Magnetic inhomogeneities inherent to spin systems with quenched random-exchange disorder

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A critical assessment of theoretical models proposed in the literature for the spin structure in amorphous $Fe_{100-x}Zr_x$ alloys (a three-dimensional ferromagnet with quenched random-exchange disorder which exhibits a re-entrant behaviour at low temperatures), based on a detailed comparison between theoretical predictions and available experimental results, is presented. The infinite ferromagnetic matrix plus finite ferromagnetic spin clusters model, originally proposed by the author, is shown to provide a coherent basis for understanding the nature and origin of magnetic inhomogeneities in this system.

THE past decade has witnessed a growing experimental evidence to the effect that magnetic inhomogeneity or the so-called 'magnetic microstructure' is an attribute *inherent* to magnetic systems as disparate as ferromagnets or antiferromagnets with or without quenched random-exchange disorder, nanocrystalline soft magnetic alloys, nanostructures, magnetic fine particles, granular giant magnetoresistance (GMR) materials, colossal magnetoresistance (CMR) manganates and frustrated pyrochlore oxides, and that the nature of magnetic inhomogeneity basically decides the magnetic behaviour of a given system[1]. Attempts to understand the origin of magnetic inhomogeneities in these systems have heavily drawn upon the existing knowledge[2–35] about the influence of spin frustration and local magnetic anisotropy on the magnetic order in the amorphous (a-) $Fe_{100-x}Zr_x$ ($7 \le x \le 12$) alloys. However, despite the fact that the a-Fe–Zr system is the most thoroughly studied quenched random-exchange ferromagnet (note that this class of ferromagnets includes both quenched random site-diluted and bond-diluted ferromagnets), such attempts have met with limited success primarily because conflicting opinions prevail about the nature and origin of magnetic inhomogeneity and about the finer details of the magnetic microstructure in a-$Fe_{100-x}Zr_x$ alloys. Now that the progress in understanding magnetic properties of a wide variety of spin systems is directly linked to the advances made in ascertaining the actual role of competing interactions in the model system a-Fe–Zr, this paper makes a critical assessment of these divergent viewpoints in the light of the existing experimental data. In this context, the results of recent neutron scattering experiments that have provided a crucial clue about the nature of magnetic inhomogeneities in a-$Fe_{100-x}Zr_x$ alloys are also briefly discussed.

Bulk magnetization[2–5,11,12], magnetic susceptibility[3,6,10,14], Mössbauer effect[19–22] and muon spin relaxation[33] data have established the following widely-accepted magnetic phase diagram (Figure 1) for a-$Fe_{100-x}Zr_x$ alloys. Barring the alloy with $x = 7$ ($x = 12$), which behaves as a spin glass (conventional ferromagnet) with a well-defined freezing temperature T_f (ordering temperature T_C), the alloys with $x = 8$–11 exhibit two transitions as the temperature is lowered from high temperatures; a paramagnetic (PM) to ferromagnetic (FM) transition at the Curie temperature T_C followed at a lower temperature T_{RE} by a transition from the FM state to the reentrant (RE) state. With x decreasing from $x = 11$, T_{RE} increases while T_C decreases such that the $T_{RE}(x)$ and $T_C(x)$ phase transition lines meet at T_f for $x = 7$. There is a general consensus that the RE state is a *mixed* state in which long-range ferromagnetic order coexists with the spin glass order but the exact nature of the ferromagnetic and spin glass order remains a highly controversial issue. The next section briefly summarizes the models proposed in the literature for describing the ferromagnetic and reentrant states in a-$Fe_{100-x}Zr_x$ alloys.

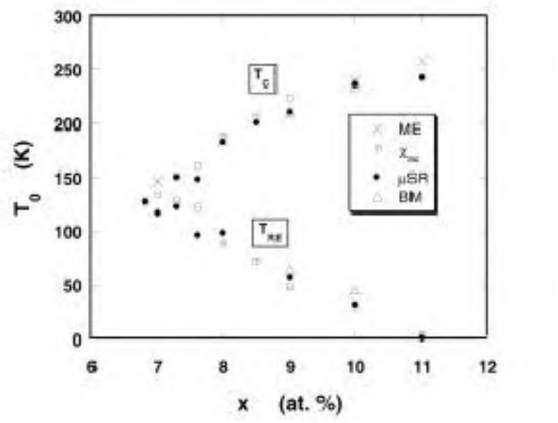

Figure 1. Magnetic phase diagram for amorphous $Fe_{100-x}Zr_x$ alloys compiled using the bulk magnetization (BM), ac susceptibility (χ_{ac}), Mössbauer effect (ME) and muon spin resonance/relaxation (μ_{SR}) data available in the literature.

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Models for magnetic microstructure

Magnetic inhomogeneities in a-$Fe_{100-x}Zr_x$ alloys have found *four different* model descriptions in the literature. The *first* approach[2,4,6,9] considers the magnetic microstructure of the samples as consisting of *spin clusters of antiferromagnetic* (AF) Fe spins and the *ferromagnetic* (FM) Fe–Zr *matrix* (in which these clusters are frozen in random orientations for $T \leq T_{RE}$) and arising from the changes in the sign of the exchange interaction due to *local* variations in the *composition* (i.e. the sample regions where Fe atoms have predominantly Fe neighbours, Fe spins are coupled antiferromagnetically whereas the Fe spins in the spatial regions where Fe atoms share Zr neighbours they are ferromagnetically coupled), as shown in Figure 2. According to the second (the so-called FM cluster-FM matrix) model, proposed by Kaul *et al.*[3,12,20,23,24] and depicted in Figure 3, the spin system for $T \leq T_C$ comprises the *infinite* three-dimensional *ferromagnetic network* (matrix) and finite spin clusters (composed of a set of *non-collinear*[20,22] but *ferromagnetically* coupled spins), which are embedded in the *spin-canted* FM matrix[3,12,20,23,24] and frozen in random directions for $T \leq T_{RE}$. Contrasted with the first picture, the spatial segregation of finite FM clusters and FM matrix in this model originates from the *local* atomic *density* fluctuations. A somewhat similar model, put forward by Kiss *et al.*[11] based on the interpretation of the magnetization–magnetic field isotherms in terms of the classical theory for *interacting* superparamagnetic particles, indicates that the FM clusters occupy the entire volume of the sample. The *third* model regards the a-$Fe_{100-x}Zr_x$ alloys to be a 'wandering axis' ferromagnet[7] (Figure 4) in which the non-collinear magnetic moments are ferromagnetically correlated but the *local* ferromagnetic axis changes throughout the sample. The *fourth* one (the so-called transverse spin-freezing model), due to Ryan *et al.*[21,32,33], envisages the spin system for $T \leq T_C$ to be composed of *ferromagnetically* correlated *longitudinal* (z-direction) spin components and strongly fluctuating *transverse* (xy) spin components (Figure 5); as the temperature is lowered below T_C, transverse spin components *cooperatively* freeze in random orientations in the xy-plane at $T \equiv T_{RE} \leq T_{xy}$ and *coexist* with *collinear* ferromagnetic order along the z-direction.

In subsequent sections, the predictions of these phenomenological models are compared with experimental findings with a view to ascertain which of the models forms a correct description of the magnetic inhomogeneities in a-FeZr alloys.

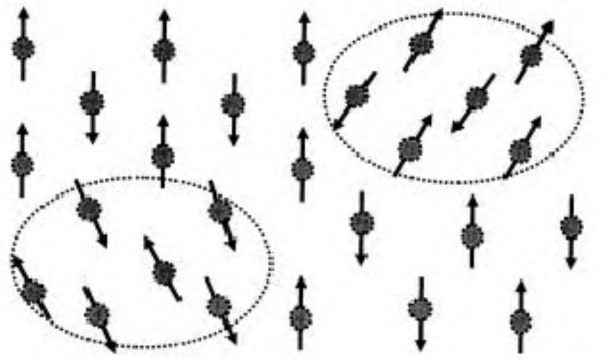

Figure 2. Schematic of spin orientation as per the antiferromagnetic spin clusters–ferromagnetic matrix model[2,4,6,9].

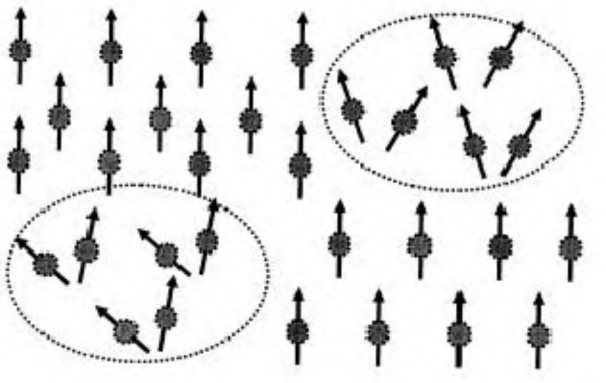

Figure 3. Schematic of spin orientation as per the non-collinear ferromagnetic spin clusters–ferromagnetic matrix model[3,12,20,23,24].

Experimental observations

A variety of experimental techniques such as bulk magnetization[2–5,7–14] (BM), 'in-field' and 'zero-field' ac susceptibility[3,6,10,12–14] (ACS), vibrating reed[15], electrical and galvanomagnetic transport[16,17], electrical noise[18], 'zero-field' and 'in-field' Mössbauer spectroscopy[19–22] (MS), ferromagnetic resonance[23,24] (FMR), small-angle neutron scattering[25–28] (SANS), inelastic neutron scattering[29] (INS), spin polarized neutron scattering[30] (SPNS), neutron depolarization[31,32] (ND), muon spin resonance[33] (SR), Lorentz electron microscopy[34] (LEM) and Kerr effect[35] have been employed in the literature to extensively study the approach-to-saturation[3,5,7,11,12] (high-field susceptibility), quantum corrections

Figure 4. Schematic of spin orientation as per the 'wandering-axis ferromagnet' model[7].

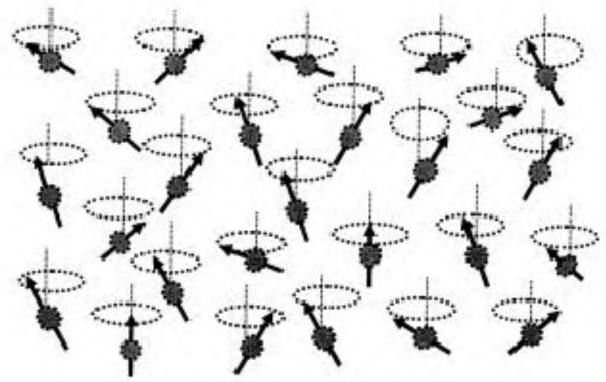

Figure 5. Schematic of spin orientation as per the transverse spin freezing model[21,32,33].

to electrical resistivity at low temperatures[16,17], magnetic excitations[3,5,12,19,20,24], magnetic irreversibilities[3–6,9,12,14], the freezing of the spin degrees of freedom[19–22,24,31–33] and the softening of spin wave modes[19,20,24,29,31] associated with the transition to the reentrant state, magnetoelastic effects[15], critical phenomena[3,8,10,12–15,17], electrical noise[18] and domain structure[34,35] in a-FeZr alloys. The following observations have been made based on these studies: (i) While the technical saturation in magnetization at all temperatures below the Curie temperature, T_C, in reentrant systems is achieved typically at fields ≤ 10 kOe, the magnetization continues to increase gradually as the external magnetic field, H, is increased to values as high as 140 kOe and the magnitude of the high-field susceptibility, χ_{hf}, remains essentially unaltered[3,5,7,11,12] as the temperature is lowered through T_{RE} (the temperature at which the transition to the reentrant state takes place). (ii) Spin wave excitations (*propagating* transverse spin fluctuations) at *low temperatures*, enhanced fluctuations in the local magnetization (*non-propagating* longitudinal and transverse spin fluctuations) over a wide range of *intermediate* temperatures and for temperatures *close* to T_C are mainly responsible for thermal demagnetization[12] of the spontaneous as well as 'in-field' magnetization. A partial substitution of Fe by Co or Ni leads to a progressive suppression of spin fluctuations[12] with the result that the contribution due to spin waves becomes more prominent over a wider range of temperature. (iii) As the temperature is lowered below T_C, the spin wave stiffness, D, increases up to a certain temperature $T \approx 3\ T_{RE}$, as it does for a conventional homogeneous ferromagnet, but upon lowering the temperature further, D decreases[3,19,20,29,31] (softening of spin-wave modes) and attains a small but finite value in the reentrant state. (iv) The weak-irreversibility phase boundary and the weak-to-strong-irreversibility crossover line in the H–T phase diagram, signalling respectively the freezing of spin degrees of freedom *transverse to* and *along* the field direction, exhibit an *observation-time-dependent*[12] temperature shift. (v) The temperature-induced growth of the finite ferromagnetic spin clusters (coexisting with infinite ferromagnetic matrix) in a-$Fe_{90+x}Zr_{10-x}$ alloys has been indirectly inferred from the bulk magnetization[3,12], Mössbauer[20], ferromagnetic resonance[23,24] and electrical noise[18] data. In conformity with this finding, the results of an elaborate analysis of the SANS data[28], taken over a wide range of Q on a-$Fe_{91}Zr_9$, provide a strong evidence for *two kinds* of spin clusters that *coexist* with the *infinite* ferromagnetic matrix for $T < T_C$ and distinguish themselves in the way they respond to the variations in temperature; while the temperature has practically no influence on the average size and log-normal size distribution of one type of spin clusters, it induces growth in both the average size and log-normal size distribution of the spin clusters of other type till the sample warms up to T_C and then disintegrates/disorders them for $T > T_C$ with the result that the average size reduces and the size distribution narrows down. In addition, the Mössbauer[20] and ferromagnetic resonance[23,24] results demonstrate that the freezing of finite spin clusters in random orientations does not begin at T_{RE} but at a temperature $T \approx 3.5\ T_{RE}$ and proceeds gradually over a wide temperature range from $T \approx 3.5\ T_{RE}$ down to the lowest temperature. By contrast, the Mössbauer[21] and SR[33] results indicate a cooperative freezing of transverse spin components at $T = T_{RE}$ and place an upper bound of 3% on the sample volume in which unordered spin clusters could exist for $T < T_C$. (vi) The spin-spin correlation length *diverges*[28] at T_C and the critical behaviour near T_C is akin[3,8,10,12,13,15,23,24] to that in a three-dimensional Heisenberg ferromagnet. However, unlike a conventional homogeneous ferromagnet, only a small (5–10%) fraction of the total spins actually participates in the ferromagnetic (FM)–paramagnetic (PM) phase transition, which implies that a small number of spins constitute the infinite FM network (matrix) while a vast majority of the spins reside in the finite FM clusters for temperatures close to T_C. (vii) Large ferromagnetic domains (typical size 50 μm), which remain unaltered in size when the temperature is lowered through T_{RE} to temperatures as low as 1K, have been directly observed by Lorentz electron microscopy[34] and Kerr-effect method[35].

In the next section, we discuss the above observations in the light of the models proposed in the literature and described in the preceding section.

Inferences

While the presence of an *infinite* ferromagnetic matrix for $T < T_C$ and hence the divergence of the spin-spin correlation length, ξ, at $T = T_C$ rules out descriptions such as the 'wandering-axis' ferromagnet[7], since in such a ferromagnet, ξ *does not diverge* at $T = T_C$. The presence of clusters, and that too in a great proportion, is in direct contradiction with the transverse spin-freezing model[21,32,33] because it considers the spin system to be magnetically *homogeneous* even on the microscopic scale. By comparison, the

coexistence of finite spin clusters with the infinite ferromagnetic (FM) matrix finds a natural place in the so-called FM cluster-FM matrix model[3,12,20,23,24] (Figure 3), which envisages the spin system for $T \leq T_C$ to be composed of the *infinite* three-dimensional *ferromagnetic network* (matrix) and finite spin clusters (composed of a set of *non-collinear*[20,22] but ferromagnetically coupled spins), which are embedded in, but either partially or completely isolated from the FM matrix by zones of frustrated spins surrounding the finite clusters. According to this model, the exchange interaction between spins in the FM matrix weakens as $T \rightarrow T_C$ while the FM coupling between the spins within the finite clusters is still quite strong due to the higher Curie temperature for the clusters. As a consequence, the spins of the clusters that are *partially isolated* from, and hence weakly interact with, not only the FM matrix but also the neighbouring clusters (the so-called strongly interacting clusters), can grow in size with temperature through two mechanisms. In one such mechanism, they can merge together because of the strong coupling between the neighboring clusters to form a bigger cluster. In the other mechanism, the cluster spins are able to polarize an increased number of spins originally belonging to the FM matrix via direct exchange interactions, and hence the clusters grow in size at the expense of the spins contained in the FM matrix. However, the temperatures in excess of T_C disorder not only the FM matrix but also the clusters and hence the cluster size decreases for temperatures above T_C. On the other hand, so far as the direct exchange interactions are concerned, the clusters which are completely isolated from the FM matrix and also from other clusters (the so-called non-interacting clusters) cannot grow in size with increasing temperature, in agreement with the above observation (v). The net result of the temperature-induced cluster growth and the existence of many isolated clusters is that a *major* fraction of total spins resides in the *finite* clusters for temperatures in the vicinity of T_C. The growth process resulting in an increasing presence of large clusters would naturally enhance long-range RKKY and dipolar interactions among clusters, because of the large magnitude of the cluster moments. Moreover, the observations (i)–(iv) can be explained only[3,12,20,23,24,28] in terms of the FM cluster-FM matrix model.

If the spins within the clusters (matrix) were antiferromagnetically (ferromagnetically) coupled, as considered in the antiferromagnetic (AF) spin cluster-FM matrix model[2,4,6,9], it is not easy for the AF cluster spins to polarize the FM matrix spins and thereby grow in size with increasing temperature because of a much higher energy cost involved in this process. Thus, the above observations do not support such a model.

Furthermore, from a recent spin polarized neutron scattering determination of the structure factor, Wildes *et al.*[30] conclude that in direct contradiction with the proposal of Ryan *et al.*[21,32,33] that FeZr glasses are collinear ferromagnets with strong spin fluctuations for $T_{xy} < T < T_C$, *non-collinear* spin components are *ferromagnetically* correlated over several atomic spacings and that the fraction of magnetic moments that are collinear with the mean ferromagnetic direction is small. This observation lends a firm support[30] to the FM cluster-FM matrix model[3,12,20,23,24].

The final picture about the nature and origin of magnetic inhomogeneities in a-$Fe_{100-x}Zr_x$ alloys that emerges from the foregoing discussion is that, due to local atomic density fluctuations, which give rise to spin frustration at the interfaces between the low density pockets and the high density bulk, the spin system consists of an *infinite* three-dimensional *ferromagnetic network* (matrix) and *finite* spin clusters (composed of a set of *non-collinear*[20,22] but *ferromagnetically* coupled spins), which are embedded in, but either partially or completely isolated from, the FM matrix by zones of frustrated spins surrounding the finite clusters, as shown in Figure 3.

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ACKNOWLEDGEMENTS. I thank all the collaborators within and outside India as well as my research students who have contributed significantly to the work presented here.