Magnetic domains in magnetostrictive Fe–Ga alloys

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Lorentz microscopy was applied to the observation of magnetic domains in iron-gallium (Fe–Ga) alloys. Results did not show any link between the magnetic domains and the magnetostriction enhancement by Ga addition, but did reveal that the drastic decrease in magnetostriction for Fe–31.2 at. % Ga was due to the presence of large scale precipitates. Magnetic domain features did not change in the alloys of A2, D0$_3$, A2+D0$_3$, A2+B2+D0$_3$, and A2+fine scale precipitates. Large scale precipitates within the slow-cooled Fe–31.2 at. % Ga affected both the distribution and wall motion of magnetic domains. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3013575]

Iron-gallium (Fe–Ga) alloys have attracted growing research interest as promising magnetostrictive materials.[1] The origin of the magnetostriction enhancement caused by Ga addition is not completely understood. Recently we have clarified the structural relationship between magnetostriction $(3/2)\lambda_{100}$ and Ga additions by transmission electron microscopy (TEM) and have found that $(3/2)\lambda_{100}$ is strongly phase dependent.[2] Single phase, either A2 or D0$_3$, shows an increase in $(3/2)\lambda_{100}$ with Ga addition. The phase mixture of A2 and D0$_3$ displays a decrease in $(3/2)\lambda_{100}$ with Ga addition, while the mixture of A2, B2, and D0$_3$ shows a slightly higher $(3/2)\lambda_{100}$ than that of single phase D0$_3$. The processes that lead to magnetostrictive strains involve the 90° rotation of magnetic domains under an applied magnetic field. Earlier attempts to measure the magnetic domain structure using magnetic force microscopy (MFM) suggest that the domain structure is highly complex and quite heterogeneous, and it was speculated that magnetic domain structure was correlated directly with the underlying phase distribution and microstructure.[3] More recent MFM studies indicated that the domain structure is highly dependent on surface preparation methods and residual strains present due to the enhanced magnetoelastic coupling of these alloys.[4] However, subsequent studies[2,5] have shown that the length scales of the microstructure and the magnetic domain structure are disparate. To determine if a correlation exists between the observed structural phase and magnetic domain characteristics, it is necessary to examine the magnetic domains on a length scale that is consistent with the phase distribution within the alloys.

An FEI Tecnai-F20XT was employed for Lorentz microscopy. The microscope is equipped with a Lorentz lens, which serves as an objective lens for the observation of magnetic domains. The normal objective lens was usually turned off for the observation. However, the normal objective lens was sometimes slightly excited to generate a magnetic field along the TEM column to force domain wall motion. The magnetic domains were observed via the defocus method. The alloys were single crystals grown by the Bridgman technique[6] and heat treated under various conditions.[7] The TEM samples were prepared by electrojetting. Detailed information is reported in Ref. 2.

Qualitatively, the scale of the magnetic domains is of the order of tens of microns. Except for slow-cooled Fe–31.2 at. % Ga, difficulties arise when trying to quantitatively link the domain size to either Ga concentration or phase distribution. These difficulties are tied to our observations that domain size and type (closure or not) depend on the local environment of the examined region such as TEM sample thickness, distance between adjacent edges (the size of perforation), and cracks around the examined region. Under an applied magnetic field (produced by the normal objective lens), the magnetic domain wall motion was not observed to occur along any specific crystallographic direction.

Figure 1 illustrates the magnetic domains in a slow-cooled (10 °C min^{-1} cooling rate) Fe–10 at. % Ga alloy with a single phase A2.[2] The magnetic domain size increases from the edge to the center of the sample due to sample thickness change. The domain distribution does not change with Ga addition for alloys that are in the single phase A2 region. The slow-cooled Fe–20.1 at. % Ga contains A2 and D0$_3$, where the D0$_3$ phase is about 30–50 nm in size [see Fig. 2(a)].[2] The magnetic domain structure of this alloy [Fig. 2(b)] reveals small magnetic domains around a crack due to its dipole field and larger magnetic domains in the rest of the sample. During the overfocus/defocus change, dark bending contour lines do not change their contrast, while the magnetic domain walls undergo dark/bright contrast change. Therefore, the dark domain walls can be distinguished from
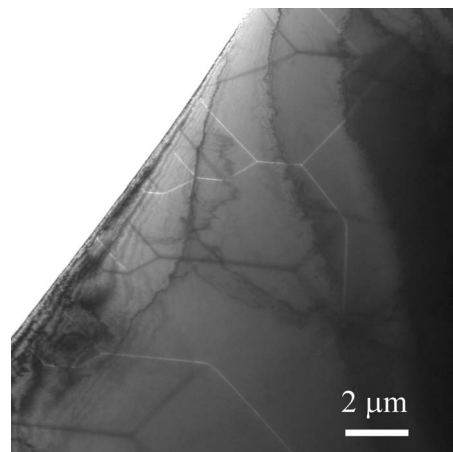

FIG. 1. Magnetic domains of Fe–10 at. % Ga cooled at 10 °C min^{-1} from 1000 °C. The domain walls are straight dark/bright lines.

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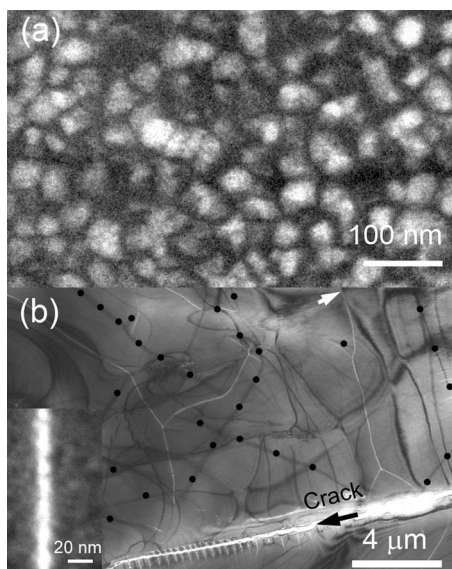
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FIG. 2. (a) Microstructure showing the phase mixture of A2 (dark), D0$_3$ (bright) (Ref. 2), and (b) the magnetic domains of slow-cooled Fe–20.1 at. % Ga. Dots are added to the dark domain walls. A high magnification micrograph revealing the details of a magnetic domain wall at the place marked with a white arrow was inserted in (b).

the bending contour lines. Figure 2 clearly shows that the magnetic domain size is several orders of magnitude larger than the domains of D0$_3$ phase for the alloy. The magnetic domain wall width of 12 nm is about one third to one fifth of the average D0$_3$ phase domain size. The smooth motion of the magnetic domain walls under an applied field indicates that they are not pinned by the D0$_3$. A similar finding is observed for Fe–25 at. % Ga, which is single phase D0$_3$ under a slow-cooled condition and composed of A2, B2, and D0$_3$ when quenched from 1000 °C.[2] As was the case with single phase A2, neither the single phase D0$_3$ nor the phase mixture of A2, B2, and D0$_3$ had any influence on the magnetic domain distribution or on the domain wall motion. No influence on either domain distribution or domain wall motion was observed for a slow-cooled Fe–29.9 at. % Ga with nanoprecipitates on antiphase boundaries.[2] The similarity in the magnetic domain distribution of these alloys is not surprising given the structural similarity of A2, B2, and D0$_3$ phases and the fact that the differences of magnetocrystalline anisotropy between ⟨100⟩, ⟨110⟩, and ⟨111⟩ directions are negligible for the phases.[8] In addition the axis of maximum magnetostriction is the same for all three phases.[8]

In contrast to the above results for alloys consisting of phases with related and comparable crystal structures, significant effects due to secondary phases on both magnetic domain distribution and wall motion were found in an Fe–31.2 at. % Ga under the slow-cooled condition. This alloy consists of precipitate-free regions and regions with large scale precipitates [Fig. 3(a)]. The former regions show large magnetic domains while the latter show small domains [Fig. 3(b)]. The dark short walls for the small domains are almost invisible due to their low contrast. No precipitates were found to contain domains or domain walls. The domain walls in the precipitate-free regions are more mobile than those in the precipitated regions under the magnetic field generated by the normal objective lens and are liable to be pinned by the precipitates (Fig. 4). Figure 4(a) shows a pair of bright (A) and dark (B) long domain walls in a precipitate-free
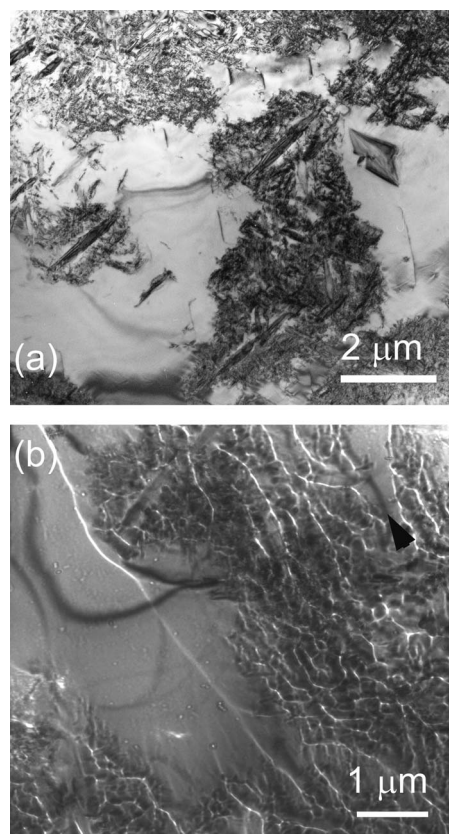

FIG. 3. (a) Bright-field image of slow-cooled Fe–31.2 at. % Ga and (b) Lorentz microscopy image showing the magnetic domain structure. One dark domain wall is arrowed. Slight surface contamination is visible as dots in the precipitate-free regions in the Lorentz microscopy image but invisible in the conventional bright-field image.

region and short domains walls in a precipitated region under condition of nearly zero field. When the normal objective lens was continuously excited [Figs. 4(b) and 4(c)], the long walls (particularly the bright wall) moved significantly from their original positions, while the short walls moved very little. A notable phenomenon is that the mobile long walls were found to be pinned by the precipitates, as shown in Fig. 4. For the samples containing less than 29 at. % Ga, the long domain walls were always mobile and easily driven to disappear (as the sample saturated). The above results suggest that the size of the secondary phase plays an important role in determining not only the magnetic domain structure but the domain wall mobility as well.

Our results reveal that the magnetic domains in the alloys of interest for magnetostrictive applications show no relationship with the underlying microstructure and therefore, provide little insight into the origin of the magnetostriction enhancement due to Ga addition. This suggests that the enhancement may come from an intrinsic source, e.g., the electronic interaction between Ga and Fe atoms. However, the proposed intrinsic model[9] is not consistent with the experimental results.[10] This inconsistency may stem from incorrect structural models (see the models used in Ref. 9 and experimentally identified phases in Ref. 2), limited number of atoms used in the computation, or other reasons. On the other hand, the current findings do not necessarily support the extrinsic model[11,12] where single or multivariant tetragonally distorted nanosized D0$_3$ domains result in macroscopic strains by field-induced variant reorientation. The large
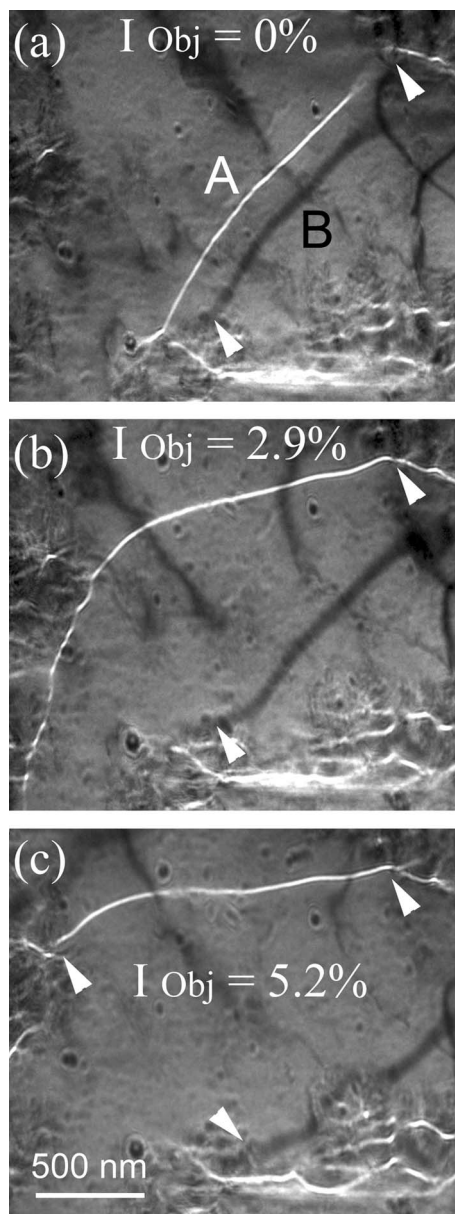
FIG. 4. Domain wall motion of slow-cooled Fe–31.2 at. % Ga under a magnetic field produced by the slightly excited normal objective lens. The lens current I_{obj} is shown in each micrograph. The locations where the magnetic domain walls were pinned are arrowed. At the bottom-right corner of the micrographs are short domain walls which are less mobile.

anisotropy. This cannot be the case since anisotropy measurements of Fe–Ga alloys have shown the anisotropy constant K_1 to be small ($\sim 10^4$ J/m^3) at low Ga compositions and near the solubility limit the anisotropy falls rapidly through zero and becomes negative.[13,14] Therefore the tetragonally distorted structural regions (clusters) are apt to be a single variant whose reorientation upon an applied field would be limited to the magnetic domain rotation concurrent with the matrix in which it is embedded.

Furthermore, the magnetic domains in bulk samples are expected to be larger than those observed in thin-foil TEM samples. A thin-foil sample has a large surface area where the dipole field is strongest.[15] This increase in magnetostatic energy due to sample geometry is then minimized by a reduction in magnetic domain size.

length scale of the magnetic domain structure, several orders of magnitude larger than the structural scale, suggests that a multivariant state would not be stabilized within a single magnetic domain unless there was large magnetocrystalline

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