研究の目的と問題の設定

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題名 | Origin of low coercivity of (Fe0.75B0.15Si0.10)100-xNbx (x = 1-4) glassy alloys

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Origin of low coercivity of \((\text{Fe}_{0.75}\text{B}_{0.15}\text{Si}_{0.10})_{100-x}\text{Nb}_x\) \((x=1-4)\) glassy alloys

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The density and the magnetization process of the melt-spun \((\text{Fe}_{0.75}\text{B}_{0.15}\text{Si}_{0.10})_{100-x}\text{Nb}_x\) \((x=1-4)\) glassy alloys have been investigated to clarify the origin of low coercivity \((H_c)\). Both \(H_c\) and the difference of the densities between the crystalline and glassy phases, which corresponds to the free volume in the glassy phase, decrease with increasing Nb content. An analysis of the magnetization process based on the law of approach to ferromagnetic saturation reveals that quasi-dislocation dipole (QDD)-type defects are the main sources of elastic stress. The results also suggest that the pinning force for magnetic domain walls generated by one QDD-type defect is independent of the Nb content, but the number density of QDDs decreases with increasing the Nb content. Therefore, it is concluded that the origin of low \(H_c\) of the glassy alloys is the low number density of QDDs which corresponds to the low number density of the domain-wall pinning sites. © 2006 American Institute of Physics. [DOI: 10.1063/1.2158587]

I. INTRODUCTION

In the last decade, a new class of Fe-based glassy alloys with a high glass-forming ability has been found.1,2 These glassy alloys have a large supercooled liquid region \([\Delta T_c=\text{crystallization temperature} (T_c) - \text{glass transition temperature} (T_g)]\) before crystallization combined with good soft magnetic properties. Recently, we reported that the glassy Fe–(Al, Ga)–(P, C, B, Si, Ge) alloys exhibit low coercivity \((H_c)\), though their saturation magnetostriiction constant \((\lambda_s)\) is relatively large.3 We also reported that the origin of low \(H_c\) of the glassy Fe–(Al, Ga)–(P, C, B, Si) alloys is the low number density of the quasidislocation dipole (QDD)-type defects due to the small free volume in the glassy phase.4

In the present study, Nb content dependence of the density and the magnetic properties of the melt-spun \((\text{Fe}_{0.75}\text{B}_{0.15}\text{Si}_{0.10})_{100-x}\text{Nb}_x\) \((x=1-4)\) glassy alloys5 are investigated. The origin of low \(H_c\) of the glassy alloy system is also discussed.

II. THEORETICAL BACKGROUND

In crystalline materials \(H_c\) is determined by dislocations and grain boundaries. In amorphous or glassy materials both kind of defects in the conventional picture do not exist. Nevertheless, the observed \(H_c\) has values of the order of magnitude 0.5–10 A/m which are considerably larger then the expected ones for the intrinsic inhomogeneities \((\approx 3 \times 10^{-3} \text{ A/m})\) or short-range order \((\approx 1 \times 10^{-4} \text{ A/m})\).6 It is therefore suggested that in amorphous alloys exist inhomogeneities acting as strong pinning sites for magnetic domain walls (DWs). These pinning sites are found to correspond to stress sources.

The stress sources are supposed to have their origin in the partial instability of the free volume below the melting temperature. The trapped free volume in an amorphous/glassy phase may exist in dispersed form as in the melt or in the form of agglomerates. However, three-dimensional clusters of vacancy type are supposed to be unstable.7 By a relaxation of the atomic network the vacancy clusters may collapse thus generating planar defects which act as the stress source.6,8 The dispersed free volume is similar to partial point defects with stress fields varying as \(r^{-3}\) (\(r\) is the distance from the stress center) whereas the planar defects are equivalent to (quasi-)dislocation dipoles with stress fields varying as \(r^{-2}\).8 Figure 1 shows a model for formation of the QDDs in amorphous/glassy alloys by agglomeration of the vacancy-type point defects.8,9 The QDDs generate short-range (but longer than DW thickness) stress fields and act as pinning sites for DWs.6,8

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FIG. 1. Schematic two-dimensional model for formation of QDDs in amorphous/glassy alloys by agglomeration of vacancy-type point defects in planar regions (Refs. 8 and 9). The QDD is characterized by dipole width \((D)\), dipole length \((L)\) perpendicular to the drawing plane and an effective Burgers vector \((\vec{b})\).
The type of the stress sources existing in amorphous/glassy alloys was investigated by means of the low of approach to ferromagnetic saturation, \( J = J_1 - \Delta J(H) + \Delta J_{param}(H) \), where \( J \) is the magnetization, \( J_1 \) is the saturation magnetization, \( H \) is the magnetic field, \( \Delta J_{param}(H) \) describes the increase of \( J \) due to the so-called spin wave paraprocess, and \( \Delta J(H) = a_J/H^p \). Since the effect of intrinsic inhomogeneities on \( \Delta J \) is negligibly small, \( \Delta J \) is due to spin inhomogeneities induced by the magnetoelastic interactions between elastic stress (\( \sigma \)) and the magnetization. The inhomogeneity term may be attributed to a certain type of the stress sources as follows:

**quasidislocation dipoles:** \( \sigma \propto r^{-2} \rightarrow H^{-1} \),

**isolated quasidislocations:** \( \sigma \propto r^{-1} \rightarrow H^{-2} \).

It was confirmed that \( \Delta J \) of many ordinary amorphous alloys obeys the \( H^{-1} \)-power law. This means that the QDDs are the main sources of the elastic stress. The range of spin inhomogeneities is governed by the so-called exchange length, \( L_{ex} = (2A/(HJ_1))^1/2 \), where \( A \) is the exchange stiffness constant. The mean dipole width can be obtained by the transition field (\( H_1 \)) from the \( H^{-1} \) law to the \( H^{-2} \) law where \( D = L_{ex} \), i.e., \( D = (2A/(HJ_1))^1/2 \).

Kronmüller and co-workers calculated \( H_c \) of a random distribution of the QDDs with the number density \( \rho_d \) based on the statistical potential theory as follows:

\[
H_c = 12G\Delta V \left[ \frac{\pi \rho_d}{30F\delta} \ln \left( \frac{\pi L_{DW}}{2\delta} \right) \right]^{1/2} J_s,
\]

where \( G \) is the shear modulus, \( \Delta V = DLb \) corresponds to the local volume contraction due to the QDDs, \( F \) is the DW area, \( \delta \) is the WD thickness, \( L_{DW} \) is the domain width, \( L \) is the dipole length, and \( b \) is the length of the effective Burgers vector, respectively.

**III. EXPERIMENTAL PROCEDURE**

The mother alloys were prepared by arc melting the mixtures of pure Fe (4N), B (2N5), Si (5N), and Nb (3N) in an Ar atmosphere. The rapidly solidified tapes with 5 mm in width and 20–30 \( \mu m \) in thickness were prepared by a single-roller melt-spinning apparatus in a reduced Ar atmosphere (20 kPa). Annealing treatment of the samples was carried out with no applied magnetic field in a vacuum. The amorphicity of the as-quenched and annealed samples was examined by x-ray diffractometry. The Curie temperature \( T_C \), \( T_g \), and \( T_x \) were measured with a differential scanning calorimeter (DSC) at a heating rate of 0.67 K/s. \( J_1 \), \( H_c \), and \( \lambda_s \) were measured by a vibrating sample magnetometer, a dc B-H loop tracer, and a three-terminal capacitance method, respectively. The density (\( \rho \)) was measured by the Archimedian method using tetrabromoethane. All the measurements were performed at room temperature.

**IV. RESULTS AND DISCUSSION**

Figure 2 shows \( T_C \), \( T_g \), \( T_x \), and \( \Delta T_x \) obtained from the DSC curves as a function of the Nb content. When \( x = 0 \), no glass transition was observed: i.e., this is not a glassy alloy but an ordinary amorphous alloy. The annealing conditions were determined from the results of DSC, i.e., for 7.2 ks at slightly below \( T_C = 673 \) K = 0.94\( T_C \) for \( x = 0 \) and for 600 s at slightly below \( T_g = 773 \) K = 0.96\( T_g \) for \( x = 1–4 \).

Figure 3 shows the Nb content dependence of \( \rho \) for the annealed amorphous/glassy and crystalline (mother alloys) samples. The density of the crystalline samples is almost constant.
The addition of Nb to the amorphous Fe-B-Si alloy increases monotonously with increasing Nb content. The density difference \( \Delta \rho = (\rho_{\text{crystal}} - \rho_{\text{glass}}) \) shown in Fig. 3 decreases considerably with increasing the Nb content from 2.9% for \( x = 0 \) to 1.1% for \( x = 4 \). Figure 4 shows (a) \( J_s \), (b) \( \lambda_s \), and (c) \( H_c \) of the annealed amorphous/glassy samples as a function of the Nb content. The addition of Nb to the amorphous Fe-B-Si alloy causes the decrease in \( J_s \) and the increase in \( \lambda_s \); however, it also brings the drastic decrease in \( H_c \).

Figure 5 shows \( J \) as a function of \( (\mu_0 H)^{-1} \) (where \( \mu_0 \) is the permeability of a vacuum) for the annealed samples. The demagnetizing-field correction was made considering the sample to be an oblate ellipsoid with the same axial ratio.\(^{10} \) The \( H^{-1} \)-power law behavior of \( \Delta J \) is observed for all the alloys in the range of \( 20 \leq (\mu_0 H)^{-1} \leq 50 \) T\(^{-1} \) (\( 20 \leq \mu_0 H \leq 50 \) mT). In the higher magnetic field range of 180 \( \leq (\mu_0 H)^{-2} \leq 400 \) T\(^{-2} \) (50 \( \leq \mu_0 H \leq 75 \) mT), \( \Delta J \) of all the alloys obeys the \( H^{-2} \)-power law. These results indicate that the QDDs are the main sources of the elastic stress similar to the already reported.\(^4 \) All the alloys have almost the same \( \langle D \rangle \) (\( \approx 15 \) nm), \( b \) (\( \approx 0.05 \) nm), and \( \delta \approx 85 \) nm). These values are almost the same as those of the Fe-(Al, Ga)-(P, C, B, Si) glassy alloys.\(^4 \)

The QDDs are formed by agglomeration of vacancy-type point defects in planar regions. Therefore, \( \rho_0 \) is proportional to \( \Delta \rho / \Delta V \) because \( \Delta \rho \) denotes the amount of the free volume in the amorphous/glassy alloys. Let us further consider that \( L \) is proportional to \( D \). Then all the alloys have almost the same \( \Delta V \). Under the above-mentioned assumptions, \( H_c^p \) can be expressed as follows:

\[
H_c^p = p (F, L_{\text{DW}}) \sqrt[3]{\Delta \rho / \lambda_s}
\]

where \( p (F, L_{\text{DW}}) \) is the prefactor depends on \( F \) and \( L_{\text{DW}} \).

Figure 6 shows observed \( H_c^p (= H_c - H_c^{\text{surf}}) \) times \( J / \lambda_s \) as a function of \( \sqrt{\Delta \rho} \). Here, \( H_c^{\text{surf}} \) denotes the contribution of the surface irregularities to \( H_c \) and is calculated from the surface roughness of the tapes.\(^4,11 \) As shown in Fig. 6, \( H_c^{\text{surf}} / \lambda_s \) increases in proportion to \( \sqrt{\Delta \rho} \). This means that the pinning force for DWs generated by one QDD-type defect is independent of the Nb content, but the number density of QDDs decreases with increasing the Nb content. Since \( H_c \) originates from the elastic stress of QDDs is proportional \( \sqrt{\Delta \rho} \). The origin of low \( H_c \) of the Fe-B-Si-Nb glassy alloys is the low number density of QDDs which corresponds to low number density of the DW pinning sites, same as the Fe-(Al, Ga)-(P, C, B, Si) glassy alloys.

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**TABLE I.** Transition field \( (\mu_0 H^t) \) from \( H^{-1} \) law to \( H^{-2} \) law, mean dipole width \( \langle D \rangle \), length of effective Burgers vector \( b \), and domain-wall thickness \( \delta \) of annealed amorphous/glassy \( \text{Fe}_{0.75} \text{B}_{0.15} \text{Si}_{0.10} \text{Nb}_x \) alloys.

<table>
<thead>
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<th>( x )</th>
<th>( \mu_0 H^t ) (mT)</th>
<th>( \langle D \rangle ) (nm)</th>
<th>( b ) (nm)</th>
<th>( \delta ) (nm)</th>
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**FIG. 6.** Coercivity \( (H_c) \) times saturation magnetization \( (J_s) / \text{saturation magnetocrystalline anisotropy constant} \( (\lambda^2) \) as a function of square root of density differences \( (\Delta \rho) \) between crystalline and amorphous/glassy phases for annealed \( \text{Fe}_{0.75} \text{B}_{0.15} \text{Si}_{0.10} \text{Nb}_x \) alloys.