MAGNETIC PHASE TRANSITIONS IN TERBIUM SINGLE CRYSTALS (*)

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ABSTRACT—New high precision measurements of the electrical resistivity and its thermal derivative for a high quality terbium single crystal along a basal direction are presented and discussed in connection with the para-antiferromagnetic and anti-ferromagnetic transitions.

In terbium, the charge distribution of 4f-electrons is toroidal in character, originating a number of interesting effects and a succession of two magnetic transitions. As the temperature is monotonically decreased Tb goes from the para- to the antiferromagnetic phase at $T_N = 229\,K$ (2nd order), the localized ionic spins ($\vec{S}_i$; site $\vec{R}_i$) presenting then an helicoidal arrangement:

$$S_i^x = S.\cos(\vec{q}.\vec{R}_i) \quad , \quad S_i^y = S.\sin(\vec{q}.\vec{R}_i) \quad , \quad S_i^z = 0 \qquad (1)$$

$\vec{q}$ characterizes the helix period along the $\vec{c}$-axis $(2\pi/q)$ and $qc/2$ gives the rotation angle of S_i from an atomic plane to the next along $\vec{c}$ ($\simeq 20°$).

A second transition, of the order-order type, takes place at $T_c = 221\,K$, where Tb goes into a simple ferromagnetic state ($\vec{q} = 0$

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in eq. 1). Spins $\vec{S}_i$ are still in the basal plane, but they now point everywhere in the same direction.

These magnetic transitions received considerable attention in the past [1], through extended measurements of thermodynamic (specific heat, magnetization, susceptibility; *no* latent heat measurements) and transport properties, namely electrical and thermal resistivity, thermopower, ultrasonic attenuation.

In spite of this, most properties did not reveal clearly the order-order transition at T_e. Electrical resistivity studies have been most informative, and for that reason we restrict ourselves here to this type of measurements.

The temperature dependence of the electrical resistivity $\rho(T)$ has been studied by Hegland et al [2] along a- and c-crystallographic directions and from $4 - 300$ K. The considerable anisotropy is apparently enhanced in the paramagnetic phase which is a rather surprising result.

(i) *Along the c-axis*, the onset of the antiferromagnetic phase at T_e originates a sharp rise in ρ, as shown in Fig. 1 (from ref. 2): The increase in ρ is due to the formation of magnetic superzones caused by the new periodicity in the system (helix structure; wave vector $\vec{q}$ along $\vec{c}$); this produces new gaps, reducing the effective

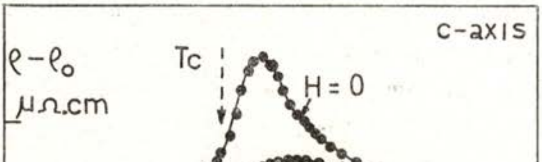

Fig. 1 — Temperature dependence of ρ for Terbium along the c-axis in transverse (along b) external magnetic field (from ref. [2]).

number of electrons to carry the current along the c-axis. Since T_c is close to the temperature at which magnetic order is suppressed (T_N), such gaps are relatively small and the anomaly $\Delta\rho/\rho$ at T_c becomes correspondingly small (≈ 0.04). Part of the jump at T_c may also come from a change in magnon dispersion. Superzone effects in the antiferromagnetic phase naturally produce the well develloped hump observed in ρ (Fig. 1). This hump can be suppressed if the helical phase is destroyed by application of an external magnetic field along a basal direction, as shown in the same figure ($H \geqslant 11\,kOe$; interlayer rotation supressed; spins $\vec{S}_i$ frozen along $\vec{H}$).

A clear identification of the Néel temperature is difficult from simple $\rho(T)$ measurements. A better insight can be gained with measurements of the temperature derivative ($d\rho/dT$), as performed by Meaden et al [3] in the vicinity of T_N.

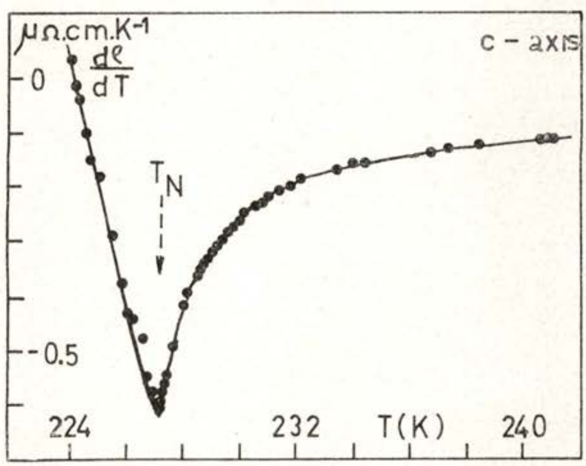

Fig. 2 — Temperature dependence of $d\rho/dT$ for Terbium along the c-axis (from ref. [3]).

As shown in Fig. 2, a sharp minimum occurs in $d\rho/dT$ at $T_N = 227.2$ K. The critical behaviour reveals a log-divergence for $0.6 \leqslant T - T_N \leqslant 3$ K, whereas for $T - T_N \gtrsim 3$ K one has a $(T-T_N)^{-1/2}$ dependence. For $T - T_N < 3$ K the specific heat also diverges pratically in a logarithmic way [3]. As will be seen later, this result has interesting consequences regarding the critical behaviour.

(ii) *Along a basal direction,* previous measurements [2] revealed a rather uninteresting behaviour in ρ: a simple knee in the curve at the Néel point and no obvious anomaly around T_c. Besides that, no experimental data seem available for $d\rho/dT$, neither attempts to analyze the critical behaviour near T_N and T_c.

In view of this we decided to perform very accurate measurements of ρ and $d\rho/dT$ along a basal direction (a-axis). For the first time, we believe, a sharp anomaly has been detected in ρ_a and $(d\rho/dT)_a$ near T_c and new information obtained for the critical behaviour of the basal-resistivity of Tb near T_N. The anomalously large $(d\rho/dT)_a$ values in the ferromagnetic phase are also analyzed here.

Fig. 3 shows our results for ρ_a and $(d\rho/dT)_a$:

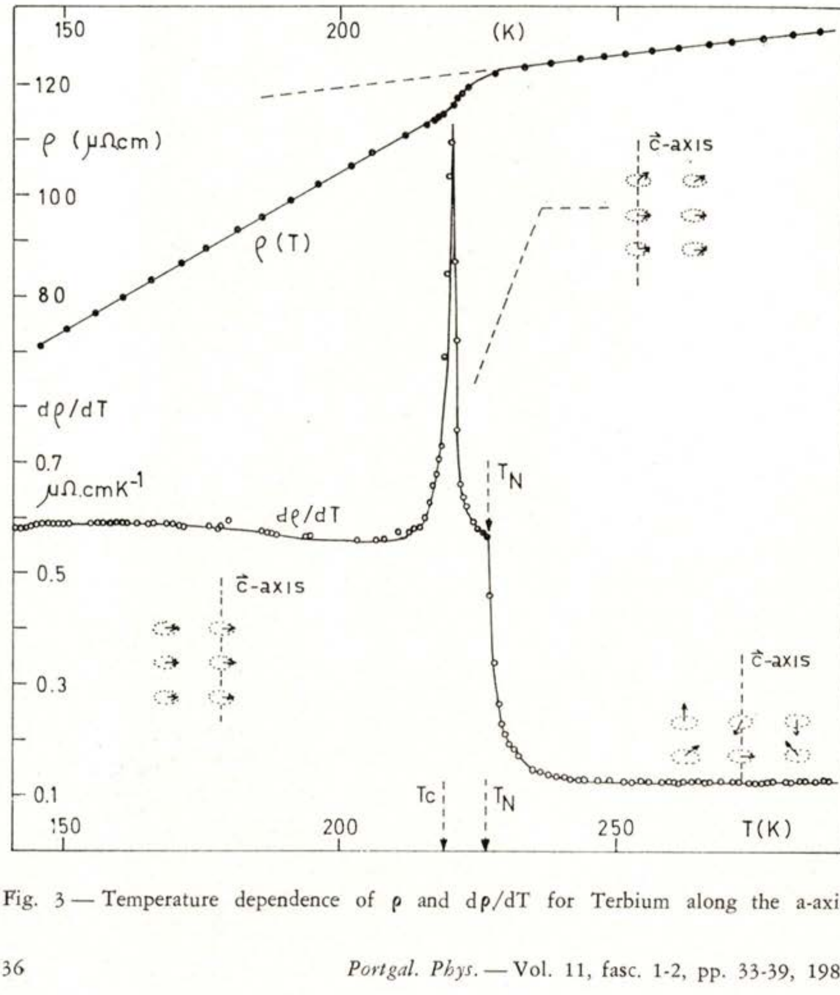

Fig. 3 — Temperature dependence of ρ and $d\rho/dT$ for Terbium along the a-axis.

The usual knee in ρ_a at the Néel point is confirmed in Fig. 3, but our data also reveal a neat kink in ρ_a at T_c. While in previous investigations the latter anomaly is hardly observed (see e.g. the specific heat C_m in ref. 4), it is exhuberantly present in our $d\rho/dT$ curve, where a very sharp peak appears just at the ferro-antiferromagnetic transition (T_c). The Néel temperature is also well defined, by the sudden and sharp decrease in $d\rho/dT$. Above T_N, $d\rho/dT$ rapidly reaches a constant value of $\sim 0.13\ \mu\Omega.cm.K^{-1}$, attributable to electron-phonon scattering.

The critical behaviour of $(d\rho/dT)_a$ near T_N has been analyzed in detail for $T > T_N$. Our data closely follow a logarithmic dependence:

$$(d\rho/dT)_a = A . \ln (T - T_N) + B \qquad (2)$$

with $A = -0.078$, $B = 0.2834$ ($\mu\Omega.cm.K^{-1}$ units), $T_N = 226.9\,K$; the fit is valid down to reduced temperatures $\sim 10^{-3}$ (i.e. $T - T_N \simeq 0.3K$). This result confirms that $(d\rho/dT)_a$ and $(d\rho/dT)_c$ have the same functional (log) dependence near T_N, as expected within the universality hypothesis. Since the specific heat practically diverges as $\ln (T - T_N)$ near the Néel point it also follows that:

$$(d\rho/dT)_a \sim C_m (T) \qquad (3)$$

This relation is expected to hold when short-range fluctuations dominate the electrical resistivity near T_N [5]: large-momentum transfer to the electrons.

A brief comment is now in order on the qualitative shapes of $(d\rho/dT)_a$ and $(d\rho/dT)_c$ near T_N (Figs. 2 and 3): whereas along c the shape is characteristic of typical antiferromagnets (e.g. cromium; $d\rho/dT < 0$), for the a-axis it rather looks like usual ferromagnetic systems ($d\rho/dT > 0$; see Fig. 4).

In physical terms, what really happens is that electrons flowing along a basal plane in Tb always 'see' the corresponding spins in the *same* direction. This is exactly a ferromagnetic-like situation, which, of course, appears reflected in the shape of the $(d\rho/dT)_a$ curve for terbium. As an illustration, Fig. 4 shows the similarities between $d\rho/dT$ anomalies for Tb near T_N (a-axis) and for gadolinium near the Curie temperature [6].

A striking feature in terbium is the anomalously high $(d\rho/dT)_a$ derivative in the ferromagnetic phase, even for $T \ll T_c, T_N$. In simple ferromagnets like Ni, Fe $(d\rho/dT)$ decreases rather fast below T_c, due

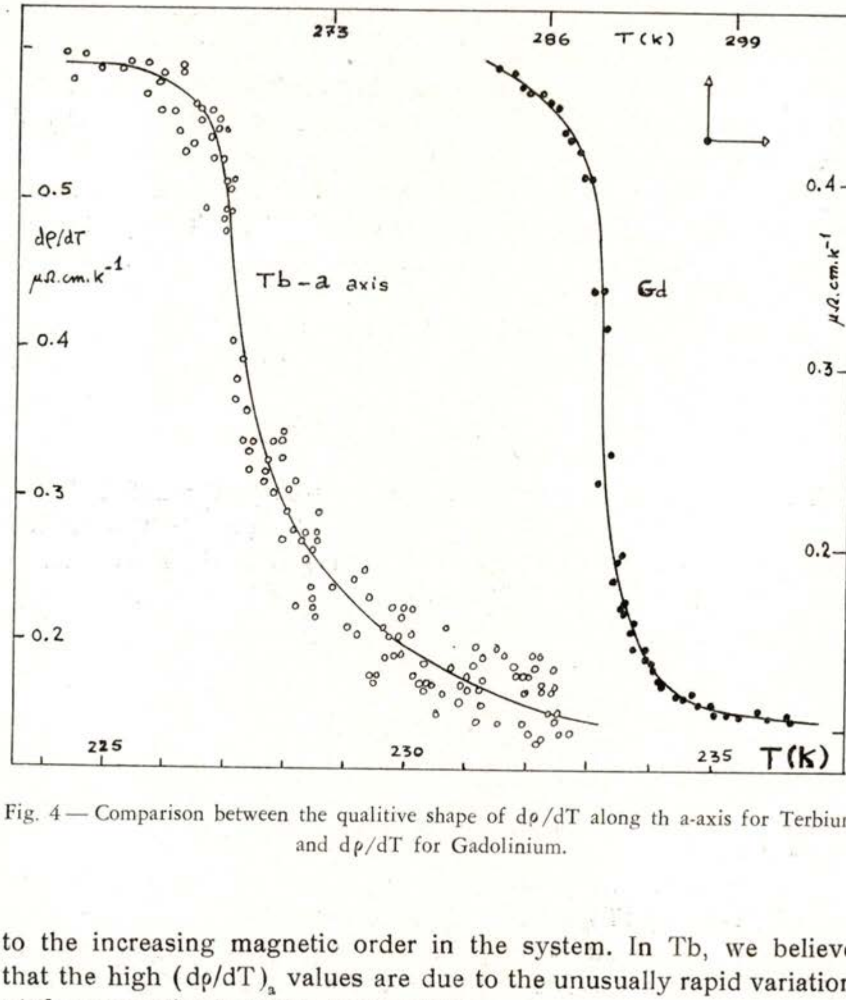

Fig. 4 — Comparison between the qualitive shape of $d\rho/dT$ along th a-axis for Terbium and $d\rho/dT$ for Gadolinium.

to the increasing magnetic order in the system. In Tb, we believe that the high $(d\rho/dT)_a$ values are due to the unusually rapid variation of the magnetization with temperature:

$$M(0) - M(T) = A \cdot T^{3/2} \cdot e^{-\Delta/kT} \tag{4}$$

with $M(0) = 325$ emu.g^{-1}, $A = 0.03595$ emu.g^{-1}.K$^{-3/2}$, $\Delta/k = 20$ K [7]. For order of magnitude calculations, let us use a mean fiel model for the magnetic resistivity:

$$\rho_m(T) = \rho_\infty [1 - (M(T)/M(0))^2] \tag{5}$$

where $\rho_\infty \simeq \rho(T_N) = 123\ \mu\Omega$.cm. We then find $d\rho_m/dT = 0.37\ \mu\Omega$.cm.K^{-1} at $T = 120$ K, and $d\rho_m/dT = 0.39\ \mu\Omega$.cm.K^{-1} for $T = 150$ K. If one adds the phonon contribution to the resistivity derivative ($d\rho_p/dT \sim 0.13\ \mu\Omega.cm.K^{-1}$) we get $d\rho/dT = 0.50$; $0.52\ \mu\Omega.cm.K^{-1}$ at $T = 120$ and 150 K, respectively, whereas experiment gives correspondingly: $d\rho/dT = 0.58$ and $0.59\ \mu\Omega.cm.K^{-1}$. In view of the approximate nature of our calculations, the results are indeed quite good.

The high-quality terbium single crystal used in this work has been purified by solid state electron transport, and our high accuracy data on $d\rho/dT$ were obtained with a quasistatic method previously described in the literature [8].

Work is now in progress to investigate the critical behaviour of the thermoelectric power and thermal conductivity in the same Tb single crystal.

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REFERENCES

[1] SEE e.g. R. J. ELLIOTT (ed.) in Properties of Rare Earth Metals, Plenum Press 1972, p. 245.

[2] HEGLAND, D. E., LEVGOLD, S., SPEDDING, F. H., *Phys. Rev.*, **131**, 158 (1963).

[3] MEADEN, G. T., SZE, N. H., JOHNSTON, J. R., in Dynamical Aspects in Critical Phenomena, Budnick and Kawatra (ed.), Gordon and Berach 1972, p. 315.

[4] JENNINGS, L. D., STANTON, R. M., SPEDDING, F. H., *J. Chem. Phys.*, **27**, 909 (1957).

[5] ALEXANDER, S., HELMAN, J. S., BALBERG, I., *Phys. Rev.*, **B13**, 304 (1976).

[6] DAMAS, A. M., RESTIVO, M. T., SOUSA, J. B., *Anais da Faculdade de Ciências do Porto,* **LIX**, 1 (1976).

[7] MACKINTOSH, A. R., *Physics Letters,* **4**, 140 (1963).

[8] SOUSA, J. B., AMADO, M. M., BRAGA, M. E., PINTO, R. P., MOREIRA, J. M., HUKIN, D., *Communications on Physics,* **2**, 95 (1977).