Neutron depolarization study of magnetic order in a-Fe_xZr_{100-x} ($x=90-93$), a-$Fe_{90}Sc_{10}$ and their hydrides

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Neutron depolarization measurements have been used to show that ferromagnetic domains exist in a-Fe_xZr_{100-x} alloys for $90 \leqslant x \leqslant 93$, and that these domains are not affected by the ordering of transverse spin components at T_{xy}. Domain sizes decrease from 1.1 μm at $x=90$, to 0.08 μm by $x=93$. Measurements in an applied field set an upper limit of $\sim$50 nm for correlations in the transverse components. By contrast, a-$Fe_{90}Sc_{10}$ shows no evidence of ferromagnetic order and is confirmed to be a spin glass. Measurements on deuterium-loaded samples show that all of the a-Fe–Zr and a-Fe–Sc alloys studied here are ferromagnetic with T_c's in the range 380–400 K, and domains $\sim$1 μm in size. © *1996 American Institute of Physics.* [S0021-8979(96)23608-5]

INTRODUCTION

Amorphous iron rich alloys of the form a-Fe_xETM_{100-x} (where ETM=Sc, Y, Zr, Hf, and $x \sim 90$) provide a rich testing ground for models of magnetic order in the presence of exchange frustration as their behavior spans the full range of possibilities from ferromagnet to spin glass with increasing frustration. Two magnetic transitions are observed in partially frustrated systems, the first, at T_c, to a collinear ferromagnetic state, followed at T_{xy} by the ordering of transverse spin components.[1] Earlier neutron depolarization work on a-Fe_xZr_{100-x} ($x=90-92$)[2] confirmed the presence of ferromagnetic order and found large domains for $x \leqslant 91$, but only weak correlations for $x=92$. However, magnetization and Mössbauer measurements suggest that significant ferromagnetic order persists to $x=93$.[1,3] Our aim here is to extend the neutron depolarization data to $x=93$, to look for possible long-range order in the transverse spin components, and to confirm that the ordering at T_{xy} does not destroy the ferromagnetic order that develops at T_c.

For comparison, we have studied a-$Fe_{90}Sc_{10}$, which is fully frustrated and exhibits a single transition to a spin glass state with no spontaneous magnetization,[1,4–6] although a large magnetization is readily induced by the application of a modest field ($\sim$1 T). Since there should be no long-ranged correlations in a spin glass, we expect no depolarization from this material, and it serves in part as an internal test of our procedures.

Finally, we have also investigated the hydrides of these alloys. The lattice expansion associated with the absorption of hydrogen lifts the exchange frustration in both a-Fe–Zr (Ref. 3) and a-Fe–Sc (Ref. 4) converting them to soft ferromagnets with ordering temperatures $\sim$400 K. However, it has been suggested that a-Fe–Sc–H may remain a spin glass,[6] and depolarization measurements provide a direct way to check for this possibility.

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EXPERIMENTAL METHODS

The alloys were prepared by arc-melting appropriate ratios of the pure elements (Fe: 99.95%, Zr: 99.8%, and Sc: 99.9% pure) under Ti-gettered argon to yield $\sim$2 g ingots. Melt spinning was carried out under a partial pressure of helium onto a copper wheel at 50 m/s. Ribbons were typically 1 mm wide, 20 μm thick, and several meters long. Thickness variations along the samples used for the depolarization measurements were found to be less than ± 1 μm. Absence of crystallinity was confirmed using Cu–K_α powder x-ray diffraction and room temperature Mössbauer spectroscopy. A LakeShore susceptibility system with a closed-cycle fridge was used to record χ_{ac} vs T down to 12 K. The samples were saturated with deuterium using an electrolytic charging method[7] and final deuterium contents were determined from the mass loss on heating to 300 °C in a Perkin–Elmer TGA-7 Thermogravimetric analyzer. This instrument was also used to determine the magnetic ordering temperature of the deuterides by operating with a small field gradient applied. The TGA was calibrated using alumel and nickel standards.

The neutron depolarization measurements were carried out on the long wavelength polarized neutron spectrometer (LONGPOL) at the High Flux Australian Reactor (HIFAR) operated by the Australian Nuclear Science and Technology Organisation (ANSTO). The incident beam, with a wavelength of 0.36 nm, is polarized vertically by scattering from saturated polycrystalline iron sheets. A similar arrangement is used as an analyzer. Typical beam polarizations were in the range 32%–36%. The instrument has been described in more detail elsewhere.[8] A cadmium plate with a 5 mm×5 mm square hole served as the sample holder. Several ribbons were laid side-by-side to cover the opening, with Cd wire covering the gaps between ribbons. One to four layers of ribbon were used, depending on the depolarization expected. The holder was mounted on the cold stage of a closed-cycle fridge which operates down to 12 K. An air-cooled electromagnet was used to apply fields of up to 80 mT in the sample plane, perpendicular to both the beam and the incident polarization.

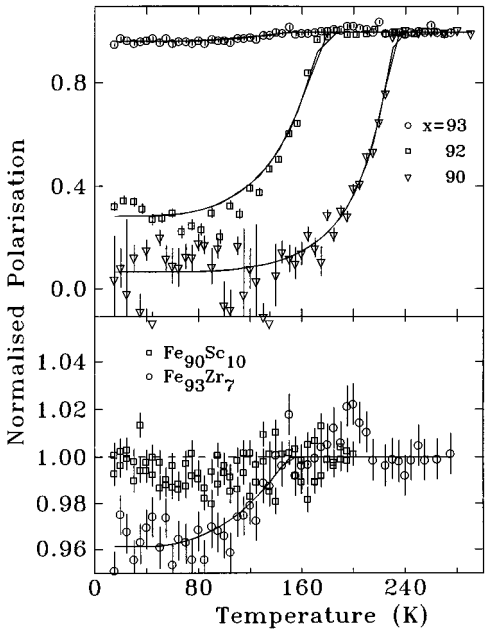

FIG. 1. Normalized depolarization signal for a-Fe_xZr_{100-x} (top) and a-$Fe_{90}Sc_{10}$ (bottom) measured in a guide field of 1 mT. Solid lines are fits assuming a temperature independent domain size. Notice that the a-$Fe_{90}Sc_{10}$ sample does not affect the beam polarization at any temperature.

RESULTS AND DISCUSSION

On passing through a uniformly magnetized region such as a domain, the neutron moment experiences a torque which causes it to precess about the magnetization direction. The total rotation increases with the internal magnetization, the thickness of the domain, and the wavelength of the neutron. If the neutron now passes through many such domains it will undergo many rotations. If the magnetization direction within each domain can be assumed to be a random vector, and the angle through which the neutron moment turns in each domain is $\ll 2\pi$, then the initial polarization is scrambled and decays according to:[9]

$$P = \exp(-\alpha\lambda^2) \tag{1}$$

with

$$\alpha = \tfrac{1}{2}c^2\langle B_\perp^2\rangle d\,\delta, \tag{2}$$

where λ is the neutron wavelength, $\langle B_\perp^2\rangle$ is the mean square domain magnetization perpendicular to the neutron polarization, d is the sample thickness, and δ is the mean domain size. c is a constant which takes the value 4.63×10^{14} $m^{-2}\,T^{-1}$ in S.I. units.

The depolarization signals for all of the samples studied here are shown in Fig. 1. It is immediately apparent that all of the a-Fe–Zr alloys cause a significant depolarization of the beam below their respective T_c's, confirming the presence of ferromagnetic order. The depolarization weakens with increasing iron content as the exchange frustration builds up and the system moves closer to becoming a spin glass. However, even at $x=93$, (shown in the lower half of Fig. 1 on an expanded scale) there is some loss of polarization. By contrast, a-$Fe_{90}Sc_{10}$ has no effect on the beam at any temperature, consistent with the spin glass nature of the order in this alloy and the absence of any ferromagnetic correlations. If we assume that the domains, once formed, do not change size, then the observed temperature dependence of the polarization must result from the normal change in spontaneous magnetization with temperature. Taking the estimated magnetization from Ref. 1, and assuming a simple mean-field temperature dependence, then the data in Fig. 1 can be fitted to Eq. (1) to obtain domain sizes and ordering temperatures. These fits (shown as solid lines on Fig. 1, and summarized in Table I) show that large domains persist to $x=92$ but that beyond this point there is a very rapid reduction in domain size. It is not clear that the 80 nm regions suggested by the fit to the $x=93$ sample are large enough to justify the use of the term ''domain'' but it is clear that some ferromagnetic correlations are present in this material, and that they are not affected by the ordering of the transverse spin components at $T_{xy}\sim80$ K.[1]

TABLE I. Summary of fitted parameters from data shown in Figs. 1 and 3. $T_c(\chi)$ and T_c(ndep) are ordering temperatures derived from bulk and depolarization measurements, respectively. δ is the mean domain size. * Note: a-$Fe_{90}Sc_{10}$ is a spin glass, so the value for T_c given here is for T_{sg}.

Alloy	$T_c(\chi)$ K	T_c(ndep) K	δ μm
$Fe_{90}Zr_{10}$	240±3	232±4	1.1±0.2
$Fe_{92}Zr_8$	182±2	176±1	1.16±0.07
$Fe_{92}Zr_8D_{13\pm2}$	376±3	362±10	1.4±0.1
$Fe_{93}Zr_7$	145±2	150±5	0.077±0.006
$Fe_{93}Zr_7D_{17\pm2}$	372±3	359±6	0.90±0.05
$Fe_{90}Sc_{10}$	95*	···	0.0
$Fe_{90}Sc_{10}D_{28\pm0.5}$	395±5	390±40	1.0±0.1

The main difference between the results presented here and those of Hadjoudj *et al.*,[2] is that we observe a strong depolarization even at $x=92$ and only the $x=93$ sample shows a weak effect. However, examination of the transition temperatures shown in Ref. 2 allows us to bring the two sets of data into agreement. T_c in a-Fe_xZr_{100-x} is a very strong function of x, decreasing at ~25 K/at.%,[1,3] and thus provides a sensitive check on sample composition. Using this scale, it is clear that their $x=90$ and 91 samples are consistent with typical values, but the T_c of their $x=92$ sample is below 150 K, a value more often associated with $x=93$. With this modified composition, our data and those of Ref. 2 are in full agreement.

At T_c a ferromagnetic state forms with a substantial fraction of each moment oriented perpendicular to the ordering axis. The moments precess rapidly about the ferromagnetic, z, axis, so that the xy components time average to zero, and the system appears to be a collinear ferromagnet. The second transition in a-Fe–Zr is associated with the freezing of the transverse degrees of freedom, and as noted above, does not lead to a destruction of the ferromagnetic order. Since the average magnetization does not increase at T_{xy}, the xy components cannot be correlated over substantial distances, but no limits have yet been placed on their correlation length. Since the neutrons are only sensitive to $B_\perp$ we can eliminate the ferromagnetic contribution to the depolarization signal by magnetizing the sample parallel to the neutron polarization

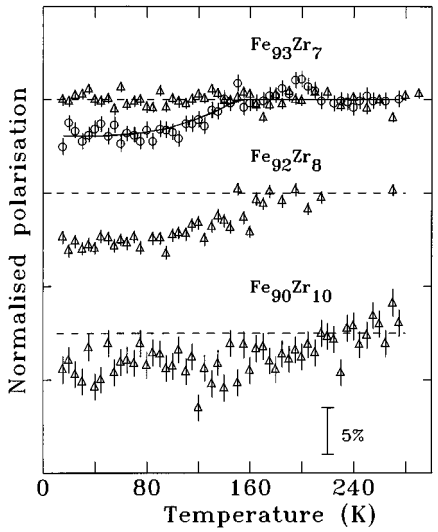

FIG. 2. Normalized depolarization signal for a-Fe_xZr_{100-x} measured in a guide field of 80 mT. The 1 mT data for a-$Fe_{93}Zr_7$ showing a 4% depolarization of the incident beam are repeated for comparison. Dashed lines indicate the base polarization of the beam.

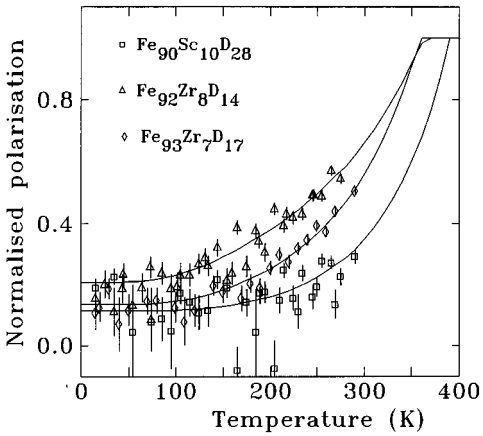

FIG. 3. Normalized depolarization signal for a-$Fe_xZr_{100-x}D_y$ and a-$Fe_{90}Sc_{10}D_{28}$ measured in a guide field of 1 mT. Solid lines are fits assuming a temperature independent domain size.

direction. A simple ferromagnet would then not depolarize the beam even below T_c. In the absence of the ferromagnetic contribution, any correlations in the transverse components should appear as a loss of polarization around T_{xy}. However, Fig. 2 shows that while there is some residual effect from the imperfect alignment obtained in the 80 mT field available, no change can be seen in either the $x=92$, $T_{xy}=46$ K, or the $x=93$, $T_{xy}=78$ K (Ref. 1) samples. Since the change in polarization is much smaller than that seen for the ferromagnetic component of the $x=93$ sample, we are able to rule out correlations in the transverse spin components longer than about half of the 80 nm length scale seen there.

Hydrogen loading is known to convert these alloys into soft ferromagnets,[3,4] and the depolarization behavior of the deuterium loaded samples shown in Fig. 3 is as expected. TGA analysis confirmed that they had ordering temperatures well above room temperature, although the values obtained are only lower limits as the samples lose deuterium rapidly as T_c is approached. Neutron depolarization shows that all of the deuterides are ferromagnetic, and fits to the curves yield ordering temperatures in agreement with the TGA data. Values for domain sizes and ordering temperatures are summarized in Table I. The data for a-$Fe_{90}Sc_{10}D_{28}$ show that it depolarized the beam more strongly than the two Zr samples, leaving no doubt that this material is ferromagnetic. It is not clear why no critical peaks were observed in the susceptibility of a similar sample.[6]

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