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LETTER TO THE EDITOR

Spin-reorientation transitions in $NdCo_5$ and critical effects on the electrical resistivity temperature derivative

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Abstract. High resolution electrical resistivity measurements (ρ dρ/dT) are used to investigate in detail the two spin-reorientation transitions of the hexagonal ferromagnetic $NdCo_5$ compound which occur at $T_{SR1} = 242$ K and $T_{SR2} = 283$ K ($T > T_{SR2}$ gives spontaneous magnetisation, M_s, parallel to the c axis; $T_{SR1} < T < T_{SR2}$ gives a conical structure; $T < T_{SR1}$ produces M_s in the basal plane). We have obtained the temperature dependence of the angle θ ($T_{SR1} < T < T_{SR2}$) using our $\rho(T)$ data. The results being in fair agreement with $\theta(T)$ derived from the anisotropy constants K_1 and K_2. However, the temperature derivative dρ/dT exhibits new (and very localised) δ-type singularities near T_{SR1} and T_{SR2}. These new anomalies in dρ/dT are interpreted in terms of lattice softening effects at T_{SR1} and T_{SR2} and associated anomalies in the C_{33} elastic constant.

The hexagonal compound $NDCo_5$ exhibits two spin-reorientation transitions (T_{SR1} = 242 K, T_{SR2} = 283 K) caused by the competition between the Nd anisotropy favouring a basal plane spontaneous magnetisation M_s (anisotropy constant $K_1^{Nd} < 0$) and that of Co favouring c axis alignment ($K_1^{Co} > 0$) (Tatsumoto *et al* 1971).

At high temperatures the magnetism is dominated by Co (strong Co–Co exchange) and M_s is set along the c axis. Below room temperature the Nd moments order considerably, making K_1^{Nd} more negative. The effective anisotropy constant $K_1 = K_1^{Co} + K_1^{Nd}$ then decreases with temperature, changing sign at a characteristic temperature T_{SR2} where M_s moves out of the c axis, through a second-order spin-reorientation transition.

Although $K_1 < 0$ would favour M_s in the basal plane, the smallness of K_1 near T_{SR2} makes imperative the effect of the second-anisotropy constant K_2, which is positive. This produces a conical structure characterised by an angle θ (between M_s and the c axis) given by (Belov *et al* 1976, Ohkoshi *et al* 1976):

$$\theta = \sin^{-1}\sqrt{-K_1(T)/2K_2(T)}. \tag{1}$$

Ultimately θ reaches $\pi/2$ at a lower temperature T_{SR1}, where M_s rapidly rotates to the basal plane through a second-order orientational transition.

Both transitions have been extensively studied in the past, mainly with magnetic (Lemaire 1966), elastic (Deryagin *et al* 1984), ultrasonic (Patterson *et al* 1986), thermal

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(Ohkoshi and Kobayashi 1977, Pourarian *et al* 1981), magnetostrictive (Algarabel *et al* 1987) and neutron diffraction techniques (Bartholin *et al* 1966). Anomalies in the electrical resistivity (ρ) have also been reported (Andreev *et al* 1982), but the relation between $\rho(T)$ and $\theta(T)$ has not been studied in sufficient detail; also, the $\mathrm{d}\rho/\mathrm{d}T$ data have not the required resolution to reveal localised effects near T_{SR1} and T_{SR2}.

In the present work we report a detailed investigation of the behaviour of the electrical resistivity in $\mathrm{NdCo_5}$, using high accuracy measurements both of ρ and its temperature derivative $\mathrm{d}\rho/\mathrm{d}T$.

A close relation between $\rho(T)$ and $\theta(T)$ is established for the conical structure. New δ-type singularities are observed for the first time in $\mathrm{d}\rho/\mathrm{d}T$ near spin-reorientation transitions, the results being analysed in terms of lattice softening at T_{SR1} and T_{SR2} and associated anomalies in the C_{33} elastic constant.

The electrical resistivity (ρ) was measured from 4.2 to 300 K with a four-wire potentiometric method giving a resolution of $1:10^6$. The temperature derivative $\mathrm{d}\rho/\mathrm{d}T$ was obtained directly using a quasistatic method with heating rates of about 100 mK $\mathrm{min^{-1}}$. A microprocessor enabled (ρ, T) values to be obtained every 20 mK and immediately calculated local values of $\mathrm{d}\rho/\mathrm{d}T$ (Moreira 1984, Sousa *et al* 1977).

The $\mathrm{NdCo_5}$ sample used here ($10 \times 1 \times 1\ \mathrm{mm^3}$) was prepared by argon arc-melting using 99.9% purity neodymium (Rare Earth Products) and 99.99% purity cobalt (Koch Light Laboratories).

Electron scanning microanalysis and metallographic analysis were performed at CEMMUP, University of Porto. These studies revealed good chemical homogeneity, a virtual absence of secondary phases, and enabled the determination of the effective composition to within 1% accuracy. Details of the x-ray analysis have been given previously (Algarabel *et al* 1987) and this again confirmed the single phase nature of our sample.

Figure 1 shows the temperature dependence of the electrical resistivity of $\mathrm{NdCo_5}$, from 4.2 K to 300 K, after subtraction of the residual resistivity (ρ_0).

For temperatures above ~120 K the electrical resistivity increases almost linearly with temperature, except between ~240 K and ~280 K where the slope decreases noticeably. As shown below, this is due to the onset of a conical ferromagnetic structure with the angle θ changing rapidly with temperature. This spin-reorientation region is here studied in detail as shown in the inset of figure 1. The arrows mark the two spin-reorientation transitions of $\mathrm{NdCo_5}$, at T_{SR1} (transition from basal plane to cone) and at T_{SR2} (cone to *c* axis).

Below ~120 K, $\rho(T)$ progressively deviates from linear behaviour, exhibiting a much faster decrease with temperature. Besides phonon quantisation, part of the effect is attributed to de-population of crystal field levels (Elliott 1954, Rao and Wallace 1970).

Using the method referred to above, we directly measured the temperature derivative of the electrical resistivity in the same temperature range, as shown in figure 2. The spin-reorientation transitions are associated with the two sharp anomalies observed in $\mathrm{d}\rho/\mathrm{d}T$ near T_{SR1} and T_{SR2}. To our knowledge, this is the first experimental observation of such localised critical features in the resistivity temperature derivative of $\mathrm{NdCo_5}$. The previously available $\mathrm{d}\rho/\mathrm{d}T$ data (Andreev *et al* 1982) had insufficient resolution to reveal such fine structure details near T_{SR1} and T_{SR2}. The present data also extend the information on $\mathrm{d}\rho/\mathrm{d}T$ from 200 K to the helium temperatures revealing a broad maximum in $\mathrm{d}\rho/\mathrm{d}T$ at around 80 K.

In addition to the usual isotropic contributions due to impurity, phonon and spin disorder scattering, the electrical resistivity also depends on the angle θ between the

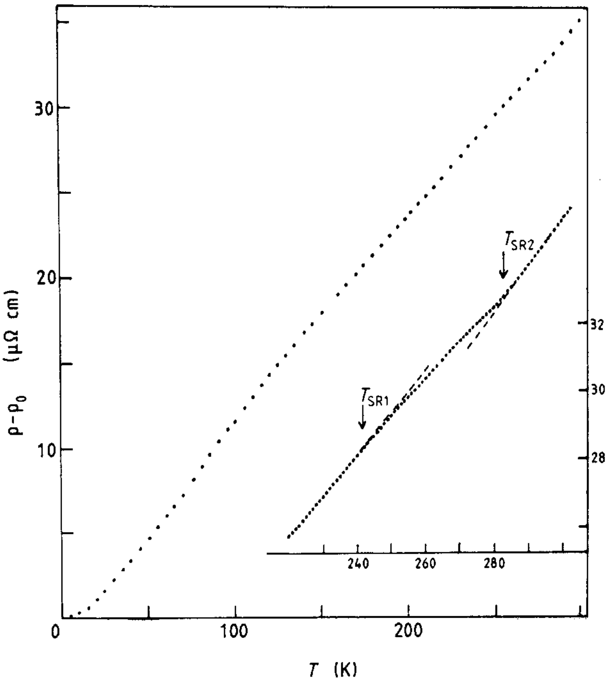

Figure 1. The temperature dependence of the electrical resistivity (ρ) of $NdCo_5$ between 4.2 K and 300 K, after subtraction of the residual (ρ_0).

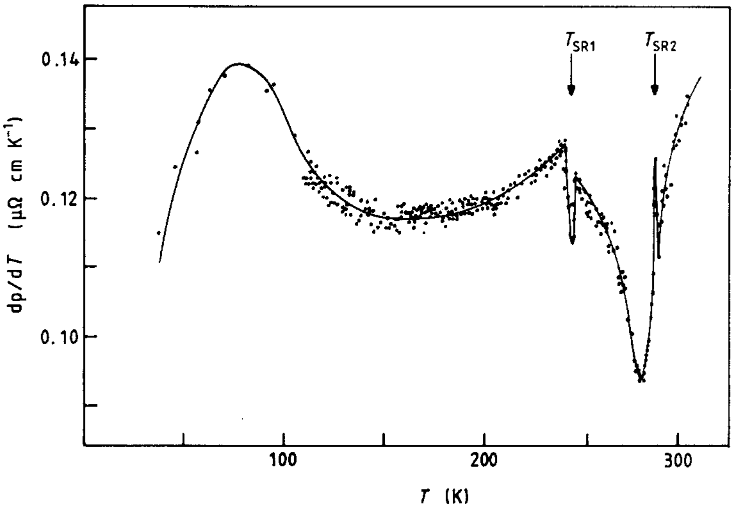

Figure 2. The temperature derivative of the electrical resistivity ($d\rho/dT$) of $NdCo_5$ as a function of T. The spin-reorientation transitions are indicated by vertical arrows.

spontaneous magnetisation M_s and the c axis. When this angle changes rapidly near a spin reorientation transition (T_{SR}) a corresponding anomaly appears in $d\rho/dT$ (Salamon and Simons 1973, Sousa *et al* 1979). From symmetry considerations ρ is an even function of θ (or $\cos\theta$) and, in general, a second-order development of $\rho(\theta)$ is sufficient to describe such a dependence:

$$\rho(T) = a(T) + b(T)\cos^2\theta \tag{2}$$

where $a(T)$ is the background (isotropic) resistivity contribution and $b \propto M_s^2(T)$. The $\cos^2\theta$ term usually represents a very small correction to the resistivity, about 1.5% in $NdCo_5$. When $T_{SR} \ll T_c$ (in $NdCo_5$ we have $T_c = 910$ K) we can neglect the temperature dependence of M_s taking b as a constant.

In $NdCo_5$ the isotropic resistivity is responsible for the quasi-linear temperature dependence of $\theta(T)$ outside the spin-reorientation region. The background resistivity in the transition region can be obtained by extrapolation of the fitted resistivity curve between 200 K and T_{SR1}: a second-order polynomial in T is sufficient for this purpose. Ascribing $\rho_n(T)$ to this background one can write

$$\rho(T) = \rho_n(T) + b\cos^2\theta. \tag{3}$$

The constant b can be estimated if we recall that for $T = T_{SR1}$ we have $\cos\theta = 1$, and thus $b = \rho(T_{SR1}) - \rho_n(T_{SR1})$. Therefore

$$(\rho(T) - \rho_n(T))/(\rho(T_{SR1}) - \rho_n(T_{SR1})) = \cos^2\theta. \tag{4}$$

This equation enables us to obtain the temperature dependence of θ between T_{SR1} and T_{SR2}, using the resistivity data. As shown in figure 3 the curve obtained is similar to the one derived from direct measurements (Bartholin *et al* 1966, Tatsumoto *et al* 1971, Klein *et al* 1975, Ermolenko 1980).

Another check on $\theta(T)$ can be made using (1) and a computer fit to the experimental points available for $K_1(T)$ and $K_2(T)$ (Ohkoshi *et al* 1976). As shown by the dotted curve in figure 3, a reasonable description of the resistivity-derived $\theta(T)$ curve (4) is obtained over the whole temperature range, the maximum deviations being less than 3°.

We now investigate higher-order effects associated with the spin-reorientation transitions and look, in particular, at the temperature derivative $\mathrm{d}\rho/\mathrm{d}T$. Assuming that relation (3) is still appropriate we obtain

$$\mathrm{d}\rho/\mathrm{d}T = \mathrm{d}\rho_n/\mathrm{d}T - b\sin 2\theta(\mathrm{d}\theta/\mathrm{d}T). \tag{5}$$

Let us consider the expected behaviour of $\mathrm{d}\rho/\mathrm{d}T$ near T_{SR1} and T_{SR2}.

For $T < T_{SR1}$ we have $\theta = \pi/2$ and $\mathrm{d}\theta/\mathrm{d}T = 0$ thus $\mathrm{d}\rho/\mathrm{d}T = \mathrm{d}\rho_n/\mathrm{d}T$. Just above T_{SR1} one can write

$$\cos\theta = c_1\sqrt{T - T_{SR1}} \tag{6}$$

where c_1 is a constant dependent on the magnitude of K_1 and K_2 (Belov *et al* 1976). This leads to

$$(\mathrm{d}\rho/\mathrm{d}T)^{+}_{SR1} = \mathrm{d}\rho_n/\mathrm{d}T - bc_1 \tag{7}$$

which shows that $\mathrm{d}\rho/\mathrm{d}T$ has a discontinuity at T_{SR1} given by:

$$\Delta(\mathrm{d}\rho/\mathrm{d}T)_{SR1} = -bc_1. \tag{8}$$

In a similar manner, for $T > T_{SR2}$ we have $\theta = 0$ and $\mathrm{d}\rho/\mathrm{d}T = (\mathrm{d}\rho_n/\mathrm{d}T)$. Just below T_{SR2} we can write

$$\sin\theta = c_2\sqrt{T_{SR2} - T} \tag{9}$$

where c_2 is a constant depending on the magnitude of K_1 and K_2 (Belov *et al* 1976). This leads to a discontinuity in $\mathrm{d}\rho/\mathrm{d}T$ at T_{SR2} given by:

$$\Delta(\mathrm{d}\rho/\mathrm{d}T)_{SR2} = -bc_2. \tag{10}$$

Between T_{SR1} and T_{SR2} (5) gives a continuous variation of $\mathrm{d}\rho/\mathrm{d}T$, essentially imposed by the temperature dependence of the quantity $\sin 2\theta$ $(\mathrm{d}\theta/\mathrm{d}T)$.

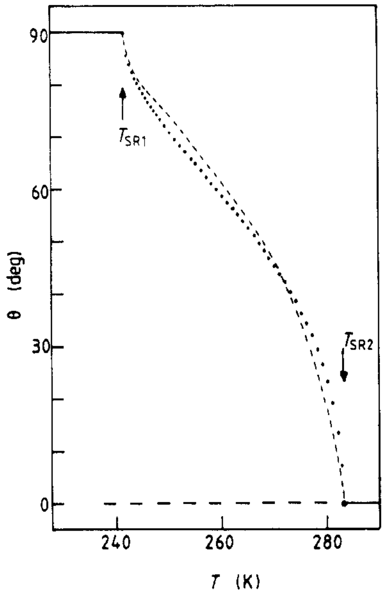

Figure 3. The temperature dependence of θ (the angle between M_s and the c axis). Broken curve—results derived from experimental ρ data, using (4); dotted curve—results derived using the anisotropy constants K_1 and K_2 using (1).

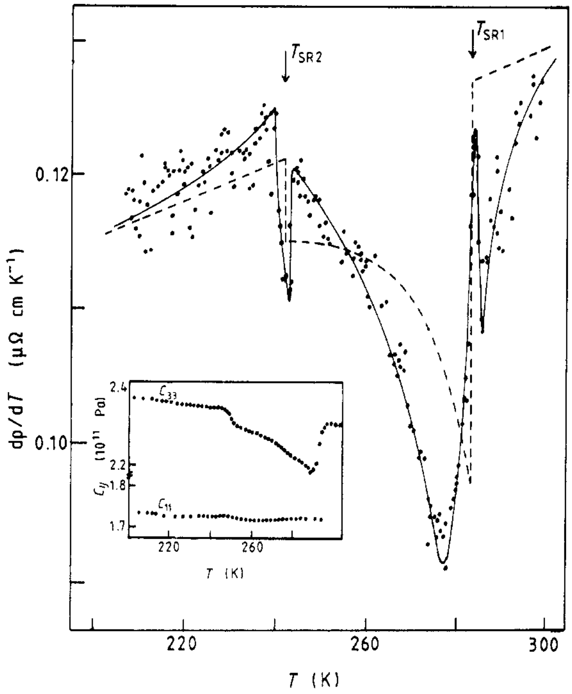

Figure 4. The temperature derivative of the electrical resistivity ($\mathrm{d}\rho/\mathrm{d}T$) of $NdCo_5$ in the vicinity of T_{SR1} and T_{SR2}. Dotted points—experimental data; broken curve—calculated curve using (5) and $\theta(T)$ derived from the anisotropy constants K_1 and K_2; inset—elastic constants of $NdCo_5$ (data from Patterson *et al* 1986).

Using (5) and $\theta(T)$ derived from K_1 and K_2 (b and $\mathrm{d}\rho_n/\mathrm{d}T$ taken from our resistivity data) we obtain the $\mathrm{d}\rho/\mathrm{d}T$ curve shown in figure 4.

The average behaviour of $\mathrm{d}\rho/\mathrm{d}T$ is roughly described by such a calculated curve and even the order of magnitude of the $\Delta(\mathrm{d}\rho/\mathrm{d}T)$ changes at T_{SR1} and T_{SR2} are included. However, the narrow δ-type singularities near T_{SR1} and T_{SR2} are not predicted and, in addition, (5) does not explain why the minimum in $\mathrm{d}\rho/\mathrm{d}T$ occurs slightly below T_{SR2} ($\sim$279 K instead of 283 K).

We believe that the localised δ-type anomalies observed in $\mathrm{d}\rho/\mathrm{d}T$ are related to the sudden softening of the crystal lattice which occurs in $NdCo_5$ at both spin-reorientation transitions (Patterson *et al* 1986). This originates finite discontinuities in the elastic constant C_{33} for longitudinal waves propagating along the c axis, as shown in the inset of figure 4 (data from Patterson *et al* 1986). This in turn causes sudden changes in the phonon velocity and phonon energy, producing small discontinuities in the phonon resistivity. δ-type singularities are therefore predicted in the temperature resistivity derivative $\mathrm{d}\rho/\mathrm{d}T$ both at T_{SR1} and T_{SR2}. The sign and magnitude of each anomaly is directly related to the step in C_{33} at each temperature. Since $\Delta C_{33}(T_{SR1}) < 0$ and $\Delta C_{33}(T_{SR2}) > 0$, we predict different signs for the δ-type anomaly in $\mathrm{d}\rho/\mathrm{d}T$.

These theoretical predictions near T_{SR1} and T_{SR2} are consistent with our experimental results for $\mathrm{d}\rho/\mathrm{d}T$ (figure 4). However, some remarks should be made on a subtle distinction between the experimental data near T_{SR1} and those near T_{SR2}. Close to T_{SR1} the $\cos^2\theta$ resistivity dependence gives a practically constant $\mathrm{d}\rho/\mathrm{d}T$ contribution (broken curve in figure 4) and thus the superimposed δ-anomaly (due to the sharp variation in $\mathrm{d}C_{33}/\mathrm{d}T$) should be direct and cleanly displayed by the experimental $\mathrm{d}\rho/\mathrm{d}T$ curve. On the other hand, near T_{SR2} there is a direct competition between two opposing effects: a deep minimum in $\mathrm{d}\rho/\mathrm{d}T$ due to the $\cos^2\theta$ dependence (dotted curve in figure 4) and a sharp maximum due to $\mathrm{d}C_{33}/\mathrm{d}T$. This interplay considerably distorts $\mathrm{d}\rho/\mathrm{d}T$ near T_{SR2}, in particular displacing the minimum in $\mathrm{d}\rho/\mathrm{d}T$ to a temperature slightly below T_{SR2}. It may also occur in some cases, that the deep minimum outweighs the other anomaly.

In conclusion, although it is difficult to separate the two opposing critical contributions to $\mathrm{d}\rho/\mathrm{d}T$ near T_{SR2} in $NdCo_5$ (we are at the limit of the experimental resolution) our data clearly reveal the predicted δ-type singularities in $\mathrm{d}\rho/\mathrm{d}T$ at spin-reorientation transitions, the effect being attributed to the discontinuities in the elastic constants due to magnetoelastic softening.

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