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LETTER TO THE EDITOR

Critical behaviour in the spontaneous resistive anisotropy near a ferromagnetic percolation threshold

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Abstract. The spontaneous resistive anisotropy (SRA) in **Pd**Ni has been measured in the vicinity of the critical Ni concentration (c_0) necessary to establish a ferromagnetic ground state. These data provide the first demonstration of a power-law relation for the SRA on the reduced composition ($c/c_0 - 1$) near c_0; the associated critical exponent governing this relationship is estimated experimentally.

The spontaneous resistive anisotropy (SRA) is a measure of the difference in the resistance of a single-domain ferromagnetic conductor in zero induction (B) when the magnetisation lies parallel ($\parallel$) or perpendicular ($\perp$) to the current direction. Its existence has been known for over a century (Thomson 1857). The SRA is usually defined by the ratio (Dorleijn (1976) who discusses various extrapolation procedures for estimating the SRA).

$$\Delta\rho/\rho_0 = [3(\rho_\parallel(B) - \rho_\perp(B))/(\rho_\parallel(B) + 2\rho_\perp(B))]_{B\to 0}. \tag{1}$$

Two complementary approaches have been introduced to interpret such data, the first based on an itinerant picture (Mott 1936, Campbell *et al* 1970; Malozemoff 1986), and the second on a localised model (Friederich and Fert 1974). Both, however, rely on the presence of two principal components, a polarising field and spin–orbit coupling at the scattering sites, so that the SRA does not simply depend on the square of the magnetisation, as the magnetic component in the resistivity often does. This is most clearly seen in localised models where this anisotropy is shown to arise from electric quadrupole (D) scattering (Friederich and Fert 1974), with

$$\Delta\rho/\rho_0 = (D/V)[\langle J_z^2\rangle - \tfrac{1}{3}J(J+1)] \tag{2}$$

when the potential scattering term $V \gg D$; $\boldsymbol{J}$ is the total angular momentum at the scattering site. This latter model has been used primarily to discuss the anisotropy induced in paramagnetic systems (dilute **Au**Ho, for example) by an externally applied polarising field. By contrast, itinerant models have been applied predominantly to the large body of data accumulated on dilute alloys based on ferromagnetic transition-metal hosts (Dorleijn 1976), where the (polarising) exchange field is large. To date, however, no investigation of the SRA in systems close to the ferromagnetic instability has been performed (Senoussi *et al* 1977†), and no specific theoretical prediction for its behaviour

† These authors also mention the problems encountered in attempting to measure the SRA near c_0 in **Pd**Ni.

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exists. Here, we report the results of the first such investigation, introduce a new technique for estimating the SRA when this ratio becomes very small, demonstrate a power-law relationship for the SRA near the critical concentration (c_0) for ferromagnetism and determine the critical exponent governing this relationship.

The system we have chosen to investigate is dilute **Pd**Ni. Here, (i) c_0 has been well established at (2.30 ± 0.05) at.% Ni by a variety of techniques (Tari and Coles 1971, Murani *et al* 1974), (ii) Ni is known to carry an orbital moment in this host (Senoussi *et al* 1977, Hamzic *et al* 1978), and (iii) it was shown in a recent Letter (Kunkel *et al* 1987) that measurements of the SRA in low applied field ($H_a \leq 0.01$ T, which introduces no significant modification to either the intrinsic spin-polarisation or the conduction-electron trajectories) could be used to infer the presence of an emerging exchange field close to the critical composition. The last paper also contains details of the sample preparation and of the AC technique (Muir and Strom-Olsen 1976) used to measure the field-induced fractional change in resistance to a few parts in 10^6.

The problems that arise in measuring the SRA accurately on approaching the critical composition are illustrated in figure 1. At 5 at.% Ni ($T_c \simeq 64$ K) the conventional extrapolation procedure† can be used to find the SRA from equation (1); the value so obtained (−2.43 ± 0.03%) agrees well with the one previous measurement in this composition range (Senoussi *et al* 1977). The result that $\rho_\perp > \rho_\parallel$ is consistant with the predictions of a localised model (Kondo 1962) based on electric quadrupole scattering from either a d^8 or d^9 configuration at the Ni site. However, as the Ni composition is lowered towards c_0, not only does the magnitude of the SRA decrease rapidly, but the 'high field' slopes can be seen to increase substantially (as confirmed in table 1), so that estimates of the SRA based on this extrapolation procedure are subject to increasing uncertainty, as is also reflected in table 1. Indeed, in our opinion, the behaviour just discussed means that such an extrapolation technique cannot be relied upon to predict correctly the sign, let alone the magnitude, of the SRA, below 3 at.% Ni. By way of contrast, the insert in figure 1 demonstrates that the low-field data is unambiguous; it indicates the presence of a small SRA, the sign of which is definitely negative. Such behaviour can also be seen to occur down to 2.4 at.% Ni (figure 2), but by 2.3 at.% Ni no anisotropy could be observed at 1.5 K with the resolution currently available to us ($\rho_\perp - \rho_\parallel \sim 10$ pΩ cm). The appearance of a non-zero SRA in 'low applied field' (<0.01 T close to c_0) has been taken as convincing evidence for the presence of a strong exchange field‡ in these samples, indicating that the 2.4 at.% sample is definitely ordered at 1.5 K, whereas the 2.3 at.% specimen is not. Such unambiguous results cannot be obtained from magnetisation measurements, where strong curvature in low temperature Arrott plots make the associated ordering temperatures difficult to identify (Murani *et al* 1974) near c_0. Our investigation of the AC susceptibility of **Pd**Ni confirms the difficulties encountered in accurately estimating not only T_c, but also the associated critical exponents. As in **Pd**Co (Hamzic *et al* 1978) (but in contrast with **Pd**Mn) the existence of

† Various field dependences (namely H^2, $H^{1/2}$, etc) have been fitted to the higher field data, but none reproduce these data as well as the simple linear form shown.

‡ As we have argued previously, the appearance of some anisotropy in low applied field (<0.01 T) is strong evidence in favour of the presence of a significant exchange field. Such small applied fields cannot appreciably modify any intrinsic spin polarisation or the conduction-electron trajectories; we suggest that it simply renders the system single domain over distances on the order of the mean free path. Applied fields can certainly induce an anisotropy in paramagnetic systems (Friederich and Fert 1974), but such fields have to be sufficiently large to induce a significant polarisation of the scattering moments, typically $H_a/T \geq 1$ T K^{-1} is required, far in excess of the values quoted above. All of the above are confirmed by the failure of small applied fields to induce an anisotropy in samples with less than 2.3 at.% Ni.

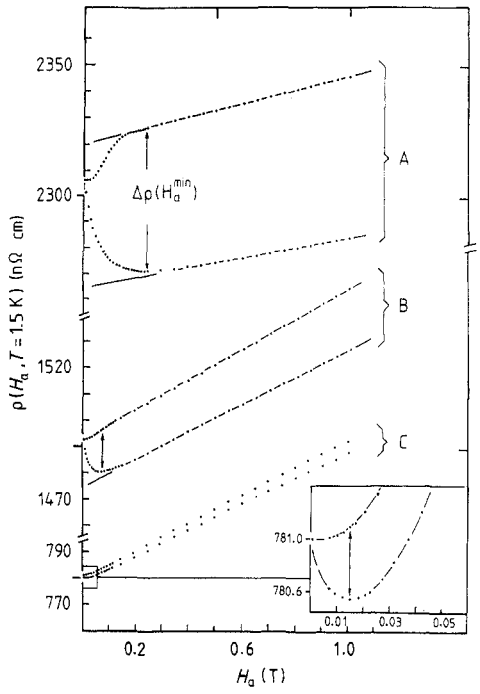

Figure 1. The resistivity measured in transverse (upper curves) and longitudinal (lower curves) applied fields at 1.5 K for three typical samples: curves A, 5 at.% Ni; curves B, 3.5 at.% Ni; curves C, 2.6 at.% Ni. The linear extrapolations used for conventional estimates of the SRA are shown, along with the anisotropy measured at the field, H_a^{min}, at which the longitudinal magnetoresistance is a minimum. Inset: the low-field behaviour of the 2.6 at.% sample on an expanded scale.

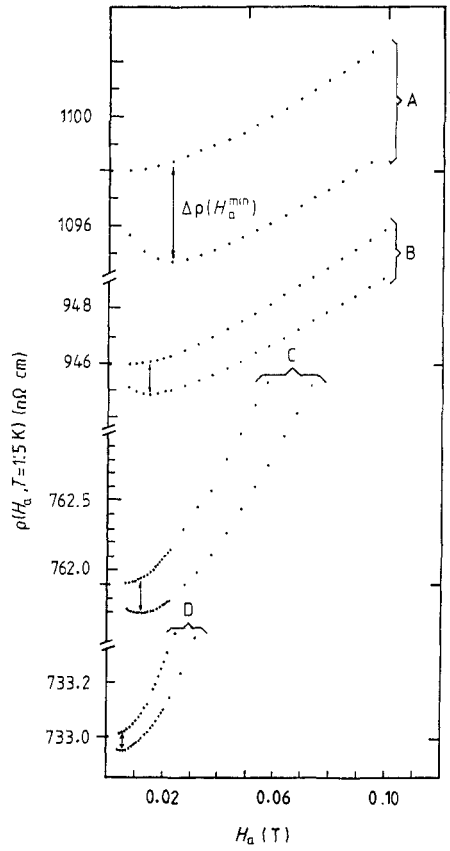

Figure 2. The low-field transverse (upper curves) and longitudinal (lower curves) magnetoresistivities at 1.5 K plotted against the applied field for samples close to the critical composition: curves A, 2.9 at.% Ni; curves B, 2.7 at.% Ni curves C, 2.5 at.% Ni; curves D 2.4 at.% Ni. The anisotropy at H_a^{min} is shown in each case.

an orbital moment at the Ni site can be correlated with the presence of a regular contribution to the susceptibility near T_c that is difficult to saturate (Hall *et al* 1984) (presumably due to anistropy resulting from single ion spin–orbit coupling). This would mean that the measured susceptibility is not dominated by the singular (critical) component, but is obscured by contributions from domain wall motion and/or coherent rotation that remains below technical saturation. Thus the presence of an orbital moment, which is an essential component in the appearance of an SRA, can inhibit the interpretation of magnetic data.

While these low temperature magnetoresistance data thus make the presence of an anisotropy with ($\rho_\perp > \rho_\parallel$) clear, the question still remains as to how the magnitude of the associated SRA can be estimated reliably. We suggest that the one consistent feature

Table 1. Summary of data on **Pd**Ni.

Concentration (at.% Ni)	SRA[a] (%)	SRA[b] (%)	$d\rho_\perp/dH_a$ (nΩ cm T^{-1})	$d\rho_\parallel/dH_a$ (nΩ cm T^{-1})	T_c[c] (K)
2.1		$<-1.4\times10^{-3}$	26(±1)	23.5(±1)	—
2.2		$<1.4\times10^{-3}$	34(±1)	31.5(±1)	—
2.3		$<1.4\times10^{-3}$	37(±1)	34(±1)	—
2.4		$-9.2(\pm2)\times10^{-3}$	42(±1)	39.5(±1)	2.25
2.5		$-3.4(\pm0.2)\times10^{-2}$	47(±1)	44(±1)	4.3
2.6		$-6.85(\pm0.4)\times10^{-2}$	52(±2)	49(±2)	5
2.7		$-1.30(\pm0.05)\times10^{-1}$	63(±2)	57.5(±2)	7
2.8		$-2.03(\pm0.05)\times10^{-1}$	67.5(±2)	63(±2)	10
2.9		$-3.2(\pm0.05)\times10^{-1}$	69(±2)	69(±2)	12.5
3.0	$-5.3\ (\pm1.3)\times10^{-1}$	$-3.55(\pm0.05)\times10^{-1}$	70(±1)	67(±1)	13.5
3.5	−1.14(±0.05)	−1.11(±0.02)	57.5(±0.5)	52(±0.5)	32
4.0	−1.72(±0.04)	−1.68(±0.03)	41.5(±0.5)	35.5(±0.5)	45
4.5	−2.12(±0.03)	−2.12(±0.01)	31.2(±0.5)	25.5(±0.5)	58
5.0	−2.43(±0.03)	−2.41(±0.01)	24.8(±0.5)	19.5(±0.5)	64

[a] From the extrapolation procedure (not able to give accurate prediction below 3.0 at.% Ni).
[b] From $\Delta\rho(H_a^{min})$.
[c] Estimated from AC susceptibility data; for the reasons outlined in the text, uncertainties range from ±0.25 K at 2.4 at.% to ±1 K at 5 at.%.

of these magnetoresistance data—the appearance of a minimum in $\rho_\parallel(H_a)$ at $H_a = H_a^{min}$—can be exploited to estimate the SRA, specifically by finding the ratio

$$\Delta\rho(H_a^{min})/\rho_0 = [\rho_\parallel(H_a^{min}) - \rho_\perp(H_a^{min})]/\rho_0. \tag{3}$$

It is obvious that the use of equation (3) provides a method for measuring the anisotropy in each sample at a unique field (H_a^{min}), thus avoiding the complications inherent in extrapolation procedures. Furthermore, as is evident from figures 1 and 2, this characteristic field H_a^{min}, at which the ratio is measured, exhibits the desirable property of approaching zero as $c \to c_0$. This means that the potential perturbing effects of an external field are minimised in this sensitive composition range. Indeed, estimates of the effects of the latter at the measuring field can be made by examining the quantity $H_a^{min} \cdot (\rho'_\perp - \rho'_\parallel)$ where $\rho'_\perp$ ($= d\rho_\perp/dH_a$) and $\rho'_\parallel$ ($= d\rho_\parallel/dH_a$) are taken from the high-field slopes. This quantity is comparable to the listed uncertainty in the SRA shown in table 1. Furthermore, the result that the low-field 'normal' magnetoresistance is weaker than its high-field counterpart means that this latter estimate represents an upper limit. All this is confirmed by our failure to observe an anisotropy at low fields in samples below c_0 (which nevertheless have comparable high field slopes). However, estimates of the SRA based on the use of equation (3) must agree with those obtained using established extrapolation techniques (at least, in situations where comparisons are possible). We have estimated the SRA using both methods in samples containing $\geqslant$3 at.% Ni where this comparison is possible. As table 1 shows, these two sets of estimates are in excellent agreement.

Having demonstrated the applicability of this low-field method in **Pd**Ni, it is now possible to use it in conjunction with data similar to those shown in figure 2 to find the SRA in samples with composition close to c_0 (where the extrapolation method is not reliable). The corresponding SRA estimates are also listed in table 1.

These latter estimates drop rapidly as the critical composition is approached, and this has prompted us to examine the possibility of a power-law relationship between the two, governed by some critical exponent Δ:

$$\Delta\rho/\rho_0 \propto (c/c_0 - 1)^{\Delta}. \tag{4}$$

We have attempted to fit equation (4) to the data in table 1 using c_0 values in the range (2.3 ± 0.05) at.%, as deduced from previous analyses (Tari and Coles 1971, Murani *et al* 1974) and confirmed by recent magnetoresistance anisotropy measurements (Kunkel *et al* 1987). Figure 3 shows the results of the first such attempt to fit this type of data: the power-law relationship does reproduce the SRA estimates at 1.5 K for concentrations close to c_0 ($(c/c_0 - 1) < 5 \times 10^{-1}$) with parameters in the range

$$c_0 = 2.25 \text{ at.\% Ni} \qquad \Delta = 2.25 \pm 0.1$$

to

$$c_0 = 2.30 \text{ at.\% Ni} \qquad \Delta = 2.0 \pm 0.1.$$

We argue for the lower estimate for c_0 (and the larger for Δ) in spite of the result that the 1.5 K data are equally well fitted by both. This argument is based on our measurements of the anisotropy as a function of temperature between 1.5 and 4.2 K. The SRA is, of course, temperature dependent, vanishing as the temperature increases towards T_c. Between 1.5 and 4.2 K we find little change in the SRA for samples with T_c above about 10 K. In the other samples there is a decrease in the SRA at 4.2 K that becomes progressively more marked for samples closer to c_0, with the anisotropy in the 2.4 at.% sample ($T_c \simeq 2.25$ K) actually vanishing at 4.2 K. The data reproduced in figure 3 on the 2.4 at.% sample was thus obtained at a temperature (1.5 K) not too far below its ordering temperature. Measurements at temperatures below 1.5 K would thus be expected to yield an increased SRA for it. Our estimates for this expected increase (based on comparisons with the temperature dependence exhibited by the SRA in other samples) is around 15%. Such data still would be compatible with the power-law relationship based on the lower c_0 estimate, but the larger c_0 value would not produce such a convincing plot.

Finally we indicate that other data we have acquired also support the estimate for c_0 given above. The identification of c_0 as a critical composition means that plots of quantities such as $(1/\rho_0)(\mathrm{d}\rho/\mathrm{d}H_a)(1/T_c)$ against composition should diverge at c_0. This is simply equivalent to stating that $T_c \to 0$ as $c \to c_0$. Measurements of T_c close to the critical composition in **Pd**Ni are difficult not only for the technical reasons outlined above, but also because T_c falls well below 1 K. The advantage of plotting the quantity indicated above is two-fold; first, it uses data acquired from the same sample, for which the SRA was measured, and second, it allows the data on paramagnetic samples to be included by simple and conventional replacement of T_c for the ferromagnetic specimens by T (the measuring temperature) in the paramagnetic alloys. Thus, not only can data for samples with composition above c_0 measured above their ordering temperature be included (e.g. $T_c < 1.5$ K, the lowest measuring temperature, would apply in the present experiment specifically to the 2.3 at.% Ni sample; furthermore, it is precisely in this composition range that an accurate evaluation of T_c becomes increasingly difficult for the reasons outlined previously), but samples with composition below c_0 can also be assessed (Loewen *et al* 1986). These data are reproduced in figure 4; they peak between 2.2 and 2.3 at.% Ni, thus providing indirect confirmation of our choice for c_0.

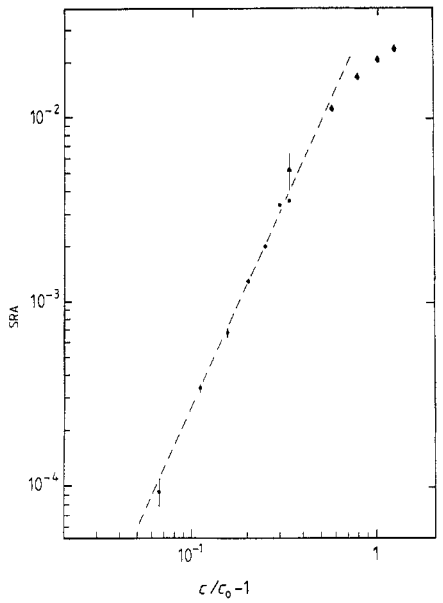

Figure 3. SRA against the reduced composition $(c/c_0 - 1)$ on a log–log plot with $c_0 = 2.25$ at.% Ni. The straight line drawn corresponds to a power-law dependence (equation (4)) with $\Delta = 2.25$.

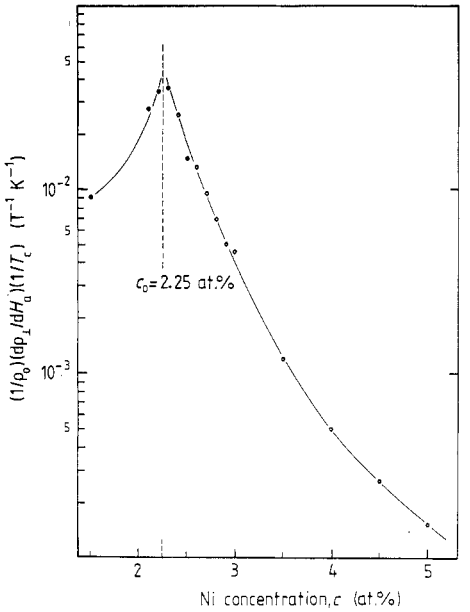

Figure 4. The scaled high-field slope against composition on a log–linear plot. The full curves are merely guides to the eye. The data for the 1.5 at.% Ni sample is estimated from Loewen *et al* (1986).

In summary, to our knowledge, no experimental measurements or theoretical prediction for critical behaviour in the SRA close to the critical composition exists. The numerical value we estimate for the exponent Δ is quite different from that predicted for, say, the Hall coefficient exponent ($g = -0.05 \pm 0.05$) near a conductivity percolation threshold (Straley 1988), and while it is close to the value predicted for the corresponding conductivity exponent ($t = 2.02 \pm 0.06$) (Gerauld *et al* 1984), such an association cannot be justified. Specifically, both of these latter exponents were calculated for systems in the vicinity of a percolation controlled conductivity threshold, whereas the present system is a good conductor over the entire composition range investigated (the conductivity is dominated by conduction-electron scattering from the screened Coulomb potential at the Ni sites, with weak magnetic scattering corrections), but is near a magnetic threshold. If this emerging ferromagnetism is treated within the framework of a percolation approach, then the localised model leads to the following prediction for the behaviour of the SRA. Using the latter model's expression summarised in equation (2), it might be argued that this ratio varies as m^2 or possibly m^3 (since we measured the 'saturated SRA' (that is, at temperatures well below T_c) a Wortis type expansion might be applicable); this yields a variation of the form $\langle S_z \rangle^{l(l+1)/2}$, i.e. m^3 for operators with quadrupolar symmetry ($l = 2$). In a percolation context, if this magnetisation m is taken simply to be proportional to the probability $P(c) = (c/c_0 - 1)^\beta$ of a given spin belonging to the infinite cluster, then with $\beta = 0.4$ (Stauffer (1979) independent of lattice type (Kertesz 1982)), one obtains $\Delta \simeq 0.8\, m^2$ or $1.2\, m^3$. This value is much smaller than that estimated from our measurements, so we suggest that an additional exponent(s) must

be involved. However, at the present time we are unable to suggest what it might be. We hope therefore that the present measurements stimulate further work in this area.

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References

Campbell I A, Fert A and Jaoul O 1970 *J. Phys. C: Solid State Phys.* **3** 595
Dorleijn J W F 1976 *Philips Res. Rep.* **31** 287
Friederich A and Fert A 1974 *Phys. Rev. Lett.* **33** 1214
Gerauld G R, Scriven L E and Davis H T 1984 *J. Phys. C: Solid State Phys.* **17** 3429
Hall N G, Roshko R M and Williams G 1984 *J. Phys. F: Met. Phys.* **14** 711
Hamzic A, Senoussi S, Campbell I A and Fert A 1978 *J. Phys. F: Met. Phys.* **8** 1947
Kertesz J 1982 *J. Physique* **42** L393
Kondo J 1962 *Prog. Theor. Phys. Kyoto* **27** 772
Kunkel H P, Wang Z and Williams G 1987 *J. Phys. F: Met. Phys.* **17** L157
Loewen C, Saran M and Williams G 1986 *J. Phys. F: Met. Phys.* **12** 43
Malozemoff A P 1986 *Phys. Rev.* B **34** 1853
Mott N F 1936 *Proc. R. Soc.* A **153** 699
Muir W B and Ström-Olsen J O 1976 *J. Phys. E: Sci. Instrum.* **9** 163
Murani A P, Tari A and Coles B R 1974 *J. Phys. F: Met. Phys.* **4** 1769
Senoussi S, Campbell I A and Fert A 1977 *Solid State Commun.* **21** 269
Stauffer D 1979 *Phys. Rep.* **54** 1
Straley J P 1988 *Phys. Rev.* B **38** 11639
Tari A and Coles B R 1971 *J. Phys. F: Met. Phys.* **1** L69
Thomson W 1857 *Proc. R. Soc.* **8** 546