Spin Glasses and Frustrated Systems — Mark van Schilfgaarde, Chairman

Magnetic ordering in Re-doped *a*-$Fe_{90}Zr_{10}$

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The magnetic properties of rhenium-doped *a*-$Fe_{90}Zr_{10}$ have been studied by magnetization, susceptibility, neutron depolarization, and Mössbauer spectroscopy and a magnetic phase diagram is presented. Rhenium leads to a very rapid loss of magnetic order and *a*-$Fe_{90-x}Re_xZr_{10}$ is a spin glass beyond a critical composition of $x_c \sim 4.5$ at. %. © *1999 American Institute of Physics.* [S0021-8979(99)26808-9]

I. INTRODUCTION

The amorphous Fe–Zr system spans the full range of correlated electron behavior in metals, from superconductivity through spin fluctuations and ferromagnetism, and ultimately into spin glasses.[1–3] As a result, it provides an ideal matrix in which to investigate the effects of doping on magnetic properties. In the binary system *a*-Fe_xZr_{100-x}, both T_c and the magnetization fall rapidly for $x>90$. We have previously demonstrated that this is due to the effects of exchange frustration[3] and that this behavior is fully consistent with the predictions of numerical simulations.[4]

Exchange frustration in the *a*-Fe–Zr system arises through competing direct exchange interactions ($\mathscr{J}$). Positive and negative $\mathscr{J}$'s are present as a result of a fortuitous sign change in the interaction at a distance corresponding roughly to iron's metallic diameter. The detailed behavior of $\mathscr{J}$ is quite complex and depends both on bondlength and on the mean electron density in the alloy.[5] Dopants can therefore modify the exchange distribution in two ways. Where there is a significant size mismatch or a large volume change associated with the dopant (e.g., with hydrogen) the dopant can simply increase the average bondlength and eliminate the short contacts that contribute to the frustration.[6,7] Where the size difference is small (e.g., most transition metals) changes in the average electron density can move the $+\rightarrow-$ crossover distance, without affecting the actual distribution of bondlengths, and so modify the distribution of exchange interactions indirectly. In general, both effects will be present to some degree.

We report here the effects of Rhenium on the magnetic properties of *a*-$Fe_{90-x}Re_xZr_{10}$, close to the ferromagnet-spin-glass crossover. This system was identified during a survey[8] of the effects of transition metal dopants on the *a*-Fe–Zr system as leading to one of the most rapid increases in frustration.

II. EXPERIMENTAL METHODS

Ribbons of *a*-$Fe_{90-x}Re_xZr_{10}$ were prepared by arc-melting appropriate amounts of the pure elements (Fe 99.97%, Zr 99.5%, and Re 99.997%) under Ti-gettered argon, followed by melt spinning in 40 kPa helium with a wheel speed of 55 m/s. For $x \leqslant 5$, good quality meter length ribbons were consistently obtained, and these were sufficient for all of the intended measurements to be made. Beyond this point, the yield was much lower, and relatively small amounts of useful ribbons were obtained. Absence of crystallinity was confirmed by Cu K_α x-ray diffraction on a conventional automated powder diffractometer, and room temperature ^{57}Fe Mössbauer spectra (for $x \leqslant 5$) obtained on a constant-acceleration spectrometer using a 1 GBq ^{57}CoRh source. Low temperature ($12\,\mathrm{K} \leqslant T \leqslant 290\,\mathrm{K}$) spectra were obtained using a vibration-isolated closed-cycle fridge, and fitted using Window's method.[9]

Magnetization and susceptibility measurements were made on a commercial system equipped with a 9 T superconducting magnet and operating between 2 and 300 K.

Neutron depolarization data were obtained using the C5 beamline of DUALSPEC, a triple-axis spectrometer, at AECL, Chalk River. Initial polarizations of ~96% at $\lambda = 0.237$ nm were achieved with Cu_2MnAl single crystals as polarizer and analyzer. Measurements were made between 11 and 300 K in a 1 mT guide field, on stacks of one or more 15 mm lengths of 20 μm thick ribbons.

III. RESULTS AND DISCUSSION

The χ_{ac} data in Fig. 1 show a rapid reduction in T_c and a clear evolution in behavior from the flat topped form characteristic of a demag-limited ferromagnet at $x=0$, to a spin-glass-like cusp at $x=8$. These changes are accompanied by declines in both the magnetisation and average hyperfine field ($\langle B_{hf} \rangle$) measured by Mössbauer spectroscopy at 12 K (Fig. 2). As can be seen from the right-hand side of Fig. 2,

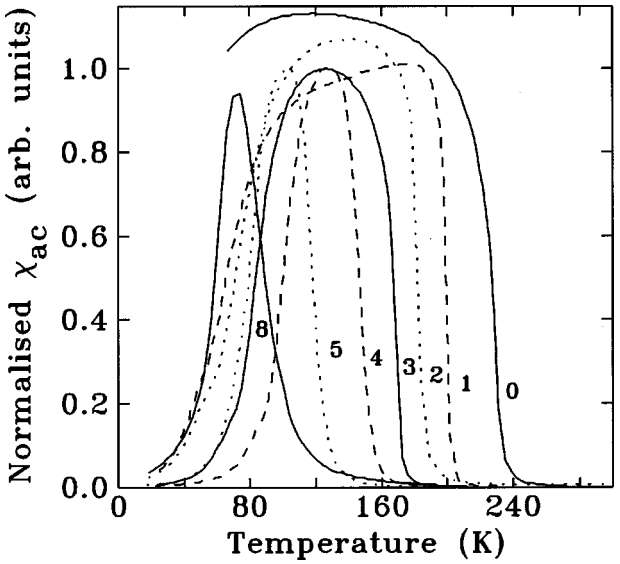

FIG. 1. χ_{ac} data for a-$Fe_{90-x}Re_xZr_{10}$ showing the rapid reduction in T_c and the evolution to a spin-glass-like cusp with increasing Re content.

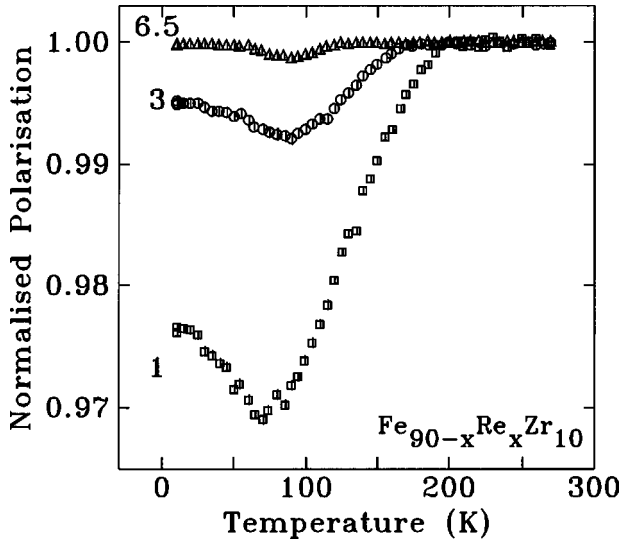

FIG. 3. Neutron depolarization data for three representative a-$Fe_{90-x}Re_xZr_{10}$ samples. Thicknesses: $x=1$, 20 μm; $x=3$, 60 μm; and $x=6.5$, 120 μm. Data are normalized for base polarization and sample thickness.

much of the decline in $\langle B_{hf}\rangle$ is due to a broadening in the distribution of $P(\langle B_{hf}\rangle)$ towards smaller fields. The temperature dependence of $\langle B_{hf}\rangle$ was followed to obtain a second estimate of the ordering temperatures for these alloys, and yielded values in agreement with those obtained from $\chi_{ac}(T)$.

While the good agreement between the χ_{ac} and Mössbauer T_c's allows us to rule out blocking of superparamagnetic clusters as the origin of the magnetic ordering of any of the alloys in this system (indeed, the Mössbauer values were in general slightly lower, despite the higher effective frequency associated with the measurement) neither technique is unambiguously sensitive to the presence or absence of long-range magnetic order. For this information we turn to neutron depolarization.[10] Here, a beam of polarized neutrons passes through the sample and the series of random precessions undergone by the neutrons as they pass through successive domains depolarizes the beam according to: $P=P_0\exp(-\alpha\lambda^2)$, where $\alpha=\frac{1}{2}c^2\langle B_\perp^2\rangle d\,\delta$, P_0 is the initial polarization of the beam, d is the sample thickness, δ is the mean domain size along the neutron flight path, $\langle B_\perp^2\rangle$ is the mean square domain magnetization perpendicular to the beam polarization, and c has the value $4.633\times10^{14}\,m^{-2}\,T^{-1}$.[11] The onset of depolarization marks the ordering temperature, and the degree of depolarization (if the domain magnetization directions are random or at least well known) reflects the domain average domain size. The data in Fig. 3 are very similar to results from ruthenium-doped a-Fe–Zr alloys,[10] in that there is a pronounced recovery in the polarization on cooling well below T_c (see, e.g., the curve for $x=1$). This recovery reflects a tendency for the sample to magnetize parallel to the neutron polarization direction which is maintained by a 1 mT guide field in the region of the sample. This variation in magnetization direction precludes determination of domain sizes. However, it is clear from the very rapid decline in the depolarization signal with increasing Re content, that the domains decrease in size as Re is added. A second feature this system shares with the Ru series is that for $x=5$ and 6.5, there is a peak in the depolarization signal, and at the lowest temperatures the sample does not depolarize the beam at all. This latter observation requires that there is no long-ranged magnetic order ($\xi<10$ nm) present in these two samples. The weak peak apparent in Fig. 3 for the $x=6.5$ sample, corresponds to a polarization change of less than 1%. The peak position and form correspond to the magnetization trace obtained on warming a zero field cooled (ZFC) sample in a 1 mT dc field, as was the case for the fully frustrated Ru-doped series. We therefore attribute it not to the formation and loss of a long-range ordered state, but rather to field cooling of the sample in the guide field used here. Furthermore, the internal field deduced for the Re-6.5 at. % is 10 mT, as was also found in Ru-3 at. %.[10]

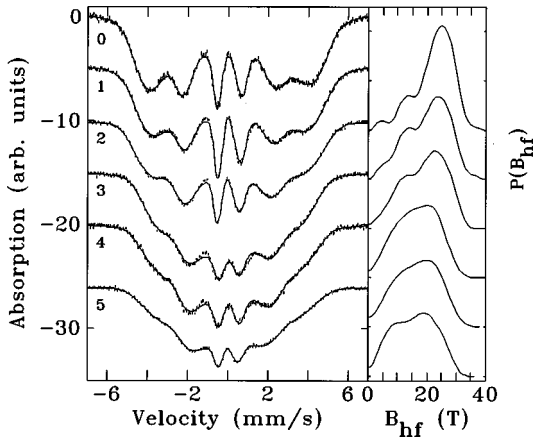

FIG. 2. Mössbauer spectra (left) and corresponding hyperfine field distributions (right) for a-$Fe_{90-x}Re_xZr_{10}$ at 12 K showing a steady reduction in magnetic splitting and a broadening of $P(B_{hf})$ with increasing Re content.

Combining the results obtained on a-$Fe_{90-x}Re_xZr_{10}$ allows us to present a magnetic phase diagram for this system

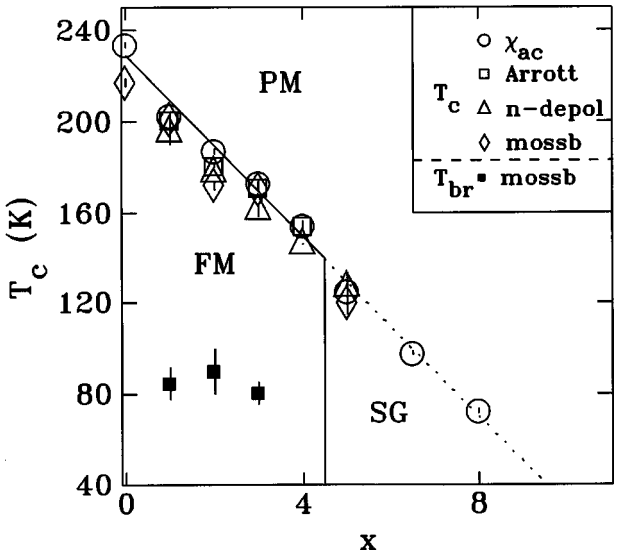

FIG. 4. Magnetic phase diagram for a-$Fe_{90-x}Re_xZr_{10}$ showing transition temperatures deduced using four different techniques. Regions are labeled: PM—paramagnetic, FM—ferromagnetic, and SG—spin glass. The absence of long-range order for $x>4$ is indicated by labeling the low temperature state as a spin glass. T_{br} is discussed in the text.

(Fig. 4). Here we have summarized all of the T_c's determined by various methods, and have included also the results of modified Arrott plots (scaled using correct Heisenberg critical exponents rather than mean-field values) which confirm the absence of long-range order for $x>4$. For $x\leqslant 4$, the system is ferromagnetic, however, T_c and the magnetization fall rapidly as x increases and the level of frustration grows. For $x>4$, the effects of the frustration are sufficient to destroy the long-range order and the system is a spin glass. These results are in full agreement with the results of both numerical simulations[4] and mean-field calculations.[12] Analysis of the temperature dependence of $\langle B_{hf}\rangle$ showed a clear break in slope at a temperature well below T_c. The position of this break is shown as T_{br} on Fig. 4. A similar feature has been seen in the Ru-doped system[10] and is also expected theoretically.[4,12] We have attributed it to the onset of transverse spin freezing at T_{xy}. However, as this has not yet been confirmed by in-field Mössbauer measurements, we continue to label this point as T_{br} for the moment.

Re additions to a-Fe–Zr lead to a very rapid (-19.8 ± 0.7 K/at. %) reduction in ordering temperature, and a total loss of long-range order by a critical composition of $x_c\sim4.5$ at. %. The magnitude of these effects is second only to those caused by Ru (-35 ± 2 K/at. % with $x_c\sim2.4$ at. %).[8,10] Both metals are larger than Fe, and Re is actually slightly larger than Ru, so a simple bond dilation argument fails immediately. However, it is clear that the loss of order is driven by frustration because a substantial iron moment persists beyond x_c, and there is evidence for transverse spin freezing below T_c for $x<x_c$ in both systems. Chemical similarities within the transition metals are quite strong, and once we avoid the 3-d series, many of which are themselves magnetic, it seems plausible to expect that the electronic effects of close neighbors should be similar enough that the contribution of size differences may be resolved. We therefore propose that the origin of the large increase in frustration caused by Re additions lies primarily with modifications in the electron density of the alloys. However, the changes are more moderate than in the case of the neighboring Ru series, perhaps as a result of the slightly greater metallic radius of Re contributing a competing dilation effect.

ACKNOWLEDGMENTS

This work was supported by grants from the Natural Sciences and Engineering Research Council of Canada, Fonds pour la formation de chercheurs et l'aide à la recherche, Québec and the Australian Research Council.

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