Low-temperature magnetization study of crystalline and glassy Fe-B alloys

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The low-temperature dependence of the magentization is reported for crystalline and glassy $Fe_{100-x}B_x$ alloys. Although the temperature coefficients reduced to the Curie temperatures are four to five times larger for the glassy alloys than the crystalline ones, all of the alloys exhibit Bloch's $T^{3/2}$ dependence. The spin-wave stiffness constant deduced from the temperature coefficient reaches a broad minimum ($D \sim 65$ meV Å^2) for $x = 14$–16. This observation seems to be consistent with the Invar-like behavior reported recently for the similar alloys.

I. INTRODUCTION

The temperature dependence of the magnetization toward $T \to 0$ K in crystalline ferromagnets has been well described by Bloch's $T^{3/2}$ law, indicating that long-wavelength spin-wave excitations determine the magnetization behavior.[1] The existence of spin waves in noncrystalline ferromagnets is conceivable and has been indeed demonstrated by various studies employing low-temperature magnetization,[2] inelastic neutron scattering,[3] and Mössbauer techniques.[4] Thus for both crystalline and glassy ferromagnets, the magnetization $M(T)$ at low temperatures varies as

$$M(T)/M(0) = 1 - BT^{3/2} - CT^{5/2} - \cdots . \quad (1)$$

The major difference between glassy and crystalline ferromagnets seems to be found in the values of B between the two cases.[4] The quantity of B for a noncrystalline ferromagnet is, in general, five to ten times larger than that for a crystalline ferromagnet. The coefficients C are found to be comparable in both cases. These results imply the following: (i) The spin-wave density of states is proportional to $\omega_q^{1/2}$ for small magnon frequency ω_q in both crystalline and glassy ferromagnets with higher density of states at low ω_q in the latter case. (ii) Exchange interactions between atoms carrying magnetic moments are greatly reduced and more localized in noncrystalline solids.

The results thus far obtained and their implications are based on a limited number of glassy ferromagnets having compositions close to $TM_{80}M_{20}$ where TM and M stand for $3d$ transitions metal(s) and metalloid(s). Recently the boron concentration range of the binary $Fe_{100-x}B_x$ glassy system has been extended from $x \sim 20$ to $12 \leq x \leq 28$.[5] Furthermore these alloys have been found to exhibit a number of properties which imply a subtle structural change from a bcc-like at $x \sim 12$ to a dense random packing structure at $x \sim 20$.[5] Since this structural change, if there is any, should be reflected in the spin-wave properties, the compositional dependence of the low-temperature magnetization has been studied in this investigation. The results for the glassy alloys ($12 \leq x \leq 28$) are compared with those for the crystalline bcc solid solutions ($0 \leq x < 12$).[6]

II. EXPERIMENTAL METHODS AND RESULTS

The sample preparation and experimental procedures are the same as described in Refs. 5 and 6. The temperature ranges covered in the present study are between 4.2 K and the Curie temperatures of the materials. The sample temperature measurement was made by using a glassy carbon resistance thermometer situated adjacent to the sample and was accurate to within ±0.1 K around 4.2 K and about ±1 K around 300 K. The sample temperature was also monitored by a copper-constantan thermocouple, which has a better accuracy in the vicinity of 300 K. The magnetization was measured with an applied field of 9 kOe by a vibrating sample magnetometer having a resolution of about 10^{-4} emu. The square-shaped samples had dimensions approximately 3 mm × 3mm × 35 μm. The applied field was in the sample plane to minimize the demagnetizing field effects.

Since the measurements were performed with an applied field H of 9.0 kOe, the effective internal field H_{eff}, which contributes to the energy gap in the spin-wave spectrum, has to be accounted for in determining the coefficients B, C, etc. in Eq. (1). The expres-

sion, Eq. (1), is then written

$$\frac{\Delta M(T)}{M(0)} = \frac{\{M(T)-M(0)\}}{M(0)} = -z\left(\frac{3}{2}, \frac{T_g}{T}\right) BT^{3/2} - z\left(\frac{5}{2}, \frac{T_g}{T}\right) CT^{5/2} - \cdots , \quad (2)$$

where z functions are defined in Ref. 1 for a given gap energy $k_B T_g (= g\mu_B H_{\text{eff}})$. By using $T_g = 1.21$ K,[7] the quantity $\Delta M(T)/M(0) z(\frac{3}{2}, T_g/T)$ is plotted in Fig. 1 against $(T/\theta_f)^{3/2}$ for $x = 10$ and 16 where θ_f is the ferromagnetic Curie temperature of the sample. The proportionality constant $B_{3/2}$ gives the value $B (= B_{3/2}/\theta_f^{3/2})$. The coefficients B and $B_{3/2}$ thus determined are listed in Table I. It is noted that the coefficient $B_{3/2}$ for $x = 20$ is close to that obtained previously by Mössbauer techniques ($B_{3/2} = 0.40$).[4] The plot $\Delta M(T)/M(0)\, T^{3/2}$ vs T was made in an attempt to determine the quantity C. However, the scatter in data was such that the value of C had an error of the same order of the magnitude of C. Thus meaningful discussion can be only made for the data of the coefficients B, which is given below.

III. DISCUSSION

The present data, as exemplified in Fig. 1, show that the magnetization of glassy Fe-B alloys obeys the $T^{3/2}$ law for $T < \frac{1}{3}\theta_f$ as observed in many other metallic glasses.[4] This contrasts with the crystalline case including the Fe-B bcc solid solutions, in which the $T^{3/2}$ law is valid only up to about $T \sim 0.15\theta_f$. This is illustrated in Fig. 1. It is instructive to compare the reduced value $B_{3/2}$ for the different compositions. For the crystalline region ($x < 12$), the coefficient $B_{3/2}$ is essentially constant at $0.11 \sim 0.12$, while it has a definite compositional dependence with a peak

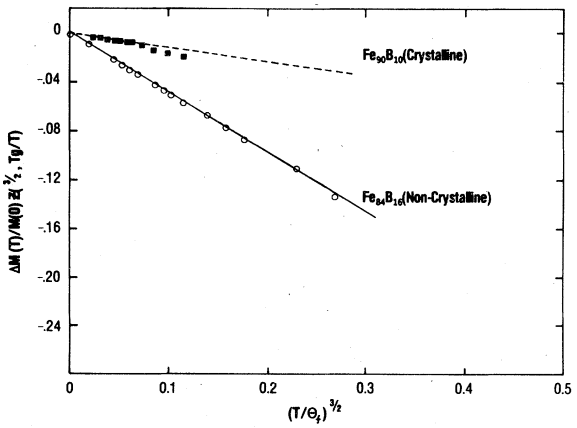

FIG. 1. Change of the magnetization as a function of $(T/\theta_f)^{3/2}$ for crystalline $Fe_{90}B_{10}$ and glassy $Fe_{84}B_{16}$ alloys. The applied field was 9.0 kOe.

TABLE I. Compositional dependence of Curie temperature θ_f, saturation magnetization $M(0)$, and coefficients B, $B_{3/2}$, and D for crystalline and glassy $Fe_{100-x}B_x$ ferromagnets.

x	θ_f (K)	$M(0)$ μ_B/Fe atom	B (10^{-5} K$^{-3/2}$)	$B_{3/2}$	D (10^{-3} eV Å^2)
0	1043	2.22	3.4 ± 0.2	0.11 ± 0.02	278 ± 30
4	978	2.19	3.6	0.11	267
6	964	2.17	3.7	0.11	267
8	944	2.15	4.1	0.12	259
10	916	2.13	4.3	0.12	254
12	509	2.14	3.1	0.35	70 ± 3
13	531	2.13	3.4	0.41	66
14	556	2.12	3.4	0.45	65
16	598	2.12	3.4	0.49	66
18	636	2.07	3.1	0.50	71
20	647	2.00	2.6	0.43	83
22	690	2.00	2.3	0.41	92
24	723	2.00	2.2	0.42	96
28	760	1.94	2.0	0.41	106

around $x \sim 18$ in the noncrystalline region. It is noted that the value of $B_{3/2}$ for $x = 18$ is quite close to the theoretical value of 0.512 calculated for $S = 1$ case in the Heisenberg model.[8] The closeness between the experimental and calculated values implies, as pointed out previously,[8] that the localized model can describe the magnetic behavior of glassy ferromagnets better than that of crystalline ones. Further experimental evidence to support this may be given by (i) the success in the mean-field analysis of the data obtained for rare-earth transition-metal base amorphous films[9]; and (ii) the fact that the critical exponents are close to those calculated for Heisenberg ferromagnets.[10–12]

In simple spin-wave theory,[13] the coefficient B is related to the spin-wave stiffness constant D through

$$B = 2.612\{g\mu_B/M(0)\}\{k_B/4\pi D\}^{3/2} . \quad (3)$$

The coefficients D calculated using this expression and the values of B in Table I are listed in the last column of the table. The compositional dependence of D and $B_{3/2}$ for all the alloys studied in this report are shown in Fig. 2.

The results can be summarized as $D/\theta_f \simeq 0.27$ and 0.12 meV Å^{2}K^{-1} for crystalline and glassy Fe-B alloys. The smaller D/θ_f ratios for the noncrystalline alloys imply shorter range of the exchange interactions, being consistent with the previous results obtained for Fe-Ni base glassy alloys.[14] While this seems to be a general trend for iron base glassy alloys, the same does not hold for noncrystalline Co-B alloys. For example, we find $D \simeq 350$ and 200 meV Å^2 for pure crystalline cobalt[15] and noncrystalline $Co_{74}B_{26}$,[16] respectively. Using the reported values of θ_f,[17] we obtain $D/\theta_f = 0.25$ and 0.29 meV Å^{2}K^{-1} respectively. These results are consistent with the findings[17] indicating that the magnetic properties of noncrystalline Co-B alloys differ very little from those of the crystalline counterparts, being quite different from the glassy Fe-B case.

It has been reported recently that the glassy Fe-B alloys with $x \sim 17$ exhibit Invar-like properties.[18,19] In the crystalline fcc Fe-Ni alloy system, the Invar anomalies can be observed for the iron concentration larger than ~65 at.%, and close to the composition at which the alloy system undergoes an $\alpha \rightleftarrows \gamma$ phase transition. The anomalies are also accompanied by a reduction of the spin-wave stiffness constant D, implying a ferromagnetic instability.[20] It appears that the present Fe-B glassy alloys in the region $x = 14–16$ provide a system resembling that of fcc Fe-Ni in the Invar region; namely, the value of D is the lowest at these boron concentrations, below

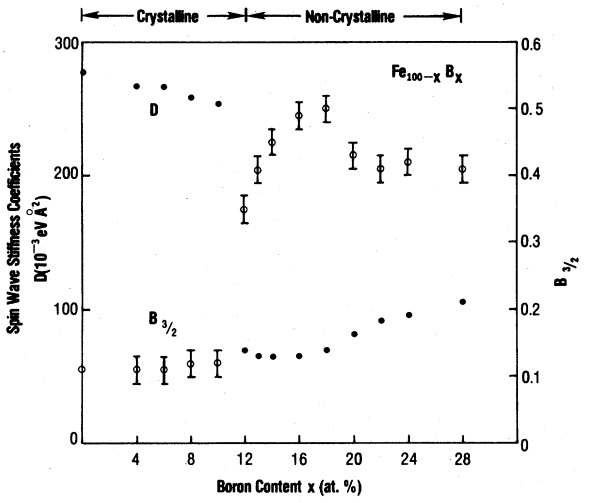

FIG. 2. Spin-wave stiffness D and temperature coefficient $B_{3/2}$ vs boron content. Error bars for D are not indicated.

which the local atomic arrangements may favor those based on a randomized bcc-like structure, as discussed previously.[5] In the crystalline region ($x < 12$), on the other hand, the observed data seem to be consistent with the previous ones.[6] For a cubic ferromagnet, the stiffness constant D is given by $D = 2SJa^2$, where S is the spin value, J is the exchange constant, and a is the cube length. Thus, for example, using the values of S, J, and a from Ref. 6, one obtains $D = 263$ meV Å^2 for $x = 0$ and $D = 251$ meV Å^2 for $x = 10$ which are quite close to the observed values (see Table I). The reduction of D with increasing x, in this case, is due to the lattice contraction and the decrease of S with the boron content.[6]

In summary, the compositional dependence of the spin-wave stiffness constant for the binary Fe-B alloys is consistent with the previous results indicating the range of the exchange interaction is reduced in the noncrystalline Fe-base alloys. The observation of the Invar-like behavior and the lower values of the spin-wave stiffness constants for the alloys containing 14–16 at.% boron resemble those properties of fcc Fe-Ni Invar alloys. This is favorable for the previously advanced notion that the noncrystalline alloys containing boron concentration near 12 at.% have a local atomic arrangement based on a bcc-like structure. While the data presented here are quantitatively compatible with the previously reported results for the crystalline case with less than 12 at.% B, they are only relatively consistent for the alloys in the noncrystalline region.

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