Distribution of Hyperfine Fields in Magnetically Anisotropic Amorphous Fe_<100-x>B_x Alloys

Distribution of Hyperfine Fields in Magnetically Anisotropic Amorphous $Fe_{100-x}B_x$ Alloys*

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Synopsis

The temperature dependence of the Mössbauer spectra of the amorphous $Fe_{100-x}B_x$ (x=14∿21) alloys has been measured. The two distributions of intensity, $I_{1,3}(H)$ and $I_2(H)$, which are associated with (±3/2-±1/2) and (±1/2-∓1/2), and with (±1/2-±1/2) transitions, respectively, are calculated from the spectra separately. The fact that the shapes of $I_2(H)$ are different from those of $I_{1,3}(H)$ at 20K gives evidence that the magnitude and direction of hyperfine fields are not independent of each other, but are correlated. The ratio of the average fields $\bar{H}_2$, derived from $I_2(H)$, to $\bar{H}$, derived from the hyperfine field distribution $P(H)$, at 20K is dependent on the boron concentration. It is considered that there exists a spatial fluctuation of composition in the samples, and that in the regions where x≃17 the magnetization changes its direction easily from parallel to perpendicular to the sample plane, under the stresses caused by decreasing temperature.

I. Introduction

In a previous paper[1], we have proposed a modified analytical method for finding the probability distribution $P(H)$ of magnetic hyperfine fields from the Mössbauer spectrum of a ferromagnetic amorphous sample. The method is useful in determining whether the magnitude of a hyperfine field and its direction are correlated or not. In most Mössbauer effect studies, the analyses of $P(H)$ have been performed based on the assumptions that the amorphous samples are isotropic and homogeneous and that the magnitude of the hyperfine field

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and its direction are not correlated. However, there is some experimental evidence conflicting with these assumptions. The magnitude as well as the direction of hyperfine fields are clearly different between the bulk and the surface of amorphous $Fe_{40}Ni_{40}P_{14}B_6$ [2]. The upper and lower surfaces of ribbons of $Fe_{40}Ni_{40}P_{14}B_6$ have different domain structures[3], and the domain structure in $Fe_{80}P_{13}C_7$ changes with removal of material from the surface by electropolishing[4]. It is, therefore, likely that there are special cases in which the magnitude and the direction of the hyperfine fields are correlated.

In another previous paper[5], we reported that the Mössbauer spectra of amorphous $Fe_{100-x}B_x$ (x=14∿21) alloys showed temperature dependence of the magnetization direction. The easy axis of magnetization at room temperature is nearly in the plane of the sample ribbon, while it tilts out of the plane below 160K. Similar behavior has also been observed in many ferromagnetic amorphous alloys[6-11]. Precise discussion of the phenomenon that the magnetization direction changes with temperature decrease has been given by van Diepen and den Broeder [11], where they explained this effect to be due to the difference between the thermal expansion coefficients of the amorphous sample and such backing materials as vacuum grease, aluminum foil and adhesive tape which held the ribbon samples in place. Since the thermal expansion coefficient of these backing materials is about an order of magnitude larger than that of the amorphous alloys, the sample is compressed in its plane. Therefore perpendicular anisotropy is induced because of the positive magnetostriction of the sample. These authors have also supposed that the domain structure of the sample under compressive stress is similar to the maze domain patterns observed at room temperature, and hence the temperature-dependent Mössbauer spectra can be explained as due to a fractional change of two types of domains (maze domains and 180° wall domains). Through computer analyses of the Mössbauer spectra in which two distribution functions of magnetic hyperfine fields appearing in (±3/2-±1/2) and (±1/2-∓1/2) absorption lines and in (±1/2-±1/2) lines were calculated independently, we noticed that the domains whose magnetization direction tilted out of the ribbon plane with temperature decrease had the boron content close to the nominal composition of x=17[5].

In the present paper, we recalculate the probability distribution $P(H)$ of hyperfine fields from the Mössbauer spectra of the amorphous $Fe_{100-x}B_x$ (x=14∿21) alloys. The analysis is performed without the assumptions that the amorphous samples are isotropic and homogeneous and that the magnitude and direction of the hyperfine field are independent. If the magnitude of the hyperfine field and its direction

are correlated, it is somewhat complicated to find the true shape of $P(H)$. The relative intensity of the six lines can be described as a function of the angle θ between γ-rays and the direction of the hyperfine field;

$$I_3(H) = P(H) \cdot 3[1+\cos^2\theta(H)]/2,$$

$$I_2(H) = P(H)\cdot 2\sin^2\theta(H), \qquad (1)$$

$$\text{and} \quad I_1(H) = P(H)\cdot[1+\cos^2\theta(H)]/2,$$

where $I_3(H)$, $I_2(H)$ and $I_1(H)$ are the area intensities of the absorption lines corresponding to $(\pm 3/2-\pm 1/2)$, $(\pm 1/2-\pm 1/2)$ and $(\pm 1/2-\mp 1/2)$ transitions, respectively. In the usual analysis of $P(H)$, θ and H are assumed to be independent of each other, so that values of $I(H)$ are simply proportional to $P(H)$. In the case of the present treatment, however, θ and H may be correlated, and the true distribution of hyperfine fields, $P(H)$, is different from the distributions of line intensities, $I_1(H)$ and $I_2(H)$. $I_1(H)$ and $I_2(H)$ are observed separately, and they give information about the relation of the magnitude and the direction of hyperfine fields. In this case $P(H)$ is obtained form $I_1(H)$ and $I_2(H)$ by the relation,

$$P(H) = I_1(H) + I_2(H)/4 \qquad (2)$$

which is derived form eq. (1). Then $P(H)$ is obtained from an analysis in which two distribution functions, $I_1(H)$ and $I_2(H)$, are calculated independently. A detailed analysis is given in ref. 1.

II. Experiments and Results

The amorphous samples of $Fe_{100-x}B_x$ (x=14∿21) alloys were produced by rapid quenching from the melt using a single-roller type quenching apparatus. The samples were ribbons of 1∿2.5mm in width and about 20∿60 μm in thickness. The amorphous state of the samples was by X-ray diffraction. Measurements of the Mössbauer effect have been performed by a conventional method in which the direction of γ-rays from the Pd(^{57}Co) source was perpendicular to the ribbon plane. In the measurements below room temperature, the sample was glued to single-sided transparent tape and was held on a copper holder in a cryostat.

Figure 1 shows the Mössbauer spectra of $Fe_{82}B_{18}$ at several temperatures. As can be seen from the figure, the intensity of the

second and the fifth absorption lines decreases monotonically with decreasing temperature. This phenomenon is the same as that reported in refs. 6-11. All the samples show very similar temperature dependence of the intensity ratio except $Fe_{86}B_{14}$. We define the "anisotropy parameter" f_A of the Mössbauer spectrum by the relation

$$\int_0^\infty I_1(H)dH : \int_0^\infty I_2(H)dH = 1 : f_A.$$

Then the intensity ratio of the observed spectrum can be expressed as $3:f_A:1:1:f_A:3$. The temperature dependence of f_A is shown in Fig.2[5]. If the value of f_A is 4, the magnetization direction is parallel to the ribbon plane, i.e., perpendicular to the direction of γ-rays, while a random distribution of the magnetization direction corresponds to $f_A=2$.

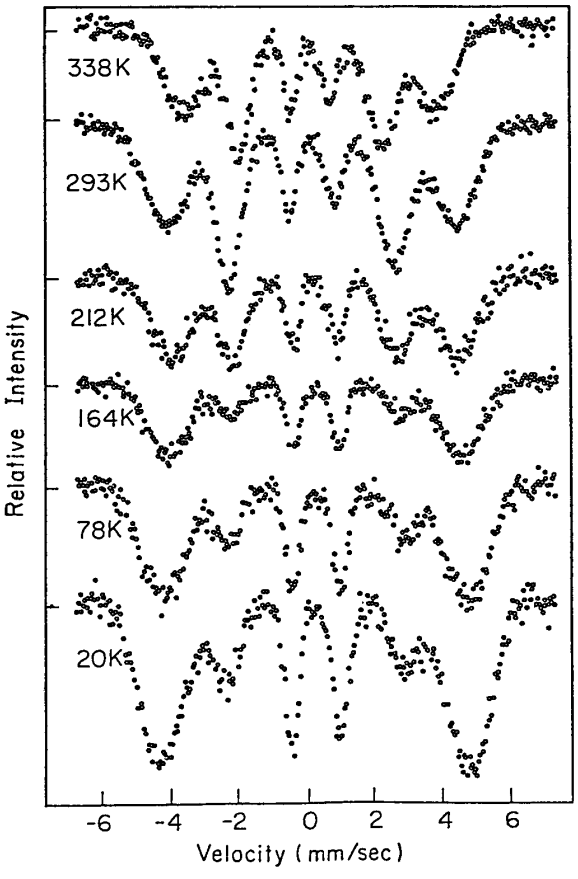

Fig. 1. Temperature dependence of Mössbauer spectrum of the amorphous $Fe_{82}B_{18}$ alloy.

The Mössbauer spectra have been analyzed with the computer by the modified method[1] in which $I_{1,3}(H)$ and $I_2(H)$ are obtainted separately, and $P(H)$ is derived for $I_{1,3}(H)$ and $I_2(H)$ using eq. (2). Figures 3(a) and (b) show $I_{1,3}(H)$, $I_2(H)$ and $P(H)$ obtained from the Mössbauer spectra at room temperature. They are normalized so that $\int_0^\infty P(H)dH : \int_0^\infty I_{1,3}(H)dH : \int_0^\infty I_2(H)dH = 1 : 1 : f_A/3$ in order to make comparison easier. The shapes of all the distribution functions of hyperfine fields are similar to those in many previous reports on the Mössbauer effect of amorphous ferromagnets. The shapes of $P(H)$ and $I_{1,3}(H)$ at room temperature are very similar in all the samples. Figures 4 (a) and (b) show the three kinds of distributions at 20K. The shapes of $I_2(H)$ are different from those $I_{1,3}(H)$ and $P(H)$, having a certain structure in the profile. The shapes of $I_2(H)$ are depressed roughly in the middle position of the peak as compared with those of $I_{1,3}(H)$. As an example of the analysis, observed and

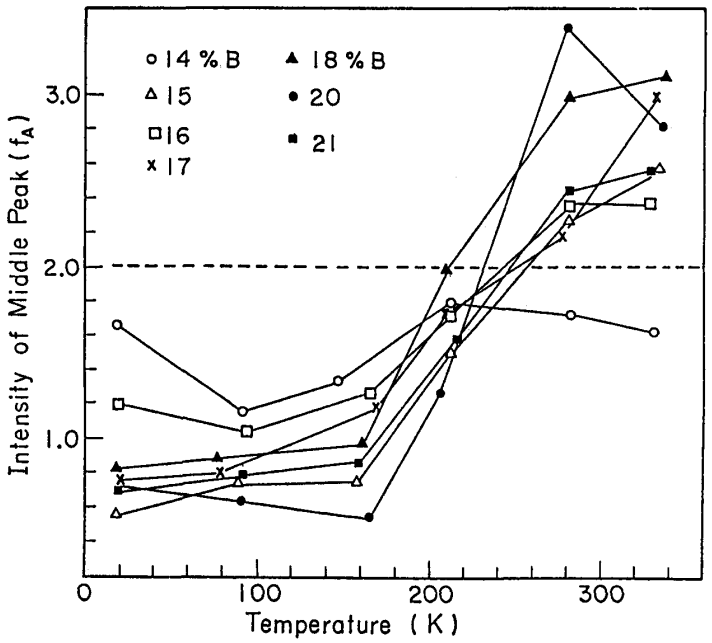

Fig. 2. Temperature dependence of "anisotropy parameter" f_A of the amorphous $Fe_{100-x}B_x$ alloys[5].

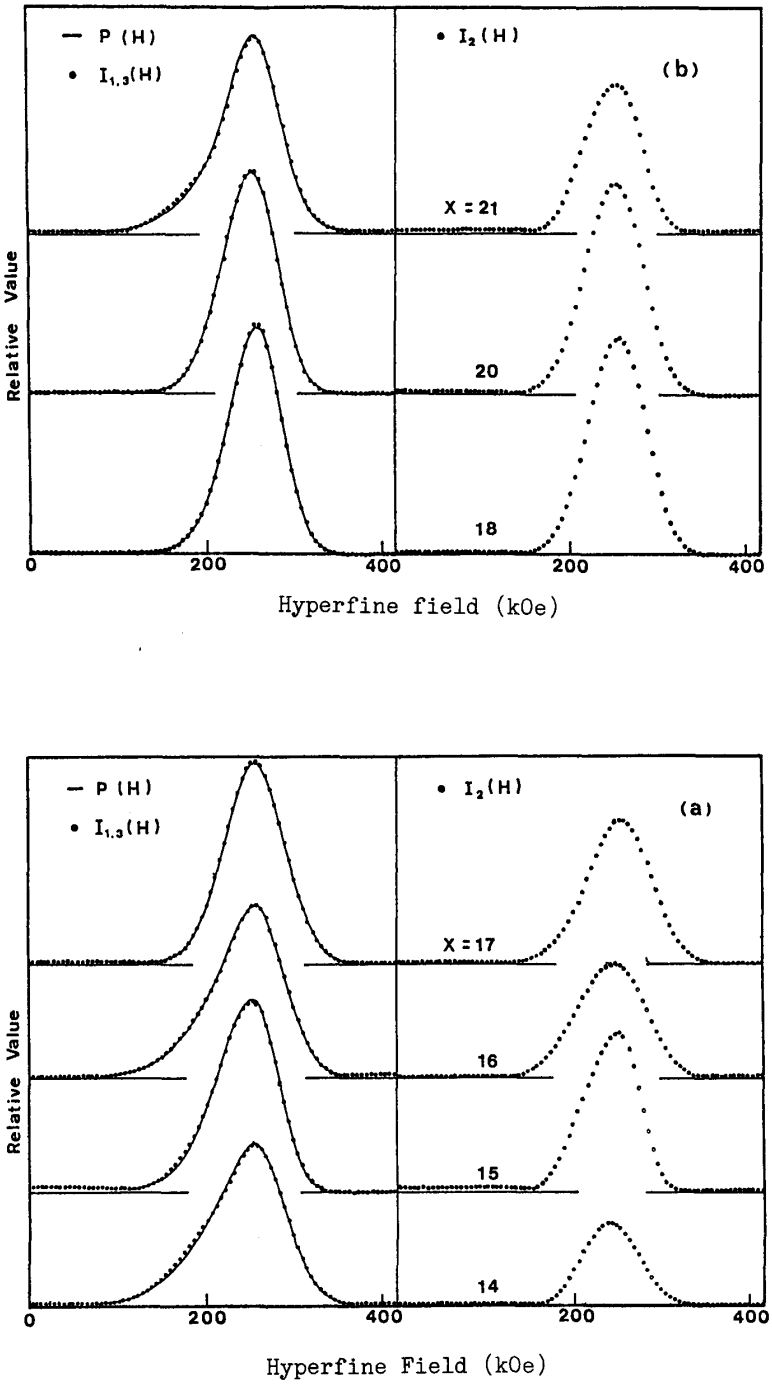

Fig. 3. Distributions of hyperfine fields and line intensities at room temperature.

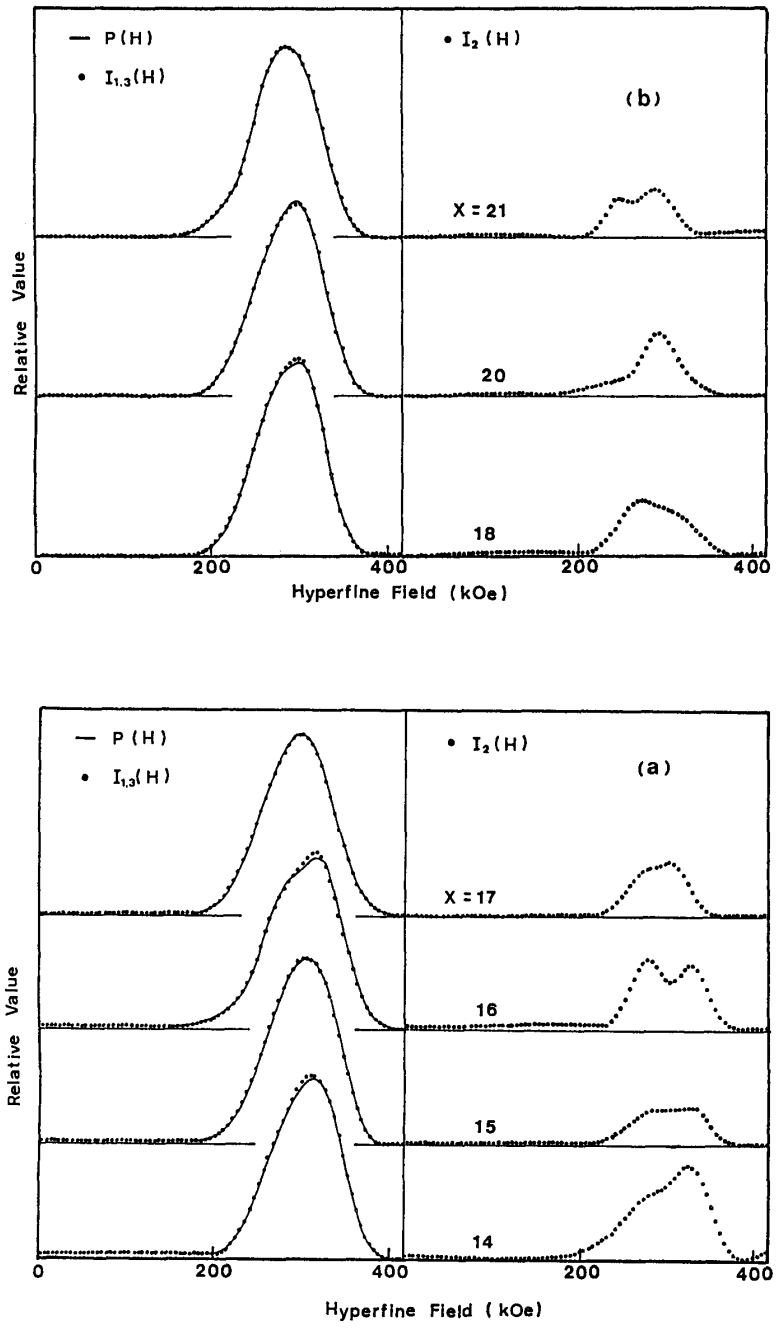

Fig. 4. Distributions of hyperfine fields and line intensities at 20K.

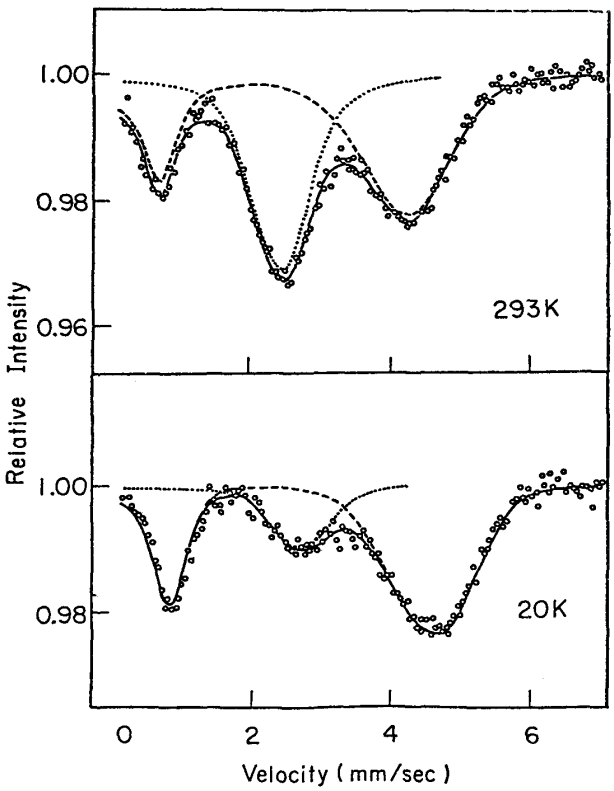

Fig. 5. Observed and calculated spectra of the amorphous $Fe_{82}B_{18}$ alloy[1]. The broken and dotted lines show the spectra calculated from $I_{1,3}(H)$ and $I_2(H)$, respectively. The solid line is the sum of the two calculated spectra.

calculated spectra of $Fe_{82}B_{18}$ are shown in Fig. 5[1]. In the figure, only the right halves of the spectra are shown in order to make observation easier in detail.

III. Discussion

In the present measurements, similar temperature dependence of the spectra to that reported in refs. 6-11 was observed, as shown in Fig. 2. However, the $Fe_{86}B_{14}$ sample did not show a strong temperature dependence of f_A. The spectrum with $f_A<2$ at room temperature indicates that the $Fe_{86}B_{14}$ sample has more volume of maze domains than of 180° wall domains, since the maze domains have a large fraction of their volume with the magnetization perpendicular to the ribbon plane. This

may imply that the $Fe_{86}B_{14}$ sample contains many regions where the domain wall motion is suppressed even though compressive stresses are developted with temperature decrease. It is not obvious, however, that this is so at the composition of 14% boron.

Though the f_A values of all the samples except $Fe_{86}B_{14}$ decrease with temperature decrease, some areas remain where the magnetization directions are parallel to the ribbon plane. The shapes of $I_2(H)$ in Figs. 4 (a) and (b) indicate the distribution of hyperfine fields in these residual regions at 20K, provided that the gradual changes of the directions of hyperfine fields at the domain boundaries can be naglected. The shapes of $I_2(H)$ are very different from those of $P(H)$ at first sight. Since the shapes of $I_2(H)$ are depressed roughly in the middle portions of the curves as compared with those of $P(H)$, it is suggested that Fe atoms which have the hyperfine fields close to the mean value have their magnetization direction greatly tilted away from the ribbon plane. However, the present method of analysis creates the problem that $I_2(H)$ with a small f_A value emphasizes information about that part of the sample where the magnetization direction is parallel to the ribbon plane, but simultaneously gives rise to ambiguity in the calculated results of $I_2(H)$ due to the relative increase of the statistical errors. In order to avoid the errors as far as possible, it is better to discuss the average hyperfine fields, $\bar{H}$ and $\bar{H}_2$, which

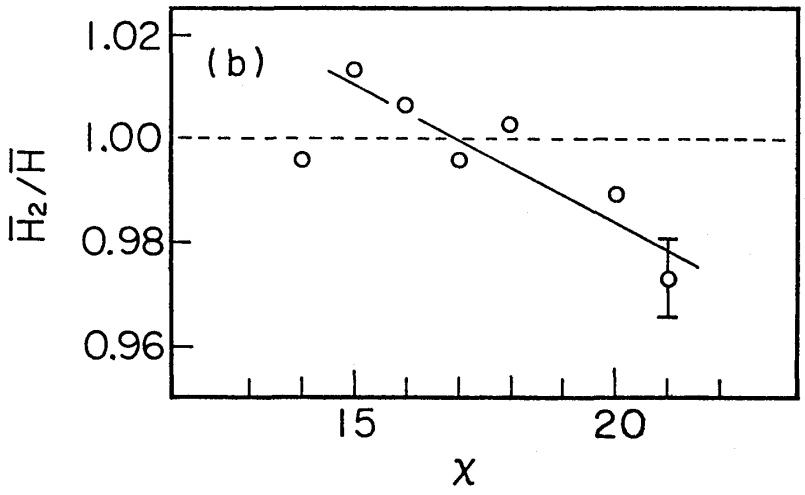

Fig. 6. Ratios of $\bar{H}_2$ to $\bar{H}$ of the amorphous $Fe_{100-x}B_x$ alloys at 20K. The solid line is to guide the eye.

are obtained from $P(H)$ and $I_2(H)$. Figure 6 shows the values of $\bar{H}_2/\bar{H}$ at 20K. The ratios of $\bar{H}_2$ to $\bar{H}$ should be unity if there is no correlation between the magnitude of a hyperfine field and its direction. However, it is clear that $\bar{H}_2/\bar{H}$ decreases with the increase of boron concentration, x, and has the value of unity at $x \simeq 17$. The $\bar{H}_2/\bar{H}$ value at $x=14$, as well as the temperature dependence of f_A, is an exception to this tendency. Many studies have reported that the hyperfine field decreases as the boron concentration increases. It is, therefore, considered that the regions where the magnetization direction changes from parallel to perpendicular to the sample plane under compressive stresses have the boron concentration near to 17% and the residual regions with the parallel direction contain volumes where the boron contents are far from 17%. The boron concentrations of the residual regions where the magnetization directions do not change are lower for $x \lesssim 17$ and higher for $x \gtrsim 17$ than the nominal concentrations. It should be noted that the eutectic point is located at 17% boron in the crystalline Fe-B phase diagram, and that it becomes more difficult to prepare the amorphous samples with the boron concentration higher or lower than 17%. From in-plane torque measurements Takahashi and Kim have claimed that the uniaxial magnetic anisotropy is due to the existence of fine crystallites of Fe_2B in the amorphous $Fe_{81}B_{19}$.[12] Though there is no evidence for the existence of the crystallites in the Mössbauer spectrum, it may be possible that the regions with the direction of magnetization parallel to the ribbon plane at 20K contain fine crystallites which tend to suppress the domain wall motion under compressive stresses.

In conclusion, the amorphous $Fe_{100-x}B_x$ alloys prepared by rapid quenching from the melt have a spatial fluctuation of composition. Though the magnetization of most regions which have boron concentration close to 17% in the sample change direction easily under stresses, the residual regions whose magnetization does not change direction under the same stresses contain volumes in which the boron concentrations are far for 17%.

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References

(1) H. Onodera, H. Yamamoto and H. Watanabe, J. Phys. Chem. Solids, 43 (1982), 1177.
(2) S. Nasu and U. Gonser, J. Phys. (France), 41 (1980), C6-690.
(3) M. Tejeder and B. Hernando, J. Phys. D, 13 (1980), 1709.
(4) M. Takahashi, T. Suzuki and T. Miyazaki, Jpn. J. Appl. Phys., 16 (1977), 521.
(5) H. Onodera, H. Yamamoto and H. Watanabe, J. Phys. (France), 40 (1979), C2-142.
(6) C.L. Chien and R. Hasegawa, J. Appl. Phys., 47 (1976), 2234.
(7) L. Takåcs, Solid State Commun., 21 (1977), 611.
(8) C.L. Chien and R. Hasegawa, Phys. Rev., B 16 (1977), 2115.
(9) C.L. Chien and R. Hasegawa, Phys. Rev., B 16 (1977), 3024.
(10) H. Franke and M. Rosenberg, J. Mag. Mag. Mat., 7 (1978), 168.
(11) A.M. van Diepen and F.J.A. den Broeder, J. Appl. Phys., 48 (1977), 3165.
(12) M. Takahashi and C.O. Kim, Jpn. J. Appl. Phys., 16 (1977), 2061.