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# Magnetic properties and hyperfine interactions of amorphous Fe-Hf alloys<sup>a)</sup>

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Alloys of  $\text{Fe}_x\text{Hf}_{100-x}$  ( $20 < x < 100$ ) were fabricated by a vapor-quench method. The amorphous state, as verified by x-ray diffraction, is realized over a wide composition range of  $20 < x < 94$ . The magnetic phase diagram was determined. The variation with Fe content of the magnetic ordering temperature ( $T_c$ ) in Fe-Hf amorphous alloys shows a maximum at  $x_p \approx 87$ . In contrast, the effective magnetic hyperfine field and therefore the Fe moment increases monotonically with  $x$ . Amorphous pure Fe is likely to be magnetic ( $T_c \sim 200$  K) with a large magnetic moment. At higher Fe concentrations ( $x > 95$ ), the samples are crystalline alloys and strongly ferromagnetic. These sharp changes of the magnetic properties are consequences of structural differences, a fact which has been observed in every Fe-early transition metal alloy system reported thus far. In general, the magnetic properties and hyperfine interactions of amorphous Fe-Hf alloys are very similar to those of amorphous Fe-Zr alloys.

## I. INTRODUCTION

At high Fe concentrations, the magnetic properties of several Fe-based amorphous systems show anomalous behavior.<sup>1-3</sup> For example, in this region, the dependence of the magnetic ordering temperature on Fe concentration is opposite to those of the magnetic hyperfine field and the magnetic moment. The understanding of this phenomenon would ultimately lead to the elusive but important magnetic properties of pure amorphous Fe. Since it is difficult to fabricate pure amorphous Fe, one resorts to systems having a stable amorphous phase up to very high iron concentration. In this respect, the studies of the magnetic properties and hyperfine interactions of amorphous Fe-Hf alloys systems over a wide composition range, particularly at high Fe concentrations are desirable. It is also of interest to compare the magnetic properties of Fe-Hf and Fe-Zr alloys since Hf and Zr are chemically very similar.

## II. EXPERIMENT

Samples of  $\text{Fe}_x\text{Hf}_{100-x}$  ( $20 < x < 100$ ) were prepared by a magnetron sputtering device. The samples, 8–12  $\mu\text{m}$  in thickness, were deposited onto liquid-nitrogen cooled substrates of various kinds. The structure of the samples was determined by energy dispersive x-ray diffraction. Samples with 20–94 at. % of iron are amorphous.

It should be noted that Zr and Hf have very similar chemical properties such that one is a common impurity of the other. The commercially available high purity grade Hf (99.8% excluding Zr) used in this study contained a few percent of Zr, which was detected in the fluorescence x-ray analysis. The hyperfine interactions were studied by a conventional Mössbauer spectrometer with a  $^{57}\text{Co}$  in Rh source. The magnetic ordering temperatures were determined by the onset of the magnetic hyperfine interaction using as-prepared samples.

## III. RESULTS AND DISCUSSIONS

### A. Magnetic properties

The variation of the magnetic ordering temperature ( $T_c$ ) with Fe content of amorphous Fe-Hf alloys is shown in Fig. 1. The data determined in this work are denoted as open circles. The solid circles are data reported by Fukamichi.<sup>4</sup> The value of  $T_c$  rises sharply with increasing Fe concentration, showing a maximum at  $x_p = 87$  and then decreases. The qualitative features of the magnetic phase diagram of Fe-Hf amorphous alloys are similar to those of the Fe-Zr system,<sup>3</sup> in spite of the fact that both the atomic weight and density of Hf are about double that of Zr. Since the atomic radii of Hf and Zr are rather close, the similar magnetic properties suggest resemblance in structure characteristics. The  $T_c$ 's of all amorphous Fe-Hf samples are below 300 K, a characteristic shared by all amorphous transition metal (Fe-TM) (TM = Ti, Zr, Nb, Ta, ...etc.)<sup>3,5-7</sup> systems studied thus far.

Several Mössbauer spectra at 4.2 K of  $\text{Fe}_x\text{Hf}_{100-x}$  near the magnetic threshold  $x_c$  are shown in Fig. 2. The samples

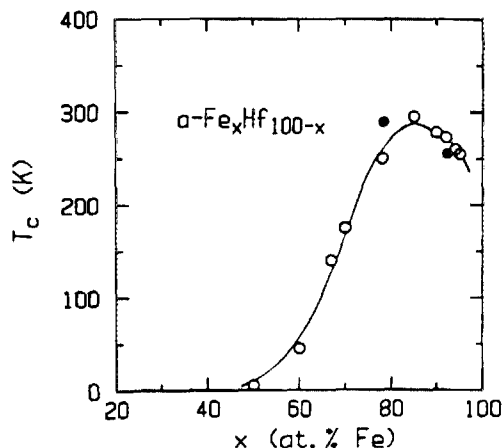


FIG. 1. Magnetic ordering temperatures of amorphous Fe-Hf alloys. The solid circles are data reported in Ref. 4.

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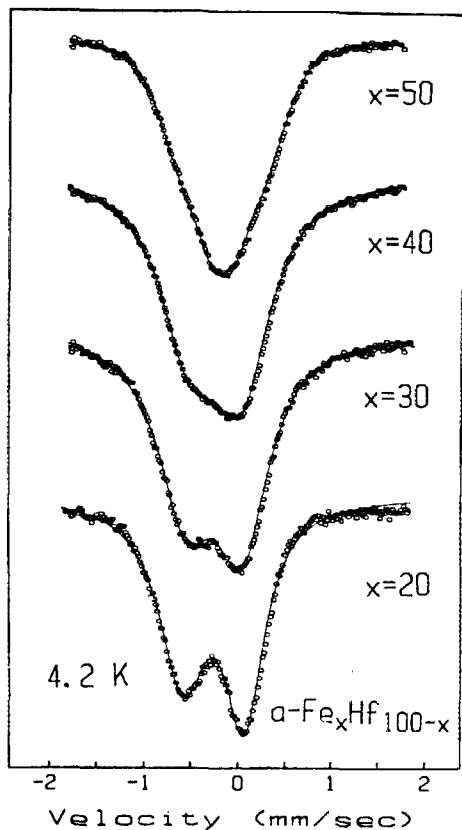


FIG. 2. Mössbauer spectra of Fe-Hf alloys at 4.2 K of  $\text{Fe}_{50}\text{Hf}_{50}$ ,  $\text{Fe}_{40}\text{Hf}_{60}$ ,  $\text{Fe}_{30}\text{Hf}_{70}$ , and  $\text{Fe}_{20}\text{Hf}_{80}$ . Only the sample of  $\text{Fe}_{50}\text{Hf}_{50}$  is magnetic.

with  $x < x_c \approx 45$  are nonmagnetic and their Mössbauer spectra show only quadrupole splittings. The sample with  $x = 50$  exhibits a small and poorly resolved magnetic spectra at 4.2 K. The magnetic threshold concentration ( $x_c$ ), considerably higher than the expectation from percolation considerations, is caused by the disappearance of the magnetic moment, as will be discussed later.

By extrapolating our data in the high iron concentration region to  $x = 100$ , the value of  $T_c$  for amorphous pure iron is estimated to be about 200 K. This is consistent with previous extrapolation made from the studies of amorphous Fe-Zr, Fe-B, and Fe-Y systems.<sup>1-3</sup> Other amorphous Fe-TM alloy systems do not show a maximum in  $T_c$ , because either the compositional range of the amorphous alloys cannot be extended to high enough Fe content (e.g., Fe-Ti)<sup>5</sup> or the values of  $T_c$  remain to be below 200 K for all available compositions (e.g., Fe-Nb).<sup>6,7</sup> All of these characteristics are consistent with a value of  $T_c \approx 200$  K for amorphous pure Fe.

The explanation for a maximum in  $T_c$  has been suggested within the frameworks of both the localized moment and the itinerant pictures of magnetism. From a localized moment standpoint,  $T_c$  provides a measure of the net magnetic exchange interaction [ $J(r)$ ], which is decreasing at high Fe concentration. This could come about by a combination of the following: a decrease in Fe-Fe distance [if  $\partial J(r)/\partial r$  is positive], a decrease in the Fe-Fe coordination number and the emergence of significant antiferromagnetic exchange interaction. A significant decrease in atomic distances or coordination number have been found to be inconsistent with most

of the structure results.<sup>8,9</sup> In the itinerant picture, an explanation for the maximum in  $T_c$  has also been proposed.<sup>10</sup> Below the maximum, the saturation magnetization and  $T_c$  vary in the same sense, as expected for weak ferromagnetism. Above the maximum, where  $T_c$  decreases and magnetization increases, the so-called strong ferromagnetism emerges. However, at still higher Fe concentrations, weak magnetism should return and the Fe moment decreases as it approaches the pure amorphous iron limit. From the measurements of the magnetic hyperfine field, this apparently does not occur in the case of Fe-Hf up to 94 at. % Fe nor in any other systems reported thus far. It should be noted that the magnetic hyperfine field and magnetic moment are proportional, as has been demonstrated in many alloy systems. On the other hand, low field magnetization measurements of the Fe-rich samples may lead to lower and erroneous values for the magnetic moment if alignment of the moments is not achieved. In such cases, the moment data can be obtained only under very high magnetic fields as has been shown in amorphous Fe-Zr.<sup>3,11</sup> Of all of these possibilities, the maximum in  $T_c$  can most consistently be explained by the emergence of the antiferromagnetic exchange interaction as the iron concentration increases.<sup>3</sup>

## B. Magnetic hyperfine interactions

The magnetic hyperfine interaction of amorphous Fe-Hf and Fe-Zr are similar. The variation of magnetic hyperfine field  $H_{\text{eff}}$  of the  $\text{Fe}_x\text{Hf}_{100-x}$  system as a function of  $x$  is shown in Fig. 3. The value of  $H_{\text{eff}}$  decreases monotonically with decreasing Fe concentration, in contrast to the maximum found in the values of  $T_c$ . The continuous diminution and the final disappearance of  $H_{\text{eff}}$ , and therefore the Fe moment, suggest that samples below  $x_c$  become nonmagnetic because of the loss of the Fe moment. Saturation magnetization measurements of several Fe-TM systems are consistent with this conclusion.<sup>1,3,5,6,12</sup> At the Fe-rich end there is no indication that the hyperfine field would decrease. Just as in the extrapolations using amorphous Fe-B and Fe-Zr, amorphous pure Fe should have a large magnetic moment and magnetic hyperfine field close to those of crystalline Fe.

For higher iron concentrations ( $x > 95$ ), the samples are

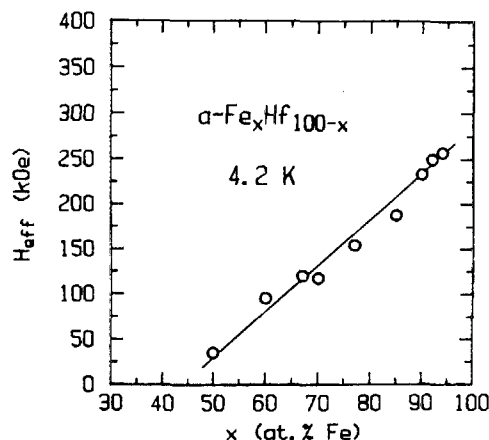


FIG. 3. Magnetic hyperfine field ( $H_{\text{eff}}$ ) of amorphous Fe-Hf alloys at 4.2 K.

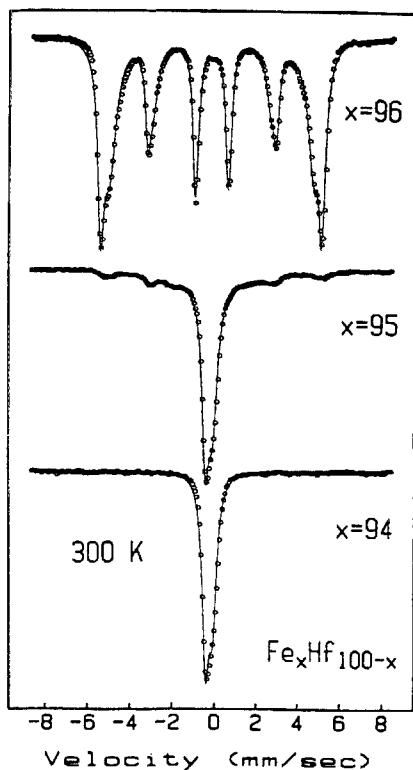


FIG. 4. Mössbauer spectra at 300 K of the Fe-Hf alloys; crystalline  $\text{Fe}_{96}\text{Hf}_4$ , amorphous  $\text{Fe}_{94}\text{Hf}_6$ , and mixed phase  $\text{Fe}_{95}\text{Hf}_5$ .

crystalline and strongly ferromagnetic with  $T_c$  several hundred degrees above those of the amorphous alloys. Room temperature Mössbauer spectra of  $\text{Fe}_x\text{Hf}_{100-x}$  alloys near the phase boundary are shown in Fig. 4. The top spectrum of crystalline  $\text{Fe}_{96}\text{Hf}_4$  shows two distinct phases ( $\alpha$ -Fe and saturated Fe-Hf crystalline alloy) both with large magnetic hyperfine fields. The spectrum of the sample with  $x = 94$  shows only an amorphous phase which is paramagnetic at room temperature. For the sample with  $x = 95$ , both

crystalline and amorphous states are present as can be clearly identified from the spectrum. X-ray diffraction provides the same conclusion. The sharply defined boundary separating the amorphous and crystalline states in the Fe-Hf system has also been observed in other Fe-TM systems.<sup>5,7</sup> The pronounced difference in magnetic properties is caused by the differences in the atomic arrangement and electronic structure between crystalline and amorphous solids.

#### IV. CONCLUSION

Amorphous Fe-Hf alloy over a wide composition range of  $20 < x < 94$  has been made by a sputtering method. The unusual magnetic behavior at high iron concentration can be explained by an increase in the antiferromagnetic exchange interaction as the iron concentration increases. The similarity of the magnetic properties and the hyperfine interaction between the Fe-Hf and Fe-Zr systems indicates that those properties are largely determined by the outer electronic shell which is the same in Hf and Zr. By extrapolation, amorphous pure Fe is likely to be magnetic with an ordering temperature of about 200 K and a high magnetic moment.

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