Anisotropy of Magnetostriction of Functional BCC Iron-Based Alloys

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This paper provides an overview of recent studies on the anisotropy of magnetostriction of functional iron-based alloys with the body-centered cubic (bcc) structure; these are potential ferromagnetic materials for use in actuators and sensors at ambient temperature. The magnetostrictive properties of these functional iron-based alloys (such as Fe–Ga, Fe–Al, and Fe–Ge alloys) are known to strongly depend on the crystallographic orientation. In these functional iron-based alloys, non-Joulian magnetostriction, in which volume is not conserved, was observed; generally, the Joulian magnetostriction in a volume is not altered by magnetic fields. As the magnetostrictive properties of these iron-based alloys are correlated to their elastic properties through the magnetoelastic effect, their elastic properties have also been investigated using single crystals of iron-based alloys. In the present paper, we discuss the characteristic features of the anisotropy of magnetostriction and inverse magnetostriction, which occur when magnetic fields and external stresses, respectively, are applied. In order to clarify the microscopic processes underlying the magnetostriction and inverse magnetostriction, the alterations in the magnetic domains by magnetic fields and external stresses were observed. The results revealed that the magnetic domain structure in the functional iron-based alloys is altered in a complex manner when applied with external fields. For example, it was demonstrated that unique motions of different types of Bloch domain walls are observed with the application of magnetic fields or external stresses along specific directions of single crystals of Fe–Ga alloys. The characteristic features of the motion of the domain walls are likely to correspond to the occurrence of magnetostriiction and inverse magnetostriction, in which magnetic fluxes are induced during alternative vibration. [doi:10.2320/matertrans.MT-M2019146]

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1. Introduction

Pure iron and iron-based alloys with the body-centered cubic (bcc) structure exhibit unique mechanical properties such as large temperature dependence and strain-rate dependence of their flow stresses in plastic deformation. In order to clarify the elementary deformation processes of these alloys, the characteristic features of the motion of dislocations have been frequently discussed.1,2) Typically, work-hardening of single crystals of high-purity iron during plastic deformation have been studied systematically; it was demonstrated that the plasticity and work hardening of high-purity iron strongly depend on the crystallographic orientation. Moreover, the deformation processes of bcc pure-iron have been discussed.3,4) Whereas the various mechanical properties of iron and steel are important in structural materials, the magnetic properties of ferromagnetic iron-based alloys are crucial when these are used as functional materials. Because the magnetic and elastic properties are considerably anisotropic in bcc pure-iron and iron-based alloys because of the magnetoelastic effect, the anisotropy of the magnetostrictive properties are investigated in conjunction with the elastic properties of the iron-based alloys.5–7) For example, the anisotropy of the magnetoelastic properties of functional iron-based alloys such as Fe–Ga, Fe–Al, and Fe–Ge alloys have been studied.8–10)

In particular, functional bcc Fe–Ga based alloys are potential magnetostrictive materials for actuators and sensors as well as for vibration energy harvesters.11,12) This is because various magnetostrictive materials have attracted significant attention with regard to their application in energy harvesting for low power wide area networks (LPWA) of internet of things (IoT).13–15) Thus, it is necessary to understand the fundamental properties of the functional iron-based alloys and relevant alloys as the magnetostriction induced by magnetic fields is complicated owing to the magnetoelasticity of those alloys.

It is generally established that the magnetization process of ferromagnetic materials corresponds to the variations in the size of the magnetic domains according to the preferable magnetic moment. Therefore, the magnetic domains in well-defined specimens are frequently observed to clarify the microscopic processes of the magnetization.5–7) For example, the 180° Bloch domain walls in bcc iron-based alloys such as silicon steels move easily when applied with external magnetic fields; consequently, the size of the magnetic domains is altered. Thus, the magnetization and magnetostriction of ferromagnetic iron-based alloys are discussed based on alterations in the magnetic domains by magnetic fields. In particular, alternative variations in the magnetic domains in the alloys by external fields generate alternative magnetic fluxes through the inverse magnetostriction (Villari effect) of the alloys.

For applying magnetostrictive iron-based alloys to vibration energy harvesting, large-sized single crystals of Fe–Ga alloys have been successfully grown by the Czochralski (CZ) method.16,17) Figure 1 shows an image of two Fe–Ga-alloy single crystal ingots grown by the CZ method; whereas one of these is 100 mm in diameter and 150 mm in length, the other is 50 mm in diameter and 280 mm in length.
The growth method of these single crystals have been established; this is notwithstanding a large temperature difference between the liquidus and solidus temperatures and the phase transformation in solid state Fe–Ga alloys with higher gallium compositions.\(^{18,19}\)

The elastic properties of single crystals of the alloy are strongly dependent on the crystallographic orientation, as illustrated by the anisotropy of Young’s modulus (Fig. 2).\(^ {17}\)

We characterized the magnetostriction and relevant properties of Fe–Ga-alloy single crystals and observed the magnetic domains in the single crystals for a better understanding of the anisotropy of the magnetostrictive properties the Fe–Ga alloys. This fundamental knowledge is necessary to consider the mechanism of vibration energy harvesting and manufacture of advanced devices. In this paper, an overview of the results of the characterization of magnetostriction and the origin of magnetostriction in functional bcc iron-based alloys, which are not only Fe–Ga alloys but also Fe–Al, Fe–Ge alloys, is presented. Moreover, the characteristic features of the anisotropy of the elastic properties and magnetic domain structures of the iron-based alloys in relation to these properties are summarized.

2. Magnetostriction of bcc Iron-Based Alloys

When a magnetic field is applied to ferromagnetic materials at ambient temperature, the shape or dimension of the materials are altered by the magnetostrictive strain along the magnetization direction. This phenomenon is called Joule effect; its characteristic features have been investigated primarily in bcc iron-based alloys. The magnetostriction of cubic materials is generally characterized using the saturation magnetostriction in the \([100]\) and \([111]\) crystallographic directions (\(\lambda_{100}\) and \(\lambda_{111}\)).\(^ {20,21}\) Approximately twenty years ago, it was observed that saturation magnetostriction along the \([100]\) direction (\(\lambda_{100}\)) of single crystals of Fe–Ga based alloys is significantly large.\(^ {9,10}\) The results revealed that the magnetostriction exhibits high anisotropy and that the magnetostriction constant \(\lambda_{100}\) is dependent on the composition. The dependence of magnetostriction on the gallium composition exhibits irregularity between approximately 17 at\% and 29 at\% Ga. This is related to the influence of the composition and heat-treatment on the partial D\(_{03}\) chemical order in the alloys with high gallium composition.\(^ {22}\)

A recent development with regard to magnetostrictive materials is the discovery of positive volume-changes (non-Joulian magnetostriction (NJM)) in Fe–Ga-alloy single crystals.\(^ {23,24}\) In these studies, the magnetostriction along various directions upon the application of magnetic fields along the \([100]\) direction of Fe–17.1 at\%Ga and Fe–26.1 at\%Ga alloys was measured. The magnetostriction is anisotropic, and the strains are evolved in the Fe–Ga-alloy single crystals, (Fig. 3(a)). These alloys exhibited a large longitudinal magnetostriction expansion strain, e.g., approximately 200 ppm, and a small transverse magnetostriction strain under a field applied along the \([100]\) direction. When the field was along the \([110]\) direction, the magnetostriction was approximately 100 ppm of the longitudinal strain. These results revealed that although the magnetostriction is anisotropic in various directions, the angular dependence of the magnetostriction is positive in all directions. That is,
the alloys expand in all directions and increase its volume (NJM); the magnetostriction normal to the alloy plane is assumed to be negligible. These results also indicate differences in the longitudinal magnetostriction along the “easy” [100] and “hard” [110] magnetic axes in the Fe–Ga alloy at room temperature. Meanwhile, two studies have reported that non-Joulian magnetostriction was not detected within experimental errors, in experiments using Fe–Ga-alloy single crystals; however, the experimental conditions here were not identical to those in the previous studies. Where it has been observed that a volume expands in the easy field directions, and the magnetostriction calculated

Whereas it has been observed that a volume expands in the easy field directions, it also provides information on the anisotropy of magnetostriction in iron-based alloys; herein, the magnetostriction is measured along different field directions. Figure 4 shows the measurement geometry of the tri-axial magnetostriction along different field directions; it also provides information on the anisotropy of the magnetostriction. Using this setup, the magnetostriction of an Fe–18 at%Ga-alloy single crystal cut from an ingot grown by the CZ method has been measured by the strain gauge method. The measured magnetostriction was compared with the value calculated based on a theory of magnetostriction for the cubic structure; here, the volume of ferromagnetic materials is assumed to be conserved during magnetostriction (that is, Joulian magnetostriction). In this magnetostriction, the saturation magnetostriction \( \varepsilon_{ij} \) in a cubic crystal is expressed by the following equation:

\[
\varepsilon_{ij} = \frac{3}{2} \lambda_{100} \left( \alpha_1^2 \beta_1^2 + \alpha_2^2 \beta_2^2 + \alpha_3^2 \beta_3^2 - \frac{1}{3} \right) + 3 \lambda_{111} (\alpha_1 \alpha_2 \beta_1 \beta_2 + \alpha_2 \alpha_3 \beta_2 \beta_3 + \alpha_3 \alpha_1 \beta_3 \beta_1) \tag{1}
\]

where \( \lambda_{100} \) and \( \lambda_{111} \) are the intrinsic saturation magnetostriction constants of the cubic material. \( \alpha_i \) and \( \beta_i \) denote the cosines that define the magnetization direction \( i \) and measurement direction \( j \), respectively. While applying the magnetic field along \( i = [100] \) (x-direction) and measuring the magnetization along \( j = [010] \) (y-direction), \( \alpha_x = 1 \), \( \alpha_y = \alpha_z = 0 \), \( \beta_y = 1 \), and \( \beta_x = \beta_z = 0 \). When the magnetic field is applied to an Fe–18 at%Ga-alloy single crystal, the angular dependence of the tri-axial magnetostriction \( \varepsilon_{ij} \) measured along the [100] direction is shown in Fig. 5. The results revealed that the measured magnetostriction had deviated from the values calculated using eq. (1).

A reason for the deviation is likely to be a difference in the volume fractions of the magnetic domains of the [100], [010], and [001] directions; this is because the fractions of the magnetic domains or closure magnetic domains are altered by the internal stress or external stresses, as illustrated in Fig. 6. If certain residual stresses remain in magnetostrictive materials owing to crystalline defects formed during crystal preparation, they produce an inhomogeneous distribution of magnetic domains in the materials. This tri-axial measurement method of magnetostriction has also been applied to the X-ray diffraction method with the values estimated using the theory. These results also demonstrated that the measured magnetostriction is different from the values calculated using the theoretical equations, indicating

Fig. 4 Measurement setup of tri-axial strain magnetostriction of a ferromagnetic cubic single crystal. 24

Fig. 5 Angular dependence of magnetostriction measured along [100] (○), [010] (●), and [001] (▲) directions, and the magnetostriction calculated along [100] (---), [010] (----), and [001] (---) directions in Fe–18 mol%Ga single crystal. 33

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non-Joulian magnetostriction and unequal fractions of the different magnetic domains.

Figure 7 summarizes the magnetostriction constants of Fe–Ga binary-alloy single crystals prepared by the Bridgman and Czochralski methods with respect to the gallium composition. The magnetostriction constants of Fe–Ga-alloy single crystals grown by the Bridgman method are represented by a broken line from references. As the Fe–Ga-alloy single crystals grown by the Bridgman method had been annealed and quenched, the scattering of the magnetostriction are small. Meanwhile, the magnetostriction constants obtained experimentally for the Fe–Ga-alloy single crystals prepared by the CZ method appear to exhibit relatively large scattering in these data; this is likely to be a result of residual stresses in the as-grown alloy single crystals.

In order to understand the origin of the large magnetostriction in Fe–Ga alloys, ab-initio simulation was carried out by several investigators. For example, the atomic structure and magnetostriction of an Fe–Ga alloy with less than 19 at%Ga was estimated using the precise full potential linearized augmented plane wave method. It was demonstrated that the extraordinary enhancement of the magnetostrictive coefficient of the Fe–Ga alloys at low concentrations have intrinsic electronic origins. Using density functional calculations, the effect of nanoprecipitation on the magnetostriction of Fe–Ga alloys was also investigated. The results indicate that the B2-like Fe–Ga clusters undergo marginal tetragonal distortion, whereas D0₃-like Fe–Ga clusters remain cubic in the Fe matrix. It was revealed that B2-like nanostructures produce negative magnetostriction, whereas D0₃-like nanostructures produce small positive magnetostriction in hypothetical inhomogeneous structures. The ground state properties and tetragonal magnetostriction of Fe–Ga alloys with different structures were estimated by the first principles calculation using the density functional theory. It was demonstrated in the study that the magnetocrystalline parameters increase with increasing gallium composition and that the magnetocrystalline energy (MAE) is related with the tetragonal distortion formed by gallium addition.

It should be noted that in the magnetostriction of ferromagnetic alloys, the magnetostrictive properties are affected by stresses. It has been demonstrated in studies on the magnetostriction of Fe–Ga alloys that in practice, the magnetization curve shifts toward the high magnetic-field side when these are subjected to stresses. This magnetization curve shift is nearly proportional to the stress and exhibits a dependence on the crystallographic orientation. In these studies, a model was proposed based on the domain observation that magnetization occurs when a certain domain structure satisfies the minimum free-energy condition; in this model, it is considered that magnetization under a low magnetic field is attained with a balance between the magnetoelastic and magnetization energies without increasing the magnetostatic energy. The influences of stresses on magnetostriction were investigated in previous studies of soft-magnetic materials such as Fe–Si alloys or silicon steels. Therefore, because the magnetic domains as well as the magnetic properties of magnetostriction are considered to be influenced by stresses owing to magnetoelasticity, it is important to characterize the anisotropy of the elastic properties and to observe the magnetic domains in functional iron-based alloys.

3. Elastic Properties of bcc Iron-Based Alloys

3.1 Characteristics of elasticity

In order to clarify the relationship of the magnetostriction of functional iron-based alloys with their elasticity, the elastic properties and related properties of the iron-based alloys have been investigated. After observing the large magnetostriction of Fe–Ga alloys, the elastic constants of Fe–Ga alloys with different compositions less than 20 at% were measured using the continuous-wave method. The measurement method of the elastic constants was similar to that for Fe–Al alloys. It was demonstrated that the shear elastic constant \( c_{44} \) decreases with increasing gallium composition in Fe–Ga alloys; meanwhile, \( c_{44} \) is almost independent of the gallium composition. Typically, the changes in the elastic constants,
c’ = (c_{11} - c_{12})/2 and c_{44}, by addition of an alloying element, which is gallium or aluminum, are demonstrated in Fig. 8.\(^{49}\)

Detailed data on the temperature dependence of the elastic constants of Fe–Ga alloys with different gallium compositions were obtained by using the resonant ultrasound technique.\(^{51}\) These results revealed that the magnetoelastic energy constant \(b_1 = -(3/2)\lambda_{100}(c_{11} - c_{12})\) corresponds to the magnetostriction of Fe–Ga alloys. In order to understand the magnetic anisotropy of Fe–Ga alloys, the magneto-crystalline anisotropy energy constants of Fe–Ga alloys with different gallium compositions were also measured. These anisotropy constants were used to interpret the composition dependence of the magnetization.\(^{32}\)

Furthermore, elastic shear modulus measurements of Fe–(12–33) at%Ga single crystals with and without a magnetic field were carried out by resonant ultrasound spectroscopy in the temperature range between 4 K and 300 K.\(^{53}\) These results indicated that the pronounced softening of the tetragonal shear modulus \(c’\) in Fe–Ga alloys arises from magnetoelastic coupling; the first peak of the tetragonal magnetostriction constant \(\lambda_{100}\) is observed in furnace-cooled Fe–17 at%Ga alloys and in quenched Fe–20 at%Ga alloys. In addition, the second peak in the tetragonal magnetostriction constant \(\lambda_{100}\) in Fe–Ga alloys appears with approximately 28 at%Ga. It was also demonstrated that the shear anisotropy \(c_{44}/c’\) is very large (over ten) in Fe–20 at%Ga (approximate composition) alloys; moreover, structural changes occur in the alloy with approximately 25 at%Ga.

To study the anisotropy of elasticity of Fe–Ga alloys, tensile tests were performed on an Fe–17 at%Ga-alloy single crystal with [110] and [100] tensile axes.\(^{54}\) The results revealed that {110} (111) slip occurs by deformation; this is consistent with the results of pure bcc iron.\(^{3,4}\) The Young’s modulus of the Fe–Ga single crystal was 160 GPa in the [110] loading direction; moreover, the Poisson’s ratio was 0.37 on the (100) face. When the tensile axis was the [100] direction, the Young’s modulus was 65 GPa and Poisson’s ratio was 0.45, on the (100) face. These indicate that the Young’s modulus and Poisson’s ratio are considerably anisotropic in Fe–Ga alloys, depending on the Ga composition. It is noteworthy that a negative Poisson’s ratio or large auxetic behavior is observed on the (100) face under a [110] loading. The large auxetic behaviors of Fe–Ga alloys were experimentally and theoretically investigated in previous studies.\(^{55-57}\) For example, the hysteretic evolution of the volume fractions of magnetic moments in different directions was considered to predict magnetomechanical data along several crystallographic directions of the [110]-oriented Fe–18 at%Ga single crystal.\(^{53}\) The auxetic behavior in Fe–Al alloys was compared with that in Fe–Ga alloys.\(^{56}\) Considering the auxetic behaviors in Fe–Ga alloys, the volume fractions of magnetic domains with different magnetic moments were also estimated by measuring the tri-axial magnetostriction of an Fe–18 at%Ga-alloy single crystal.\(^{58}\)

Moreover, the tetragonal magnetostriction and the magnetostrictive contribution to the Poisson’s ratio in Fe–Ga-based ternary alloys were investigated to understand the unique magnetoelasticity in magnetostrictive materials.\(^{59-61}\) In the study, a 3D magnetoelastic model was proposed based on the general expressions for magnetostriction-induced and stress-induced anisotropies.\(^{59}\) The measured anisotropy was observed to be unequal to the intrinsic anisotropy when the bulk magnetostriction of the material was constrained. Furthermore, a comparative study of the tetragonal magnetostriction constants, (3/2)\(\lambda_{100}\), and magnetoelastic coupling, \(b_1\), of binary Fe–(0–35) at%Z (Z = Al, Ga, Ge, and Si) and ternary Fe–Ga–Al and Fe–Ga–Ge alloys was carried out.\(^{60}\) In this study, the rhombohedral magnetostriction was also evaluated from the elastic constant \(c_{44}\). The results revealed that characteristic variations in the tetragonal magnetostriction are observed for gallium compositions between 15 at% and 20 at%.\(^{60}\)

### 3.2 Magnetoelastic strains

In general, the symmetric strain tensor is used to express elastic strains in cubic materials within the framework of linear elasticity. However, the influences of the non-linear elasticity may not be omitted in ferromagnetic materials owing to magnetoelasticity; this is because the fractions of magnetic domains with different moments are influenced by the magnetic field and stress applied to the materials. Although this phenomenon appears complex, an attempt was made to describe the different contributions to the total strain. For example, the total strain tensor \(\varepsilon^T\) was expressed according to the following equation:\(^{7}\)

\[
\varepsilon^T = \varepsilon^{ext} + \varepsilon^{def} + \varepsilon^Q + \varepsilon^{el},
\]

where \(\varepsilon^{ext}\) is the external strain resulting from the external force and surface force, and \(\varepsilon^{def}\) is the so-called internal elastic strain tensor owing to defect structures such as point defects or dislocations. Here, \(\varepsilon^Q\) denotes the spontaneous magnetostrictive strain tensor owing to spin ordering, and \(\varepsilon^{el}\) denotes the strain tensor related to the inhomogeneous spontaneous magnetic deformation. Thus, the description of the total strain tensor appears to be ambiguous, even if the magnetostriction (expressed as \(\varepsilon^Q + \varepsilon^{el}\)) is estimated from experimental results.
Recently, the influence of magnetoelastic strains on total strains were investigated using Fe–Ga alloys with different Ga compositions; here, the magnetoelastic coupling effect was considered.\textsuperscript{62,63} For example, the total strain ($\varepsilon$) in magnetostrictive materials is dependent on the stress $\sigma$ and magnetic field $H$; moreover, the strain is expressed by superposing the elastic and magnetoelastic strains, as follows: \textsuperscript{53}

$$
\varepsilon(\sigma, H) = \frac{\sigma}{E_s} + \lambda(\sigma, H),
$$

where $E_s$ is the Young’s modulus and $\lambda$ is the magnetostrictive strain. This was obtained from the stress-induced anisotropy and magnetoelastic anisotropy caused by magnetic field and stress. This energy-based model indicates that the total strains in magnetostrictive materials subjected to an external load are non-linear. Designs of actuators and sensors for magnetic materials.\textsuperscript{71–73} It should be noted that while observing the magnetic domains in soft-magnetic materials, the observed magnetic domain structure is occasionally influenced by a magnetic microscanning probe of MFM.\textsuperscript{70} Although these methods have been applied to observe magnetic domain structure in Fe–Ga alloys, the results of MFM should be thoroughly discussed.\textsuperscript{74–80}

In order to discuss the origin of the unique elasticity of Fe–Ga alloys, in general, the phonon dispersion is analyzed using inelastic neutron scattering. The phonon dispersion curves of bcc Fe–(10.8–22.5) at%Ga alloys have been measured as a function of the gallium composition, by inelastic neutron scattering techniques.\textsuperscript{65,66} The results demonstrated that the phonon frequencies of each branch decrease significantly as the gallium composition increases. The gallium composition dependence of the shear elastic constant $c' = 1/2(c_{11} - c_{12})$ was estimated from the slope of a certain branch; it was observed to agree with the results obtained by sound velocity measurements. For the sample with the higher concentration (22.5 at% Ga), new branches appeared; this indicated an effect associated with an increase in the number of atoms per unit cell.\textsuperscript{65} Moreover, in order to understand the origin of the large magnetostriction of Fe–Ga alloys, small angle neutron scattering was used to analyze nanoclusters in the Fe–Ga alloys with the A2 structure.\textsuperscript{67} These also indicated that nanoclusters formed in the matrix enhances the magnetostriction of the Fe–Ga alloys.

Utilizing magnetostriction data of Fe–Ga-alloy single crystals, the vibration characteristics of certain structures can be simulated. The vibration of a gyrosensor has been simulated using a finite element model and magnetostriction data of Fe–Ga alloys.\textsuperscript{68} However, it is observed that microscopic stresses are evolved in the crystalline grains of polycrystalline Fe–Ga alloy in a complicated manner. In a study involving stress analysis in an Fe–18 at%Ga alloy by white synchrotron radiation, it was demonstrated that external stresses are heterogeneously evolved in the crystals.\textsuperscript{69}

4. Observation of Magnetic Domains

It is important to observe the magnetic domains in ferromagnetic materials to better understand the microscopic processes of the magnetostriction and magnetostriction of materials. There are several methods such as the Bitter method, the electron reflection technique, and mechanical microscanning techniques such as magnetic force microscopy (MFM) and magneto-optical microscopy for observing magnetic domains.\textsuperscript{70} These methods have been used for characterizing magnetic domain structures mainly in soft magnetic materials.\textsuperscript{71–73} It should be noted that while observing the magnetic domains in soft-magnetic materials, the observed magnetic domain structure is occasionally influenced by a magnetic microscanning probe of MFM.\textsuperscript{70} Although these methods have been applied to observe magnetic domain structure in Fe–Ga alloys, the results of MFM should be thoroughly discussed.\textsuperscript{74–80}

Because the magnetic domains in soft magnetic materials are negligibly altered by magneto-optical Kerr microscopy, this method has been applied to observe variations in the magnetic domain structure of Fe–Ga alloys subjected to magnetic field and stress. The domain structure on the (001) plane of an Fe–16 at%Ga-alloy single crystal with (100) and (110) orientations was observed (Fig. 9). The microscope contrasts were obtained by the longitudinal Kerr effect at oblique incidence of polarized light in two directions. The images of the initial magnetic domains on the (001) plane of the Fe–Ga alloy without external magnetic field were observed by Kerr microscopy in the X-Kerr and Y-Kerr modes; herein, the illuminating directions of polarized lights were in the x- and y-directions, respectively. These images indicate that the magnetic domains on the surface of the sample can be classified into four types; moreover, 90° Bloch domain walls were frequently observed in this alloy.

4.1 Domain structure by applying a magnetic field

Figure 10 shows the magnetostriction curves of the Fe–Ga-alloy single crystal with (001) plane, obtained by applying increasing and subsequently decreasing magnetic fields along the [010] direction. These results reveal that this single crystal was elongated by the magnetic field along the
[010] direction. Whereas the magnetostrictive strains are marginally altered by magnetic fields up to 250 Oe, they are significantly increased by magnetic fields between 250 Oe and 1000 Oe. The magnetostriction curve obtained by decreasing the magnetic field is almost identical to that by increasing the magnetic field. This is related to the soft-magnetic properties of these alloys. A nonlinear relationship between the magnetostriction and the magnetic field has also been observed, although the nonlinear relationship of the magnetostriction with the magnetic field is dependent on the alloy conditions.

At certain magnetic fields denoted in the above magnetostriction curve, the magnetic domain structures were observed by Kerr microscopy. Figure 11 shows images of sequential magnetic domains observed in the alloy with the (001) plane along the [010] magnetic field direction. When magnetic fields up to 220 Oe was applied to the alloy, the zigzag-like 180° domain walls almost disappeared; meanwhile, the domain structures separated by the stripe-like 90° domain walls remained. At 220 Oe, the magnetostrictive strains were still small, as shown in Fig. 11. These results indicate that the motion of the zigzag-like 180° domain walls negligibly affected the magnetostriction. By applying higher magnetic fields up to 1100 Oe, a single magnetic-domain structure was obtained. It was observed that the magnetic field values in these stages differ marginally across the samples. These differences are considered to arise from the initial domain structure in each sample because the magnetic domain structure is straightforwardly influenced by the sample conditions.

Meanwhile, Fig. 12 shows the magnetostriction curves of the alloy with a (001) plane along the [110] magnetic-field direction; these were measured with increasing and subsequently decreasing magnetic fields. The results reveal that the magnetostrictive strains are small under magnetic fields up to approximately 300 Oe, along the [110] direction; moreover, a large negative magnetostrictive strain appears under magnetic fields between 300 Oe and 1000 Oe. This indicates that this alloy was contracted by the magnetic fields; this may have resulted from the auxetic magnetoelastic characteristics of the Fe-Ga alloys. The magnetostriction curve measured under an increasing magnetic field is similar to that under a decreasing magnetic field, indicating that the magnetic domains are almost reversible under an applied magnetic field.

Referring to the magnetostriction curves of the sample with a (001) plane along the [110] direction, the magnetic domain structures were observed under different magnetic fields, as shown in Fig. 13. When magnetic fields of up to 480 Oe were applied to the sample along the [110] direction, the 180° magnetic domain walls moved to form the magnetic domains separated by the 90° magnetic domain walls. Under an applied magnetic field of over 1000 Oe, the magnetic domain structures separated by the 90° domain walls remained. It is noteworthy that the movement processes of the magnetic domain walls under the magnetic field applied along the [110] direction corresponds to alloy shrinkage. This phenomenon is considered to have arisen from the auxetic elastic properties of Fe–Ga alloys.

4.2 Model of domain structures

It is established that in general, when a magnetic field is applied to soft magnetic materials such as silicon steels, the Bloch magnetic domain walls move to increase the domain
size; here, the magnetization is almost parallel to the applied field. The magnetic domains in Fe–Ga alloys are considered to behave similarly under magnetization, although the shapes of the magnetic domains and the evolution processes of the domains with magnetic fields may differ between silicon steels and Fe–Ga alloys.

The population of 90° domain walls in the Fe–Ga alloys is significant; moreover, the motion of 90° domain walls corresponds to magnetostriction in these alloys. In addition, it is noteworthy that the 90° domain walls moved owing to external stress. This is because characteristic strains remain near the 90° domain walls in ferromagnetic iron-based alloys. It is considered that the strains near the domain walls are caused by the different magnetic moments perpendicular to the two magnetic domains neighboring the 90° domain walls. Meanwhile, there is less strain near the 180° domain walls because the moments of the two domains are opposite at the 180° domain walls. Thus, the magnetic domains divided by the 180° domain walls and 90° domain walls in Fe–Ga alloys evolve with the magnetization along the direction of the applied field. However, this phenomenon may be relatively marginal in the evolution of the fundamental domain structures in grain-oriented silicon steels; this is owing to the small population of 90° domain walls in these silicon steels. Thus, the difference of strain fields between the 180° domain walls in silicon steels and the 90° domain walls in Fe–Ga alloys may be a result of the magnetoelastic properties; this indicated that the softening of the tetragonal shear modulus is more pronounced in Fe–Ga alloys.

Figure 14 illustrates a simple model illustrating the alterations in the magnetic domain structures in common bcc-Fe-based alloys such as grain-oriented silicon steels with low tetragonal shear, when subjected to a magnetic field. Herein, 180° domain walls are frequently observed; moreover, magnetization appears to occur mainly by the movement of 180° domain walls.

Meanwhile, a model of the alteration in the magnetic domain structures in the (001) plane of Fe–Ga alloys applied with magnetic fields along the [010] direction is shown in Fig. 15. Here, the 180° domain walls move in the weak magnetic fields, and then, the 90° domain walls move with the application of stronger magnetic fields. These characteristics of the magnetic domains in Fe–Ga alloys are related to the magnetoelastic properties discussed in previous studies. It is noteworthy that only 90° domain walls are present in Fe–Ga alloys under a moderate magnetic field. This is because magnetic fluxes are generated by the inverse magnetostriction (Villari effect) by external stress; this is utilized in vibration energy harvesting. Therefore, it is important to control the primary movement of 90° domain walls in the inverse magnetostriction using the moderate magnetic field, to efficiently utilize the inverse magnetostriction in Fe–Ga alloys. The importance of the relationship between the magnetostriction and magnetic domain have also been discussed in studies on the other iron-based alloys.

Fig. 13 Sequential images of magnetic domains in Fe–16 at%Ga along the [110] direction, captured in the X-Kerr mode.

Fig. 14 Model showing alterations in magnetic domain structures induced by external magnetic field in general bcc-Fe based alloys.
Besides of the observation of magnetic domains, in order to understand the atomic structure of Fe–Ga alloys, a few studies on the structure analysis\textsuperscript{84--86} were performed. For example, the results showed that atomic short-range ordering is observed in Fe–Ga alloys between 13 and 20.3 at\% Ga for both slow-cooled and quenched samples. In addition, the large magnetostriction in Fe–Co alloys have been also investigated\textsuperscript{87--90} Although it is challenging to clarify the magnetic domains in these Fe–Co alloys, information obtained from observation of the magnetic domains and structure analysis in the Fe–Ga alloys is likely to be effective for designing and controlling the magnetostriction of magnetostrictive materials.

5. Summary

The anisotropy of the magnetostriction in functional iron-based alloys such as Fe–Ga, Fe–Al, and Fe–Ge alloys were overviewed in this paper. Several physical phenomena which is related to the magnetostriction were also reviewed. The magnetostriction depends on the crystallographic orientation and the alloying element composition. Nonetheless, none-Julian magnetostriction, in which the volume is not conserved under a magnetic field, is occasionally detected in the magnetostriction of those alloys.

The anisotropy of magnetostriction is distinctly related to the anisotropic elasticity of iron-based alloys. The tetragonal shear ($c_{11} - c_{12}$) decreases, and $c_{44}$ is almost unaltered with increasing gallium composition. This corresponds to the large magnetostriction of Fe–Ga alloys. The gallium composition dependence of the tetragonal shear is in agreement with the result of phonon dispersion by inelastic neutron scattering.

The results observed by magneto-optical Kerr microscopy provide information on the alteration in the magnetic domains on the (001) plane of Fe–16 at\% Ga alloy single crystals applied with a magnetic field. The magnetic domains are fundamentally classified into magnetic domains based on four magnetic moments on the (001) plane under zero magnetic field. The magnetic domains are separated mainly by 90° domain walls and 180° domain walls. When a magnetic field is applied along the [010] direction of a Fe–Ga alloy, the zigzag-like 180° domain walls move first. After the 180° domain walls almost disappear, the stripe-like 90° domain walls move, by which large magnetostriction occurs.

A model for the influence of a moderate magnetic field on the magnetic domain structure in inverse magnetostriction of the Fe–Ga alloys was considered; this was done because the 90° domain walls move due to the external stress under a moderate field. This indicates that an adequate moderate magnetic field effectively induces alterations in the magnetic flux induced by cyclic stress.

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