Magnetic cluster relaxation in amorphous Fe-Zr alloys

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The spin-glass and the ferromagnetic transitions of amorphous $Fe_{100-x}Zr_x$ ($7 \leq x \leq 12$) alloys in a small applied field (13 mT) were investigated by use of ^{57}Fe Mössbauer spectroscopy. The observed linewidth increase over a wide (≈ 30 K) temperature range both *below* and *above* the transition temperature gives clear evidence of a magnetic relaxation.

The magnetic ordering at the paramagnetic-to-spin-glass (PM-SG) transition temperature (T_G) and at the paramagnetic-to-ferromagnetic-to-spin-glass (PM-FM-SG) transitions (T_c, T_f) in reentrant spin-glass systems (RSG) is not understood, although it has been the subject of extensive investigations.

The Mössbauer linewidth as a function of temperature near T_G (i.e., at the transition from the paramagnetic to the spin-glass state) does not exhibit a clear-cut break, suggesting less sharp transitions than observed at conventional second order magnetic transitions (e.g., Refs. 1–3). Similarly, the existence of a real paramagnetic to ferromagnetic phase transition at T_c of the reentrant magnetic systems is debated.[4–6] In these systems both sharp[2,7,8] or less sharp[4] transitions have been reported (as it is indicated by the temperature dependence of the Mössbauer linewidth). In the insulating $Eu_xLa_{1-x}S$ system, where (for different x values) the reentrant and the spin-glass regime is also detected, both behaviors have been observed.[2] These results were interpreted either in terms of a distribution of the transition temperatures[1,6] or by spin-relaxation broadening[2,9] and superparamagnetic fluctuations.[5]

Amorphous $Fe_{100-x}Zr_x$ ($7 \leq x \leq 12$) alloys are highly suitable systems with which to study the transition from the paramagnetic to the spin-glass ($x = 7$) or to the ferromagnetic state both in the reentrant ($8 \leq x \leq 11$) and in the ferromagnetic regime ($x = 12$) because all these transitions can be observed[10] in a narrow (6 at.% Zr) composition range and the presence of compositional modulation[11] or spatially ordered segregation[12] suggested for *Au*Fe is unlikely. The PM-FM transition has been studied in detail, but the results remain controversial. Below T_c, large (10–50 μm) magnetic domains are observed[13] by use of Lorentz transmission electron microscopy. For the magnetic phase transition, values of the critical exponents ranging from the usual[14] to the unusual[15] were obtained. The temperature dependence of the ac susceptibility was also interpreted as supporting the establishment of a long-range ferromagnetic order[16] or indicating a mictomagnetic behavior.[17] Small-angle neutron-scattering measurements demonstrated[18] that conventional long-range ferromagnetic order was not achieved at any temperature below T_c. The presence of relatively large (≈ 400 Å) spin clusters that do not disorder above T_c was also confirmed.

Mössbauer investigations in moderate applied fields (≈ 3 T) show evidence of magnetic inhomogeneities above and below T_c. In $Fe_{91}Zr_9$, the value of the induced iron hyperfine field was[19] about 9 T at T_c under an applied field of $B_{ext} = 3$ T. Similar data are also reported[20] for $Fe_{92}Zr_8$. The strong applied-field dependence of the average hyperfine field $\langle B_{hf} \rangle$ was attributed[19] to exchange coupled magnetic clusters. At $T = 0.98T_c$, the average cluster moment was also determined using the theory[21] for noninteracting or weakly interacting superparamagnetic particles. It was found[19] that the cluster moment indicates an average cluster size of ≈ 300 atoms corresponding to a cluster dimension of ≈ 15–20 Å, far smaller than the neutron-scattering value[18] of ≈ 100–200 Å.

In these experiments[19,20] a relatively large field (≈ 3 T) applied perpendicularly to the sample surface was required to overcome the demagnetizing field (≈ 0.7 T) and possible domain effects. The magnetic field has a significant influence on the SG state and thus a considerable perturbation of the possibly complicated (e.g., wandering axis[20]) FM state cannot also be ruled out. The aim of the present paper is to investigate whether magnetic fields smaller by more than two orders of magnitude can also influence the magnetic transition. For this reason, Mössbauer-spectroscopy investigations in a small magnetic field ($B_{ext} = 13$ mT generated in the plane of the sample by a small permanent magnet) are reported for melt-spun $Fe_{100-x}Zr_x$ ($7 \leq x \leq 12$) amorphous ribbons in the 4 K $< T <$ 300 K temperature range.

For the evaluation of the spectra the binomial distribution method[22] was used in which the shape of the binomial distributions is adjusted to the spectra and linear correlation is assumed between the hyperfine field, isomer shift, and quadrupole splitting. The effect of the uncorrelated part of the isomer shift and quadrupole splitting distributions was taken into account in the linewidth of the individual six line patterns, which, together with the intensity of the 2–5 lines, was determined by iteration.

Besides the x-ray and Mössbauer-spectroscopy studies magnetization measurements were performed by a vibrating sample magnetometer which confirmed the same magnetic behavior reported in Refs. 10 and 17. PM-SG transition is detected only for $x = 7$, PM-FM-SG transi-

tion (i.e., reentrant behavior) is observed for $x=8$, 9, and 10 while a single PM-FM transition is found for $x=12$. The transition temperatures (T_c, T_f, T_G) are also in good agreement with the literature values.

The Mössbauer data as shown in Fig. 1 give the same T_c values and T_G as the magnetic measurements. The temperature of the transition is obtained from the temperature break of the full linewidth at the half maximum, 2Γ, as determined from the fit of a single Lorentzian to the spectra. In the case of the PM-FM transition, we observe a well-defined, sharp T_c as in Ref. 2. On the other hand, the PM-SG transition is not sharp[1,2] but extends over an appreciable temperature range (~ 50 K). An independently prepared second sample with the same nominal composition has reproduced well this behavior. The relatively large 2Γ values in the paramagnetic state are caused by the isomer shift and quadrupole distributions.

The application of a small magnetic field, $B_{ext}=13$ mT, in the plane of the samples has caused a further, dramatic rounding of the temperature dependence of the width $2\Gamma(T)$ for both the PM-SG and the PM-FM transitions. A considerable increase of 2Γ was found both below and above the transition temperatures in a $\approx \pm 30$ K range. The effect is considerably smaller for the PM-FM transition of the $x=12$ alloy, which does not show the reentrant behavior. The observed rounding is rather surprising since at conventional second order magnetic transitions relaxation effects associated with pretransition fluctuations can only be observed on hyperfine time scales for a very narrow range of temperature.[2] In the only similar experiment[23] on Ni–1 at. %Fe^{57} the increase of 2Γ due to $B_{ext}=15$ mT was observed exclusively in the ($T_c \pm 2$ K) temperature range corresponding to the $|T-T_c|/T_c \approx 3\times 10^{-4}$ value. It is remarkable that in our field-dependent experiments the small field has a considerable effect also well below the transition temperature where its magnitude is certainly negligible compared with the exchange field.

The typical behavior of the Mössbauer spectra in $B_{ext}=0$ and $B_{ext}=13$ mT are show in Fig. 2 for the reentrant $Fe_{92}Zr_8$ ($T_c=167$ K). Below ≈ 130 K ($T/T_c \approx 0.8$) the deduced static hyperfine field distribution $p(B)$ is the same with and without B_{ext}. The only effect of the applied field—between T_f and this temperature—is the increased intensity of the 2–5 lines corresponding to a nearly perfect magnetic polarization alignment in the sample plane. Above this temperature the shape of $p(B)$ has also changed significantly: The probability of the low field part decreased, that of the high field part increased or a new ≈ 12 T component reappeared. This rather large and practically temperature independent hyperfine field can only be induced by a 13 mT external field if B_{ext} influences significantly the magnetic cluster relaxation.

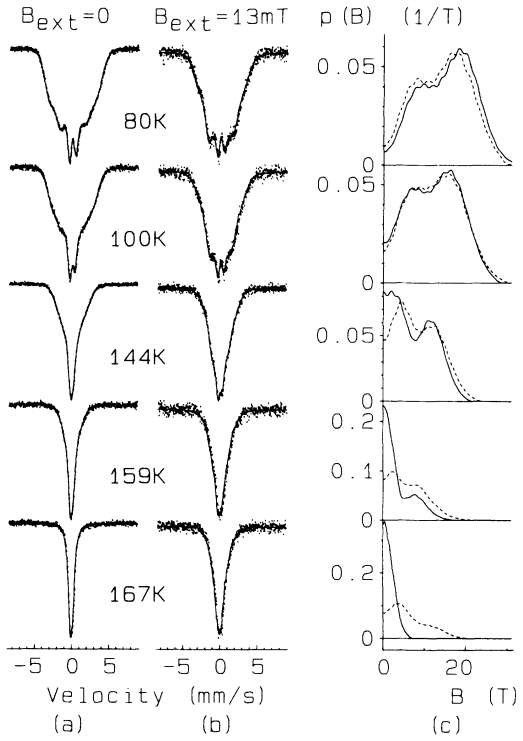

FIG. 2. Typical Mössbauer spectra at different temperatures and static hyperfine distributions of the RSG $Fe_{92}Zr_8$ without [(a) and solid line in (c)] and with applied field [(b) and dashed line in (c)], respectively. Note the change in the scales of $p(B)$.

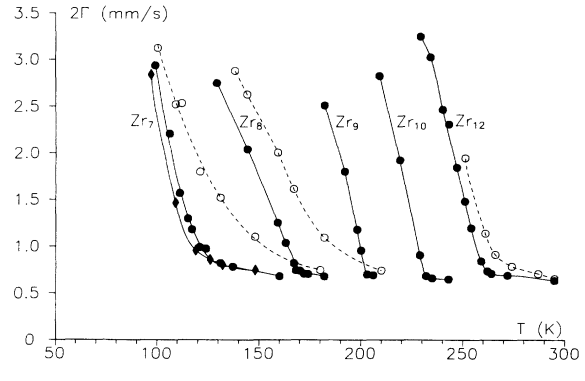

FIG. 1. Temperature dependence of the full linewidth at half maximum, 2Γ, for amorphous $Fe_{100-x}Zr_x$ [$x=7$ (two independent samples), 8, 9, 10, and 12] in zero field (solid symbols) and in $B_{ext}=13$ mT (for $x=7$, 8, and 12: open circles), respectively. The solid and dashed lines are guides to the eye.

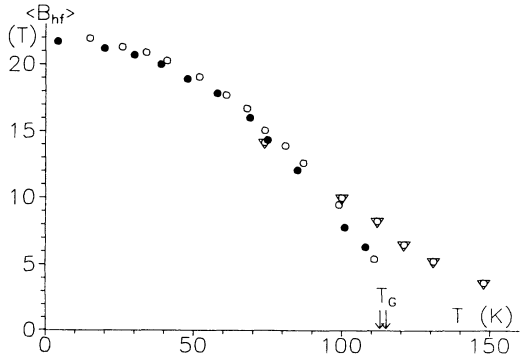

FIG. 3. Temperature dependence of the average hyperfine field of the SG $Fe_{93}Zr_7$ in $B_{ext}=0$ (for two samples: open and solid circles) and in $B_{ext}=13$ mT (open circle in triangle). The arrows show the PM-SG transition temperatures.

A comparison of the static and dynamic evaluation of $B_{ext}=0$ Mössbauer spectra for the same composition suggests[24] similar conclusions. The observed changes extend over the whole $p(B)$ shape, that is, the effect of small magnetic field cannot be localized to specific iron environments. The small field affects the PM-SG transition in a similar manner; the temperature dependence of the average hyperfine field for $Fe_{93}Zr_7$ with and without B_{ext} is shown in Fig. 3. The cluster size, when estimated from the temperature dependence of the average hyperfine field above T_G according to the model outlined in Ref. 21, is found to be approximately 2000 Fe atoms. This value corresponds to a somewhat larger cluster size (≈ 30 Å) than that determined[19] in ≈ 3 T applied field but it is still smaller than the neutron correlation length.[18]

The present experiment proves the existence of magnetic clusters in a wide temperature range near the magnetic transition temperature. In this respect there is no qualitative difference between the paramagnetic–spin-glass and the paramagnetic-ferromagnetic transitions of the reentrant spin glass and the ferromagnetic Fe-Zr systems, but the clustering is weaker for larger Zr content. Our results strongly support the view[18] that these alloys do not exhibit conventional long-range-ordered ferromagnetism below their T_c.

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