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Resistivity anomalies in ferromagnetic RNi_5 (R = Tb, Dy or Er) compounds

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Abstract. Anomalies in the transport properties on RNi_5 single crystals are presented. These anomalies are characterized by the appearance of a peak, in some cases, and hysteresis in the electrical resistivity just below the Curie temperature. A qualitative analysis suggests that these features could be correlated with spin fluctuations in these compounds. The presence of two crystalline-electric-field levels close to each other as the lowest-energy states and the nature of these levels could play a crucial role in these phenomena.

1. Introduction

The intermetallic compounds RNi_5 (space group, $P6/mmm$) crystallize in the hexagonal $CaCu_5$-type structure and have been extensively investigated because they exhibit strong 4f crystalline-electric-field (CEF) anisotropy [1–4]. In this series the nickel is non-magnetic and there is only one magnetic R^{3+} ion in the primitive unit cell.

The magnetic and thermodynamic properties of these compounds are quite well understood within a simple mean-field model involving exchange, CEF and quadrupolar interactions. However, not much attention has been paid to the transport properties. Only the temperature and magnetic field dependences of the electrical resistivity on the Van Vleck paramagnetic $PrNi_5$ have been studied, showing that the quadrupolar contribution to the resistivity plays an important role in the observed anisotropy [5].

In order to analyse the possible anisotropy of the electrical resistivity in other RNi_5 compounds, we have measured the temperature dependence of the electrical resistivity on single crystals of the heavy-rare-earth compounds $TbNi_5$, $DyNi_5$ and $ErNi_5$ and the magnetoresistivity of $TbNi_5$. We have also measured the thermal variation in the magnetic susceptibility along different crystallographic directions as well as the specific heats of $TbNi_5$ and $DyNi_5$.

The measurements presented in this paper were performed on several single crystals of $TbNi_5$, $DyNi_5$, and $ErNi_5$, which were prepared by the Czochralski method in a cold-crucible induction furnace. For each compound a unique ingot has been subsequently spark cut in order to obtain three parallelepipeds with their sides perpendicular to the [100], [010] and [001] axes of the orthohexagonal cell. Typical dimensions of the samples were 6 mm × 1.5 mm × 1.5 mm.

Measurements were all performed in different installations at the Laboratoire Louis Néel in Grenoble. The electrical resistivity and the magnetoresistivity were measured by the AC four-point method with $\Delta\rho/\rho = 10^{-4}$ between 1.5 and 50 K and magnetic fields up to

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6 kOe [5]. The susceptibility was determined from the measurement of the integral of the induced EMF when the sample is extracted from a pick-up coil in a constant magnetic field in a temperature range between 1.5 and 30 K. The sensitivity of the system is 10^{-2} emu. The specific heat was studied using the AC calorimetry technique between 1.5 and 30 K [6].

2. Experimental results

2.1. $TbNi_5$

The low-temperature behaviour of the zero-field resistivity of $TbNi_5$ is depicted in figure 1. Apart from the anisotropy between the crystallographic directions already evidenced in previous measurements on $LaNi_5$ [5], it is worth noting two major features in the resistivity of this compound:

(i) Along the [100] easy-magnetization direction the experimental variation presents a normal behaviour as expected for ferromagnetic ordering with a Curie temperature $T_C = 22.7$ K, showing a slight minimum for temperatures higher than T_C.

(ii) Along the [001] hard-magnetization direction it shows an important increase near T_C, giving rise to a peak. This anomaly is also characterized by the appearance of thermal hysteresis of almost 4 K, which is a maximum of 21.2 K and 17.4 K for increasing temperature and decreasing temperature, respectively.

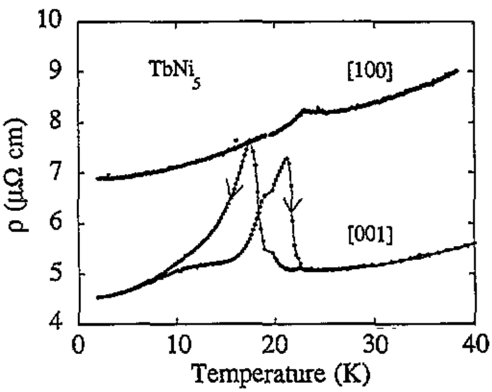

Figure 1. Temperature dependences of the electrical resistivity of $TbNi_5$ in a zero magnetic field along the [100] and [001] directions of the orthohexagonal cell.

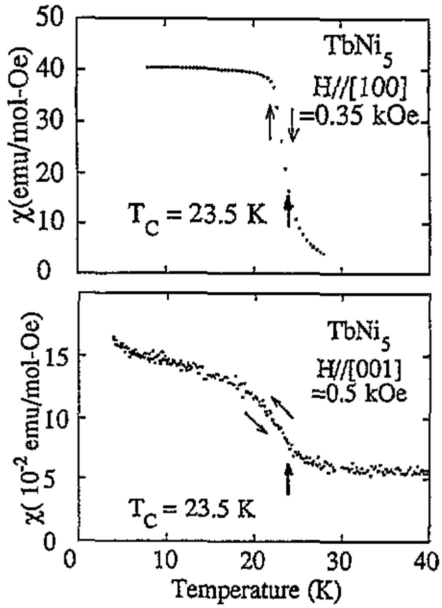

Figure 2. Thermal variation in the susceptibility of $TbNi_5$ along the [100] and [001] directions.

The susceptibility measurements when the magnetic field is applied along the [100] and [001] directions are shown in figure 2. For both axes an anomaly is observed around the ordering temperature $T_C = 23.5$ K which does not show any thermal hysteresis. On the other hand, figure 3 presents the magnetic part C_{mag} of the specific heat as a function of the temperature for $TbNi_5$. This variation exhibits a λ-type anomaly at $T_C = 22.4$ K, the temperature corresponding to the inflection point of C_{mag} above its maximum. In the same way as for the susceptibility, the specific-heat dependence is characterized by the absence of thermal hysteresis around T_C, because no differences are found between the heating and cooling curves (see inset of figure 3).

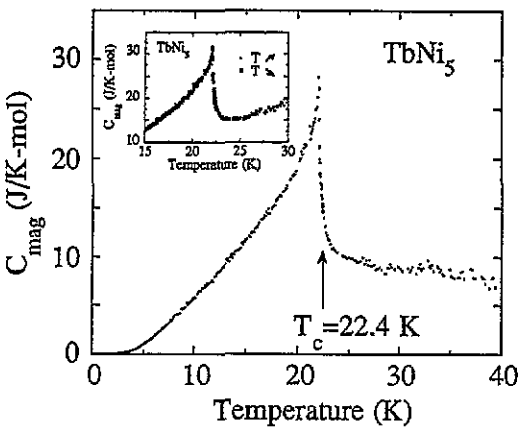

Figure 3. Temperature dependence of the specific heat of $TbNi_5$. The inset shows in detail the absence of hysteresis.

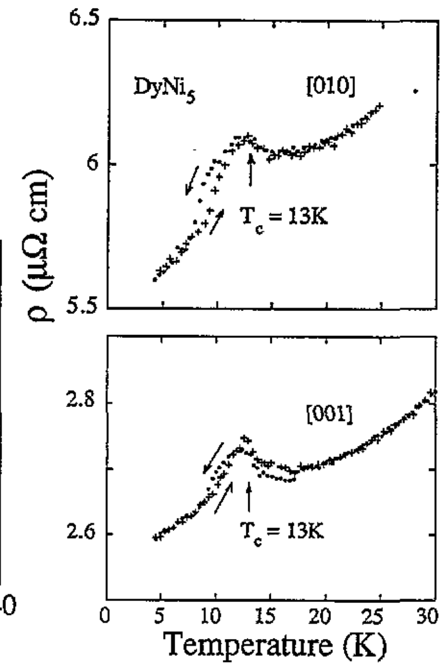

Figure 4. Temperature dependences of the electrical resistivity of $DyNi_5$ in a zero magnetic field along the [010] and [001] directions.

On comparison of the values obtained from the different experiments, the actual Curie temperature can be fixed as $T_C = 23.0 \pm 0.6$ K; then the positions of the two peaks observed in the resistivity along the [001] hard axis correspond to the values $0.92T_C$ and $0.76T_C$ for increasing temperature and decreasing temperature, respectively.

2.2. *$DyNi_5$*

In figure 4 is shown the thermal variation in the resistivity of $DyNi_5$ perpendicular and parallel to the [001] hard-magnetization direction. It presents an anomaly around the Curie temperature $T_C = 13$ K in both directions. The maximum is much less pronounced than for $TbNi_5$ and contrary to this they are quite similar for the [010] and [001] axes in the ordered range. The maxima of the heating and cooling curves occur at the same temperature, but a thermal hysteresis appears on the left-hand side of the peaks. According to the magnetic properties of this compound, this hysteresis centred around 9 K has been attributed to a change in the easy-magnetization axis in the basal plane, in fact, in zero internal field, the

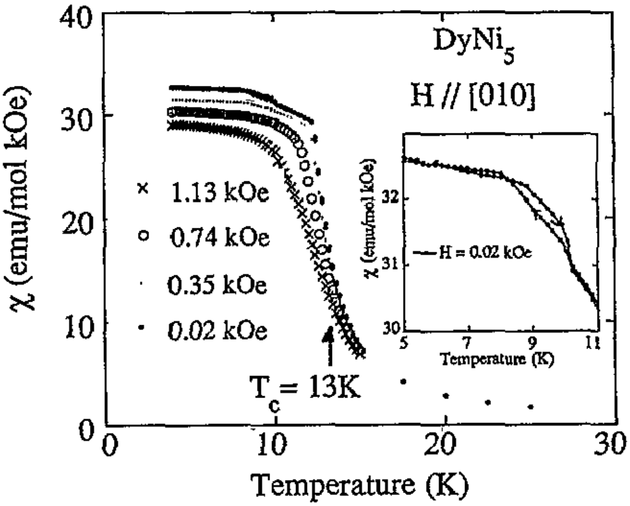

Figure 5. Thermal variation of the susceptibility of $DyNi_5$ along the [010] direction. The inset shows the observed hysteresis at around 9 K corresponding to the change in the easy-magnetization axis for the applied magnetic field $H = 0.02$ kOe.

easy direction changes on heating from the [010] to the [100] axis of the orthohexagonal unit cell [7]. Moreover, in the paramagnetic range, hysteresis is observed along the [001] direction, but its origin is not yet understood at the moment.

Figure 5 presents the susceptibility measurements when the magnetic field is applied along the [010] direction. The inflection point corresponds to the Curie temperature $T_C = 13$ K. In addition, at around 9 K, thermal hysteresis is detected, associated (as commented on above) with the change in easy-magnetization direction (see inset of figure 5). This hysteresis decreases when the magnetic field increases and finally disappears above 0.5 kOe.

The magnetic part C_{mag} of the specific heat is presented in figure 6. A clear λ-type anomaly is found at the ordering temperature $T_C = 12.8$ K. This value is in good agreement with those obtained from resistivity and susceptibility measurements. As occurs in these properties, C_{mag} does not show any thermal hysteresis around the Curie temperature (see inset of figure 6).

2.3. $ErNi_5$

In figure 7 is depicted the thermal variation in the electrical resistivity of $ErNi_5$ along the [010] hard and [001] easy directions. Along the former, a slight anomaly is observed, without hysteresis, at around $T_C = 9.2$ K, similar to that of $TbNi_5$ along the easy axis, and the resistivity curve shows a slight minimum at temperatures just higher than the Curie temperature. Along the [001] direction, no anomaly is detected at this temperature.

2.4. Magnetoresistivity of $TbNi_5$

In order to understand the particular resistivity behaviour detected for $TbNi_5$, we considered that it would be interesting to measure the magnetoresistivity of this compound. Figure 8 presents the thermal variations in the resistivity in several magnetic fields applied along the [010] direction with the current parallel to the [001] hard axis. This experiment clearly shows that:

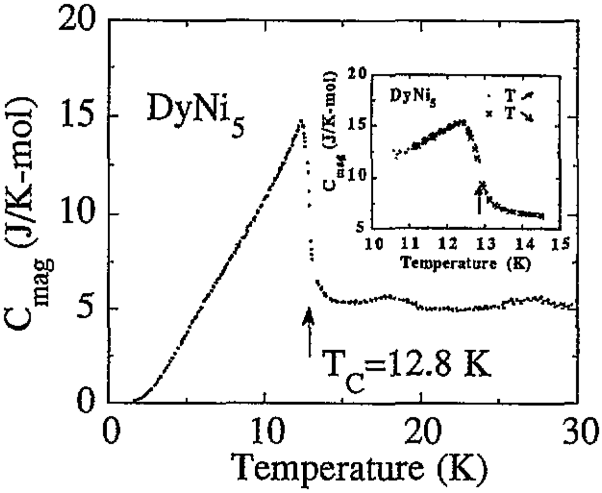

Figure 6. Temperature dependence of the specific heat of $DyNi_5$. The inset shows in detail the absence of hysteresis.

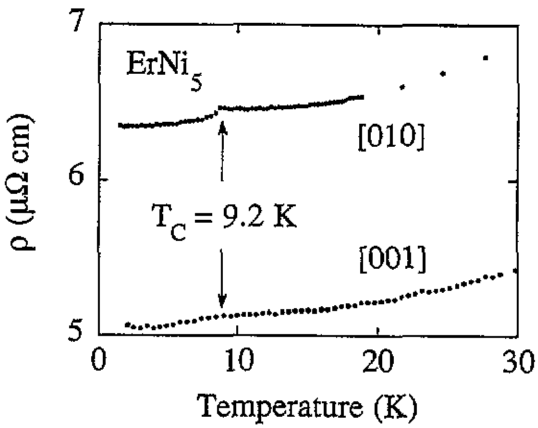

Figure 7. Temperature dependences of the electrical resistivity of $ErNi_5$ in a zero magnetic field along the [010] and [001] directions.

(i) the amplitude of the peaks decreases when the magnetic field rises irrespective of whether the temperature increases or decreases and

(ii) the thermal hysteresis decreases and disappears for magnetic fields higher than 1.4 kOe.

Furthermore, up to magnetic fields of 2.0 kOe, the high-temperature edge of the peak obtained during heating occurs at the same value of around 22 K; then it reaches values of 20 K and 17.5 K when the magnetic field is increased to 2.4 kOe and 2.9 kOe, respectively. However, the behaviour of $\rho(T)$ on cooling seems to be more complicated because it is much more magnetic field dependent than on heating.

In figure 9 is shown the magnetic field dependence of the resistivity $\rho(H)$ when the temperature is fixed at $T = 18.5$ K close to the maximum (this temperature was reached on cooling from the paramagnetic phase). The anomaly disappears for magnetic fields higher

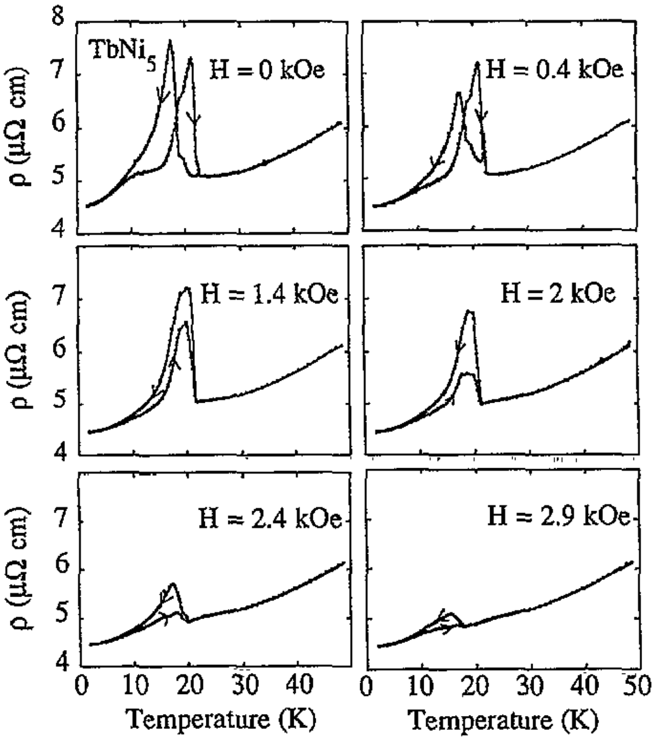

Figure 8. *Temperature dependences of the electrical resistivity of $TbNi_5$ in different magnetic fields* $H = 0$, 0.4, 1.4, 2, 2.4 and 2.9 kOe along the [010] direction when the current is parallel to the [001] direction.

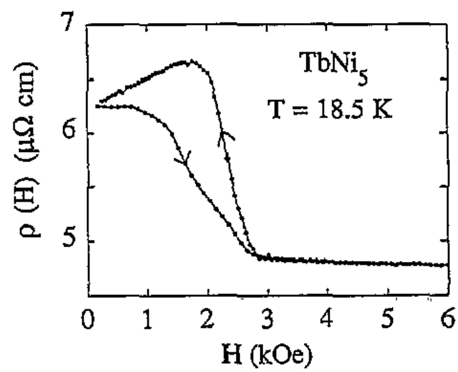

Figure 9. Magnetic field dependences of the electrical resistivity of $TbNi_5$ at 18.5 K when the current is parallel to the [001] axis and the magnetic field is parallel to the [010] axis.

than 3 kOe, in agreement with the result shown in figure 8. The variation in $\rho(H)$ at 18.5 K is also characterized by the appearance of a magnetic field hysteresis which gives rise to a maximum at about $H = 1.7$ kOe when the magnetic field decreases.

3. Analysis and discussion

Although the magnetic behaviour of the ferromagnetic compounds $TbNi_5$, $DyNi_5$ and $ErNi_5$ seems to be quite similar near the Curie temperature from the susceptibility and specific heat, the electrical resistivities of these compounds exhibit important differences. It turns out that the experimental electrical resistivity of $TbNi_5$ near the Curie temperature is quite unique, even though other properties do not seem to be influenced in the same way at this temperature. The resistivity is characterized by a temperature range near T_C where a pronounced peak is observed, accompanied by a large thermal hysteresis along the [001] hard-magnetization direction.

Similar behaviour has been observed for pure rare-earth metals in which this feature has been associated with the incommensurable character of the magnetic structures and attributed to the appearance of a Brillouin superzone [8]. However, this explanation is unlikely in the case of the $TbNi_5$ structure, which is *a priori* simple ferromagnetic considering the properties of the whole series [9].

On the other hand, the resistivity of $DyNi_5$ along and perpendicular to the [001] hard-magnetization direction presents an intermediate behaviour between the case of $TbNi_5$ and that of a normal rare earth such as $ErNi_5$, because quite a small peak is detected, which is much less pronounced than that observed for $TbNi_5$. Furthermore, the resistivity of $DyNi_5$ presents a hysteresis at about 9 K, the temperature at which the change in the magnetization easy axis occurs [7].

These facts, together with the destructive character of the magnetic field observed for $TbNi_5$, seem to suggest that these anomalies near the Curie temperature are due to magnetic fluctuations, which may make an important contribution to the temperature-dependent part of the resistivity [10, 11]. Although the reason for this enhancement in the spin fluctuation scattering mechanism is not yet fully understood, we believe that it could be related to the CEF level schemes. In both compounds $TbNi_5$ and $DyNi_5$, the two lowest energy levels are close to each other, the separation in energy between the ground state and the first excited state being 5.8 K and 9 K for $TbNi_5$ and $DyNi_5$ respectively (table 1) [12], and the magnetic moments order ferromagnetically in the basal plane. This latter feature provokes a quasi-degeneracy between the [100] and [010] axes, as confirmed by the magnetization curves of both compounds [12]. In this way, when magnetic order has begun to be established, the magnetic moments fluctuate with a time scale which is quite similar to the relaxation time of conduction electrons; so this mechanism of scattering becomes important, leading to an enhancement in the dispersion of the conduction electrons when the current is parallel to the hard axis [001] (giving rise to a peak which disappears when we apply a field, because this aligns the moments along the direction of the magnetic field).

The different amplitudes of the peaks observed for $TbNi_5$ and $DyNi_5$ could be associated with the nature of the ground state, which is a singlet in the former and a doublet in the latter (see table 1), indicating that the relaxation time is quite dependent on the nature and separation between the ground state and the first excited state [13]. For this reason, it would be very interesting to perform dynamic measurements, such as muon spectroscopy, in order to determine the relaxation time and the energy distribution of fluctuations [14].

On the contrary, for $ErNi_5$ the first excited state is much higher in energy (about 21 K; see table 1) [15], and the magnetic moments are along the [001] direction. This compound exhibits an Ising-type behaviour, and this could explain the absence of any anomaly similar to that observed for $TbNi_5$ and $DyNi_5$.

On the other hand, other mechanisms such as the quadrupolar contribution to the spin-disorder resistivity could have an important influence on the magnetoresistivity, as was

Table 1. CEF states on $TbNi_5$, $DyNi_5$ and $ErNi_5$ in the $|J, M\rangle$ basis. Ψ_0 is the eigenfunction of the groundstate. E_n and Ψ_n are the energy position and eigenfunction of the next two excited states, respectively. The data are taken from [12] for $TbNi_5$ and $DyNi_5$ and [15] for $ErNi_5$.

Compound	Ψ_0	E_1 (K)	Ψ_1	E_2 (K)	Ψ_2
$TbNi_5$	$\|6, 0\rangle$	5.8	$\|6, \pm 1\rangle$	28.2	$\|6, \pm 2\rangle$
$DyNi_5$	$0.055\|\frac{15}{2}, \pm\frac{13}{2}\rangle$ $-0.990\|\frac{15}{2}, \pm\frac{1}{2}\rangle$ $+0.130\|\frac{15}{2}, \mp\frac{11}{2}\rangle$	9.0	$-0.016\|\frac{15}{2}, \pm\frac{15}{2}\rangle$ $+0.965\|\frac{15}{2}, \pm\frac{3}{2}\rangle$ $-0.262\|\frac{15}{2}, \mp\frac{9}{2}\rangle$	21.2	$-0.522\|\frac{15}{2}, \pm\frac{7}{2}\rangle$ $+0.853\|\frac{15}{2}, \mp\frac{5}{2}\rangle$
$ErNi_5$	$0.998\|\frac{15}{2}, \pm\frac{15}{2}\rangle$ $-0.070\|\frac{15}{2}, \pm\frac{3}{2}\rangle$ $+0.190\|\frac{15}{2}, \mp\frac{9}{2}\rangle$	20.9	$\pm 0.980\|\frac{15}{2}, \pm\frac{13}{2}\rangle$ $\mp 0.190\|\frac{15}{2}, \pm\frac{1}{2}\rangle$ $\pm 0.090\|\frac{15}{2}, \mp\frac{11}{2}\rangle$	67.3	$\mp 0.910\|\frac{15}{2}, \mp\frac{11}{2}\rangle$ $\pm 0.390\|\frac{15}{2}, \pm\frac{1}{2}\rangle$ $\pm 0.150\|\frac{15}{2}, \pm\frac{13}{2}\rangle$

observed for $PrNi_5$ [5]; however, it does not lead to the appearance of a sharp peak in $\rho(T)$ near the critical temperature.

Finally it is important to mention that the existence of hysteresis in the resistivity curves is not related to the first-order character of the ferromagnetic transition, as confirmed by means of the specific-heat data. The nature of the magnetic phase transition is actually second order, although from the viewpoint of the resistivity the short-range spin fluctuations depend strongly on the phase changes which take place at the critical temperature, because differences are found from paramagnetic-to-ferromagnetic and from ferromagnetic-to-paramagnetic transitions. Further theoretical investigations would be fruitful in order to check the exact role played by the CEF level scheme on these strong spin fluctuation effects suggested by the transport properties.

Acknowledgments

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