Magnetization and AC-Susceptibility of Fe-Zr Amorphous Alloys

Magnetization and AC-Susceptibility of Fe-Zr Amorphous Alloys*

Naoaki Saito**, Kazuaki Fukamichi*** and Yasuaki Nakagawa

Institute for Materials Research.

(Received January 25, 1990)

Synopsis

Measurements of magnetization and ac-susceptibility of Fe-Zr amorphous alloys over the wide composition range have been carried out. The spin glass like behavior appears in two different regions of composition. The spin glass behavior in Fe-rich Fe amorphous alloys (around 90 at.% Zr) is due to the frustration of antiferromagnetic coupling between spins. The other behavior around 50 at.% is due to the dilution of atoms with magnetic moment. They are explained by the local environment effect.

I. Introduction

Fe-Zr amorphous alloys have been investigated by many authors because these alloys are typical Fe - metallic glasses with dense random packing structure[1] and show interesting magnetic properties.[2]

The concentration dependence of the Curie temperature T_C of Fe-Zr amorphous alloys indicates a maximum around 20 at.% Zr. It should be noted that T_C rapidly decreases, a magnetic freezing state appears at low temperature, and a ferromagnetic order tends to disappear with decreasing Zr concentration in the Fe-rich side.

We have deduced the reentrant spin glass behavior of Fe-rich Fe-Zr amorphous alloys from the measurements of magnetization in both low and high magnetic fields,[3] magnetic viscosity,[4] ac-susceptibility[4,5]

---

* The 1847th report of Institute for Materials Research.

** Present Address : Division of Energy Foundation, Electrotechnical Laboratory, Tsukuba 305.

*** Department of Materials Science, Faculty of Engineering, Tohoku University, Sendai, 980.

and Mössbauer spectrum. Moreover, we have found that 7.6 at.% Zr alloy, the most Fe-rich alloy we have obtained, shows a simple spin glass behavior at low magnetic field; the ferromagnetic percolation limit situates around 8 at.% Zr. The alloys containing less than 8 at.% Zr, including the pure Fe amorphous alloy, would exhibit the spin glass behavior. On the other hand, in the high concentration of Zr, T_C decreases toward 50 at.% Zr with increasing Zr concentration, and the spin glass state appears near this region.

The Fe-Zr amorphous alloys have not been studied in detail except in Fe-rich region. In order to clarify the magnetic behavior over the entire concentration range, we have prepared the Fe-Zr amorphous alloys in the wide concentration range and made systematic measurements of magnetization and ac-susceptibility.

II. Experimental Methods

Fe-Zr amorphous alloys with 7.6 - 10.5 at.% Zr were prepared in argon atmosphere by melt-spinning and the alloys with 14.1 - 57.2 at.% Zr were obtained by dc-sputtering. The alloy compositions were confirmed by chemical analysis.

Magnetization measurements were performed by using a vibrating sample magnetometer in high field up to 143 kOe produced by a Bitter magnet.

The ac-susceptibility was measured in the ac-field with a frequency of 85 Hz and an amplitude of 0.1 Oe by a mutual inductance Hartshorn method. The measurement was performed during heating after zero field cooling on the condition that the earth's field was shielded by a Permalloy cylinder.

III. Experimental Results and Discussion

Figure 1 shows the magnetization process up to 143 kOe for Fe-Zr amorphous alloys at 4.2 K after cooling in zero field. The magnetization value at high field decreases monotonically with increasing Zr concentration. The magnetization of 7.6 at.% Zr alloy increases with increasing field even in high field and shows an unusual hysteresis loop. We called it the high-field hysteresis in our previous paper,[3)] which is characteristic of the spin glass behavior of Fe-rich Fe-Zr amorphous alloy.

Magnetization process for the alloys with intermediate Zr con-

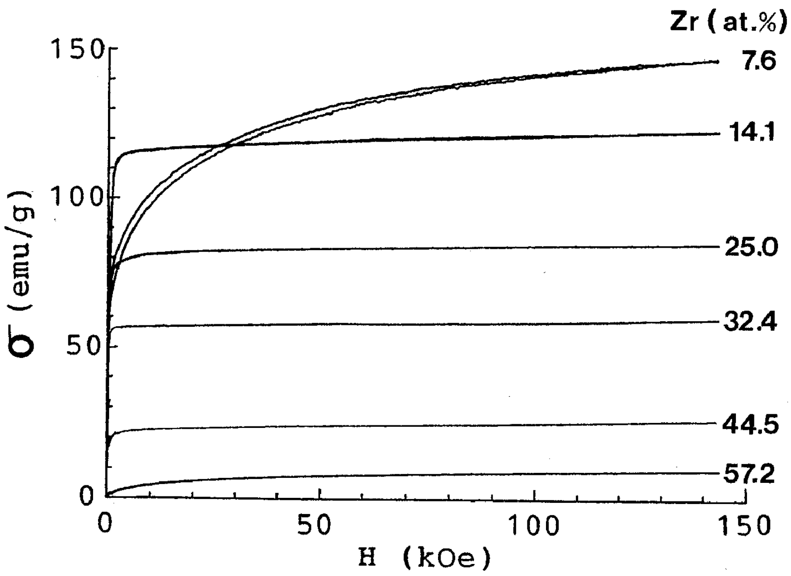

Figure 1 High field magnetization of Fe-Zr amorphous alloys at 4.2 K.

centration looks like a usual soft ferromagnetic alloy without hysteresis loop. No spontaneous magnetization is observed for 57.2 at.% Zr at 4.2 K. The magnetization process is often analyzed by the law of approach to saturation magnetization:

$$\sigma = \sigma_S (1 - a/H - b/H^2) + \chi_{HF} H ,$$

where σ_S is the saturation magnetization and χ_{HF} the high-field susceptibility. The terms of a/H and b/H^2 are concerned with the local and uniform anisotropy, respectively. It is considered that uniform anisotropy is absent in these alloys because these have a random structure. Therefore, we tried to determine parameters σ_S , a and χ_{HF} without b-term. Figure 2 shows the concentration dependence of the parameters. With decreasing Zr concentration, σ_S increases rapidly from around 60 at.% Zr and decreases slowly through a maximum around 15 at.% Zr. This decrease of σ_S in the Fe-rich region agrees with our recent results obtained by Mössbauer spectroscopy, being consistent with the local environment effect described later. In Fe-rich region, both χ_{HF} and a-term rapidly increase with decreasing Zr content; these behaviors are connected with the spin glass behavior of Fe-rich Fe-Zr amorphous alloys.

In the entire concentration range, the high-field susceptibility of these alloys is much larger than that of usual ferromagnets such as Fe or Co, suggesting that there are some small clusters with the spin glass behavior even in the alloys with intermediate Zr concentration.

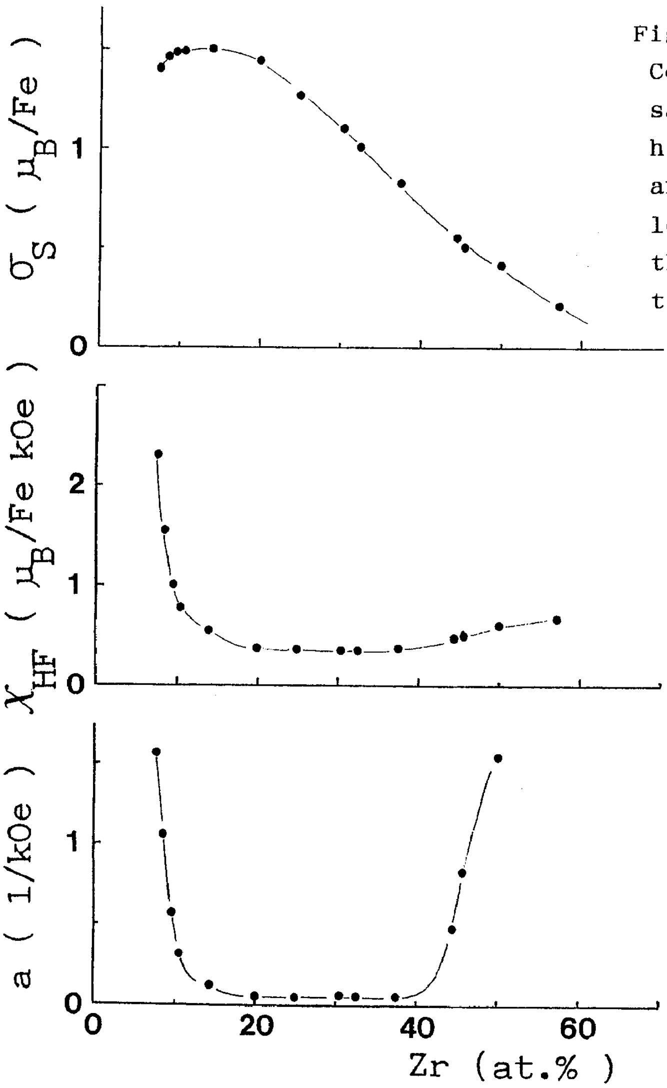

Figure 2
Concentration dependence of saturation magnetization σ_S, high-field susceptibility χ_{HF}, and a parameter a related with local anisotropy obtained by the law of approach to saturation magnetization.

The ac-susceptibility of the Fe-rich alloys exhibits a typical spin glass behavior, as reported in the previous papers.[4,5] Figure 3 shows the temperature dependence of ac-susceptibility for the alloys with 14.1 - 37.6 at.% Zr. The drastic feature at high temperature side is due to the ferromagnetic - paramagnetic transition. The Curie temperature T_C can be determined from the inflection point of these curves. This value agrees with that evaluated by analysis of magnetization curves using the Arrott-Noakes plot.

The fall off of ac-susceptibility below 4 K is observed for 14.1 at.% Zr. This is attributed to a reentrant spin glass behavior of Fe-rich Fe-Zr amorphous alloy. On the other hand, no reentrant behavior is observed for the alloys with 20.0 - 32.4 at.% Zr; these are ferromagnetic down to 1.5 K. It should be mentioned, however, that the temperature dependence of ac-susceptibility of usual ferromagnets is

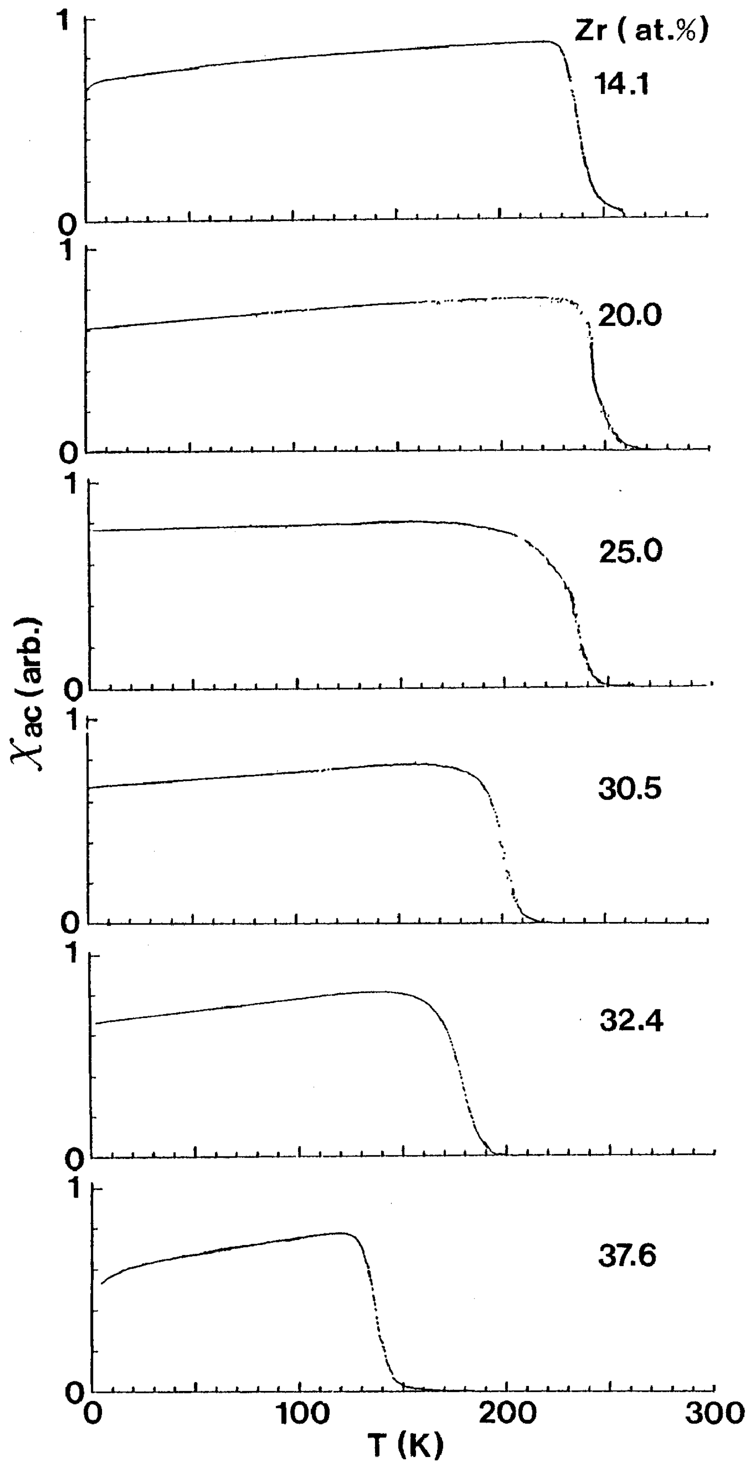

Figure 3 Temperature dependence of ac-susceptibility of Fe-Zr amorphous alloys with 14.1, 20.0, 25.0, 30.5, 32.4 and 37.6 at.% Zr.

quite flat, owing to the demagnetization effect. However, we see the ac-susceptibility slightly decreases with decreasing temperature in the ferromagnetic region; this may be explained by the existence of the clusters mentioned above.

Figure 4 shows the temperature dependence of ac-susceptibility for alloys with 44.5 - 57.2 at.% Zr. The strong fall off of ac-susceptibility at low temperature is observed in the alloys with 44.5 and 45.5 at.% Zr, suggesting that a magnetic freezing state appears. The freezing temperature T_F is defined as the inflection point at a lower temperature. With increasing Zr concentration, the ferromagnetic region becomes narrow, then only one peak is observed for 49.8 and 57.2 at.% Zr. The value of ac-susceptibility of 49.8 at.% Zr has the same order of magnitude as that of 44.5 or 45.5 at.% Zr, but that of 57.2 at.% Zr is much smaller than these values. Therefore, it is

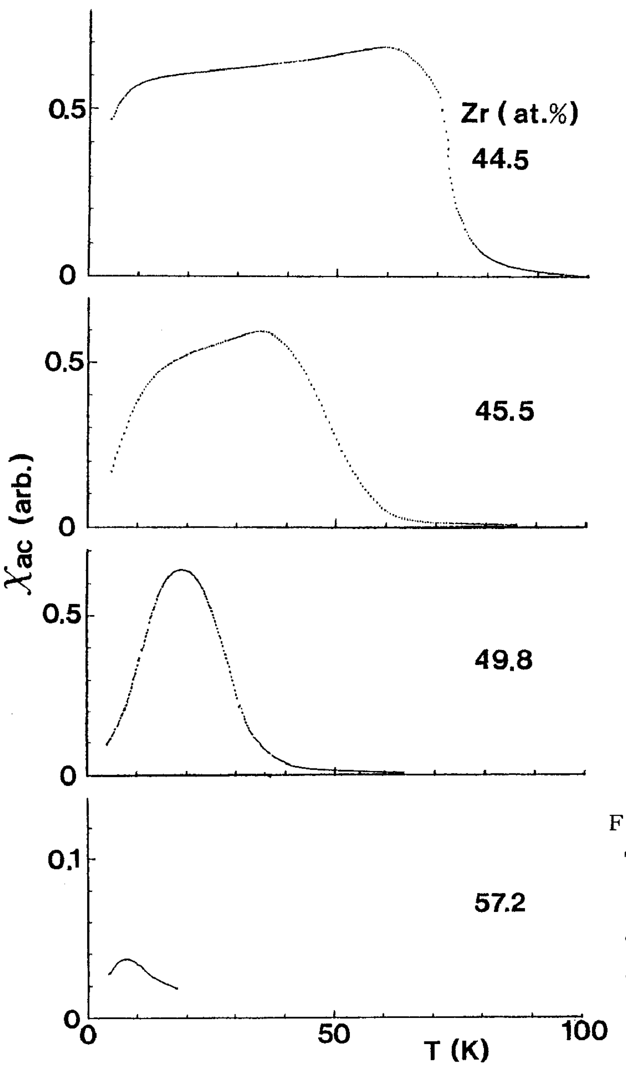

Figure 4
Temperature dependence of ac-susceptibility of Fe-Zr amorphous alloys with 44.5, 45.5, 49.8 and 57.2 at% Zr.

considered that the peak of the curve of 49.8 at.% Zr is attributed to a reentrant transition near the percolation concentration and that of 57.2 at.% Zr is due to a simple spin glass transition.

Figure 5 shows the magnetic phase diagram of Fe-Zr amorphous alloys. In the Fe-rich region, T_C drastically decrease with decreasing Zr concentration and the magnetic freezing state appears at low temperature. The alloys with 8.6 - 10.5 at.% Zr show the reentrant spin glass behavior. The ferromagnetic percolation limit situates around 8 at.% Zr. The simple spin glass behavior is observed in 7.6 at.% Zr. It is expected that also alloys containing less than this concentration show the spin glass behavior.

From the theoretical point of view, these magnetic behaviors are interpreted by the local environment effect. It is pointed out by Y. Kakehashi[6] that the Fe local moment has small amplitude and antiferromagnetically couples to the neighboring Fe local moments in the environment with more than 10 nearest neighboring Fe. The spin glass in Fe-rich Fe-Zr amorphous alloys is considered to be due to the frustration of antiferromagnetic coupling with random structure. We at-

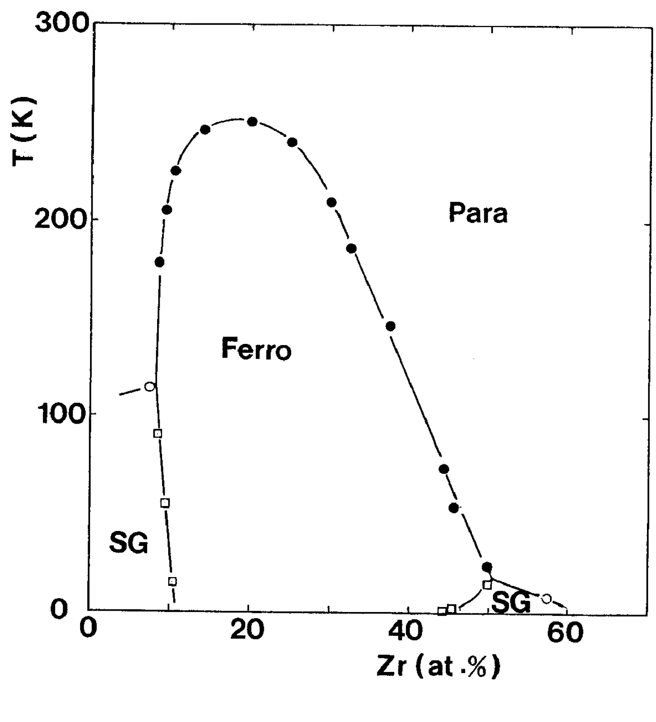

Figure 5 Magnetic phase diagram of Fe-Zr amorphous alloy.
● : the Curie temperature;
○ : freezing temperature (simple spin glass);
□ : freezing temperature (reentrant spin glass).

tribute the reentrant behavior in Fe-rich alloys to the coexistence of ferromagnetic region and frustrated region led by the statistical concentration fluctuation.

The reentrant behavior appears for the alloys with 44.5, 45.5 and 49.8 at.% Zr. The ferromagnetic percolation limit in this case situates around 50 at.% Zr. The spin glass one appears for the alloy with 57.2 at.% Zr. Frequently the spin glass is observed in dilute magnetic alloys. It is due to the indirect interaction between isolated magnetic atoms. By increasing the concentration of magnetic atom, magnetic atoms become to contact with each other and the direct interaction becomes dominant. Then one arrives at the critical concentration for ferromagnetic or antiferromagnetic order. Often the reentrant behavior appears near this critical concentration. The value of percolation limit is sensitively dependent on the nearest neighbor coordination number. It is estimated to be 10 - 20 at.% magnetic atom for the amorphous alloy. But the value for the Fe-Zr amorphous alloys is 50 at.% Fe, being much higher than usual. This is due to the fact that the concentration dependence of average moment of Fe shown in fig 2 is much steeper than that of simple dilution. Similar results were found in other Fe-based amorphous alloy systems such as Fe-B. It is estimated that the Fe atom having nearest neighbor Fe atoms of less than half of coordination number looses its local moment for the Fe-Zr amorphous alloys. The spin glass behavior of 50 at.% Fe alloy is attributed to the diminishing of atoms with local moment.

Fe-Zr amorphous alloys with intermediate Zr concentration seem ferromagnetic, but they show much larger high-field susceptibility as compared with usual ferromagnets and exhibit a slight decrease in ac-susceptibility with decreasing temperature in the ferromagnetic region. According to the model mentioned above, we attribute these behaviors to the existence of small clusters of frustrated spin glass or diluted spin glass.

IV. Summary

Measurements of high field magnetization and ac-susceptibility have been performed on Fe-Zr amorphous alloys in the concentration range from 7.6 to 57.2 at.% Zr. The saturation moment per Fe atom shows nonlinear dependence on concentration. It is considered to be due to the local environment effect which leads the nonlinear magnetic coupling between Fe local moments. Two kinds of spin glass behavior appear in Fe-Zr amorphous alloys. It is considered that the spin

glass behavior in Fe-rich alloy is due to the frustration of antiferromagnetic interaction. The other behavior around 50 at.% Zr is due to the dilution of atoms with local moment.

Acknowledgment

The authors are indebted to Dr. H. Hiroyoshi for helpful discussions and to staff members of High Field Laboratory for Superconducting Materials, Tohoku University, for the operation of the Bitter magnet.

References

1) Y. Waseda. Prog. Mater. Sci. 26 (1981) 1.
2) H. Hiroyoshi and K. Fukamichi. Phys. Lett. 85A (1981) 282.
3) H. Hiroyoshi, N. Saito, K. Fukamichi and Y. Nakagawa. Sci. Rep. RITU A33 (1986) 58.
4) N. Saito, K. Fukamichi and Y. Nakagawa. Sci. Rep. RITU A34 (1989) 235.
5) N. Saito, H. Hiroyoshi, K. Fukamichi and Y. Nakagawa. J. Phys. F 16 (1986) 911.
6) Y. Kakehashi. J. Phys. Colloque 49 (1988) C8-1079.