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The magnetism of $TbNi_5$ at low fields

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Abstract. Magnetic susceptibility and magnetization measurements were made on a polycrystalline sample of $TbNi_5$. The compound is ferromagnetic for $T < T_h \approx 17$ K at any applied magnetic field. For temperatures between T_h and $T_N = 23$ K and magnetic fields smaller than a critical field $H_c = 400$ Oe, the compound is helimagnetic. When $H > H_c$ or $T < T_h$, the anisotropy in the basal plane increases, and the compound becomes ferromagnetic.

1. Introduction

The compound $TbNi_5$ crystallizes in the hexagonal $CaCu_5$ structure (space group $P6/mmm$) [1]. In this compound, the main contribution to the magnetization comes from the 4f electrons of the terbium ions, and the contribution of the nickel to the magnetism is very small because its 3d shell is almost filled up by the 5d electrons of the terbium ion. On the basis of magnetization [2, 3], inelastic neutron scattering [4], μSR [5], torque magnetometry [6], and NMR [7] measurements, the magnetic and thermodynamic properties of this compound have become well understood within a mean-field model involving exchange and crystalline-electric-field magnetoelastic and quadrupolar interactions. There is a magnetic order–disorder transition at 23 K with the magnetic moments ordered ferromagnetically in the basal plane of the hexagonal structure. The compound presents a strong axial magnetic anisotropy and a very small planar anisotropy. However, all of the measurements to date were made either in a high field at low temperature or in zero field in the paramagnetic phase. From resistivity measurements in zero field, Blanco *et al* [8] have confirmed the transition to the paramagnetic phase at 23.2 K, but they also observed that the thermal variation of the resistivity, along the [001] hard-magnetization direction, shows a sharp maximum with a temperature hysteresis of about 5 K between 16 K and 21 K. This peak disappears as the magnetic field is increased. They also observed an anomaly of the DC susceptibility in fields smaller than 500 Oe close to the order–disorder temperature, but no thermal hysteresis was observed.

In this paper we present AC susceptibility and magnetization measurements of polycrystalline $TbNi_5$ at low magnetic fields, together with a discussion of their significance.

2. Experimental procedure

A polycrystalline $TbNi_5$ compound was prepared by melting stoichiometric amounts of the constituent materials in an argon arc furnace. To improve the homogeneity, the resulting metallic button was turned over and melted several times. The purity of the elements is 99.99% for terbium and 99.999% for nickel. X-ray diffraction analysis shows that the

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prepared specimen has the $CaCu_5$-type crystal structure, and there is no evidence for the presence of any impurity phase.

The magnetic AC susceptibility and magnetization measurements were performed on a Lake Shore AC Susceptometer/DC Magnetometer System (Model 7225) in the temperature range 4.5–60 K and in magnetic fields up to 1500 Oe.

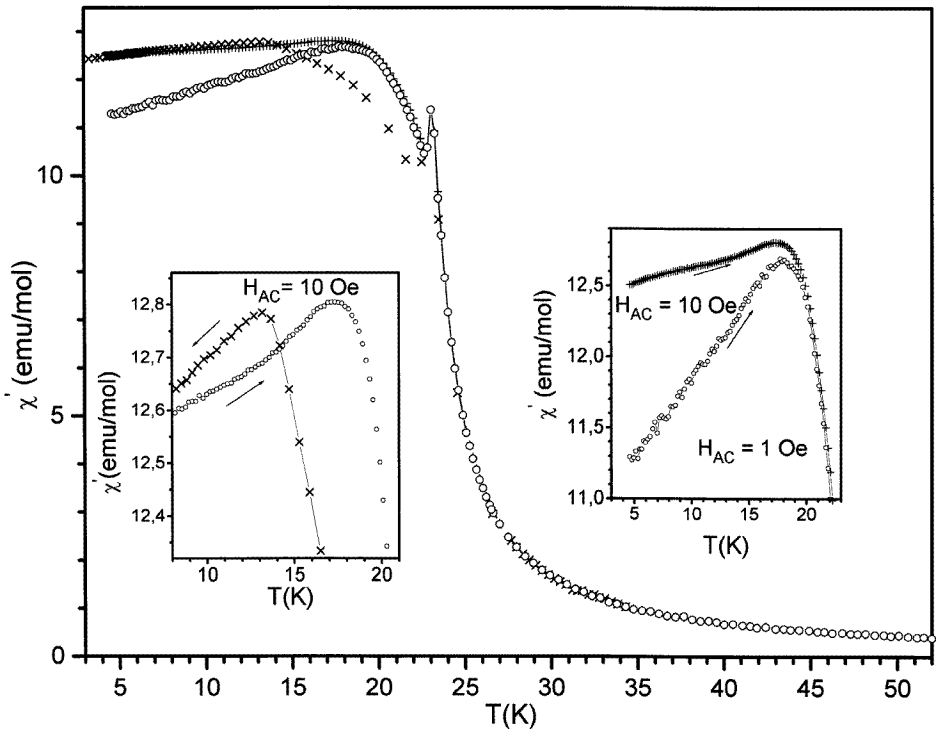

Figure 1. The temperature dependence of the real AC susceptibility of $TbNi_5$; the cooling curves are with $H_{AC} = 10$ Oe and the warming curves are with $H_{AC} = 1$ Oe and 10 Oe. The behaviours of the curves for $T < 20$ K are shown in detail in the insets. The left-hand inset shows the warming and cooling curves for $H_{AC} = 10$ Oe. The right-hand inset shows the susceptibilities for $H_{AC} = 1$ Oe and 10 Oe.

3. Results

The thermal variation, in zero DC field, of the real part of the AC susceptibility (χ') is shown in figure 1. There is a sharp peak at 23.5 K associated with a magnetic order–disorder transition. The behaviour is the same as that observed for the DC susceptibility by Blanco *et al* [8]. We see that this peak shows no thermal hysteresis and no AC field dependence. The insets show the details of the behaviour of the susceptibility for $T < 20$ K. From the left-hand inset one can see that the thermal hysteresis is of about 4 K. The right-hand inset shows the susceptibility variation for two amplitudes of the AC field at the same frequency (5 Hz). The two curves presented are very different for $T < 17$ K. In the range 17 K–19 K the difference becomes small, and for higher temperature there is no appreciable difference.

Figure 2 presents the thermal variation of the AC susceptibility in an applied magnetic

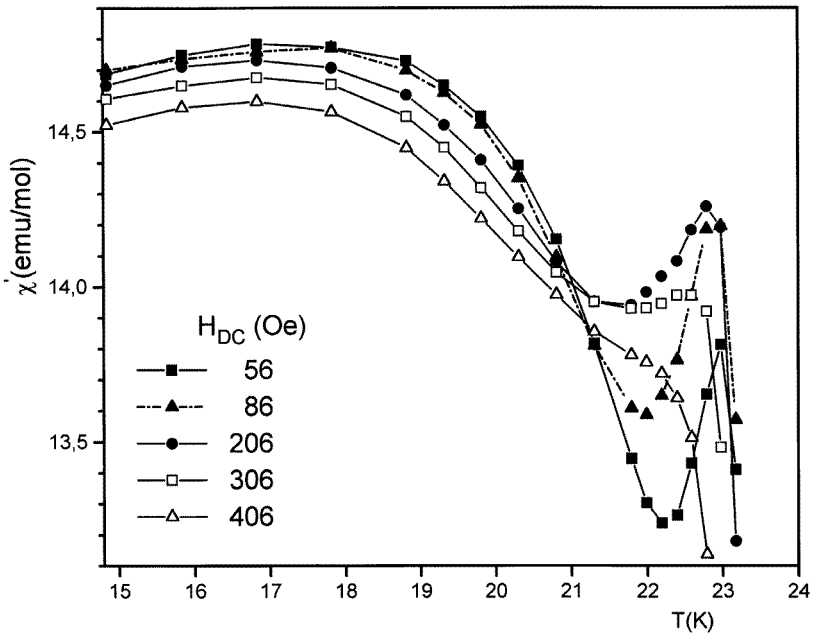

Figure 2. The thermal variation of the AC susceptibility for $TbNi_5$, at different magnetic fields.

field. One can observe that as the field increases, the peak shifts towards lower temperatures, and disappears at a critical field of about 400 Oe. Such behaviour is usually observed in antiferromagnetic materials.

Figure 3 presents the temperature dependence of the imaginary part of the AC susceptibility (χ''). In the paramagnetic phase, χ'' is nearly zero. With the onset of the ordered phase at 23 K, χ'' increases as the temperature decreases, and reaches a maximum (χ''_{Max}) at 22 K. For lower temperatures, χ'' decreases, and at around 17 K starts to increase again, reaching about $2\chi''_{\mathrm{Max}}$ at 4.2 K.

Figure 4 shows M^2 versus H/M (Arrott plots) for different temperatures and for fields smaller than 1400 Oe. The data indicate that the order–disorder temperature is around 23 K. At low fields, the M^2 versus H/M variation depends whether the temperature is smaller or greater than 18.8 K.

4. Discussion

The thermal variations of the real part of the AC susceptibility (this work) and of the DC susceptibility (reference [8]), show no anomaly at 23 K. We can therefore conclude that they are not due to relaxation effects.

The changes in the thermal variation of χ' and χ'' and in the Arrott plot occur at about the same temperature, $T = 17.5$ K. This suggests that there is a magnetic phase change at this temperature.

The results in this work, and the strong changes in the thermal variation of the resistivity observed by Blanco *et al* [8], can be explained as resulting from there being a helimagnetic phase between $T_h = 17$ K and $T_N = 23$ K when $H < 400$ Oe.

As terbium, dysprosium, and $TbNi_5$ have hexagonal crystallographic symmetry, a hard-

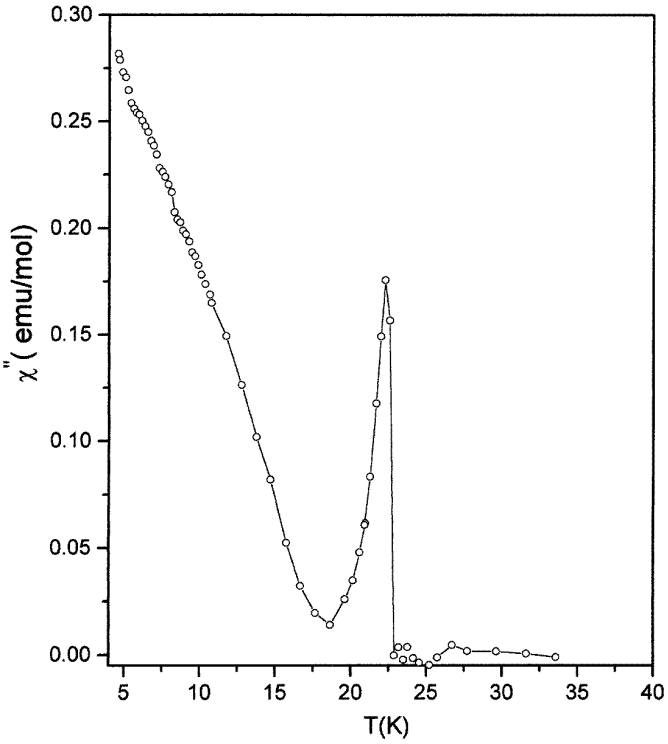

Figure 3. The temperature dependence of the imaginary AC susceptibility of $TbNi_5$, with $H_{AC} = 10$ Oe.

magnetization *c*-axis, a strong axial anisotropy, and a small planar anisotropy, we use in this work some terbium and dysprosium results as references to achieve a better understanding of our results.

The field dependence of the susceptibility peak at 23 K shows an antiferromagnetic peak behaviour for fields smaller than $H_c = 400$ Oe. The maximum at $T_h = 17$ K of the susceptibility curve at zero DC field in the warming curve, and at 13 K in the cooling curve, can be attributed to a transition from the ferromagnetic ($d\chi/dT > 0$) to the helical phase ($d\chi/dT < 0$), as in terbium [9]. The thermal hysteresis of 4 K is about the same as is observed in the resistivity measurements [8]. The strong increase in the thermal variation of the resistivity, along the [001] hard-magnetization direction [8], can be associated with a helimagnetic transition. Mackintosh [10] and Miwa [11] have shown that when the magnetic lattice periodicity in the *c*-direction differs from the ionic one, as is the case for a helimagnetic structure, extra planes of energy discontinuity are introduced in the Brillouin zone structure. These new magnetic energy gaps, along the *c*-axis, wipe out areas of the Fermi surface with *c*-axis projections, and strongly increase the resistivity in the *c*-direction, leading to the so-called 'superzones'.

The ferro–helimagnetic transition at $T_h \approx 17$ K is also responsible for the strong increase in χ'', as the temperature decreases, and for the increase in the H_{AC}-field dependence of χ' (characteristic of a ferromagnetic phase [12]).

A critical field of $H_c = 400$ Oe suppresses the helical structure (figure 2), and eliminates the superzone energy gaps near the Néel point. In the resistivity measurements [8], this field

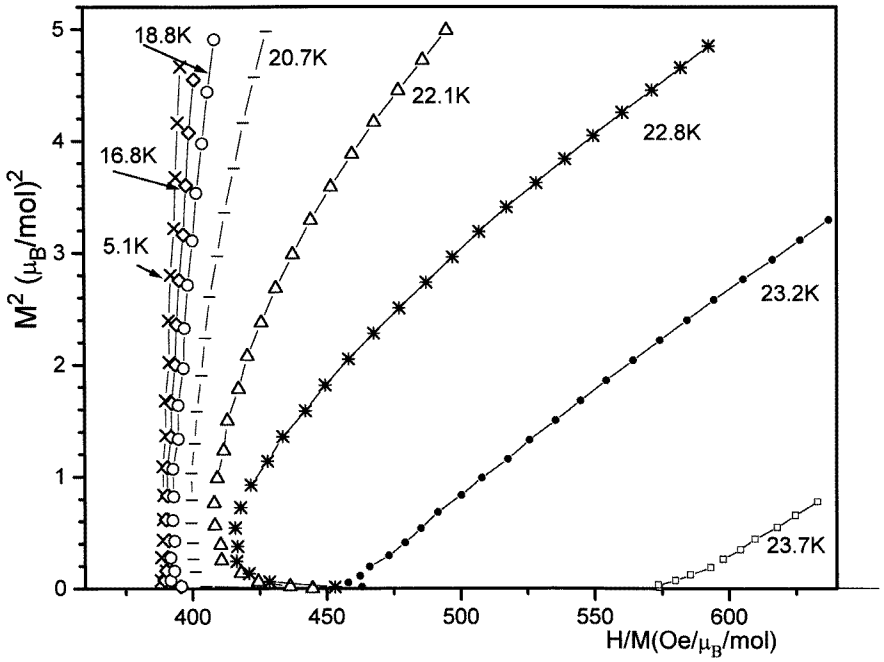

Figure 4. An Arrott plot for $TbNi_5$ at different temperatures.

is about 3 kOe. This discrepancy can be explained by the widely differing demagnetizing fields related to the different sample geometries. The sample used for the resistivity measurements is a parallelepiped which [8] has a high demagnetization factor. The same order of difference was observed in the magnetization and resistivity measurements of metallic terbium [13].

To analyse the Arrott plot of $TbNi_5$, we must remember that, in the case of homogeneous ferromagnetic materials in the ordered phase, one can write for a given temperature [14]:

$$H/M = 1/\chi_i + N + AM^2 \tag{1}$$

where N is the demagnetization factor, and χ_i and A are constants that are proportional to the anisotropy constants K_1, K_2 and inversely proportional to the spontaneous magnetization M_S. In this work, the magnetization measurements have been made on demagnetizable samples. For $T < 16.8$ K and $H < 400$ Oe, the constant A and the magnetization M are so small that the first two constant terms in equation (2) are the dominant ones. This means that the variation of M^2 with H/M is a vertical line, as observed in figure 4. For higher fields, the M-dependent term must be considered. So, $TbNi_5$ has a ferromagnetic behaviour for $T < 16.6$ K. At higher temperatures and for $H > 400$ Oe, the compound shows an M^2 versus H/M variation typical of a ferromagnet. When $H < 400$ Oe, we observe that H/M decreases with increase in the field. To understand this result, we must remember that, in a helimagnetic phase, the magnetic susceptibility at low field [15] is proportional to $[\cos(\varphi)]^{-1}$, where φ is the mean angle between the spins of two terbium ions situated in two planes perpendicular to the c-axis. The magnetic field deforms the helices, so $\varphi = \varphi + \Delta H$. Therefore, as the intensity of the magnetic field increases, φ increases, and hence $\cos\varphi$ and, consequently, $1/\chi_i$ decrease also. So, H/M decreases with increasing field, as is experimentally observed.

Blanco *et al* [8] have concluded that the anomaly in the thermal variation of the resistivity of $TbNi_5$ was due to magnetic fluctuations, because no anomaly was observed in the susceptibility and specific heat curves. From our work we see that the anomaly in the susceptibility is observed only in small magnetic fields and in the 17 K–23 K temperature range. It is important to note that terbium, which shows at T_h a strong resistivity variation [13], presents only a very small change in the specific heat variation [16] at this temperature, which was observed only after a detailed study around T_h. The specific heat curve in reference [8] is not so detailed as to give information about the anomaly at 17 K.

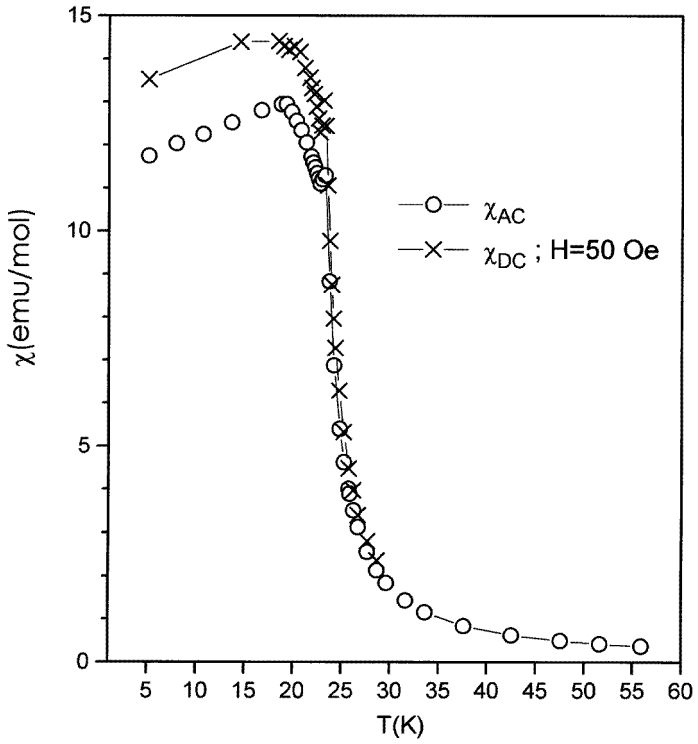

Figure 5. The temperature dependence of the DC susceptibility, $H_{DC} = 50$ Oe, and the AC susceptibility, with $H_{AC} = 10$ Oe, in zero DC field.

Figure 5 shows that for $T < 23$ K the AC susceptibility is smaller than the DC susceptibility (calculated from the slope of the magnetization curves as the DC field goes through zero). For $T > 23$ K the two susceptibilities coincide. There are then irreversible contributions to the low-field susceptibility in the ordered phase. In the helical one this can be a consequence of some magnetic structure that is not perfectly helimagnetic present in this range of temperature. This fact, and the presence of thermal hysteresis in the variation of the susceptibility, suggest, as in the case of dysprosium [17], that in this compound there are antiferromagnetic domains which appear as a result of uniform spiralling of the ferromagnetic coupled planes, but in the opposite sense. These domains would be separated by thin ferromagnetic domain boundaries of different volumes that are responsible for the difference observed.

5. Conclusion

The results indicate that for $T < T_h$, where $T_h = 17$ K, at any magnetic field, the compound $TbNi_5$ is ferromagnetic, and for $T_h < T < T_N$, where $T_N = 23$ K, if the field is smaller than a critical one, $H_c = 400$ Oe, the compound is helimagnetic. When $T < T_h$, the magnetic properties come from ferromagnetic interactions between the terbium-ion first neighbours in the same plane (perpendicular to the c-axis). As the temperature increases in a field smaller than H_c, the interaction between terbium ions in different planes becomes dominant. In this case the interactions between Tb ions in the first- and second-neighbour planes are of the same order. This comes from the fact that in a helimagnetic system, when there is no external field, the energy can be expressed [18] as

$$E/N = -J_1 \cos\varphi - J_2 \cos 2\varphi - J_3 \cos 3\varphi - \cdots \tag{2}$$

where J_1 is the exchange coupling constant, per pair of atoms, between adjacent planes perpendicular to the c-axis, and J_2 is the one between next-nearest-neighbouring planes, and so on. N is the number of equidistantly spaced equivalent planes of spins coupled ferromagnetically, and φ is the angle between the spins of two adjacent planes. When only J_1 and J_2 are considered, the minimization of the energy in (2) gives a helimagnetic solution for $J_2 < 0$ and $|J_1/4J_2| < 1$. So, in a helical phase, J_1 and J_2 are of the same order.

In the helical phase, the anisotropy in the basal plane is so small that a small change in the direction of the magnetic moment has little influence on the free energy; therefore the helical structure is the most stable. When the anisotropy becomes stronger because of the decreasing temperature or the increasing field, the ferromagnetic configuration becomes the most stable. The results also show that the helical phase in $TbNi_5$ is not ideal, because we must consider the existence of antiferromagnetic domains of uniform spiralling of ferromagnetically coupled planes, in the opposite sense.

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References

[1] Warnick J H and Geller S 1959 *Acta Crystallogr.* **12** 662
[2] Nait Saada A 1980 *PhD Thesis* Grenoble University
[3] Escudier P, Gignoux D, Givord D, Lemaire R and Murani A P 1977 *Physica* B **86–88** 197
[4] Goremychkin E A, Mühle E, Ivanitskii P G, Krotenko V T, Pasechnik M V, Slisenko V V, Vasilkevich A A, Lippold B, Chistyakov O D and Savitskii E M 1984 *Phys. Status Solidi* b **121** 623
[5] Dalmas de Réotier P, Sanchez J P, Yaouanc A, Harris S W, Hartmann O, Karlsson E, Wäppling R, Gignoux D, Gorges B, Schmitt D, L'Héritier P, Weidinger A and Gubbens P C M 1990 *Hyperfine Interact.* **64** 389
Dalmas de Réotier P, Yaouanc A, Gubbens P C M, Gignoux D, Gorges B, Schmitt D, Hartmann O, Wäppling R and Weidinger A 1992 *J. Magn. Magn. Mater.* **104–107** 1267
[6] Algarabel P A, Morellon L, Ibarra M R, Schmitt D, Gignoux D and Tari A 1993 *J. Appl. Phys.* **73** 6054
Morellon L, Algarabel P A, Ibarra M R, del Moral A, Gignoux D and Schmitt D 1996 *J. Magn. Magn. Mater.* **153** 17
[7] Carboni C, Gignoux D and Tari A 1995 *Phys. Rev.* B **52** 9486
[8] Blanco J A, Gignoux D, Schmitt D, Tari A and Zhang F Y 1994 *J. Phys.: Condens. Matter* **6** 4335
[9] Derevyanko V V and Finkel V A 1985 *Sov. Phys.–Solid State* **27** 1405
[10] Mackintosh A R 1962 *Phys. Rev. Lett.* **9** 90
[11] Miwa H 1962 *Theor. Phys. (Kyoto)* **28** 208
[12] Kneller E 1962 *Ferromagnetismus* (Berlin: Springer)

[13] Hegland D E, Legvold S and Spedding F H 1963 *Phys. Rev.* **131** 158
[14] Brommer P E and Franse J J M 1990 *Ferromagnetic Materials* vol 5 (Amsterdam: Elsevier) p 323
Grossinger R, Sun X K, Eibler R, Buschow K H J and Kirchmayr H R 1986 *J. Magn. Magn. Mater.* **58** 55
[15] Herpin A 1968 *Theorie du Magnetisme* (Institut National des Sciences et Techniques Nucleaires) p 506
[16] Jennings L D, Stanton R M and Spedding F H 1957 *J. Chem. Phys.* **27** 909
[17] del Moral A and Lee E W 1974 *J. Phys. F: Met. Phys.* **4** 280
[18] Nagamiya T, Nagata K and Kitano Y 1962 *Prog. Theor. Phys.* **27** 1253