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## The spontaneous resistive anisotropy in PdFe above the ferromagnetic percolation threshold

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**Abstract.** A detailed study of the anisotropy in the magnetoresistance of PdFe alloys containing between 0.35 and 2.2 at.% Fe at 1.5 and 4.2 K is presented. Using these data, the composition dependence of the spontaneous resistive anisotropy at Fe concentrations above that necessary to establish a ferromagnetic ground state is established and compared with that reported recently in PdNi. The 'exponent' governing this concentration dependence appears to be different for systems in which the impurities either carry or lack an orbital moment.

### 1. Introduction

Pd is an incipient ferromagnet in which the effects of exchange enhancement suppress the usually rapid oscillations in the RKKY spin polarization to quite large distances from the polarizing site (i.e. a moment-bearing impurity). This leads to the well known giant moment phenomenon [1] in alloys such as PdFe and PdCo, as well as to the establishment of a ferromagnetic ground state above a critical concentration  $c_0 \simeq 0.01$  at.% Fe [1, 2] owing to a percolating backbone of ferromagnetically coupled giant moments. By contrast, such effects are absent in PdNi [3]; Ni impurities do not exhibit giant moments, and the critical concentration is correspondingly larger ( $c_0 \simeq 2.25$  at.% Ni [4, 5]).

Measurements of the spontaneous resistive anisotropy (SRA)—essentially the difference in the resistance of a single-domain ferromagnetic conductor (extrapolated to zero induction  $B$ )—have been instrumental in revealing subtle differences in the characteristics of impurity magnetic moments in a variety of hosts [6–8]. In particular, the concentration dependence of SRA data has been interpreted as indicating the presence (Co and Ni) or absence (Fe) of an orbital component in the moment at essentially isolated impurity sites in Pd [6]. Recently, a detailed study of the SRA in PdNi was reported [5, 9], and this anisotropy was shown to display critical behaviour as a function of the reduced concentration i.e.  $SRA \propto (c/c_0 - 1)^\Delta$  with  $\Delta \simeq 9/4$ . Here we report the results of a similar study in PdFe which concludes that the 'exponent'  $\Delta$  is quite different in this system. While no theoretical study carried out to date indicates whether or not the SRA is indeed a critical quantity (either as a function of concentration  $c - c_0$  near the critical concentration at  $T = 0$  or as a function of temperature near the Curie temperature  $T_C$ ), these data nevertheless

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raise the intriguing possibility that the SRA displayed by systems with or without orbital moments *may* belong to different universality classes. Such a result would be in marked contrast with that for the magnetic properties.

## 2. Experimental details

Magnetoresistance measurements were carried out at 1.5 and 4.2 K on six PdFe samples containing between 0.35 and 2.2 at.% Fe; these same samples had been the subject of a recently reported AC susceptibility study, so that details of the preparation technique, homogenization treatment, etc, have been given previously [2]. These specimens, of approximate dimensions 35 mm  $\times$  2 mm  $\times$  0.1 mm, were mounted in series on a high-thermal-conductivity copper block which was immersed in a liquid-helium Dewar positioned between the pole faces of an electromagnet. Rotating this magnet around a vertical axis enabled data to be collected with the applied field parallel or perpendicular to the direction of the sample current, directed along the largest dimension. The differential resistivity ratio  $[\rho(H_a, T) - \rho(0, T)]/\rho(0, T)$  of each specimen was measured by a low-frequency (37 Hz) AC method [9, 10] by using exciting currents of typically 30–50 mA, enabling this ratio to be measured to a few parts in  $10^6$ . The maximum applied field  $\mu_0 H_a$  available with the pole-piece separation necessary to accommodate the present experiment was 1 T. Absolute resistivities were found from  $\rho(0, T)$ , which was also measured using a four-probe DC potentiometric technique with an associated error of  $\pm 0.5\%$  due principally to shape-factor uncertainties.

## 3. Results and discussion

### 3.1. General features and considerations

The recently reported susceptibility study on this system [2] revealed that the 1.4 at.% Fe specimen displayed nearly 'ideal' critical behaviour, i.e. asymptotic critical exponents  $\gamma = 1.37 \pm 0.01$ ,  $\beta = 0.39(5) \pm 0.01(5)$ , and  $\delta = 4.5 \pm 0.15$  which were very close to values predicted for the isotropic three-dimensional Heisenberg model using renormalization group techniques ( $\gamma = 1.386$ ,  $\beta = 0.365$  and  $\delta = 4.80$  [11]), and an exchange coupling distribution with little width. The features of the magnetoresistance in this sample are also the most straightforward to analyse and discuss; consequently they are presented first.

Figure 1 reproduces the longitudinal ( $\parallel$ ) and transverse ( $\perp$ ) magnetoresistivities of the 1.4 at.% Fe sample at 1.5 and 4.2 K. Despite the fact that dilute PdFe is often cited as an example of a soft ferromagnet (the coercive field  $\mu_0 H_c$  at 4.2 K in well annealed specimens has been estimated [2, 12] as 17  $\mu$ T (0.35 at.% Fe), 1  $\mu$ T (0.4 at.% Fe) and 160  $\mu$ T (1.4 at.% Fe)), magnetization measurements on samples near either end of the composition range studied here indicate that true saturation is achieved only in applied fields  $\mu_0 H_a$  well in excess of 1 T [13–15]. The data shown in figure 1 are consistent with these findings; at both 1.5 and 4.2 K,  $\rho_{\parallel}(H_a)$  increases rapidly at low fields, but, above 0.1 T,  $d\rho_{\parallel}/dB_a$  is negative, a result that is consistent with a (small) non-collinear spin component. The behaviour of  $\rho_{\perp}(H_a)$  at 4.2 K is qualitatively similar; however, at 1.5 K,  $\rho_{\perp}$  exhibits a weak minimum

near  $\mu_0 H_a \simeq 0.8$  T, above which  $d\rho_{\perp}/dB_a \simeq 4 \times 10^{-3} \mu\Omega \text{ cm T}^{-1}$ . This latter value is comparable with the magnitude of Kohler terms ('normal' magnetoresistance) reported in various Ni-based alloys [16], while the observation that  $d\rho_{\parallel}/dB_a$  remains negative at 1.5 K is qualitatively consistent with reports that the Kohler term is weaker in this orientation [16]. Nevertheless the data in figure 1 indicate clearly that the SRA defined as

$$\Delta\rho/\rho_0 = [(\rho_{\parallel} - \rho_{\perp})/\rho_0]_{B \rightarrow 0} \quad (1)$$

is positive, in agreement with previous estimates in this system [6]. Difficulties arise, however, when attempts are made to obtain quantitative estimates for this ratio.

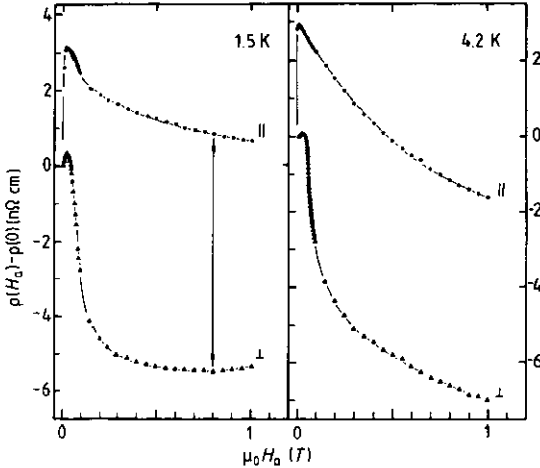


Figure 1. The field-dependent resistivities  $\rho(H_a) - \rho(0)$  of the 1.4 at.% Fe sample in the longitudinal (curve ||) and transverse (curve  $\perp$ ) configurations at 1.5 and 4.2 K. The vertical arrow indicates  $\Delta\rho(H_a^{\min})$ .

Traditionally the SRA has been found by extrapolating the variation in  $\rho_{\parallel}$  and  $\rho_{\perp}$  with  $B$  above technical saturation to  $B = 0$ , thus obtaining  $\Delta\rho$  [16,17]; frequently fields up to  $B \simeq 5$  T have been applied. While the arguments used to justify an extrapolation based on  $B$  (rather than on  $\mu_0 H_a$ ) remain valid [16], the use of high fields in experiments of the present type are not. Specifically, if  $c_0$  does indeed represent the critical concentration for a low-temperature percolation-controlled transition, the application of large fields at concentrations below  $c_0$  can induce a resistive anisotropy, as is observed in paramagnetic systems [18]. The required balance that needs to be achieved therefore is to use fields high enough to reveal any 'incipient' anisotropy but sufficiently small to leave the ground-state spin configuration unaltered. Typically in ferromagnets this might involve the use of fields which simply render the sample effectively single domain over distances of the order of a conduction electron mean free path [5,9]. The situation is analogous to measuring, for example, the non-linear magnetization of spin glasses.

In the previously reported study of the PdNi system [5] these criteria were met by measuring

$$\Delta\rho(H_a^{\min}) = \rho_{\parallel}(H_a^{\min}) - \rho_{\perp}(H_a^{\min}) \quad (2)$$

where  $H_a^{\min}$  is the applied field at which the transverse magnetoresistance  $\rho_{\perp}$  passes through a minimum (actually, in PdNi,  $\rho_{\parallel}$  displayed a minimum as the sign of the SRA is reversed there). Furthermore in that study it was also shown that

(i) the two estimates for the SRA—based on the use of equations (1) and (2)—agreed in all cases where a comparison was possible (an agreement also found in ferromagnetic Fe–Zr amorphous samples [19]) and

(ii) the field  $H_a^{\min}$  at which these measurements were carried out exhibited the desirable characteristic that  $H_a^{\min} \rightarrow 0$  as  $c \rightarrow c_0$ , so that near the critical concentration the potentially perturbing effects of applied external fields on the evolving spin configuration and the conduction electron trajectories were essentially minimized. [5, 9].

Figures 1 and 2 enable a comparison of these two techniques to be carried out for the 1.4 at.% Fe sample. Figure 2 reproduces the data acquired at 1.5 K; here, however, these are plotted as a function of both  $(\mu_0 H_a)^2$  and  $B^2$ . The field is found from

$$B = \mu_0(H_a + M) - DM \quad (3)$$

in which the magnetization  $M$  is taken to be the zero-temperature saturation magnetization estimated at  $6.85 \text{ J T}^{-1} \text{ kg}^{-1}$  for a 1.4 at.% Fe sample [20]. The appropriate demagnetizing factors  $D_{\parallel}$  and  $D_{\perp}$  were found by approximating the sample by an ellipsoid with principal axes equal to the dimensions of the specimen, and evaluating the corresponding elliptic integral [2, 21]. There is, consequently, minimal difference between  $\rho_{\perp}(B)$  and  $\rho_{\perp}(H_a)$  since  $D_{\perp} \approx \mu_0$ , and a slight difference between  $\rho_{\parallel}(B)$  and  $\rho_{\parallel}(H_a)$  since  $D_{\parallel}$  is small ( $D_{\parallel} \approx 5 \times 10^{-4} D_{\perp}$ ), so that  $M(\mu_0 - D_{\parallel}) \approx 0.1 \text{ T}$ . In ferromagnets, plots of  $\rho$  versus  $B^2$  have been widely utilized since this representation often linearizes the higher-field data [16], as indeed it does here ( $d\rho_{\perp}/d(B^2) \approx 2 \times 10^{-3} \mu\Omega \text{ cm T}^{-2}$ ), thus enabling reliable extrapolation to  $B = 0$  to be made. This does not hold for the  $\rho$  versus  $H_a$  plots in figure 1 as the curvature evident—particularly in  $\rho_{\perp}$  at 1.5 K—renders a simple linear extrapolation to  $H_a = 0$  inappropriate (a similar situation results if these data are replotted against  $B$  [22]).

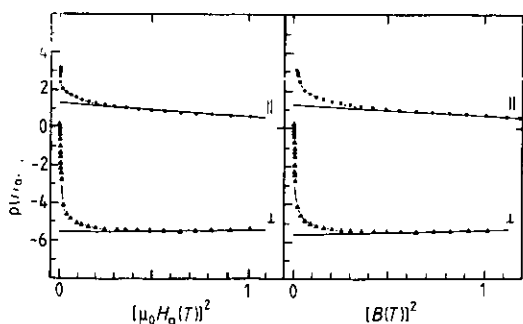


Figure 2. The field-dependent resistivities  $\rho(H_a) - \rho(0)$  of the 1.4 at.% Fe specimen at 1.5 K in the longitudinal and transverse configurations plotted against  $(\mu_0 H_a)^2$  and  $B^2$ .

The data shown in figure 2 yield estimates for the SRA at 1.5 K of  $(2.9(6) \pm 0.08) \times 10^{-3}$  using the extrapolation based on  $B^2$ , and of  $(2.9(3) \pm 0.08) \times 10^{-3}$  using the extrapolation based on  $H_a^2$ , while from figure 1, with  $H_a^{\min} = 0.08 \text{ T}$ , the SRA from equation (2) is  $(2.7(5) \pm 0.05) \times 10^{-3}$ . The listed errors arise from uncertainties in the higher-field slopes in figure 2 while, in figure 1, field points adjacent to  $H_a^{\min}$  are used to give a measure of the uncertainty in  $\Delta\rho(H_a^{\min})$ . The conclusions drawn from these results are firstly that the estimates for the SRA based on equations (1) and (2)

are in good agreement, and secondly that an extrapolation based upon  $H_a^2$  is quite adequate (basically because the magnetization  $M$  is small in these dilute specimens) and will consequently be used for the remaining samples†.

### 3.2. Samples containing 1.6–2.2 at.% Fe

The data acquired for the four specimens containing 1.6–2.2 at.% Fe are qualitatively similar to those for the 1.4 at.% Fe sample, with only slight differences, as noted below. Estimates for the SRA based on extrapolation from higher-field data for these samples are summarized in table 1. In both the 1.6 and the 1.8 at.% Fe samples,  $\rho_{\perp}$  displays a minimum—as in the 1.4 at.% Fe sample—so that estimates of the SRA based on  $\Delta\rho(H_a^{\min})$  for them are also included in table 1. The samples containing 2.0 and 2.2 at.% Fe do not exhibit a minimum in  $\rho_{\perp}$ ; this indicates a somewhat higher degree of non-collinearity in them (certainly no large increase in the Kohler term is expected between 1.8 and 2.0 at.% Fe—based on the tabulated residual resistivity  $\rho_0$ ). Unfortunately the lack of availability of detailed magnetization data in this concentration range precludes direct confirmation of this suggestion (although the estimated coercive fields are considerably larger at 2.0 at.% Fe than at 1.4 at.% Fe [2]).

### 3.3. The 0.35 at.% Fe data

Figure 3 presents magnetoresistance data at 1.5 K on the most dilute sample measured in the present study, the 0.35 at.% Fe specimen. This sample orders at 7.5 K and, despite the presence of considerable exchange bond disorder which precludes the assignment of reliable asymptotic exponent values [2], the transition is to a ferromagnetic state. Nevertheless the principle feature evident is a strong negative component in both  $\rho_{\parallel}$  and  $\rho_{\perp}$ , demonstrating the occurrence of considerable non-collinearity well below  $T_C$  at this dilution. At 4.2 K ( $T/T_C \simeq 0.56$ ), this negative contribution dominates to such an extent that no meaningful estimate for the SRA is possible;  $\rho_{\perp} - \rho_{\parallel} \simeq 0.5$  n $\Omega$  cm at 1 T while  $\rho_{\parallel}(0) - \rho_{\parallel}(1 \text{ T}) \simeq \rho_{\perp}(0) - \rho_{\perp}(1 \text{ T}) \simeq 18$  n $\Omega$  cm. Figure 3 shows that, while this negative contribution is still substantial at 1.5 K ( $T/T_C \simeq 0.2$ ;  $\rho_{\parallel}(0) - \rho_{\parallel}(1 \text{ T}) \simeq \rho_{\perp}(0) - \rho_{\perp}(1 \text{ T}) \simeq 4$  n $\Omega$  cm), estimates for the SRA using both methods discussed above are possible. At this lower temperature both  $\rho_{\parallel}$  and  $\rho_{\perp}$  display minima, the former at  $\mu_0 H_a \simeq 0.75$ –0.8 T and the latter at a slightly lower field of  $\mu_0 H_a \simeq 0.6$  T, and from the minimum in  $\rho_{\perp}$  the SRA from equation (2) is estimated at  $(7.5 \pm 0.6) \times 10^{-4}$ . This is appreciably less (although not quite outside the limits of experimental uncertainty) than the value found from extrapolation,  $(8.9 \pm 0.8) \times 10^{-4}$ . These SRA values are considerably smaller than those found at higher concentrations but are comparable in magnitude with those measured in PdNi very close to the critical concentration  $c_0$  [5].

Above the minima noted above, we have  $d\rho_{\perp}/d(B^2) \simeq 8 \times 10^{-4} \mu\Omega \text{ cm T}^{-2}$  while  $d\rho_{\parallel}/d(B^2) \simeq 2 \times 10^{-4} \mu\Omega \text{ cm T}^{-2}$ ; these are considerably lower than the corresponding estimate for the 1.4 at.% Fe sample scaled for changes in  $\rho_0$  [16] and thus provide indirect evidence for a continuing negative component in the magnetoresistance at 1 T in this sample.

† This is a useful result since no extensive magnetization field data are available for these other specimens, thus preventing an estimate for  $B$  from being made.

Table 1. Parameters deduced from the magnetoresistance data. The residual resistivity  $\rho_0$  is taken as  $\rho(H_a = 0, T = 1.5 \text{ K})$  and is subject to an uncertainty of  $\pm 0.5\%$  in absolute value, principally because of shape-factor uncertainties.

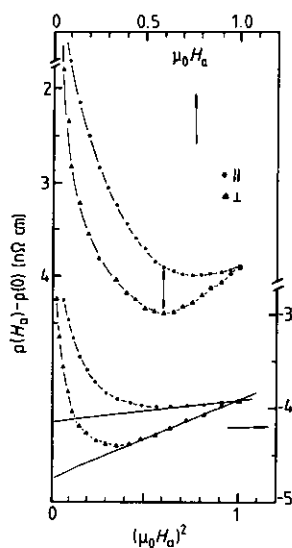
Fe in sample (at. % Fe)	$\rho$ ( $\mu\Omega \text{ cm}$ )	1.5 K			4.2 K		
		$H_a^{\text{min}} (T)$	SRA <sup>1</sup>	SRA <sup>2</sup>	$H_a^{\text{min}} (T)$	SRA <sup>1</sup>	SRA <sup>2</sup>
0.35	0.67(2)	0.6	(7.5 $\pm$ 0.6) $\times 10^{-4}$	(8.9 $\pm$ 0.8) $\times 10^{-4}$	—	—	—
1.4	2.31(7)	0.8	(2.7(5) $\pm$ 0.05) $\times 10^{-3}$	(2.9(3) $\pm$ 0.08) $10^{-3}$	—	—	—
1.6	2.70(1)	0.5	(3.8(7) $\pm$ 0.05) $\times 10^{-3}$	(3.9(3) $\pm$ 0.08) $\times 10^{-3}$	—	—	(2.7(1) $\pm$ 0.08) $\times 10^{-3}$
1.8	2.99(2)	0.45	(4.0(8) $\pm$ 0.05) $\times 10^{-3}$	(4.0(8) $\pm$ 0.08) $\times 10^{-3}$	0.75	(3.6(5) $\pm$ 0.5) $\times 10^{-3}$	(3.8(3) $\pm$ 0.08) $\times 10^{-3}$
2.0	3.36(6)	—	—	4.6(6) $\pm$ 0.08) $\times 10^3$	0.75	(3.7(0) $\pm$ 0.05) $\times 10^{-3}$	(3.8(2) $\pm$ 0.08) $\times 10^{-3}$
2.2	3.55(4)	—	—	(5.0(1) $\pm$ 0.08) $\times 10^{-3}$	—	—	(4.6(5) $\pm$ 0.08) $\times 10^{-3}$

<sup>1</sup> Estimated using equation (2).

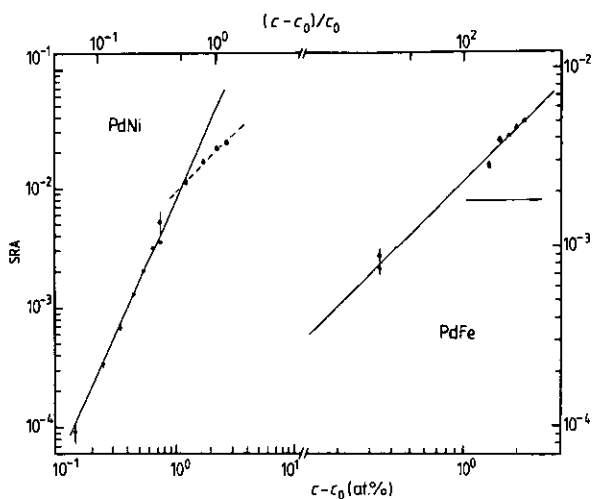
<sup>2</sup> Estimated from the extrapolation procedure (i.e.  $\rho$  versus  $H_a^2$ ).

## 3.4. Concentration dependence of the SRA

The SRA data summarized in table 1 vary by close to an order of magnitude over the concentration range examined, falling markedly with decreasing Fe concentration. Following the recent demonstration that the SRA in PdNi followed a power-law dependence on reduced concentration [5], a similar relationship has been explored for the present PdFe data. Figure 4 reproduces a plot of the SRA from table 1 against  $c - c_0$  on a double-logarithmic scale (with the previously acquired PdNi data included for comparison). Unfortunately, unlike the situation in PdNi, the critical concentration  $c_0$  for the occurrence of a ferromagnetic ground state (an infinite percolating backbone of ferromagnetically coupled giant moments) is not as well established for PdFe. However, in a recent report [2] of a detailed AC susceptibility study of the present samples, a comprehensive compilation of the concentration dependence of the ordering temperatures of PdFe samples containing between  $10^{-4}$  and 25 at.% Fe was presented, and the suggestion subsequently made that  $c_0$  may be as low as  $10^{-2}$  at.% Fe. This lower limit has been used in constructing figure 4. It should be emphasized, however, that the results reported below are *not* crucially dependent on the value for  $c_0$ , and the plot shown in figure 4 is not substantially altered if  $c_0$  is increased by an order of magnitude to  $10^{-1}$  at.% Fe.



**Figure 3.** The field-induced change in resistivity  $\rho(H_a) - \rho(0)$  for the 0.35 at.% Fe sample at 1.5 K in the longitudinal (curve ||) and transverse (curve  $\perp$ ) configurations. The upper data are plotted against  $\mu_0 H_a$  (in teslas), with  $\Delta\rho(H_a^{\min})$  being shown by the vertical arrow. The lower data are plotted against  $(\mu_0 H_a)^2$  (in teslas squared).



**Figure 4.** The SRA at 1.5 K of PdNi samples and PdFe samples plotted on a double-logarithmic scale against  $c - c_0$  on the lower axis, and against  $(c - c_0)/c_0$  on the upper axis. The full line corresponds to  $\Delta = 9/4$  and the broken line to  $\Delta = 1$  for PdNi, while for PdFe the full line yields  $\Delta = 1$ . The data points were acquired from  $\Delta\rho(H_a^{\min})$  ( $\blacktriangle$ ) and from extrapolation ( $\bullet$ ).

The straight line drawn in this last figure confirms that a power-law relationship of the form

$$\Delta\rho/\rho_0 \propto (c - c_0)^\Delta \quad 0.34 \text{ at.\% Fe} \leq c - c_0 \leq 2.1 \text{ at.\% Fe} \quad (4)$$



is a reasonably good representation of the PdFe data, although the fit is not as convincing as that shown for PdNi (which covers a comparable concentration range, i.e.  $0.15 \text{ at.}\% \text{ Ni} \leq c - c_0 \leq 1.25 \text{ at.}\% \text{ Ni}$ ). More important, however, is the result that the 'exponent'  $\Delta$  is quite different for the two systems; we estimate that for PdFe,  $\Delta = 1.0 \pm 0.05$  while, for PdNi,  $\Delta = 9/4 \pm 0.1$  (an increase in  $c_0$  the  $10^{-1} \text{ at.}\% \text{ Fe}$  reduces to  $\Delta$  estimate to around 0.9 in PdFe). As mentioned in the introductory section, it has not, as yet, been determined theoretically whether the zero-temperature SRA should exhibit a critical dependence on the reduced concentration  $c - c_0$  near a critical percolation concentration  $c_0$ . By contrast, the experimental data on both PdFe and PdNi are consistent with critical behaviour. The SRA in both systems follows a power-law relationship above a critical concentration  $c_0$  which is well established in Pd-Ni ( $c_0 = 2.25 \pm 0.05 \text{ at.}\% \text{ Ni}$  [4, 5, 23]) and very plausible in PdFe; furthermore, over the appropriate regime the SRA varies by more than two orders of magnitude in PdNi and nearly an order of magnitude in PdFe. Thus, despite the current lack of theoretical guidance, not only do these data suggest that this transport coefficient does behave critically but also they raise the possibility that the universality class characterizing the SRA is different for systems with or without orbital moments. This would be an intriguing result, particularly as the critical magnetic behaviour of these systems near  $T_C$  is in the same class.

There are, of course, several concerns raised by this interpretation that need to be answered. Immediately above the power-law regime with  $\Delta = 9/4$  in PdNi, there is a region in which these data can be represented by a similar compositional dependence, but with  $\Delta = 1$  (the broken line in figure 4). This region, however, covers a very restricted concentration interval ( $1.25 \text{ at.}\% \text{ Ni} \leq c - c_0 \leq 2.75 \text{ at.}\% \text{ Ni}$ ), far narrower than that in PdFe, but a consideration of previous data† on PdNi suggests that this simply represents transitional behaviour between the 'critical' region below  $1.25 \text{ at.}\% \text{ Ni}$  and the peak in the SRA near  $10 \text{ at.}\% \text{ Ni}$ . Previous data on PdFe [6] are inconclusive as to whether the behaviour reported here continues at higher Fe concentrations. A second, more important point is related to the choice of variable plotted on the abscissa in figure 4. It might be argued that the dimensionless ratio  $(c - c_0)/c_0$  is more appropriate than the reduced concentration  $c - c_0$ ; this dimensionless ratio is shown therefore on the *upper* scale in this figure.

Obviously, for this upper scale the choice for  $c_0$  is important, and the values shown for PdFe have been calculated with  $c_0 = 10^{-2} \text{ at.}\% \text{ Ni}$ . The concerns raised by this revised plot against the dimensionless concentration ratio are that the PdFe data now fall into a regime ( $(c - c_0)/c_0 \sim 1$ ) where the PdNi measurements indicate  $\Delta \simeq 1$ , which might be taken as suggesting that the behaviour reported for PdFe does not represent true critical behaviour. While such considerations are always present when dealing with *any* critical effects, the following three points lend support to the interpretation offered above.

- (i) Alternative choices for  $c_0$ , particularly  $c_0 = 10^{-1} \text{ at.}\% \text{ Fe}$ , would shift the PdFe data well into the region identified as 'critical' in PdNi.
- (ii) The composition region with  $\Delta \simeq 1$  is truly restricted in PdNi; this region is much more extended in PdFe, as is the 'critical' region in PdNi.
- (iii) Finally, and probably most important, the magnitudes for the SRA themselves

† Our previous investigations of PdNi did not involve concentrations above  $5 \text{ at.}\% \text{ Ni}$  [5, 9] and earlier studies [6, 7]—while showing considerable scatter in SRA values—indicate that the SRA peaks near  $10 \text{ at.}\% \text{ Ni}$  at a value of about  $3.5 \times 10^{-2}$ .

(decreasing to below  $10^{-3}$  in both systems) are quite comparable; this clearly supports the identification of the behaviour in PdFe as critical.

Despite this indirect evidence, measurements at lower Fe concentrations might still seem appropriate. Such measurements would, however, encounter substantial difficulties due to the rapid decrease in  $T_C$  and the attendant increasing influence of applied fields on the spin polarization and configuration. Thus the concentration range covered in figure 4 would be difficult to extend in PdFe and we suggest therefore that confirmation for the interesting possibilities raised by these data be sought in other systems without orbital moments, but which possess substantially higher values for  $c_0$ . We are currently investigating a number of systems which might satisfy these criteria.

To conclude, we discuss various model predictions for the SRA. Since isolated Fe atoms do not possess orbital moments—a conclusion drawn from the observation that the SRA vanishes smoothly in the limit of low Fe concentrations [6–8]—the presence of a non-zero SRA as the Fe concentration increases results presumably from spin–orbit coupling in the impurity-induced exchange-split Pd *d bands*. Despite their obvious applicability, band models do not appear capable of a quantitative explanation of the above data, at least with the current results that are available. By contrast, and rather unexpectedly, localized models do provide a quantitative basis for explaining these data. In localized models the dominant contribution to the anisotropy in scattering cross section comes from conduction electron–electric quadrupole coupling [18, 24]; at low temperatures where a Wortis [25] expansion for the *z* component of local spin operators is valid, quantities that transform with quadrupolar symmetry ( $l=2$ ) should behave as [26]

$$[(S_i^z)^2 - S(S+1)/3] \sim m^{l(l+2)/2} = m^3. \quad (5)$$

Within the framework of a percolation model (i.e. a ferromagnetic ground state emerging from the formation of an infinite percolating cluster of giant moments, as some theoretical approaches have justified on the basis of the modification of conventional RKKY coupling by exchange enhancement effects in these systems [27]), the magnetization  $m$  is simply given by the probability  $P(c)$  that a spin is part of this infinite cluster. With

$$P(c) = [(c - c_0)/c_0]^\beta \quad (6)$$

and  $\beta$  estimated at 0.4 [28], and independent of lattice structure [29], localized models predict that

$$\Delta = \beta l(l+1)/2 \quad (7)$$

which yields  $\Delta = 1.2$  for  $l = 2$ . While this prediction is higher than the value of  $1.0 \pm 0.05$  determined for PdFe, the agreement is sufficiently close—particularly when uncertainties in  $c_0$  are considered—to warrant further investigation in other systems. This ‘near’ agreement between localized model predictions and measurements on an itinerant system is particularly intriguing as the one localized moment system so far investigated (PdNi, where  $\Delta \simeq 9/4$ ) behaves quite differently.

Further detailed measurements and itinerant model predictions based on recently introduced and highly promising LSDA/LMTO techniques [30] offer interesting future possibilities.

## Acknowledgments

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