Magnetic properties of W-Fe alloys

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A recently discovered spin-glass interaction in W-Mn alloys suggested the investigation of W-Fe alloys. Indeed, similarly to the Mn alloys, the crystalline Fe alloys display a spin-glass transition with a spin-freezing temperature which increases essentially linearly with Fe content up to 31 at.% Fe, where the alloys become amorphous and antiferromagnetic. Above 52 at.% the amorphous alloys become ferromagnetic and above 68 at.% phases of the alloy separate with the appearance of crystalline WFe_2 and Fe in an amorphous matrix.

It was recently reported that although Mn has a local moment in amorphous (*a*) Mn-Zr (Ref. 1) and crystalline Mn-Ti (Ref. 2), these alloys do not exhibit a spin-glass interaction. A study of ternary *a*-(Cu-Zr-Mn) alloys established[2] that the absence of spin-glass interaction is caused by the mixing of Mn *d* states with the partially filled Zr or Ti *d* bands. This implies that while in Cu-Mn, Ag-Mn, etc., there exists a distribution of exchange wide enough and strong enough to lead to spin freezing into a random configuration, the distribution may be wide enough in Mn-Zr and Mn-Ti alloys but the overall exchange is too weak as a result of the *d*-band mixing to induce ordering. This model led to the discovery of a new spin-glass system; W-Mn, where the spin-glass transition can exist despite the partially filled tungsten *d* band because the density of *d* states at the Fermi level is minimal.[2] This result prompted the present investigation of W-Fe alloys.

All the W-Fe films were sputtered from a cathode made by wrapping various numbers (from 4 to 54) of Fe wires 0.025 cm in diameter around a tungsten rod. The films were deposited on sapphire at 260 K with a sputtering power of 15 W (1500 V, 10 mA). The structure of the films was established by x-ray diffraction and their composition was determined by x-ray fluorescence analysis calibrated by atomic absorption analysis. The deposited films were then scraped with a sapphire slide to avoid magnetic contamination. The susceptibility of the resulting flakes was measured in a high dc magnetic field (12.8 kOe) using the Faraday method, in a low dc field (3 Oe) susceptometer using a superconducting quantum interference device (SQUID), and in low ac field (4 Oe) at 10 kHz.

Since the magnetic properties of W-Mn alloys were similar to those of Cu-Mn alloys (Ref. 2) and since both Cu-Mn (Ref. 3) and Cu-Fe (Ref. 4) alloys are archetypical spin-glasses, it is not surprising that, as shown in Fig. 1, W-Fe alloys display a spin-glass transition. Because of the similarity between W-Fe and W-Mn alloys, this study will deal briefly with the spin-glass properties of W-Fe alloys and stress only the different magnetic properties of the two systems.

The W-Fe alloys with Fe content up to 31 at.% are, similarly to the W-Mn alloys,[2] a crystalline solid solution of Fe in W with a (110) preferred orientation with respect to the substrate. Using the single diffraction peak of the films the susceptibility of which is described in Fig. 1, as an interpolation between the W and Fe(110) peaks yields an Fe concentration of 13.3 at.%, which is good agreement with the 14.2 at.% obtained by atomic absorption analysis. As shown in Fig. 1, the susceptibility (χ) peaks as a function of magnetic field for W—14.2 at.% Fe are quite similar to those shown in Fig. 3 of Ref. 2 for W—12.8 at.% Mn. Consequently, one can conclude from

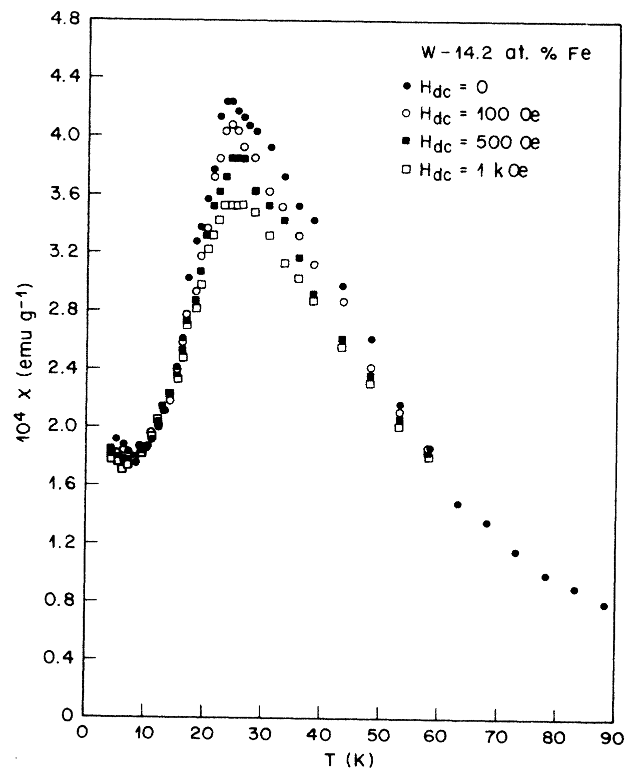

FIG. 1. Temperature dependence of the ac susceptibility as a function of applied dc magnetic field.

TABLE I. Magnetic properties of two representative W-Fe alloys.

Fe content (at. %)	p_{eff} (μ_B)	$-\theta$ (K)	χ_0 (10^{-6} emu/mole)	$\chi_{300\,K}$ (10^{-6} emu/mole)	ΔT^a (K)
14.2[b]	4.47	−55	34	1481	150–300
	4.75	−46	−101		100–300
	5.34	−30	−434		70–300
36.4[c]	2.96	16	380	1680	30–300

[a]Temperature range of Curie-Weiss fit.
[b]Crystalline alloy with spin-glass transition is shown in Fig. 1.
[c]Amorphous alloy with no spin-glass transition.

these data that such W-Fe alloys undergo a spin-glass transition. Furthermore, measurements in a SQUID susceptometer show the same irreversibility when cooled in a field of 3 Oe and time dependence of the magnetization below the freezing temperature (T_{SG}) which was observed[2] in the W-Mn alloys. The high-field (12.8 kOe) properties of the alloy discussed in Fig. 1 are listed in Table I. It is clear that these results are essentially similar to those for W-Mn alloys: the best fit ($\Delta T = 150$–300 K) which yields a positive value for the temperature-independent contribution χ_0 results in a value of p_{eff} appreciably larger than the critical value required[2] for a spin-glass interaction ($\simeq 3.9\mu_B$) and in a small value for χ_0. The large positive value of the Curie-Weiss temperature (θ) is however in better agreement with Cu-Mn alloys than with W-Mn alloys[2] which are characterized by $\theta \simeq 0$. The concentration dependence of T_{SG} is shown in Fig. 2 and although the T_{SG} values for Fe are approximately twice as high as for Mn, dT_{SG}/dC_{Fe} is again $\simeq 1$. The scatter of the T_{SG} values of Fig. 2 and the smearing of the peak in Fig. 1 are undoubtedly due to the fact that the films are sputtered from fairly inhomogeneous cathodes, which results in a composition spread of about $\pm 10\%$ from the average composition quoted for each film.

Despite the fact that W-Fe spin-glasses are characterized by an average ferromagnetic interaction (large positive θ), similarly to W-Mn alloys, they become essentially antiferromagnetic when the iron concentration exceeds 31 at. %. Simultaneously, the W-Fe films become amor-

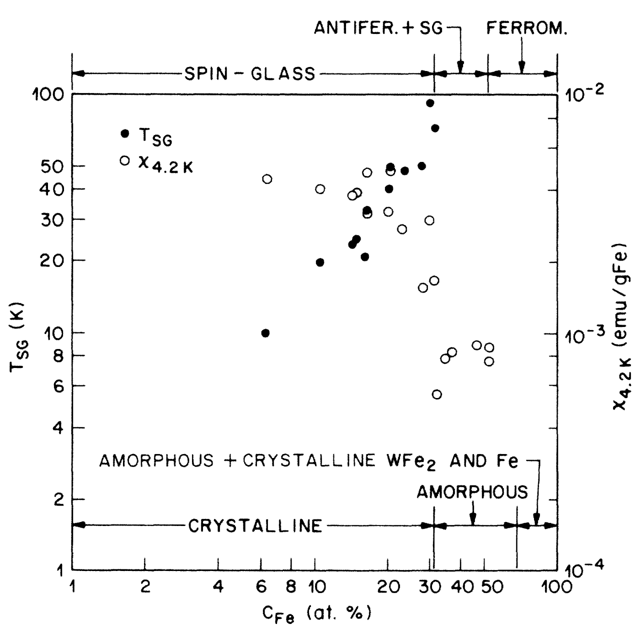

FIG. 2. Dependence of the spin-glass freezing temperature (T_{SG}) and of the susceptibility per gram of Fe at 4.2 K on Fe concentration.

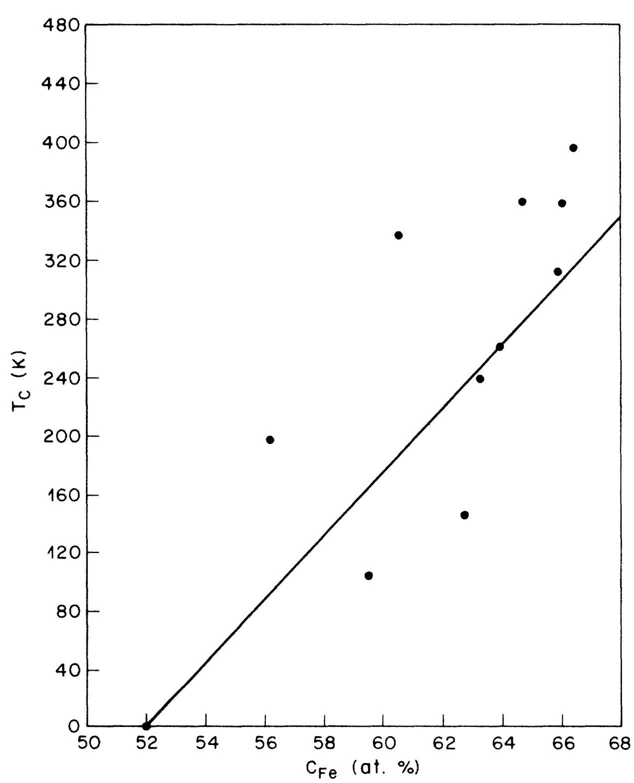

FIG. 3. Dependence of the Curie temperature (T_c) of amorphous W-Fe alloys on Fe concentration. The solid line is described in the text.

phous but as previously discussed for ternary Cu-Zr-Mn (Ref. 2) and Au-Si-Mn (Ref. 5) alloys the disappearance of the spin-glass transition is most probably not linked with the disappearance of crystallinity. The transition from the spin-glass state to antiferromagnetism is evidenced in both θ and χ. Indeed, as shown in Table I, the 36.4 at. % Fe alloy is characterized by a value of p_{eff} smaller than the critical value required for a spin-glass interaction ($\simeq 3.9\mu_B$), θ has become negative and χ_0 has a large positive value similar to that measured on Cu-Zr-Mn (Ref. 2) and Mn-Zr alloys (Ref. 1), which do not exhibit a spin-glass transition. Furthermore, one also observes a sharp decrease of the normalized susceptibility (Fig. 2) in the vicinity of the critical Fe concentration ($\simeq$30 at. %). As indicated in Fig. 2, some of the alloys in the 31–52 at. % Fe-concentration range exhibit a small susceptibility peak which is indicative of a weak spin-glass interaction superimposed on mostly antiferromagnetic interactions.

Finally, above 52 at. % Fe the amorphous W-Fe alloys become ferromagnetic. The Curie temperatures (T_c) for these alloys are plotted in Fig. 3 as a function of Fe concentration. The solid line in Fig. 3 was drawn through the point (52 at. % Fe; $T_c = 0$) since as shown in Fig. 2 this was the highest Fe concentration for antiferromagnetic alloys and with a slope such that it would intercept the ordinate axis for 100% Fe at the T_c of crystalline Fe (1043 K). Despite the scatter, this line represents a reasonable average of the data if one realizes that the worst deviations are still within the previously mentioned $\pm 10\%$ spread in composition. Furthermore, this compositional spread produces a spread in T_c, which in turn causes appreciable errors in the determination of the representative average T_c for a given film. At any rate, the T_c values shown in Fig. 3 are representative of amorphous W-Fe alloys up to 68 at. % Fe. Above this concentration, the alloys phase separate into a mixture of crystalline WFe_2 and Fe inside an amorphous W-Fe matrix. These crystalline phases are in agreement with those expected from the equilibrium phase diagram for the crystalline W-Fe system.[6]

In conclusion, W-Fe alloys with $C_{\text{Fe}} \lesssim 31$ at. % display very similar spin-glass properties to those of W-Mn alloys. Surprisingly, despite the average ferromagnetic interaction present in these alloys they become antiferromagnetic for $C_{\text{Fe}} \gtrsim 31$ at. % just like the W-Mn alloys. Above 52 at. % Fe they become amorphous ferromagnets with T_c increasing up to 68 at. % Fe which is the upper limit for fully amorphous alloys.

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