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Percolation controlled behaviour of the spontaneous resistive anisotropy near the critical concentration in CuNi

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Abstract. Measurements of the spontaneous resistive anisotropy (SRA) at low temperature in $Cu_{100-x}Ni_x$ are reported. The latter indicate a power-law dependence of this parameter on the reduced concentration in the vicinity of the critical composition for the formation of a ferromagnetic ground state. These magnetoresistance data indicate a critical composition of 44.5 at.% Ni, and fitting the concentration dependence of the SRA to a power-law behaviour yields a critical exponent $\Delta \simeq \frac{9}{4}$, in agreement with the one earlier estimate for Ni in Pd.

While the behaviour of various properties near a thermal phase transition are relatively well established [1] (the variation of the magnetization and susceptibility in the immediate vicinity of a paramagnetic to ferromagnetic transition, in particular, have been measured extensively, and such data analysed to extract critical exponent estimates for comparison with model predictions), the corresponding response around a percolation controlled (zero-temperature) critical point is, by contrast, less well understood. Within this latter context, the characteristics of the spontaneous resistive anisotropy (SRA)—despite having been observed initially in the mid-nineteenth century [2]—are even less well established. The SRA is defined through the ratio

$$\frac{\Delta\rho}{\rho_0} = \left[\frac{\rho_\parallel(B) - \rho_\perp(B)}{\rho_0}\right]_{B\to 0} \tag{1}$$

and characterizes the difference between the longitudinal ($\rho_\parallel$) and transverse ($\rho_\perp$) magnetoresistance of a single-domain ferromagnet extrapolated to zero induction B [3]. Its existence relies on the presence of both spin–orbit coupling and a polarizing field at scattering sites [4].

In recent papers, the measured 'zero'-temperature SRA was shown to exhibit a percolation related, power-law dependence on the reduced composition, viz.

$$\frac{\Delta\rho}{\rho_0} \propto \left(\frac{c - c_0}{c_0}\right)^{\Delta} \tag{2}$$

where c_0 represents the critical composition for the appearance of ferromagnetism at $T = 0$. Furthermore the exponent Δ appearing in (2) was shown to display different values ($\Delta \simeq 2.25$ for Ni in Pd [5], but $\Delta \simeq 1$ for Fe in both Pd [6] and in $(Pd_{1-x}Fe_x)_{95}Mn_5$ [7]), thus raising intriguing questions concerning the universality class for Δ: specifically, whether this exponent value depended on the SRA originating from either an orbital

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component in the total moment *localized* at the scattering site (Ni in Pd) or from spin–orbit coupling in the (exchange split) d *bands* (Fe in Pd). Indeed, the paucity of available data renders the use of the term 'universality class' contentious; furthermore, no theoretical predictions for such behaviour exist currently.

Here we provide definitive evidence for the universality of the exponent Δ, at least for systems in which the SRA appears to be described by a localized model in which the spin–orbit coupling is linked to a local moment with an orbital contribution. The system studied was CuNi near the equiatomic concentration; in this system the critical composition c_0 is near 45 at.% Ni [8] enabling low values for the reduced composition $(c - c_0)/c_0$ to be achieved directly, an appropriate requirement for the reliable estimation of Δ with the assumed asymptotic validity of (2).

Sixteen samples with concentrations ranging from 45 to 55 at.% Ni were prepared from high-purity starting materials using conventional arc melting techniques. Buttons of the highest and lowest compositions were prepared initially, and intermediate concentrations were made subsequently by melting together appropriate quantities of these two end members. Homogeneity was ensured at each stage in this process by inverting and remelting each button repeatedly. Specimens of approximate dimensions $(4 \times 0.2 \times 0.02)$ cm^3 suitable for transport measurements were cut from cold-rolled sheets, after which they were etched, washed and then annealed for 48 h at 950 °C prior to quenching into iced water. The latter treatment minimizes the potential for Ni clustering [9]. Magnetoresistance measurements were performed at 4.2 and 1.5 K using a previously described ac technique [10] of high precision; $\rho_\parallel$ and $\rho_\perp$ were measured with current along the longest dimension in applied fields $\mu_0 H_a$ up to 3 T applied parallel to the two largest sample dimensions sequentially.

A number of significant problems are encountered in attempting to measure the SRA near the critical composition c_0, and these are illustrated in figure 1. At 55 at.% Ni ($T_c \sim 85$ K) conventional extrapolation procedures [3] enable the SRA to be obtained directly and accurately, and is consistent in sign and magnitude with previous available data [11]. However, as the Ni concentration is reduced towards c_0 two features emerge: (i) the magnitude of the SRA decreases rapidly, and (ii) the high-field slopes on which this extrapolation procedure relies not only remain large but also some nonlinear variation becomes evident, complicating the choice of field region from which this extrapolation is made; this makes the extrapolations themselves subject to increasing uncertainty. These latter combine to render this procedure inappropriate for determining the sign—let alone the magnitude—of the SRA below 46 at.% Ni. This problem has been resolved in a manner similar to that reported previously for **Pd**Ni [5], viz. by utilizing a characteristic feature present in the *low*-field data: the latter, presented in figure 2, are unambiguous, in contrast to the high-field data discussed above. In particular, the magnetoresistive anisotropy, measured at that field H_m in which the longitudinal magnetoresistance exhibits a maximum, indicates clearly not only the sign but also (as confirmed below) the magnitude of the SRA. The analogous feature utilized in the **Pd**Ni system was the presence of a minimum in $\rho_\parallel$. Furthermore, as discussed previously for this latter system [5], the appearance of a finite difference $(\rho_\parallel - \rho_\perp)$ in *low applied* field ($\leqslant$0.5 mT close to c_0) is unequivocal evidence for the existence of a significant *exchange* field at the measuring temperature. These data contrast markedly with those from other measurements near c_0 in such systems, viz. strongly curved Arrott plots making the identification of T_c difficult [8], and related uncertainties in field-dependent ac susceptibility measurements (which will be the subject of a subsequent publication; the estimates of the ordering temperature, T_c, from the latter are, however, summarized in table 1). This point is emphasized in figure 3 in which the low-field data on the 45.5 and 45.0 at.% Ni samples are shown at both 4.2 and 1.5 K; these data indicate the

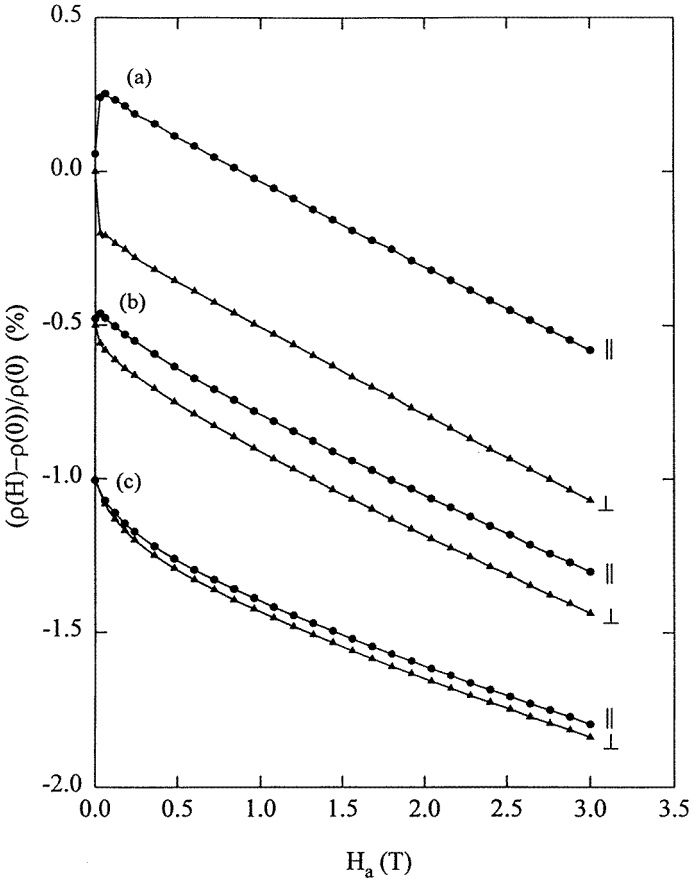

Figure 1. The magnetoresistance at 1.5 K as a function of applied field in $Cu_{100-x}Ni_x$, with $x = 55$ (a), $x = 48$ (b) and $x = 45$ (c) with curves (b) and (c) offset by -0.5 and -1% respectively.

presence of an anisotropy in the magnetoresistance at H_m in both samples at 1.5 K, viz.

$$\frac{\Delta\rho(H_m)}{\rho_0} = \left[\frac{\rho_\parallel(H_m) - \rho_\perp(H_m)}{\rho_0}\right] > 0. \tag{3}$$

From this we infer the presence of an exchange field at that temperature (H_m being <1 mT there) indicating *both* samples are ordered at that temperature. By contrast, the 4.2 K data exhibit a non-zero $\Delta\rho(H_m)$ in the 45.5 at.% specimen alone, indicating that the ordering temperature T_c for this sample is above 4.2 K whereas that for the 45.0 at.% specimen is not. Our ac susceptibility measurements confirm this, but are—separately—less conclusive.

Having established the presence of a low-field anisotropy in samples containing 45 at.% or more Ni at 1.5 K, it is then necessary to confirm that the anisotropy ratio estimated through the use of (3)—that is by measuring $\Delta\rho(H_m)$, the anisotropy at that field H_m at which $\rho_\parallel$ exhibits a maximum—provides a reliable value for the SRA. As was done previously [5], this is accomplished by comparing the results obtained from (1) and (3) for samples containing more than 49 at.% Ni, for which the conventional extrapolation procedure appears to be

Table 1. Estimates of the SRA at 1.5 K in $Cu_{100-x}Ni_x$.

at.% Ni	SRA[a] (%)	SRA[b] (%)	SRA[c] (%)	T_c (K)[d]
45.0	—	—	0.0011(4)	1.7(3)
45.5	—	—	0.0034(4)	4.2(1)
46.0	—	—	0.015(2)	5.6(1)
46.5	—	—	0.033(2)	8.1(2)
47.0	—	0.06(4)	0.030(3)	9.8(2)
47.5	—	0.10(5)	0.056(2)	13.1(3)
48.0	—	0.11(6)	0.084(2)	20.1(4)
48.5	—	0.14(2)	0.11(1)	22.2(6)
49.0	0.14(1)	0.14(1)	0.13(1)	25.1(6)
49.5	0.22(2)	0.22(2)	0.16(1)	29.4(6)
50.0	0.17(1)	0.17(1)	0.16(1)	33.3(7)
51.0	0.23(1)	0.22(2)	0.20(1)	39.1(8)
52.0	0.31(2)	0.30(2)	0.23(1)	55.4(11)
53.0	0.36(2)	0.36(2)	0.35(1)	62.8(13)
54.0	0.45(2)	0.46(2)	0.43(1)	78.4(20)
55.0	0.46(2)	0.48(2)	0.46(1)	83.2(20)

[a] From the extrapolation procedure. Note that entries in this table of the type 0.14(1) represent the shorthand form for 0.14 ± 0.01.

[b] From extrapolation of the difference, $\Delta\rho(H) = \rho_{\parallel}(H) - \rho_{\perp}(H)$, to $H = 0$; this extrapolation is based on the essentially linear variation of $\Delta\rho(H)$ with H at high field (see, for example, [12]).

[c] From the low-field feature, $\Delta\rho(H_m)/\rho_0$.

[d] The ordering temperature, T_c, estimated from ac susceptibility data.

applicable. These results are summarized in table 1. An inspection of this table confirms that these two estimates for the SRA agree well at higher concentration (>49 at.% Ni), although close examination of these tabulated values indicates that ratios determined via the use of (3) are generally slightly lower than those based on the extrapolation procedure from the high-field regime. This is not unexpected: high applied fields not only modify the conduction electron trajectories but they also enhance the intrinsic spin polarization of the system, and the latter—despite the intent of the extrapolation process—leads to enhanced SRA estimates. Those found from (3) are obtained, by contrast, in much lower fields. Thus we conclude that the ratio $\Delta\rho(H_m)/\rho_0$ does provide a reliable estimate for the SRA, not only above 49 at.% Ni where comparison with conventional techniques are possible, but also below this concentration where the usual extrapolation procedure becomes unreliable for reasons discussed above. Indeed, since the field H_m at which the anisotropy is measured can be seen to fall to zero as the Ni concentration is lowered towards the critical composition c_0, the perturbing effects of an *externally* applied field are minimized near c_0, thus making SRA estimates based on the use of (3) *more* reliable in the critical regime (as concluded for **Pd**Ni [5]).

The SRA values summarized in table 1 exhibit a strong concentration dependence, decreasing by more than two orders of magnitude as the Ni composition falls from 55 to 45 at.%. This result suggests that the power-law behaviour summarized in (2) may provide an appropriate representation of these data near c_0. However, fitting these data to such a form requires a prior knowledge of the critical composition c_0. The metallurgical sensitivity of the CuNi system is well documented, so that it is difficult to define a universal value for c_0: existing estimates range from 43 to 50 at.% Ni depending on the particular preparation technique [9]. This constraint does not apply to samples annealed and quenched

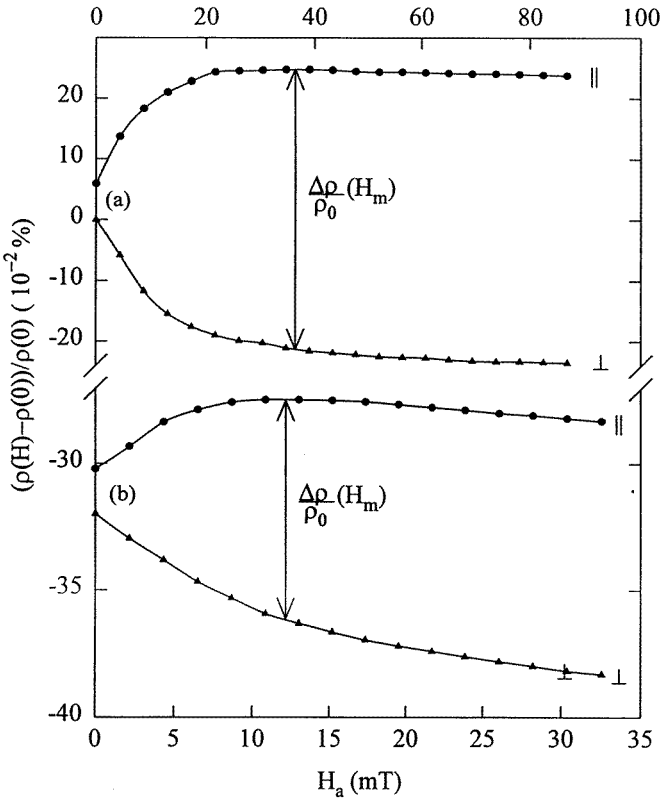

Figure 2. The low-field magnetoresistance at 1.5 K as a function of applied field in $Cu_{100-x}Ni_x$, with $x = 55$ (a) and $x = 48$ (b). Curve (b) is offset by -0.32% for clarity.

in a consistent way, as was done here. Specifically, the magnetoresistance data presented in figure 3 indicate a critical composition of marginally less than 45 at.% Ni for the present alloy series: the latter is supported by the behaviour of the ac susceptibility of similarly prepared specimens. Fits to (2) have thus been made by plotting the tabulated SRA values on a double-logarithmic scale against the reduced composition $(c - c_0)/c_0$ for a number of c_0 values close to 45 at.%; the Δ value is found from the slope of the line drawn (the power law). As is usual with exponent estimates, the range of data to be fitted is difficult to determine unequivocally, with associated uncertainty in the *asymptotic* exponent value. Figure 4 reproduces the results of such a plot using $c_0 = 44.5$ at.% Ni: the solid line drawn yields

$$\Delta = 2.2(1) \qquad \text{for } (c - c_0)/c_0 < 10^{-1} \tag{4}$$

although

$$\Delta = 2.0(1) \qquad \text{for}(c - c_0)/c_0 < 2 \times 10^{-1}$$

is also acceptable. These Δ values are in excellent agreement with the range found for this exponent in **Pd**Ni [5]; as in this latter system we argue that the larger Δ value provides—in our opinion—a better representation of the present data in the critical region, and this dependence is shown by the solid line in figure 4. Data beyond $(c - c_0)/c_0 \simeq 10^{-1}$ would thus fall outside the true, asymptotic critical regime.

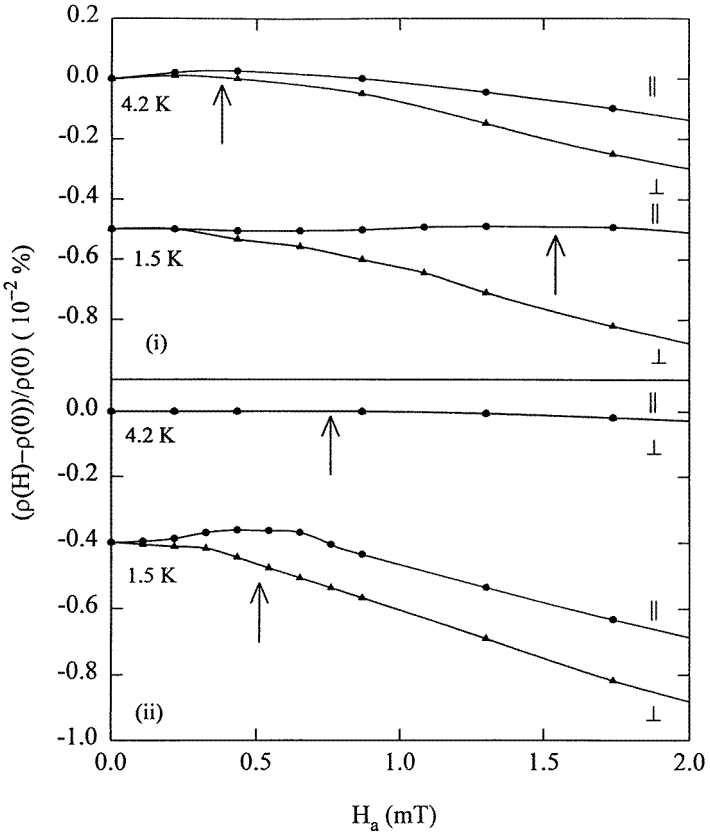

Figure 3. (i) The low-field magnetoresistance at 1.5 and 4.2 K as a function of applied field in $Cu_{54.5}Ni_{45.5}$; (ii) the low-field magnetoresistance at 1.5 and 4.2 K as a function of applied field in $Cu_{55}Ni_{45}$. The curves at 1.5 K are offset slightly for clarity. The vertical arrows indicate the estimated location of the low-field feature in $\rho(H)$.

In summary, the zero-temperature spontaneous resistive anisotropy in CuNi has been shown to exhibit a percolation controlled, power law dependence on reduced composition near the critical concentration $c_0 \simeq 44.5$ at.% Ni. The exponent governing this behaviour, $\Delta = 2.2(1)$, found in this system is in excellent agreement with the value of 2.25 ± 0.1 found previously in **Pd**Ni [5]. This agreement supports the assertion that the exponent describing such behaviour in systems with an orbital component in the total moment localized at Ni sites is indeed universal.

Finally we reiterate the lack of agreement between the measured and 'predicted' exponent values noted previously [5]. In a localized model approach, the SRA originates primarily from conduction electron–local electric quadrupole scattering [4]. In such a model (the predictions of which admit direct comparisons; the itinerant model does not [13]), the zero/low-temperature *spontaneous* resistive anisotropy (i.e. measured well below the ferromagnetic ordering temperature T_c) is expected to behave as [14, 15]

$$\frac{\Delta\rho}{\rho_0} \propto m^{\ell(\ell+1)/2} \tag{5}$$

with $\ell = 2$ for operators with quadrupolar symmetry. Further, at a percolation controlled

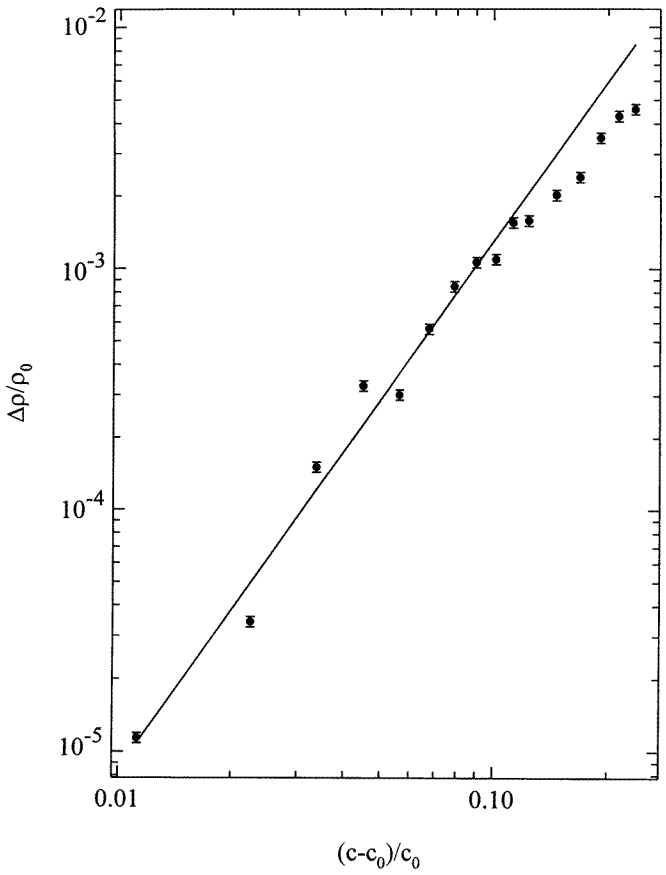

Figure 4. The SRA as a function of reduced composition with $c_0 = 44.5$. The superimposed line is a fit to equation (2) as described in the text, with $\Delta = 2.2(1)$.

transition, the spontaneous magnetization m is given by [16]

$$m \propto \left(\frac{c - c_0}{c_0}\right)^{\beta} \tag{6}$$

with β in the range (for a lattice with dimension $d = 3$) between 0.34 and 0.47 [16]; combining these yields

$$\frac{\Delta\rho}{\rho_0} \propto \left(\frac{c - c_0}{c_0}\right)^{\beta\ell(\ell+1)/2}. \tag{7}$$

Thus the 'exponent' Δ should be given by

$$\Delta = \beta\ell(\ell + 1)/2 \tag{8}$$

assuming a predicted value of between 1.02 and 1.41, roughly a factor of two smaller than the measured value of $\frac{9}{4}$. Both the suggested universality of this exponent and the discrepancy between its measured and predicted values warrant further investigation.

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