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C. L. Chien
Johns Hopkins University

K. M. Unruh
Johns Hopkins University

A. Levy
Johns Hopkins University

Sy_Hwang Liou
University of Nebraska-Lincoln, sliou@unl.edu

J. P. Stokes
Johns Hopkins University

See next page for additional authors

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Authors

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Magnetic, Mössbauer, and resistivity studies of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ alloys*

C. L. Chien, K. M. Unruh, A. Levy, S. H. Liou, and J. P. Stokes

The Johns Hopkins University, Baltimore, Maryland 21218

R. J. Gambino and K. Fukamichi**

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

Amorphous metal-metal samples of $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ have been studied by bulk magnetic, Mössbauer spectroscopy and resistivity measurements. The values of T_C have been determined to be 285 K and 295 K for $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$, respectively. Both the magnetic isotherms and Mössbauer measurements indicate that these samples are ferromagnetics. The isomer shifts and the quadrupole interactions are much smaller than those of amorphous Fe-metalloid systems. The P(H) are broad and extend to small H values. Negative temperature coefficients of the resistivity are observed at low temperatures followed by a resistivity minimum at temperatures slightly above room temperature. These results are significantly different from those of amorphous Fe-metalloid systems with comparable Fe contents.

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INTRODUCTION

A large number of studies have been made in recent years on amorphous transition metal (TM)-metalloid (M) systems, particularly those near the eutetic composition (1). Relatively fewer studies have been made on the amorphous metal-metal systems. Recently, it has been shown that Fe-Zr and Fe-Hf can be made amorphous by either liquid-quench techniques near the eutetic points, or by vapor deposition methods over a much wider composition range (2 - 7). In this work, we show the results of Mossbauer spectroscopy as well as bulk magnetic and resistivity measurements of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$. When appropriate, the present results are compared with the better known results of amorphous TM-M alloys with similar compositions.

EXPERIMENTAL

Samples of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ were made by sputtering on polymer substrates with a film thickness of about 1 μm. The compositions of the films were independently confirmed by microprobe analyses. Mossbauer spectroscopy measurements using a ^{57}Co in Rh source were made on absorbers of about ten layers. A standard four-probe technique was employed for the resistivity measurements.

RESULTS AND DISCUSSIONS

A. Magnetic Ordering Temperature

The magnetic ordering temperature (T_C) of each sample was determined by bulk magnetic measurements and by Mossbauer spectroscopy. In the magnetic measurements, T_C was determined by the intercept of the $(4\pi M)^2$ versus T plot at $(H/4\pi M) = 0$ as shown in Fig.1 by the arrows. In the Mossbauer spectroscopy measurement, a constant velocity thermal scan method was used under no external field (8). Using this method, as the sample is cooled from the paramagnetic state, the count rate increases sharply at T_C due to the onset of the magnetic hyperfine interaction as shown in Fig.1. The values of T_C = 285 K for $Fe_{74}Zr_{26}$ and T_C = 295 K for $Fe_{81}Hf_{19}$ determined by the two methods are in excellent agreement (within 2 degrees).

These values of T_C for amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ are much lower than those for crystalline alloys and compounds of comparable compositions as well as those of amorphous Fe-metalloid alloys with comparable Fe contents (1).

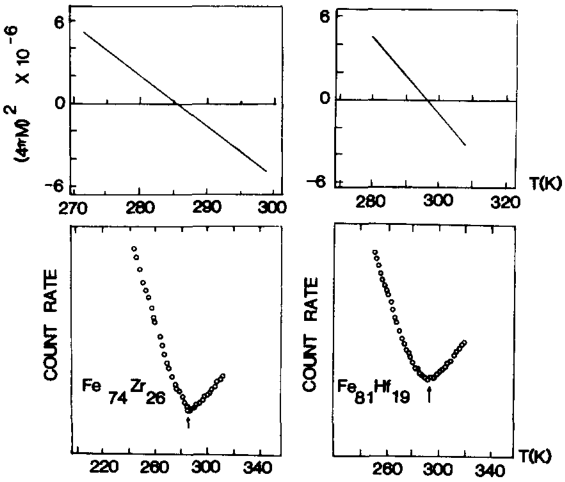

Figure 1 : Determination of T_C of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ from bulk magnetic measurements (upper) and Mossbauer spectroscopy measurements (lower).

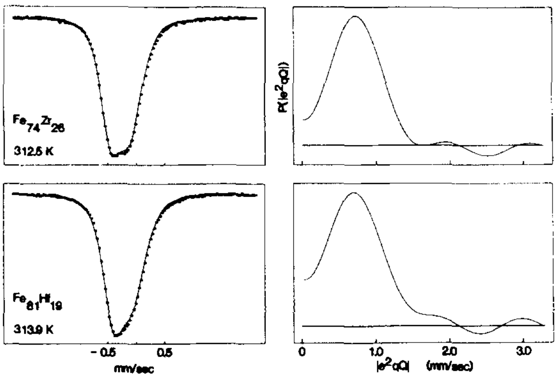

Figure 2 : Mossbauer spectra of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ at $T > T_C$. The deduced distributions of quadrupole interactions $P(|e^2qQ|)$ are shown on the right.

B. Paramagnetic Spectra

Above T_C, the spectra of $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ are shown in Fig.2. The distribution of electric quadrupole interactions $P(|e^2qQ|)$ deduced from these spectra are also shown in Fig.2. The details of the analyses are described elsewhere (9).

It is useful to compare the results of the present metal-metal systems to those of TM-M systems with similar TM concentrations (9). One notes the quadrupole splittings of the present systems are small so that the splitting is poorly resolved. The mean quadrupole splitting is at least a factor of 2 to 3 smaller than those of amorphous TM-M alloys. Buschow and Smit observed an even less resolved quadrupole spectrum in amorphous $Fe_{85}Zr_{15}$, except that the asymmetry in the spectrum is opposite to that of the present one (6). The isomer shifts at 300 K of $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ are respectively - 0.05 mm/sec and - 0.12 mm/sec relative to crystalline α-Fe at 300 K. TM-M with low ordering temperatures (less than 300 K, so that the isomer shift can be determined accurately from the quadrupole spectra) have isomer shifts of the opposite sign with values from about + 0.1 mm/sec to + 0.2 mm/sec (9). It is clear that the electronic structure of the Fe atoms and the electric quadrupole interaction at the Fe sites are rather different in amorphous metal-metal and amorphous metal-metalloid systems.

It has been observed in a large number of TM-M systems that the $P(|e^2qQ|)$ shows a very small and often nearly zero probability at $|e^2qQ| = 0$, i.e. $V_{zz} = 0$ (9). Amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ however, as shown in Fig.2, exhibit significantly different results in that there is a sizable probability at $V_{zz} = 0$. In the atomic structure of amorphous Fe-Zr and Fe-Hf alloys, there are a sizable number of Fe atoms having high site symmetry with near zero electric field gradients. According to the calculations by Czjzek et al, a near-zero probability at $V_{zz} = 0$ is a general result for a random packing structure (10). This raises the question as to whether random packing is an accurate model for amorphous Fe-Zr and Fe-Hf alloys.

C. Magnetic Hyperfine Interaction

Mossbauer spectra of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ at 4.2 K are shown in Fig.3. Due to substrate effects at low temperatures, as commonly observed in amorphous samples deposited on substrates, the magnetic moments in the samples tilt substantially out of the sample plane. The line intensities of the #2 and #5 peaks are consequently suppressed (1). However, with the application of a small external magnetic field of about 1 kOe in the sample plane, the moments are aligned toward the sample plane so that the #2 and #5 peak intensities are greatly increased as shown in Fig.3. The ease with which the moments respond to a small field indicates that both of these samples are soft ferromagnets. This is consistent with the magnetic data described below.

It is evident from Fig.3 that the spectral line intensities are not symmetric with respect to the isomer shift. The #1 peak has a larger intensity than that of the #6 peak, and similarly so for the #3 and #4 peaks. This is caused by a correlation of the hyperfine field and the isomer shift, as has been observed in a number of amorphous systems (11). Therefore in deducing the field distribution P(H) contained in the spectrum, we assume a correlation of $IS = IS_0 + \beta|H|$, where IS stands for isomer shift and β is a constant. As shown by the calculated spectra, this simple linear correlation accounts very well for the asymmetrical intensities. The constant β is about $(8 \pm 1) \times 10^{-4}$ mm/sec-kOe. The basis for the correlation can be found in the concentration dependences of the average hyperfine field and isomer shift. Both of these quantities increase with Fe concentration (6), thus accounting for the correlation as well as the positive sign of β.

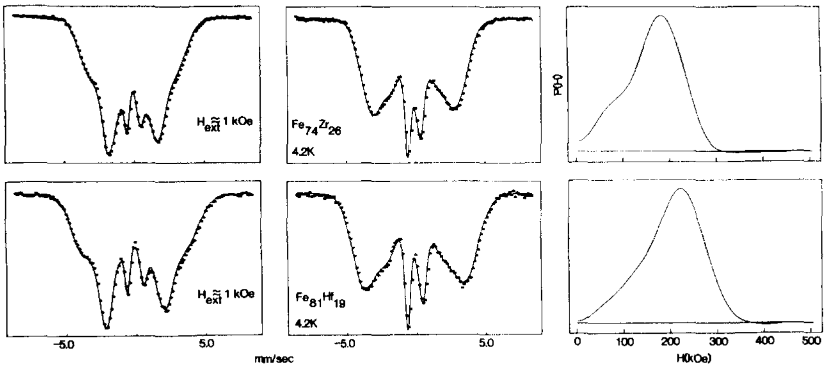

Figure 3: Mossbauer spectra of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ at 4.2 K with and without an external magnetic field of about 1 kOe applied parallel to the sample plane. The deduced distributions of hyperfine fields P(H) are shown on the right.

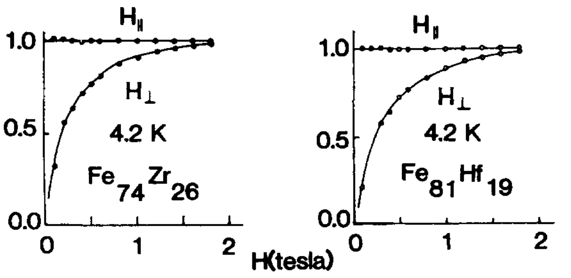

Figure 4 : Reduced magnetization at 4.2 K of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ as function of external field applied parallel and perpendicular to the sample plane.

The deduced P(H) of $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ are shown in Fig.3. Unlike the amorphous Fe-metalloid alloys, these P(H) are broad and extend to small H values. The Fe atoms which exhibit very small fields can be those that are weakly coupled. They can therefore provide the mechanisms for the unusual low field magnetic characteristics (12) and the negative temperature coefficients of the resistivity.

The peak values of P(H) are 190 kOe and 228 kOe for $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ respectively. Because the P(H) extends towards H = 0, the values of H_{mean}, defined as $\int H\,P(H)dH$ are smaller, with values of 160 kOe and 203 kOe.

D. Magnetic Properties

The reduced magnetization of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ at 4.2 K as a function of the applied field is shown in Fig.4. $H_{\parallel}$ and $H_{\perp}$ refer to the magnetic field being applied parallel and perpendicular to the sample plane. It is evident that the samples can be magnetized to saturation under a small $H_{\parallel}$. These high field measurements demonstrate that both samples are ferromagnetic. The magnetizations ($4\pi M$) for $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ are respectively 6.9 kG and 10.8 kG. These values scale reasonably well with the values of H_{mean} from P(H).

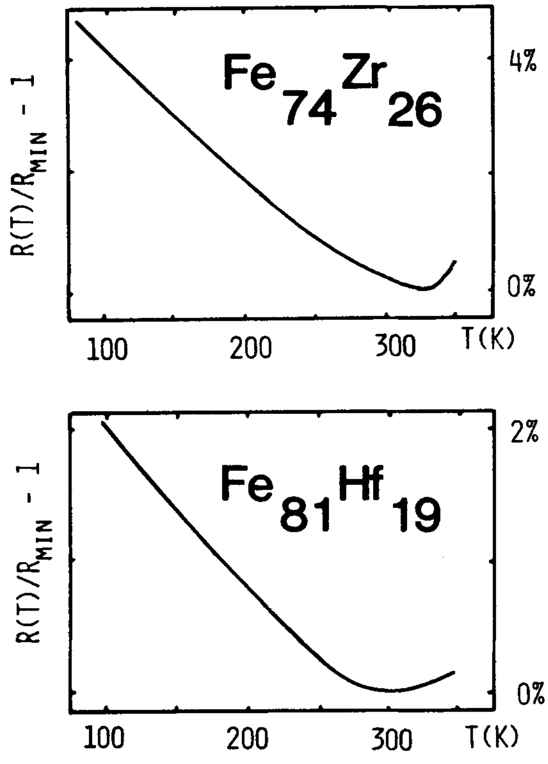

Figure 5 : Temperature dependence of the resistivity of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ near the resistivity minima.

E. Resistivity

From 4.2 K to about 300 K, the resistivity of both amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ decreases monotonically with increasing temperature. The negative temperature coefficients are exactly opposite to the expection for metallic systems. The negative temperature coefficients have also been observed in samples with similar compositions from 4.2 K to 300 K (6). However, as one extends the temperature range to slightly above room temperature, it becomes apparent that in both $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ there is a resistivity minimum, as shown in Fig.5. These results should be contrasted with amorphous Fe-metalloid systems with comparable Fe contents where the Kondo-like resistivity minima occur only at low temperatures (1). The actual mechanisms responsible for the resistivity minima in amorphous metals remain a controversial subject. For the present samples, the resistivity minima are likely to be caused by the scattering due to weakly coupled moments which are suggested by the P(H) analyses.

Conclusions

Although the magnetic properties, isomer shifts, quadrupole interactions, hyperfine field distributions and resistivity behavior of amorphous $Fe_{74}Zr_{26}$ and $Fe_{81}Hf_{19}$ are similar, they are significantly different from those of amorphous Fe-metalloid alloys. The properties of the Fe based amorphous alloys are determined to a large extent by the species of the non-transition metal elements.

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** Present address: Research Institute for Iron, Steel and Other Materials, Tohoku University, Sendai, Japan.

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