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LETTER TO THE EDITOR

Anomalous behaviour of the electrical resistivity of $Y_2Fe_{14}B$

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Abstract. Accurate measurements of the electrical resistivity (ρ) and its temperature derivative ($d\rho/dT$) in the ferromagnet $Y_2Fe_{14}B$ have been made from 4–750 K, and the results have been compared with those previously obtained for $Nd_2Fe_{14}B$.

In both cases $\rho(T)$ increases rapidly at low temperatures, approaching quasi-saturation well below the Curie point (T_c). The weak resistivity singularity near T_c is anomalous, giving *negative* $d\rho/dT$ values. Small humps also occur in $\rho(T)$ just below T_c.

The critical analysis of these results indicates that the low-temperature anomaly is likely to be associated with spin-fluctuation effects, whereas the hump effects below T_c are related to the giant Invar effect in these compounds.

Recently we reported [1] the analogous behaviour of the electrical resistivity (ρ) and its temperature derivative $d\rho/dT$ in a ferromagnetic single crystal of $Nd_2Fe_{14}B$, with measurements from 4–750 K ($T_c = 592$ K). The resistivity exhibits a fairly large and rapid initial increase with temperature (up to $T \sim 0.5\ T_c$), followed by an approach to quasi-saturation at temperatures well below the Curie point. The ferroparamagnetic transition then only produces small anomalies in ρ and $d\rho/dT$.

The results show that the magnetic scattering of the conduction electrons hardly changes with temperature above $\sim 0.5\ T_c$. In contrast, the rapid initial increase of ρ at low temperatures leads to a sharp peak in $d\rho/dT$ around 75 K ($T/T_c \simeq 0.13$). In a previous work [1] we suggested that such an anomalous initial increase of ρ could be due to the thermal disorder of the Nd magnetic moments and to the excitation of successive Nd^{3+} crystal-field levels up to saturation. A simple crystal-field formula could account for the low-temperature behaviour of ρ and $d\rho/dT$, without, however, explaining the high-temperature data. Suggestions were made regarding suitable experiments to clarify the situation.

A relevant experiment has now been performed, with a detailed study of $\rho(T)$ and $d\rho/dT$ in the ferromagnet $Y_2Fe_{14}B$, from 4–750 K. The results obtained were unexpected (in the light of our previous publication) and may lead to a different interpretation of the resistivity behaviour in $R_2Fe_{14}B$ compounds.

As shown in figure 1 the anomalous behaviour of the electrical resistivity previously observed in $Nd_2Fe_{14}B$ [1] is also present in $Y_2Fe_{14}B$, with much the same characteristics: a rapid initial increase of ρ with T, followed by quasisaturation well below T_c. If we

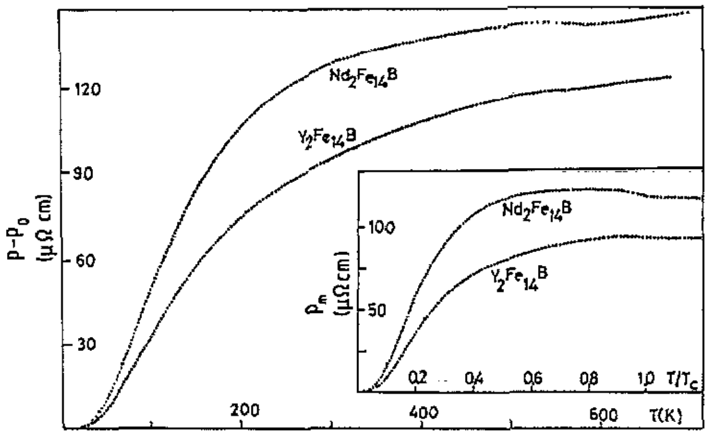

Figure 1. Electrical resistivity in $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$. Inset: magnetic resistivity versus reduced temperature T/T_c in $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$.

extract the phonon contribution from $\rho(T) - \rho_0$ (see below) the saturation effect in the magnetic part of the resistivity (ρ_m) becomes more evident, as shown in the inset to figure 1. Since Y is non-magnetic, the effect cannot be due to 'rare earth crystal field effects' or to 'rare earth spin-disorder effects'. It should therefore be associated with the Fe magnetic moments, although for $Nd_2Fe_{14}B$ we cannot exclude a superimposed contribution from the Nd crystal-field levels at low temperatures.

Figure 2 shows the experimental derivative $d\rho/dT$ as a function of temperature for $Y_2Fe_{14}B$ and $Nd_2Fe_{14}B$ where the great similarities of both cases are confirmed.

The general temperature dependence of the electrical resistivity of $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$ resembles the $\rho(T)$ behaviour observed in weak magnets dominated by spin fluctuations [2–4]. We should mention, in particular, the cases of $CeFe_2$ [5] and UAl_2, UPt_3 [6], showing a rapid initial increase of ρ (AT^2 dependence) followed by saturation at higher temperatures. In accordance with the spin-fluctuation theories [2–3] a peak is observed in the resistivity derivative at a characteristic spin-fluctuation temperature T_{sf}.

Although the available treatments of spin-fluctuations in weak magnets may not be applicable to the complex systems treated here, the strong (direct) 3d exchange interactions among the Fe atoms in the layered Fe regions favour the dominance of itinerant magnetism. Thus, large spin fluctuations could be thermally excited, leading to strong electron scattering at temperatures considerably below T_c.

We should mention the great similarity between the $\rho(T)$ saturation behaviour in $R_2Fe_{14}B$ and in binary compounds such as UFe_2 ($T_c = 162$ K) and UNi_2 ($T_c = 24$ K) [7].

An interesting feature of the Curie point anomaly in $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$ is the negative sign of $d\rho_m/dT$ (inset to figure 2), in contrast with the case of pure Fe where $d\rho_m/dT > 0$. Such anomalous behaviour in $R_2Fe_{14}B$ compounds is better displayed by the magnetic resistivity curves $\rho_m(T)$ obtained by subtracting from $\rho(T)$ the phonon resistivity $\rho_{ph}(T)$. This was calculated with a Bloch–Grüneisen formula, using a Debye temperature $\theta \simeq 414$ K [8] and fitting $d\rho_{ph}/dT$ to our experimental value of $d\rho/dT$ at temperatures sufficiently above T_c.

It becomes clear that ferromagnetic order produces an increase in the resistivity, the effect resembling the characteristic humps usually observed in antiferromagnets below T_N due to superzone gap effects [9]. However, both $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$ are true ferromagnets, with the thermal averaged magnetic moments $\langle \boldsymbol{\mu}_i(\mathrm{Fe}) \rangle$, $\langle \boldsymbol{\mu}_j(\mathrm{Nd}) \rangle$ always

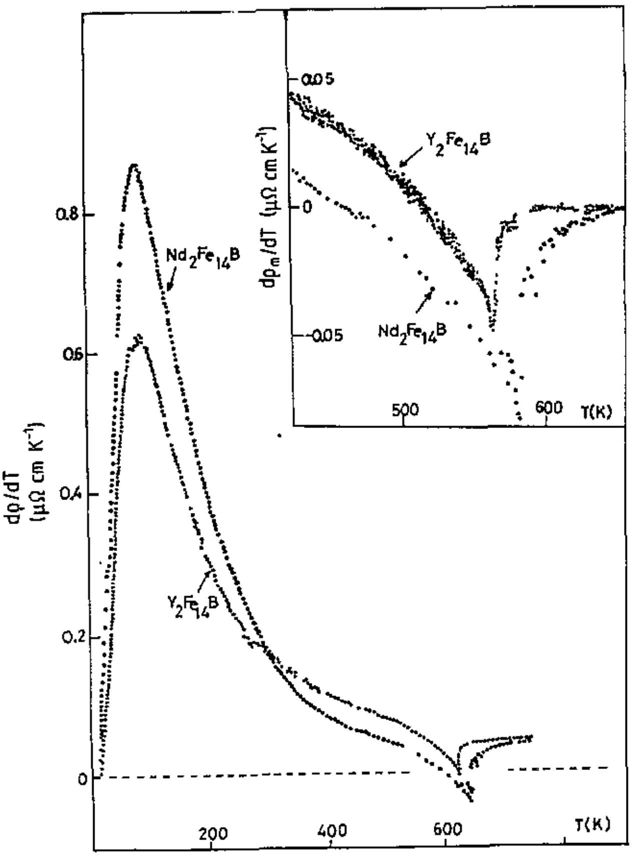

Figure 2. Temperature derivate of the electrical resistivity $d\rho/dT$ in $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$. Inset: temperature derivate of the magnetic resistivity $d\rho_m/dT$ as a function of reduced temperature for $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$.

parallel and pointing in the same direction in all lattice sites (i, j). Therefore, superzone gap effects do not occur. A qualitatively similar hump effect in the electrical resistivity below T_c has been reported for $Nd_{15}Fe_{77}B_8$ [10], but it is virtually absent in $Nd_5Fe_{62}B_{33}$ [10].

The anomalous resistivity humps below T_c are probably due to the large Invar effect observed in these compounds [11, 12]. As is well known, in Invar alloys the increase in volume below T_c is associated with a large softening of the elastic constants [13, 14]. In particular, the bulk modulus, B, goes through a deep minimum at a temperature T^* below T_c, so that between T^* and T_c we have $dB/dT > 0$.

As initially pointed out by Viard and Gavoille for the case of Fe_3 Pt Invar alloys [15], these changes in B (in some cases up to ~50% [13]) directly affect the scattering of the conduction electrons by phonons, through the usual expression for $\rho_{ph}(T)$ at high temperatures $(T > \theta)$:

$$\rho_{ph}(T) = (A/B)(1 - gw)T.$$

Here A is constant, B is the bulk modulus, g is a constant close to unity and w is the lattice expansion [15].

Analyses along the same lines have also been presented for the cases of $La(Fe, Si)_{13}$ [16] and $La(Fe, Al)_{13}$ [17] compounds.

Since for $R_2Fe_{14}B$ we have $w \ll 1$ [12], one can write the approximation:

$$\rho_{ph}(T) \simeq \text{constant}(1/B)T.$$

From this it follows that an increase of ρ_{ph} with decreasing T (hump effect) in fact occurs when

$$(1/B)\mathrm{d}B/\mathrm{d}T > 1/T.$$

This requires $\mathrm{d}B/\mathrm{d}T > B/T > 0$, a condition which is satisfied in $R_2Fe_{14}B$ compounds when T is close to T_c.

The relative change in ρ_{ph} can be directly expressed in terms of $\Delta B/B$:

$$\frac{\rho_{ph}(T) - \rho_{ph}^{(0)}(T)}{\rho_{ph}(T_c)} = \frac{B(T_c) - B(T)}{B(T_c)}$$

where $\rho_{ph}^{(0)}(T)$ is the extrapolation (below T_c) of the linear paramagnetic phonon resistivity.

From the maxima in the humps in the magnetic resistivity curves $\rho_m(T)$ we can get the approximate values $\Delta B/B(T_c) \geqslant 20\%$ and $\Delta B/B(T_c) \geqslant 4\%$ for $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$ respectively (we neglect, however, the spin-disorder thermal dependence in such estimates).

We can now summarize the main results of our work.

(i) Crystal field effects [1] cannot explain the observed results in $Y_2Fe_{14}B$.

(ii) The anomalous behaviour of ρ and $\mathrm{d}\rho/\mathrm{d}T$ in $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$ is mainly associated with the presence of the Fe ions.

(iii) The initial rapid growth of $\rho(T)$ and the approach to saturation imply considerable short-range effects well below T_c. A suggestion is advanced with regard to the possible role played by enhanced spin-fluctuations in these 3d dominated (Fe) ferromagnets.

(iv) The small humps in $\rho(T)$ below T_c are probably associated with the huge Invar effect in $R_2Fe_{14}B$ compounds which affects the phonon resistivity near T_c.

Work is now in progress to investigate the specific role played by the different rare earth ions (R) in $R_2Fe_{14}B$ compounds. In particular one would expect differences in the magnitude of the resistivity humps, as also happens with the Invar effect [11, 12].

The thermoelectric power (S) is also being investigated over the $R_2Fe_{14}B$ series. Preliminary data for $Nd_2Fe_{14}B$ show rather anomalous behaviour: with large S values at intermediate temperatures ($|S| > 10\ \mu V\ K^{-1}$ at $T \sim 200$ K) and a complex temperature dependence below about 100 K, the thermopower goes through a maximum, then a minimum, before approaching zero at low temperatures. Superzone-like gap effects are also observed in S just below T_c.

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