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LETTER TO THE EDITOR

The spontaneous resistive anisotropy as a measure of exchange bond disorder

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Abstract. We present new measurements of the low-field resistive anisotropy (LFRA) in amorphous $\text{Fe}_{91}\text{Zr}_9$, with and without hydrogen loading, together with a summary of similar data previously acquired for $\text{Fe}_{90}\text{Zr}_{10}$. The temperature dependence of these data below the ferromagnetic ordering temperature T_c is compared with that predicted for the quadrupolar term $(\langle S_z^2 \rangle - S(S+1)/3)$ by localized models incorporating exchange bond disorder, and the magnitude of this dependence is shown to scale with bond disorder.

While the influence of exchange bond disorder on the magnetic properties of interacting spin systems has been extensively investigated both experimentally and theoretically [1, 2], much less effort has been directed towards understanding the associated transport properties. The particular transport coefficient that we wish to focus on in this letter is the spontaneous resistive anisotropy (SRA), which basically measures the difference between the longitudinal (ρ_{\parallel}) and transverse (ρ_{\perp}) magnetoresistance of a single-domain ferromagnet extrapolated to zero induction [3], viz

$$\frac{\Delta\rho(B)}{\rho_0} = \frac{3(\rho_{\parallel}(B) - \rho_{\perp}(B))}{\rho_{\parallel}(B) + 2\rho_{\perp}(B)}. \quad (1)$$

Although the existence of such an anisotropy has been recognized for well over a century [4], an understanding of its varied characteristics, as revealed through numerous experiments, is currently somewhat incomplete [5–7]. As with many other aspects of magnetism two extreme approaches for interpreting SRA data exist, based on localized [8, 9] and itinerant [10, 11] models, both of which have met with varying degrees of success. In either approach the observation of an anisotropy relies on the presence of two essential ingredients, an internal or external field to polarize the spin system and spin–orbit coupling (and thus a non-zero orbital component) at scattering sites. The origin of the SRA is however more easily understood and calculated using localized models, as the following shows. Any orbital components are necessarily associated with non-spherical charge distributions, and a multipole moment analysis of the latter reveals that the lowest-order contribution to the asymmetry in the magnetoresistance arises from *electric quadrupole* scattering effects (electric dipole terms vanish in a central field). To lowest order the Hamiltonian \mathcal{H} for conduction electron scattering from state k to k' can thus be expressed as [8, 9]

$$\mathcal{H} = \sum_{k, k'} \left[V + JS \cdot \sigma - \frac{D}{k_F^2} \{ (S \cdot k)(S \cdot k') - S(S+1)k \cdot k' / 3 \} \right] \alpha_{k'}^{\dagger} a_k \quad (2)$$

with V representing the screened Coulomb potential, $JS \cdot \sigma$ the well established *magnetic dipole/exchange* coupling between a localized spin (S) and a conduction electron (spin σ) while the final term describes scattering from the electric quadrupolar moment (D). Though exchange scattering can be anisotropic in single crystals [12], the first two contributions in \mathcal{H} are isotropic in amorphous or polycrystalline conductors, so that in the limit of $V \gg |J|, |D|$, the asymmetry in the magnetoresistance assumes the form [8]

$$[\rho_{\parallel}(B) - \rho_{\perp}(B)]/\rho_0 \simeq (D/V) \left[\langle S_z^2 \rangle - S(S+1)/3 \right] \quad (3)$$

which displays the expected $(3 \cos^2 \theta - 1)$ angular dependence associated with quadrupolar effects with axial symmetry about the field direction [5].

The particular localized model that has been adopted to calculate these quadrupolar asymmetries is based on an effective field Ising Hamiltonian [13, 14] which includes a uniform external field h_a and a random spin-spin exchange coupling J_{ij} between *localized* spins with quantum number S . Assumptions regarding the exchange coupling distribution are used to characterize the disorder in the system, which, for a Gaussian distribution, lead to a set of coupled equations for the thermal and exchange averaged magnetization $m = \langle \langle S_z \rangle_T \rangle_J$ and the parameter $q = \langle \langle S_z^2 \rangle_T \rangle_J$ [15]

$$\begin{aligned} m(h_a, T) &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} S B_S(x) \exp\left(-\frac{\alpha^2}{2}\right) d\alpha \\ q(h_a, T) &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} S^2 B_S^2(x) \exp\left(-\frac{\alpha^2}{2}\right) d\alpha. \end{aligned} \quad (4)$$

In equation (4) the argument x of the Brillouin function $B_S(x)$ is defined as $x = S(\bar{J}_0 m + \bar{J} \alpha \sqrt{q} + h_a)/k_B T$ while $\eta = \bar{J}_0/\bar{J}$ represents the ratio of the first to second moment of the exchange coupling distribution. The use of such a model requires the thermal average in equation (3) to be replaced by the double average $\langle \langle S_z^2 \rangle_T \rangle_J$ and that the ratio η be kept larger than 1.2 so that a paramagnet to ferromagnet transition alone occurs on reducing the temperature (then *possible* complications associated with replica symmetry breaking and limitations imposed by the Ising character of the model on discussions of *transverse* spin ordering are irrelevant).

Before presenting the results of these model calculations, it is appropriate to review the experimental data with which they will be compared. These data consist of new measurements of the resistive anisotropy in amorphous $\text{Fe}_{91}\text{Zr}_9$ (with and without hydrogen loading) along with similar measurements reported previously on $\text{Fe}_{90}\text{Zr}_{10}$ [16]. Details of sample preparation, measuring and hydrogenation techniques have been given previously [16, 17], and while it is not possible for us to determine the H/Zr ratio this is *not* important as far as the present comparison is concerned. What is important is the well catalogued increase in the ferromagnetic ordering temperature $T_c(\propto \bar{J}_0)$ accompanying hydrogenation [18] and the suppression of the re-entrant spin-glass transition temperature ($\propto \bar{J}$) evident at lower temperatures in the composition range studied here [19]: specifically the introduction of hydrogen increases the ratio $\eta = \bar{J}_0/\bar{J}$.

The measurements summarized in figures 1 and 2 are actually of the low-field resistive anisotropy (LFRA) given by the ratio

$$\frac{\Delta\rho(H_a)}{\rho_0} = \frac{\rho_{\parallel}(H_a) - \rho_{\perp}(H_a)}{\rho_0} \quad (5)$$

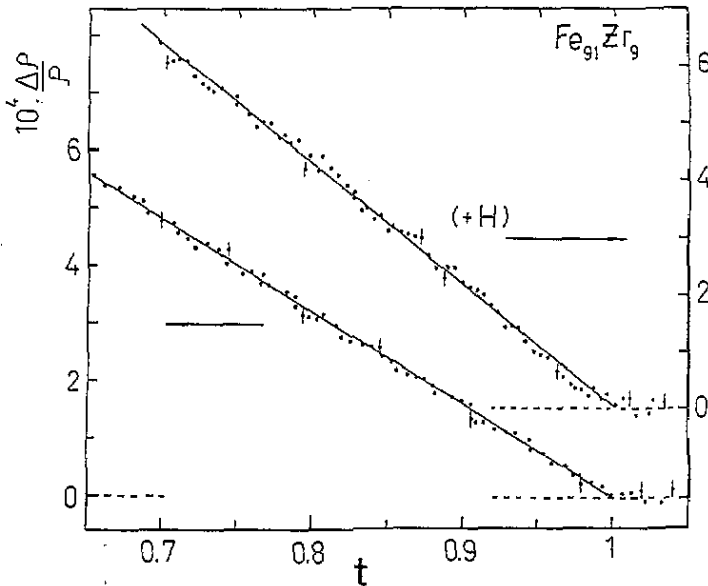


Figure 1. The low-field resistive anisotropy (LFRA), $\Delta\rho/\rho_0$, measured in an applied field of 5 mT, plotted against reduced temperature T/T_c for the $\text{Fe}_{91}\text{Zr}_9$ sample, with and without hydrogen loading. The lines drawn yield the slopes A listed in table 1.

for small applied fields $\mu_0 H_a$. This ratio differs in two important ways from the better known SRA of equation (1) in that the latter utilizes not only an extrapolation involving B , but also bases this extrapolation on the variation of ρ_{\parallel} and ρ_{\perp} above technical saturation [20]. In characterizing the behaviour of a spin system near a phase transition we strongly favour the use of the LFRA, since the application of large fields near T_c can smear the transition by altering the intrinsic spin polarization significantly. The data shown in figures 1 and 2 were acquired in the remanent field of a rotatable electromagnet (the field being applied along the two larger sample dimensions); the magnitude of this remanent field, $\mu_0 H_r \sim 5$ mT, is insufficient to enhance the intrinsic spin polarization at these temperatures, nor will it modify the conduction electron trajectories in any significant way. The essential role played by this field is to establish a single-domain structure over a length scale comparable to the mean free path, a point discussed in more detail below.

The particular points evident from the data shown in figures 1 and 2 which we wish to emphasize are that the LFRA can be expressed as

$$\frac{\Delta\rho(H_r)}{\rho_0} = A(\eta) \left(\frac{T_c - T}{T_c} \right) \quad (6)$$

indicating that this ratio increases essentially linearly with decreasing temperature below T_c , and that the coefficient $A(\eta)$ increases substantially following hydrogenation (see table 1). These are precisely the features reproduced by the model calculations.

Figure 3 depicts calculations of the quadrupolar term ($\langle S^2 \rangle_T$) - $S(S+1)/3$ plotted as a function of reduced temperature $t = T/T_c$ (a natural parameter of the model used) for various values of the ratio η . A comparison of the results displayed in figures 1-3 (and summarized in table 1) allow the following conclusions to be drawn.

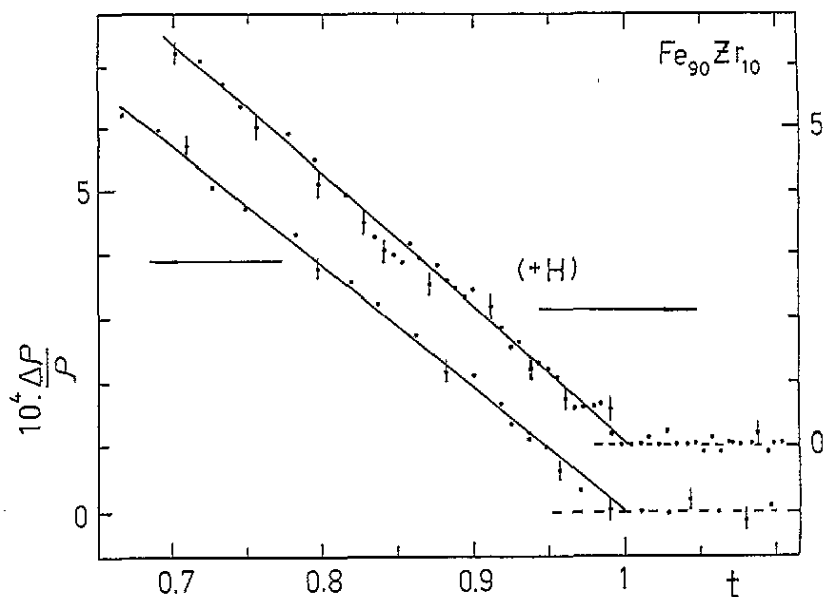


Figure 2. The LFRA, $\Delta\rho/\rho_0$, measured in 5 mT, plotted against reduced temperature for the $\text{Fe}_{90}\text{Zr}_{10}$ specimen.

Table 1. Parameters deduced from low-field resistive anisotropy (LFRA) and AC susceptibility (χ_{AC}) measurements. (Note that while relative temperatures can be determined with high precision (~ 1 mK), absolute temperatures (and hence T_c values) are uncertain to typically $\pm(0.5-1)\%$ for each measurement.)

Sample	LFRA		χ_{AC}		
	$10^4 A$	T_c (K)	T_c (K)	δ	$(\gamma + \beta)^a$
$\text{Fe}_{91}\text{Zr}_9$	16 ± 1	203	200	4.85 ± 0.15	1.75
$\text{Fe}_{91}\text{Zr}_9\text{+H}$	21 ± 1	210	206	4.8 ± 0.15	1.75
$\text{Fe}_{90}\text{Zr}_{10}$	19 ± 1	235	237	4.55 ± 0.15	1.75
$\text{Fe}_{90}\text{Zr}_{10}\text{+H}$	23 ± 1	260	—	—	—

^a The data are consistent with Heisenberg model exponents ($\gamma = 1.386$, $\beta = 0.365$) [17].

(i) The linear temperature dependence established experimentally for the LFRA immediately below T_c is consistent with localized model calculations for quadrupolar scattering [21].

(ii) The coefficient $A(\eta)$ of this linear temperature dependence is predicted to increase with increases in the ratio η (decreasing bond disorder); this result is again consistent with the established influence of hydrogen in these FeZr metallic glasses, discussed above.

In making the above comparisons there are several points that need to be kept in mind.

(a) It is the LFRA ratio $\Delta\rho(H_T/\rho_0)$ which is compared with the quadrupolar term; by taking such a ratio the influence of detailed band structure effects are removed as they contribute in a comparable way to both numerator and denominator.

(b) As is evident from equation (3) the relationship between the LFRA ratio and the quadrupolar term also involves the ratio of the two coupling constants (D/V), a ratio which is assumed to be unchanged following hydrogenation. We believe this to be reasonable for

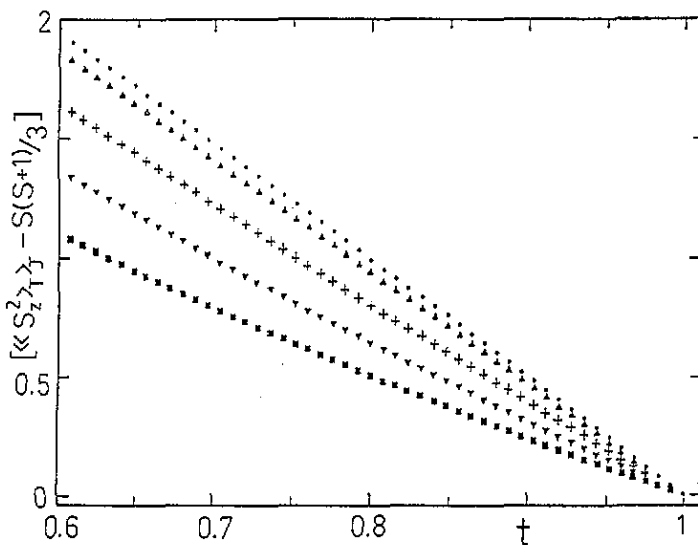


Figure 3. The calculated variation of the quadrupolar term ($\langle S_z^2 \rangle_T - S(S+1)/3$) as a function of reduced temperature $t = T/T_c$ for η values of ∞ (\bullet), 5 (\blacktriangle), 2.5 ($+$), 1.67 (\blacktriangledown) and 1.25 (\blacksquare). Here a spin value $S = 5/2$ has been employed with a reduced field $h_a = 5 \times 10^{-5}$ (corresponding to an applied field of 5 mT when $T_c = 200$ K).

the present experiments in which the degree of hydrogenation was intentionally kept small. As mentioned above, no direct means of measuring the hydrogen uptake is available to us; nevertheless the increase ΔT_c in the ordering temperature T_c following loading in the present experiment is far less ($\Delta T_c \sim 7$ K) than that accompanying complete charging ($\Delta T_c \sim 200$ K [18]) or that introduced in related previous experiments ($\Delta T_c \sim 30$ K [17], table 1). In the present experiment the room temperature resistivity of the sample was also measured before and after charging; these measurements not only monitor the resistivity of the samples in the *paramagnetic* state but they were also carried out at essentially equal temperature intervals above T_c . The result that this resistivity remained essentially unaltered between 120–125 $\mu\Omega$ cm (with experimental error of $\sim 5\%$ arising primarily from shape factor uncertainties) suggests that ρ_0 —and hence V —is reasonably constant. As far as D is concerned, no direct means of estimating it are known to us; qualitatively however it might be argued that the introduction of low-atomic-weight hydrogen should weaken spin-orbit effects and the associated anisotropy rather than accentuate them by the 25–30% observed.

(c) The claim that the applied field $\mu_0 H_r \simeq 5$ mT in which the LFRA ratios were measured renders the samples (both with and without hydrogen loading) single domain over distances comparable to a conduction electron mean free path is difficult to substantiate qualitatively. There is, however, indirect evidence supporting this claim. The Curie temperatures T_c were estimated in two ways: from the temperature at which the anisotropy vanishes, and from the detailed analysis of AC susceptibility data [17]. As can be seen from table 1 the agreement between the two is good, which confirms that the polarizing field in the present experiment is indeed the exchange field, which collapses at T_c . The coercive field H_c is also expected to vanish in the vicinity of T_c , and while the domain structure in these samples is controlled by a number of factors [22], one of the more important is the magnetocrystalline anisotropy, a measure of which is provided by H_c . No detailed study of the temperature dependence of H_c immediately below T_c has been carried out for the present samples; however the

$\text{Fe}_{91}\text{Zr}_9$ specimen was specifically chosen for its low coercive field at 77 K (estimated at 0.05 mT [17], falling slightly to 0.02 mT following charging). The experimental conditions employed in the present experiment in which the LFRA was measured in fields *at least* two orders of magnitude larger than the coercive field support the above claim. The same is true for the AC susceptibility data measured in zero field and an applied field of 5 mT; these show little change following hydrogenation over the region of interest (150–190 K).

Model calculations—which do not incorporate domain effects—indicate that the quadrupolar term should be field independent immediately below T_c for small fields (see figure 4). Such a test would provide more definite support for the above claim, but is difficult to implement with current apparatus (the remanent field of the electromagnet is not easily changed). However, a corollary to the above result would be that should the sample become multidomain over a mean-free path then the LFRA should fall *below* the linear model prediction, a result not observed in the present experiment†.

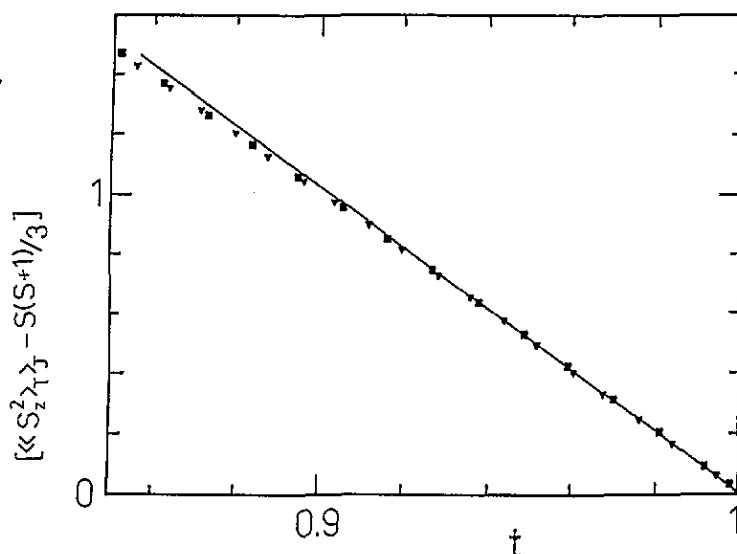


Figure 4. The calculated quadrupolar term ($\langle\langle S_z^2 \rangle_T \rangle_J - S(S+1)/3$) plotted against reduced temperature t for an η value of 2.5. For a Curie temperature $T_c = 200$ K such calculations correspond to applied fields $\mu_0 H_a$ (in mT) of 13.3 (■) and 3.4 (▼); the solid line corresponds to equation (6).

(d) In a recent paper, published prior to calculating the quadrupolar term [16], a qualitative argument was presented to explain the linear increase in the LFRA with decreasing temperature below T_c . This argument was based on the fluctuation-dissipation theorem which, in the present context, can be written as

$$\langle\langle S_z^2 \rangle_T \rangle_J = \langle\langle S_z \rangle_T^2 \rangle_J + \chi T = q + \chi T. \quad (7)$$

Provided that the field dependent susceptibility χ is not strongly temperature dependent below T_c (as is observed [17]) then, for $\eta > 1.25$, the temperature dependence in equation (7)

† Recently acquired data on FePt in which H_c increases quite rapidly below T_c exhibits such a deviation in low field.

is determined from

$$\langle\langle S_z^2 \rangle_T \rangle_J \simeq q \simeq m^2 (= \langle\langle S_z \rangle_T \rangle_J^2) \quad (8)$$

so that in low applied fields

$$\langle\langle S_z^2 \rangle_T \rangle_J \simeq \left(\frac{T_c - T}{T_c} \right)^{2\beta} = \frac{T_c - T}{T_c} \quad (9)$$

in mean field ($\beta = \frac{1}{2}$), a result which is confirmed ($\langle\langle S_z^2 \rangle_T \rangle_J \simeq m^2$) by model calculations. The behaviour of the quadrupolar term, and hence by inference the LFRA, can then be represented by

$$\Delta\rho \propto m^2. \quad (10)$$

While equation (10) yields an *approximately* correct representation of the *temperature* dependence of $\Delta\rho$, such an association should be viewed with considerable caution, for the following reasons. Firstly, neither ρ_{\parallel} nor ρ_{\perp} ($\Delta\rho = \rho_{\parallel} - \rho_{\perp}$) exhibits such a dependence on magnetization *individually*, a result that would be expected if the low-field magnetoresistance near T_c was dominated by *magnetic/exchange* scattering [23,24] (indeed such a situation would lead to $\rho(0) - \rho(H_i) \propto m^2$ in *all* orientations for polycrystalline samples, so that $\Delta\rho = \rho_{\parallel} - \rho_{\perp} = 0$ [25]). Secondly, while (10) mimics the correct *temperature* dependence it does not reproduce the correct *orientational* dependence of $\Delta\rho$ about the field direction [5]. Both of these latter points are meant to emphasize that the LFRA measured in the present experiment originates from electric quadrupole and not magnetic dipole scattering. Equation (10) is misleading in this respect.

In summary, measurements of the LFRA in amorphous $\text{Fe}_{91}\text{Zr}_9$ (with and without hydrogen loading) are presented along with a summary of previously acquired data on $\text{Fe}_{90}\text{Zr}_{10}$. These data are compared with the behaviour predicted by localized models for the quadrupolar term ($\langle\langle S_z^2 \rangle_T \rangle_J - S(S+1)/3$), from which it is concluded that such model calculations appear to reproduce both the temperature and exchange bond dependence of this anisotropy. From a model standpoint, while it might appear that an itinerant approach might provide a more appropriate description of these high-Fe-concentration metallic glasses, the behaviour of the resistive anisotropy—like the asymptotic critical exponents [17]—is in fact quantitatively consistent with a localized model.

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