_{AF}=15 K. A much sharper λ-like first-order transition follows at T=13.8 K. No evidence for a transition at this temperature is seen in the transport data. However, there is a change of slope in the temperature dependence of the dc magnetization which can be more clearly seen by plotting dM/dT as a function of temperature (see Fig. 5). The separation of these two transitions is more obvious in

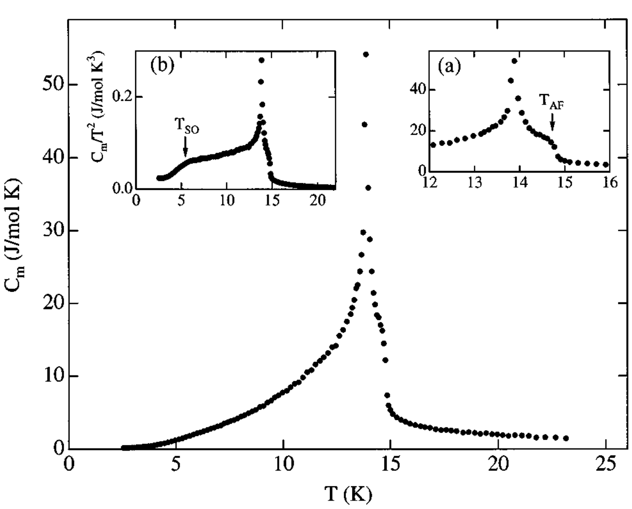

FIG. 4. Magnetic contribution of the specific heat (C_m) versus temperature for a $TbNi_2B_2C$ crystal. The inset (a) shows an expanded scale of C_m vs T to highlight the two anomalies at 13.8 and 15 K. In inset (b), C_m/T^2 is plotted against the temperature (T) to highlight the anomaly due to the spin reorientation at 5 K.

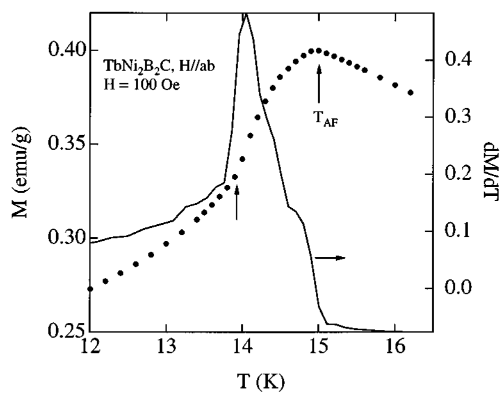

FIG. 5. Magnetization (M) versus temperature (filled circles) for a $TbNi_2B_2C$ crystal with an applied magnetic field of 100 Oe along the *ab* plane. The derivative, dM/dT is shown as the solid line.

the inset (a) of Fig. 4 where C_m versus T is plotted on an expanded temperature scale. The third transition is a subtle anomaly with a broad peak around 5 K. This is more clearly seen if C_m/T^2 is plotted against the temperatures as shown in the inset (b) of Fig. 4. We identify this feature with the strong anomaly observed in the magnetic susceptibility at the same temperature. A number of comments can be made at this stage; firstly the change in $C_m(T)$ at the antiferromagnetic (AFM) ordering temperature (T_{AF}) is relatively small compared to the λ-like peak which occurs at lower temperature (13.8 K). It is possible that the paramagnetic moments undergo a transition to an amplitude-modulated AFM structure in the narrow temperature range 13.8 K$<T<T_{AF}$ before going into another AFM spin arrangement at lower temperatures. This speculation stems from the studies of $HoNi_2B_2C$ where a similar shoulder in the specific heat[17] is found to originate from a modulation of the magnetic moments in neutron-diffraction measurements.[8–10] It is anticipated that in this kind of scenario, the specific-heat jump ΔC_m at the transition is reduced to some fraction of that expected in a paramagnetic to equal-amplitude spin AFM state, which is either commensurate or incommensurate. The magnetic entropy at the AFM ordering temperature (15 K) is only 9.3 J/mol K and reaches about 10.0 J/mol K at 20 K, which is only half the value expected for the free Tb^{3+} ion with J=6, where $R \ln(2J+1)$=21 J/mol K. This is a sign of the strong influence of the crystalline electric-field (CEF) effects present in this material which will be discussed in more detail below.

The results of the magnetization versus field measurements for applied fields both parallel to the *ab* plane and *c* axis of the crystal are presented in Figs. 6(a) and 6(b). For both the orientations, the magnetization is linear with respect to field for $T>100$ K. For applied fields parallel to the *ab* plane, the magnetization shows anomalies below T_{AF}, going through distinct metamagnetic transitions. These transitions are very clear at T=1.8 K, occurring at 0.6, 13, and 21 kOe. The magnetization eventually shows saturation reminiscent of a ferromagnet and hysteresis effects for increasing and decreasing fields. The saturation moment at 1.8 K is calculated to be 9.18μ_B, which is slightly smaller than the free ion moment of Tb^{3+}(9.72μ_B). The first metamagnetic transition at 0.6 kOe disappears for temperatures above 6 K. This temperature coincides with the temperature range at which the

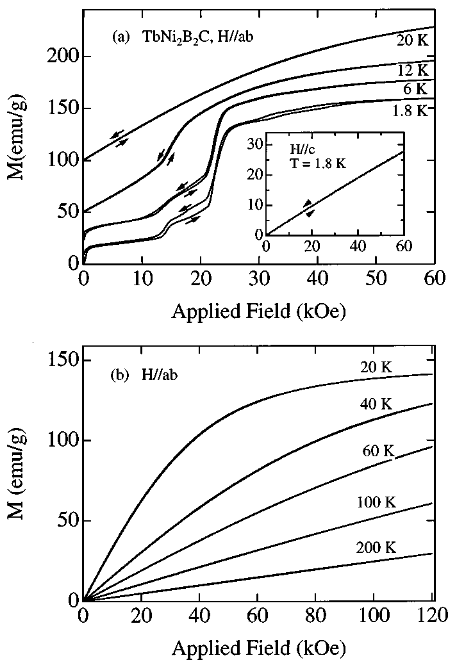

FIG. 6. Magnetization M versus applied field for a $TbNi_2B_2C$ crystal at the indicated temperatures for $H\|ab$. The curves in (a) for $T=6$, 12, and 20 K are shifted along the y axis for clarity with 25, 50, and 100 units, respectively. The inset shows the magnetization at $T=1.8$ K for the same crystal with $H\|c$. Arrows indicate the increasing and decreasing field directions.

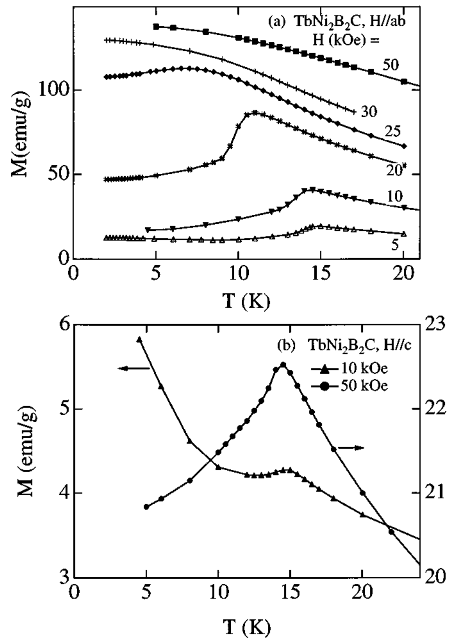

FIG. 7. Magnetization (M) versus temperature for a $TbNi_2B_2C$ crystal at various magnetic fields (a) $H\|ab$ and (b) $H\|c$. In (b), M(emu/g) is plotted on the right axis for $H=50$ kOe. The lines along the curve are a guide to the eye.

magnetic transition due to the spin reorientation occurs. It is quite possible that the first metamagnetic transition is caused by the spin reorientation effects. Above T_{AF}, the magnetization shows saturation or nonlinear behavior for higher fields up to $T=100$ K. The saturation moment at 20 K is nearly the same as at 1.8 K. In contrast, for $H\|c$, the magnetization is proportional to the field at all temperatures, as expected for a simple antiferromagnet or a paramagnet. A representative curve is shown in the inset of Fig. 6(a). The value of the magnetization for $H\|c$ is much smaller than that for $H\|ab$ at a given temperature, indicating that the Tb moments are confined mainly in the ab plane.

The temperature variation of magnetization for higher fields ($\geqslant 5$ kOe) at low temperatures is shown in Fig. 7(a) for $H\|ab$ and in Fig. 7(b) for $H\|c$. For $H\|ab$, the low-temperature magnetic transition is suppressed for $H\geqslant 5$ kOe and the peak corresponding to the Tb ordering becomes dominant. The ordering temperature of Tb moments (T_{AF}) remains the same for $H\leqslant 10$ kOe and then decreases sharply as the applied field is increased. For 20 kOe, the T_{AF} decreases from 15 to around 10 K. As the field is further increased, the peak due to the antiferromagnetic ordering of the Tb moments disappears and is replaced by a response indicating the presence of ferromagnetic correlation between the Tb moments at low temperature. A similar suppression of T_{AF} with the applied fields has been observed in the case of (Er,Ho)Ni_2B_2C.[16,17] In the case of $ErNi_2B_2C$, the variation of T_{AF} with the field follows the equation, $T_{AF}=T_0-AH^2$, where T_0 and A are constants. For $TbNi_2B_2C$, a better fit can be obtained if the H^2 term is replaced by an H^4 term, and the constants obtained are $T_0=14.22$ and $A=-2.6\times 10^{-17}$ K/Oe^4. When the applied field is parallel to the c axis, the magnitude of the magnetization is much smaller and shows the peak due to Tb ordering (T_{AF}) for all field values ($\leqslant 55$ kOe) at the same temperature (15 K). For fields $\leqslant 10$ kOe, the low-temperature magnetic transition (5 K transition) appears enhanced compared to the peak due to Tb ordering, however, for 50 kOe, only the peak due to the Tb ordering is seen.

Figure 8 shows the temperature variation of the magnetic susceptibility with temperature for an applied field of 50 kOe parallel to both ab and c directions. The susceptibility is highly anisotropic, with a Curie-Weiss behavior for $H\|ab$ and a weak temperature dependence of $H\|c$. The powder average is calculated using the equation $\chi=2/3\chi\|ab+1/3\chi\|c$ and closely resembles the $\chi\|ab$ data due to the weaker contribution from $\chi\|c$. The anisotropy remains up to 300 K which is evident from the inverse susceptibility plot in Fig. 6(b). All the three susceptibilities in the temperature range of 150 to 300 K were fitted to the Curie-Weiss equation,

$$\chi=\chi_0+N\mu_B{}^2p_{\rm eff}^2/3k(T-\theta_p),$$

where $p_{\rm eff}$ is the effective magnetic moment on the Tb^{3+} ions. The fitting yields the values of $p_{\rm eff}=10.48$, 8.02, and $9.91\mu_B$ for $H\|ab$, $H\|c$, and the powder average, respectively. The corresponding values for the Curie-Weiss temperatures (θ_p) are 6.01, -8.3, and -0.53 K, respectively. The values for the Tb moment obtained from the fit to the data are slightly higher than the free ion value ($9.72\mu_B$) for the $H\|ab$ and powder average and lower for $H\|c$. The values of both $p_{\rm eff}$ and θ_p are found to depend to some extent on the temperature range over which the fits are made. The large anisotropy in susceptibilities may be present due to the CEF effects, as observed in other members of the RNi_2B_2C (R=Tm, Er, Ho, and Dy) family of compounds. Analysis of the crystal-field effects in this compound are in progress.

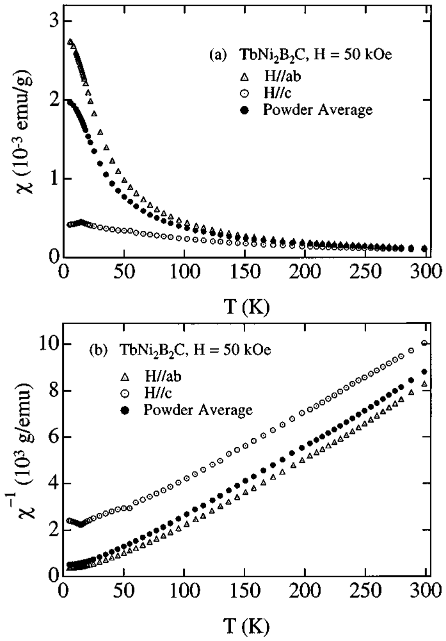

FIG. 8. (a) Anisotropic magnetic susceptibility versus temperature for a $TbNi_2B_2C$ crystal with an applied field of 50 kOe for $H\|ab$ and $H\|c$. (b) Inverse of the magnetic susceptibility in (a) versus temperature. The closed circles represent the powder average calculated from the $\chi\|ab$ and $\chi\|c$.

The results of resistance measurements in applied fields are shown in Fig. 9 where the resistance variation is plotted against the temperature for different applied fields both parallel and perpendicular to the c axis. The resistance was measured along the ab plane in all the cases. For $H\|ab$, the kink at $T_{AF}=15$ K due to the magnetic ordering of Tb moments shifts towards lower temperatures as the applied field is increased. No noticeable features are observed in the resistance curve down to 2 K for applied fields $\geqslant 20$ kOe. This confirms the results of the magnetization measurements where a similar shift in T_{AF} is observed with applied fields. As the field is increased, the resistance decreases at a given temperature confirming the ferromagnetic nature of the correlation between Tb moments within the ab planes. When the field is applied parallel to the c direction there is an increase in the resistance with the applied field, as expected for a simple antiferromagnet. There is no shift in T_{AF} for fields of up to 50 kOe, similar to the results obtained in the magnetization measurements in the same direction. Since the compound exhibits metamagnetic transitions when the field is applied along the ab plane, the magnetoresistance was measured as a function of field at constant temperatures above and below T_{AF}. Figure 10 shows the magnetoresistance along the ab plane at $T=2$ K with the magnetic field applied along the ab plane. The resistance increases as the field is increased and goes through two transitions at 13 and 21 kOe before falling off sharply to a nearly constant value for higher fields. The field values of 13 and 21 kOe correspond well with the field values at which metamagnetic transitions are observed in the magnetization data [Fig. 6(a)]. These two features in the $\rho(H)$ data are observed for temperatures of up to 10 K. No anomaly is observed in the resistance measurements which can be associated with the first metamagnetic transition at 0.6 kOe. The large negative value of the magnetoresistance for $H>25$ kOe provides evidence that the second transition corresponds to the metamagnetic transition to a ferromagnetic state. The magnetoresistance shows hysteretic behavior for increasing and decreasing fields, which is evident even at 20 K, as shown in the inset of Fig. 8.

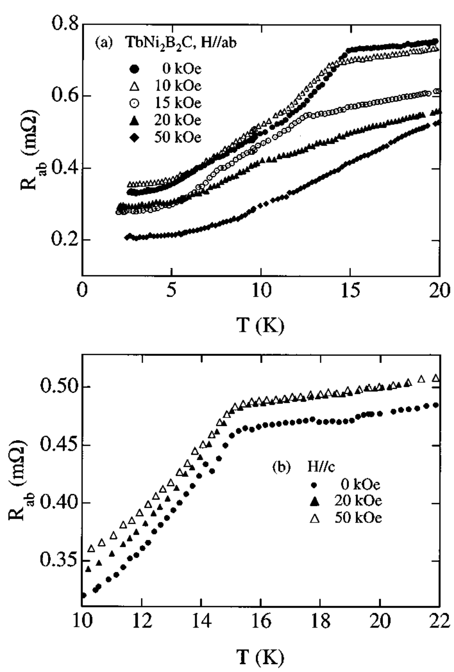

FIG. 9. Magnetoresistance in the ab plane at the indicated field values versus temperature for (a) $H\|ab$ and (b) $H\|c$ for a $TbNi_2B_2C$ crystal.

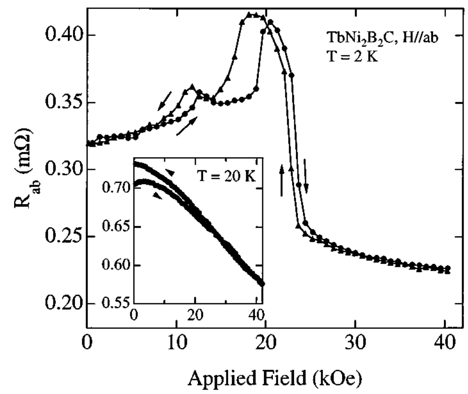

FIG. 10. Magnetoresistance in the ab plane of a $TbNi_2B_2C$ crystal at $T=2$ K versus applied magnetic field for $H\|ab$. The inset shows the magnetoresistance at 20 K for the same crystal. The arrows indicate the increasing and decreasing field directions.

$TbNi_2B_2C$ exhibits an additional magnetic transition at 5 K in the magnetically ordered state. The magnetization value (see Fig. 2) at the 5 K transition is one order of magnitude higher than that at T_{AF}. This enhanced magnetization value suggests that the Tb moments reorients with a net magnetic moment at low temperatures. This reorientation may be one of the possibilities which prevents the Cooper pair formation and superconductivity in $TbNi_2B_2C$. The anisotropic magnetic properties exhibited by $TbNi_2B_2C$ can be attributed to the CEF effects. Similar anisotropic magnetic properties are observed for other members of the series with $R=$Tm, Er, and Ho.[15–17] The CEF effects force the magnetic moments to lie in one direction either along the c axis as in the case of $TmNi_2B_2C$ or in the ab plane as in the case of the rest of the compounds. From our results, it appears that the metamagnetic transitions and the field-induced ferromagnetic states at low temperature are characteristic of these compounds. This may be due to the fact that the moments which are forced to lie along a particular direction by the CEF effects align along the magnetic field when the field value becomes strong enough to split the CEF levels. The decrease in T_{AF} with the application of magnetic fields may also be a consequence of these metamagnetic transitions. This is evident in the case of $TbNi_2B_2C$ where no change in T_{AF} is observed until 10 kOe since these fields are smaller than the field value for which the second metamagnetic transition takes place. The disappearance of T_{AF} coincides with the field values corresponding to the third metamagnetic transition which produces a ferromagnetically ordered state. These observations are further confirmed by the resistance measurements in magnetic fields. The ferromagnetic state is evident from the magnetization measurements as well as the magnetoresistance measurements. The origin of the first metamagnetic transition at 0.6 kOe is not clear. However, it is quite possible that this transition is associated with the magnetic transition observed at 5 K. This is further confirmed by the fact that the peak in susceptibility due to the spin reorientation at 5 K is strongly suppressed in applied fields greater than that (0.6 kOe) at which the first metamagnetic transition is observed. Neutron-diffraction measurements which are underway, will be helpful in illustrating the exact magnetic phase diagram of this compound.

In conclusion we have shown that $TbNi_2B_2C$ is not superconducting above 300 mK, but shows interesting magnetic properties. The ac susceptibility and low-field dc magnetization measurements for both $H \| c$ and $H \| ab$ show two transitions; one corresponding to the magnetic ordering of Tb moments (T_{AF}=15 K) and another at low temperatures (5 K), which we believe is due to a spin reorientation. The resistance measurements do not show any anomalies corresponding to the low-temperature transition. Heat-capacity measurements reveal a third magnetic transition with large λ-type anomaly in addition to the features corresponding to the AFM ordering of the Tb moments and the spin reorientation. The magnetic susceptibility follows a Curie-Weiss behavior when the field is applied parallel to the ab plane and a much weaker temperature dependence for $H \| c$. Magnetization versus field isotherms reveal the metamagnetic transitions at low temperatures and a field-induced ferromagnetic state. The magnetic ordering temperature of the Tb moments is found to depend on the magnetic field for $H \| ab$. Magnetoresistance measurements give further evidence for the temperature dependence of the T_{AF} and the metamagnetic transition to the ferromagnetic state. For $H \| c$ the compound shows a linear M-H behavior expected for a normal AFM compound. These data suggest that the Tb moments are confined to the ab planes and that these moments are arranged in a number of different configurations as a function of temperature and/or applied magnetic field.

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