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Exchange interactions in hydrogen-induced amorphous YFe₂

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Abstract

The magnetic properties of hydrogen-induced amorphous (HIA) YFe_2H_x (x = 1.8, 3.0 and 3.4) alloys have been investigated by means of ⁵⁷Fe Mössbauer spectroscopy and atomic pair distribution analysis. The exchange integral (*J*) estimated from the temperature dependence of the average ⁵⁷Fe hyperfine field shows a tendency to decrease with decreasing the average Fe–Fe interatomic separation. Moreover, an abrupt drop of *J* is evident at an average Fe–Fe interatomic separation of about 0.25 nm, which is the empirical threshold where the magnetic interaction between Fe moments changes from ferromagnetic to antiferromagnetic. The absence of a spin-glass state in HIA YFe₂(H) is well understood by the enhancement of *J* through the volume expansion induced by the absorption of hydrogen. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Two major methods for producing amorphous alloys are liquid-quenching and vapor-quenching. The vast majority of studies on amorphous alloys have employed these two methods. A third alternative for preparing an amorphous phase in metallic systems is by solid state reactions such as hydrogenation of intermetallic compounds [1]. We have recently demonstrated that such hydrogen-induced amorphization takes place in the C15 Laves phase YFe₂ [2].

Amorphous Fe–Y alloys have been studied extensively [3-5] due to their peculiar magnetic behavior which includes both paramagnetic to spin-glass transitions and large magnetovolume effects. Amorphous YFe₂ prepared by conventional vapor-quenching is known to show a paramagnetic to spin-glass transition at around 50 K [3]. However, our hydrogen-induced amorphous (HIA) YFe₂ alloy is a good ferromagnet at room temperature. It has been

found in our previous investigation [6] that the exchange integral estimated from the temperature dependence of the mean ⁵⁷Fe hyperfine field is reduced clearly by removing hydrogen from the HIA–YFe₂(H) alloy, suggesting the effect of volume on the exchange interactions. Since the reduction of the interatomic separation between Fe–Fe atomic pairs in amorphous ferromagnets could result in considerable numbers of antiferromagnetic bonds along with the majority ferromagnetic interactions which lead to exchange frustrations, the absence of the spin-glass state from our HIA–YFe₂(H) alloy is well understood if the extent of the volume expansion induced by hydrogen is clarified. In this paper we present a quantitative study on the hydrogen-induced volume effect on the exchange interactions in the HIA–YFe₂(H) alloy.

2. Experimental

Amorphous $YFe_2H_{3,4}$ was prepared by heating the crystalline YFe_2 powders under H_2 of 1.0 MPa to 550 K at a constant heating rate of 0.17 K/s. The precursor crystalline

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alloy ingot of YFe₂ was prepared by argon-arc melting followed by homogenization at 973 K for 1 week. The homogenized ingot was pulverized (<100 mesh) before amorphization. Amorphous YFe₂H_{3.0} and YFe₂H_{1.8} were also prepared by annealing the amorphous YFe₂H_{3.4} alloy under a reduced pressure of ~1 Pa at 383 K for 1.2 ks and 18 ks, respectively. The chemical compositions of these three amorphous alloys are based on the chemical analysis performed on a Horiba EMGA-621 hydrogen analyzer. ⁵⁷Fe Mössbauer spectra were obtained using a conventional constant-acceleration spectrometer with a ⁵⁷CoRh source. Microbeam electron diffraction intensity profiles were acquired on imaging plates using a transmission electron microscope operated at 300 kV; the beam size was about 100 nm.

3. Results and discussion

100 95

90

85

100

98

96 94

100

98

96

94

100

90

80

-8

Relative transmission (%)

Fig. 1 shows ⁵⁷Fe Mössbauer spectra acquired on hydrogen-induced amorphous YFe_2H_x (x = 1.8, 3.0 and 3.4) at room temperature. The result obtained from the C15 Laves phase YFe_2 before hydrogenation is also shown for comparison. The solid lines indicate fits to the spectra and the distributions of the hyperfine parameters for the HIA YFe_2H_x alloys are shown alongside the spectra. Since the magnetization easy axis of C15–YFe₂ is [111], two sex-

 B_{hf} (T)

30

P(B_{hd}) or P(QS)

1.5

10 20

19.6 T

<B_> = 14.9 T

0.5

QS (mm/s)

<QS> =

0.47 mm/s

<B_{hf}> =

c-YFe₂ (C15)

a-YFe₂H_{3.4}

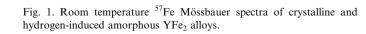
a-YFe₂H₃₀

a-YFe₂H_{1.8}

Velocity (mm/s)

tets with an area ratio of 3:1 were necessary to fit the spectrum; the hyperfine fields $(B_{\rm h\,f})$ of the major and minor subspectra are 18.5 T and 18.1 T. The hyperfine parameters of all the HIA YFe₂H_x alloys are highly distributed, typical of amorphous magnetic materials. The average hyperfinefield ($\langle B_{\rm h\,f} \rangle$) of HIA YFe₂H_x shows a tendency to decrease with decreasing the hydrogen content and HIA YFe₂H_{1.8} becomes paramagnetic at room temperature, suggesting the effect of hydrogen on the magnetic ordering temperature in HIA YFe₂H_x (x = 1.8, 3.0 and 3.4).

The temperature dependence of the ⁵⁷Fe hyperfine field was examined in order to clarify the effect of hydrogen on the thermo-magnetic behavior of HIA YFe_2H_x (x = 1.8-3.4). Fig. 2 shows a typical example of the temperature dependence of the 57Fe Mössbauer spectra, which was acquired on HIA YFe₂H_{3,4} in a temperature range of from 12 to 250 K. A systematic change in the hyperfine-field distribution, $P(B_{\rm hf})$, is seen and the average hyperfine-field tends to decrease with increasing temperature. The $\langle B_{\rm hf} \rangle$ values obtained from the Mössbauer spectra of HIA YFe₂H_x (x = 1.8-3.4) and the unhydrogenated C15-YFe₂ in the temperature range from 12 to 300 K are plotted against temperature in Fig. 3. The Curie temperatures of these alloys determined from susceptibility measurements are also plotted in this figure (i.e., the plots at $\langle B_{\rm hf} \rangle = 0$). Both the mean hyperfine field at 12 K and the Curie temperature appear to be reduced by reducing



4

at 295 K

8 0

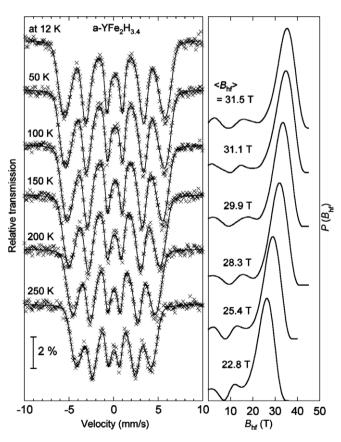


Fig. 2. Mössbauer spectra and hyperfine-field distributions of hydrogen-induced amorphous ${\rm YFe_2H_{3.4}}$ at various temperatures.

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