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Study of the magnetic viscosity of amorphous Fe–Zr alloys in the spin-glass state

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Abstract. Magnetic viscosity was measured for amorphous $\text{Fe}_{100-x}\text{Zr}_x$ ($7 \leq x \leq 12$) alloys in the spin-glass state as a function of magnetic field ($0 < H < 500$ Oe) and temperature ($4.2 < T < 60$ K). The viscosity field, H_v , deduced from these measurements is independent of H and decreases with increasing temperature as $(1/T)^{2.0 \pm 0.3}$ for all the alloys. The activation volume, V_a , calculated from H_v varies with the temperature as $T^{3.0 \pm 0.3}$. The reduced coercivity, $h_c = H_c/2\pi M_s$ (H_c being the coercivity and M_s the saturation magnetization) as a function of D_a , the characteristic size calculated from the activation volume, falls approximately to a common curve for all the alloys. This dependence decreases with increasing D_a as $(1/D_a)^{2.0 \pm 0.2}$. Such a size dependence of the coercivity hints at a curling-type nucleation mechanism of domains in the spin-glass state of the amorphous Fe-rich Fe–Zr alloys.

1. Introduction

The iron-rich Fe–Zr amorphous alloys have attracted considerable interest because of their anomalous magnetic properties at low temperatures [1]. The $\text{Fe}_{100-x}\text{Zr}_x$ alloys on the iron-rich side can be amorphized in a very limited concentration range ($7 \leq x \leq 12$). The alloy with the critical concentration ($x_c = 7$) shows a paramagnetic (PM) spin-glass (SG) transition at $T_g = 105$ K. The compositions with $7 < x < 10$ display two magnetic transitions: PM to ferromagnetic (FM) at the Curie temperature, T_c , increasing with x from T_g , and FM to SG state at the spin-freezing temperature, T_f , decreasing with x from T_g . The $x = 12$ alloy behaves like a ferromagnet with a single magnetic transition at the Curie temperature, T_c [2, 3].

One of the magnetic anomalies in these systems is the very rapidly increasing magnetic hardness on cooling. The magnetic hardness also increases very rapidly with decreasing Zr content [2] as deduced from coercivity measurements. Irreversible magnetization changes are also observed below T_g or T_f . Although these alloys have long been studied, controversies in the interpretation of the peculiar magnetic properties remain. Some authors use a homogeneous model in which the simultaneous presence of FM and antiferromagnetic (AF) exchange interactions is assumed, causing exchange frustration [4]. In the other group of models, an inhomogeneous magnetic structure is postulated. According to these models, AF [2, 5, 6] or non-magnetic [7] or FM [8] clusters embedded in the FM matrix are responsible for the anomalies. The reason for the assumption of AF exchange interactions in these alloys is the fluctuating Fe–Fe separation in the amorphous state around a value close to a critical Fe–Fe separation ($d_{\text{Fe}} = 2.55 \text{ \AA}$) where the direct exchange integral between

Fe–Fe atoms is known to change sign, strongly decreasing with decreasing Fe–Fe separation [9].

Despite the abundance of magnetic studies for the Fe-rich Fe–Zr amorphous alloys, there is only a single model which attempts to describe quantitatively the sharp temperature and composition dependence of the magnetic hardness. In the early form of the model [2, 5, 6], the presence of AF clusters embedded in a FM matrix was assumed, which could be considered as non-magnetic inclusions below their Néel temperatures. The magnetic hardness was then attributed to pinning to these inclusions, the coercivity being proportional to the volume fraction of the AF clusters. The assumption of AF exchange interactions inside the clusters is essential in this model in order to explain the temperature dependence of coercivity. In a recent revised version of this model [7], the regions having greater Fe density than a critical value (Fe-rich clusters) are assumed to contain Fe atoms in low-spin state with a moment of about $0.5\mu_B$ while the remaining matrix having a lower Fe density is assumed to be FM with the Fe atoms in high spin-state with a moment of about $2.8\mu_B$. The dominant domain wall pinning is then assumed to be caused by the high-density Fe-rich clusters (considered as 'non-magnetic inclusions' because of their low moments) instead of the AF clusters as in the former version. However, in the new version the temperature dependence of coercivity had to be explained by a new concept: the lattice parameter increases with increasing temperature because of thermal expansion, decreasing the size of the Fe-rich clusters. The very rapidly decreasing coercive field by hydrogen doping of Fe-rich Fe–Zr alloys are explained similarly by the introduced lattice distortions [10].

Both versions of the model result in an exponential increase of the coercivity, H_c , with decreasing temperature expressed in the form of $H_c \sim \exp(-\beta T)$, a feature related to the upper tail of the assumed Gaussian distribution for the Fe concentration [2]. The microscopic origin of the temperature coefficient in the exponent, β , is, however, not clear. β is a free parameter which is found to depend on composition. On the other hand, as far as the first version of the model is concerned, there are serious doubts about the existence of AF exchange interactions in the Fe-rich Fe–Zr amorphous alloys [11, 12]. In this paper we propose a new mechanism of the coercivity for the Fe-rich Fe–Zr amorphous alloys on the base of systematic magnetic viscosity measurements.

2. Experimental

The $\text{Fe}_{100-x}\text{Zr}_x$ ($7 \leq x \leq 12$) amorphous alloy ribbons ($1 \text{ mm} \times 12 \mu\text{m}$) prepared by melt-spinning in Ar atmosphere were confirmed to be amorphous by x-ray diffraction and Mössbauer spectroscopy. Up to 20 pieces, each about 4 mm long, weighing about 2.5 mg, were glued together by water-glass for coercivity and magnetic-viscosity measurements. The magnetization was measured by a Foner-type vibrating sample magnetometer (VSM) using a He flow cryostat. The accuracy of the temperature measurement was better than $\pm 0.1 \text{ K}$. The magnetic field was applied in the ribbon plane along the ribbon axis resulting in a maximum demagnetization field of the order of a few Oe. This could be neglected with respect to the much higher fields applied ($25 < H < 500 \text{ Oe}$), therefore corrections for the demagnetization field were not performed.

3. Results and discussion

Magnetic viscosity means the time dependence of magnetization in a constant or zero applied field, a typical feature showed e.g. by spin-glasses. The magnetic viscosity of

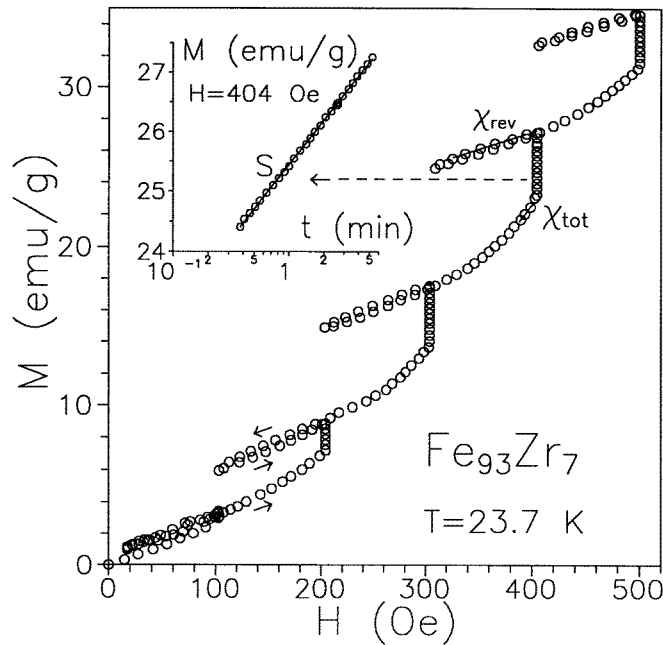


Figure 1. Magnetization as a function of external magnetic field measured with a constant rate of 100 Oe min^{-1} on the virgin curve, stopping at five field values for about 5 min for $\text{Fe}_{93}\text{Zr}_7$ at $T = 23.7$ K. Inset: Magnetization versus time at $H = 404$ Oe. S , χ_{rev} and χ_{tot} are defined as slopes to the respective curves (details in the text).

the $\text{Fe}_{100-x}\text{Zr}_x$ amorphous alloys ($x = 7-9$) was measured on the virgin-curve side of the hysteresis loop after cooling the sample in zero field to the desired temperature from above T_g or T_f . Figure 1 shows the magnetization as a function of applied field for $\text{Fe}_{93}\text{Zr}_7$ at $T = 23.7$ K, taken quasistatically with a constant velocity of 100 Oe min^{-1} and stopping at five field values for about 5 min. At every field value, the magnetization varies as $M(t) = M_0 + S \ln t$ for long enough times, where S is the viscosity constant and M_0 is a time-independent magnetization, as shown by the inset of figure 1 for $H = 404$ Oe. Note that the relative change of the magnetization can exceed even 10% over a time interval of five minutes. When the field is reduced and increased again, the magnetization changes in a quasireversible manner. The magnetization curves in decreasing and increasing the field are not exactly superposed indicating that a small irreversible relaxation also takes place in fields smaller than the measuring field. However, this relaxation can be neglected with respect to that observed in the measuring field. This way the total (χ_{tot}) and reversible (χ_{rev}) susceptibilities were determined from the respective slopes of the $M-H$ curve shown in figure 1. Similar measurements were performed for the whole Fe-Zr amorphous alloy series at several temperatures below T_g or T_f , giving similar logarithmic variation of the magnetization with time for every composition, temperature and field.

The viscosity constant, S , was found to be proportional to the irreversible susceptibility (χ_{irr}), calculated from $\chi_{\text{irr}} = \chi_{\text{tot}} - \chi_{\text{rev}}$, for every composition in the whole temperature and field range used. This proportionality is illustrated in figure 2 for $\text{Fe}_{93}\text{Zr}_7$ at 23.7 K as a function of magnetic field. Thus, S can be expressed in the well known form [13] as $S = H_v \chi_{\text{irr}}$, where H_v is the temperature and composition dependent viscosity field which is, however, found to be independent of the magnetic field. This is shown in the inset

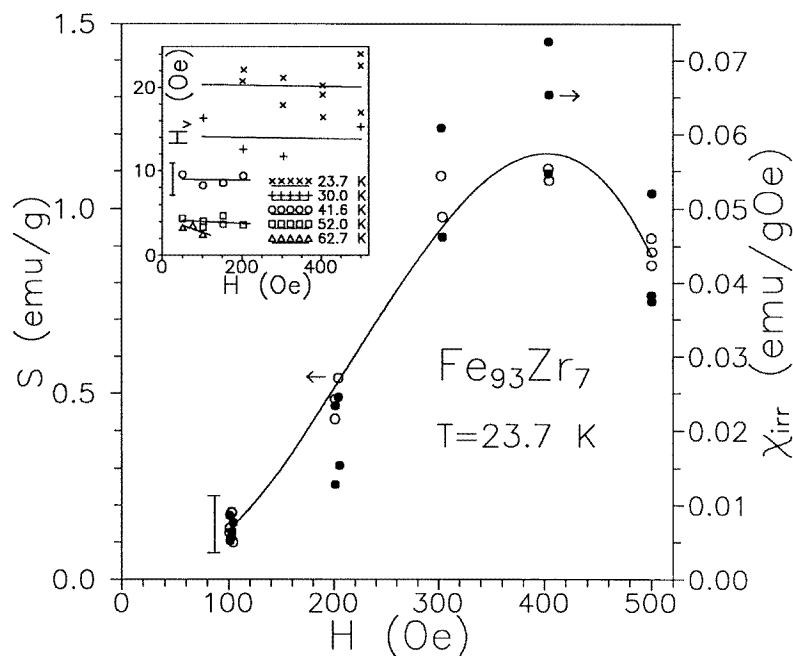


Figure 2. Viscosity parameter (circles) and irreversible susceptibility (dots) as a function of external magnetic field for $\text{Fe}_{93}\text{Zr}_7$ at $T = 23.7$ K. Inset: Viscosity field versus magnetic field for some temperatures indicated. Full lines are guides to the eye.

of figure 2 for $\text{Fe}_{93}\text{Zr}_7$ in the whole temperature range investigated. Due to the limited accuracy of the measurements, noise and scatter is a serious problem. In order to establish a definite trend, the measurements were repeated several times for certain compositions at certain temperatures which is the reason of having several points for one temperature and one applied field in figure 2.

During an irreversible magnetization change with time at a given field, H_0 , in a magnetic viscosity measurement, the thermal energy acting at the magnetization in a confined volume can be expressed by an effective field, $H_{\text{eff}} = H_0 + \Delta H$, which would have the same effect on the magnetization [14]. Here ΔH is associated with field fluctuations around H_0 and can be related to the former introduced viscosity field, H_v , as $\Delta H = H_v \ln(1 + t/\tau_0)$ where τ_0 is the timescale of thermal fluctuations of the order of 10^{-11} s. For long times ($t \gg \tau_0$), $\Delta H = H_v \ln t$ holds. The confined volume called the activation volume, V_a , corresponds to the region where the magnetization change is initiated by thermal fluctuations. Accordingly, V_a can be calculated as $V_a = kT/M_s H_v$ where k is the Boltzmann constant and M_s is the saturation magnetization. If the magnetization change is initiated by nucleation, then V_a is identical with the nucleus volume and is independent of the external magnetic field. In this case, H_v is also independent of the prehistory of the magnet and the external field. On the other hand, if the magnetic hardness is caused by domain wall pinning, V_a and H_v should not be independent of the magnetic field [15].

The fact that for the Fe-rich Fe–Zr amorphous alloys, H_v is found to be a temperature-dependent material parameter, independent of the magnetic field used (figure 2), suggests that the magnetization changes during isothermal ageing below T_g or T_f by nucleation rather than by domain wall pinning. Figure 3 shows the temperature dependence of H_v ,

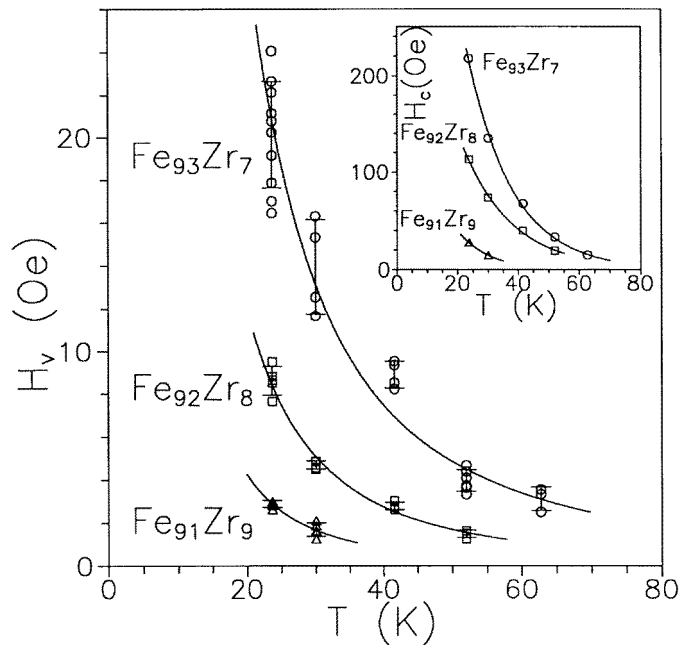


Figure 3. Viscosity field as a function of temperature for $Fe_{93}Zr_7$, $Fe_{92}Zr_8$ and $Fe_{91}Zr_9$. Full curves are fitting curves of the form $H_v \sim (1/T)^n$ where $n = 1.96, 2.14$ and 2.29 , respectively. Inset: Coercivity versus temperature for $Fe_{93}Zr_7$, $Fe_{92}Zr_8$ and $Fe_{91}Zr_9$. Full curves are exponential fitting curves used in [2].

which is found to vary for all the three alloys investigated as $(1/T)^{2.0 \pm 0.3}$ to a rather good approximation. The temperature dependence of the coercivity, H_c , shown in the inset of figure 3 for the three alloys, is similar to that of H_v . The limited temperature range available does not permit a unique decision between the exponential decay of H_c with temperature observed in [2] and an inverse power-law behaviour but the former description seems to give better results.

Since the viscosity field, H_v , is inversely proportional to the activation volume, V_a , the coercivity of the Fe-rich Fe-Zr amorphous alloys is correlated with the activation volume, i.e. the size of a nucleus reversing magnetization. To obtain this correlation quantitatively, the temperature dependence of V_a , calculated from H_v , has to be determined. In calculating V_a , the slight temperature dependence of M_s is taken into account on the basis of the temperature dependence of the hyperfine field for these alloys, being proportional to the saturation moment [16]. As shown in figure 4, V_a is found to increase as $T^{3.0 \pm 0.3}$ with increasing temperature for each alloy to a good approximation. The characteristic size of a nucleus can be well characterized by the diameter, D_a . It is found to increase linearly (as $T^{1.0 \pm 0.1}$) with increasing temperature for each alloy to a rather good approximation, as shown in the inset of figure 4. The composition dependence of D_a , increasing with the Zr concentration for a given temperature below T_g or T_f follows the same trend as the size of the magnetic clusters introduced for describing the superparamagnetic-type behaviour of the magnetization versus field curves above T_c for the same alloys [17]. It is suggested that the magnetic clusters act as activation volumes, i.e. nuclei reversing magnetization in the Fe-rich Fe-Zr amorphous alloys. Thus both the magnetization [17] and the magnetic viscosity measurements indicate, independently of each other and in rather different temperature and

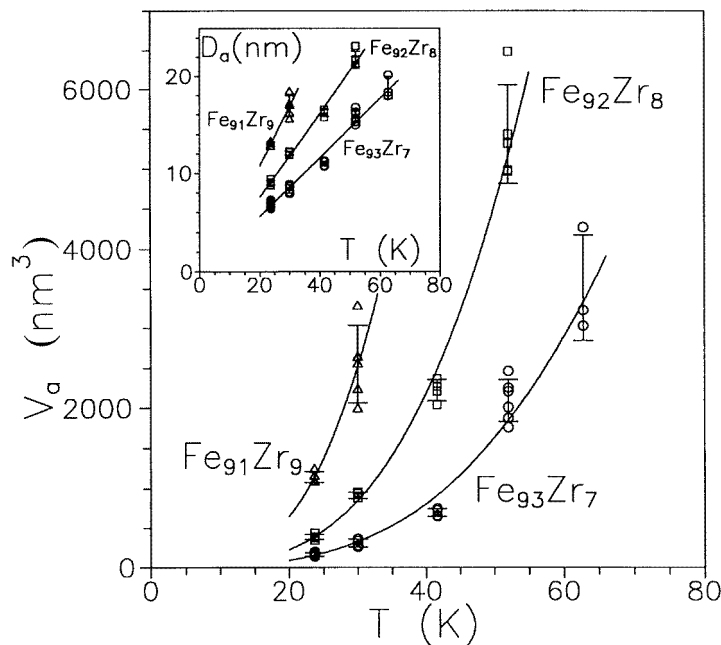


Figure 4. Activation volume as a function of temperature for $\text{Fe}_{93}\text{Zr}_7$, $\text{Fe}_{92}\text{Zr}_8$ and $\text{Fe}_{91}\text{Zr}_9$. Full curves are fitting curves of the form $V_a \sim T^n$ where $n = 3.15, 3.28$ and 3.35 , respectively. Inset: Diameter of activation volume (nucleus size) versus temperature for $\text{Fe}_{93}\text{Zr}_7$, $\text{Fe}_{92}\text{Zr}_8$ and $\text{Fe}_{91}\text{Zr}_9$. Full lines are fitting curves of the form $D_a \sim T^n$ where $n = 1.05, 1.09$ and 1.12 , respectively.

magnetic field ranges, that the magnetic structure of these alloys is inhomogeneous.

In the theory of fine particles, the mechanism of the magnetization reversal can be judged in special cases from the dependence of the coercivity on the characteristic size of the granular system. Curling is known to be a reversal mechanism with a marked size dependence of the coercivity, in contrast to homogeneous rotation and fanning where the coercivity is size-independent [18]. Since the coercivity, H_c , is found to depend on the diameter of the activation volume for the Fe-rich Fe–Zr amorphous alloys, curling is suspected as the coercivity mechanism. Therefore, a reduced coercivity, $h_c = H_c/2\pi M_s$, used in the theory of curling, is plotted for the three alloys studied as a function of D_a in figure 5. The reduced coercivity, h_c , is found to decrease as $(1/D_a)^{2.0 \pm 0.2}$ with increasing nucleus size to a rather good approximation, which is exactly the size dependence theoretically expected for curling. The h_c – D_a curves for $\text{Fe}_{93}\text{Zr}_7$ and $\text{Fe}_{92}\text{Zr}_8$ practically coincide, showing that h_c is determined dominantly by the nucleus size (independently of composition and temperature). The h_c – D_a curve for $\text{Fe}_{91}\text{Zr}_9$ also lies near the master curve but does not fall exactly on it, probably because of the largest systematic error in the magnetic quantities for this composition.

The curling mode is known to occur for D_a higher than a certain value depending on the cluster shape and anisotropy [18]. Below this value, homogeneous rotation takes place. By considering the temperature dependence of D_a (inset of figure 4), the curling reversal mechanism cannot hold at low temperature. In case of the Fe-rich Fe–Zr amorphous alloys, this limit does not seem to be reached. However, Hiroyoshi *et al* reported on the temperature dependence of coercive field for an Fe–Hf amorphous alloy (behaving very similarly to the

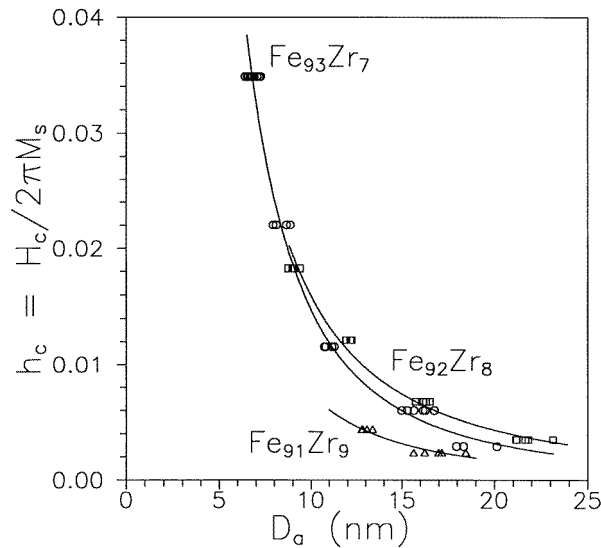


Figure 5. Reduced coercivity as a function of nucleus size for $\text{Fe}_{93}\text{Zr}_7$, $\text{Fe}_{92}\text{Zr}_8$ and $\text{Fe}_{91}\text{Zr}_9$. Full curves are fitting curves of the form $h_c \sim (1/D_a)^n$ where $n = 2.22$, 1.88 and 2.13 , respectively.

Fe–Zr amorphous alloys) which shows coercive fields approaching constant value as $T \rightarrow 0$ [19]. This change of behaviour from a strong to a slight temperature variation of the coercive field on decreasing temperature may reflect the change of the reversal mechanism discussed above.

4. Conclusion

On the basis of systematic magnetic viscosity measurements we suggest that the coercivity is dominantly determined by the size of magnetic inhomogeneities (clusters present in the alloys) via a curling-type reversal mechanism for the $\text{Fe}_{100-x}\text{Zr}_x$ ($7 \leq x \leq 12$) amorphous alloys below the spin-freezing temperature. The smaller the magnetic cluster, the more energy has to be supplied by the external magnetic field in order to rotate the spins inside the cluster from their parallel positions during magnetization reversal, explaining the very rapid increase of the coercivity both with decreasing temperature and increasing Fe concentration. The underlying physics for the existence of such magnetic inhomogeneities in these alloys and the observed temperature and composition dependence of their size (inset of figure 4) is a delicate question. The magnetic inhomogeneities could be associated with regions containing iron atoms in high-spin state, surrounded by regions with iron atoms in low-spin state. The shape anisotropy of the magnetic clusters is thought to play an important role in the spin freezing [17, 20].

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