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Magnetic force microscopy investigation of the static magnetic domain structure and domain rotation in Fe-*x* at. %Ga alloys

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Magnetic domain structures and domain rotation process in Fe-*x* at. %Ga (x=12, 19, and 25) alloys have been studied by magnetic force microscopy. Our results show that the missing connection between magnetic domains and the underlying microstructure [Xing and Lograsso, Appl. Phys. Lett. **93**, 182501 (2008)] is (i) a perpendicular magnetic anisotropy; and (ii) domain irregularities/fluctuations induced by a quenched random-field condition: both of which may have direct relations with dispersed low-symmetry nanoprecipitates [Cao, *et al.*, Phys. Rev. Lett. **102**, 127201 (2009)].
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Galfenol (Fe–Ga) alloys have attracted much attention due to combined merits of high magnetostriction, low saturation fields, and good mechanical properties.[1] Recently, it was shown that quenched Fe-*x* at. %Ga (x=5 ~ 33) alloys have two saturation magnetostriction λ_{sat} peaks: one at x =19 that approaches 400 ppm and another at x=28 of about 450 ppm: both over tenfolds that of pure α-Fe crystal.[2] The results demonstrate that the magnetostriction of α-Fe can be dramatically enhanced by the substitution of nonmagnetic elements. A similar finding was previously reported by Hall, who found that the addition of Al to α-Fe enhanced λ_{100} by a factor of ~4.[3]

The origin of giant magnetostriction in Galfenol has been controversial for some time. Initially, it was believed that the enhanced magnetostriction was related to the presence of Ga–Ga atomic pairs along the [100] direction, which acted as magnetic and magnetoelastic defects.[4] More recently, a structurally heterogeneous model was proposed,[5,6] where the giant magnetostriction of Galfenol is extrinsic and caused by a macroscopic strain generated by a magnetic-field-induced reorientation of tetragonally-distorted $L1_2$ nanotwins within an A2 matrix. The essential aspects of this model are supported by both high resolution transmission electron microscopy (i.e., presence of DO_3-like nanoprecipitates in an A2 matrix[7,8]) and neutron diffuse scattering (i.e., slitting of [300] reflection of as-grown Fe-19 at. %Ga, indicating tetragonal or lower symmetry[9]). However, a recent combined Lorentz microscopy and transmission electron microscopy study has indicated that the magnetic domains have no relation with the underlying microstructure.[10] It was argued that the very different scales of magnetic domains and nanoprecipitates require a large magnetocrystalline anisotropy, which seems absent in Galfenol following investigations by Rafique *et al.*[11]

In this paper, we have employed magnetic force microscopy (MFM) to study the static magnetic domain structure and domain rotation process under step-increases in the magnetic field applied to Fe-*x* at. %Ga crystals. Three compositions of x=12 (in the A2 phase region), x=19 (on the boundary of A2 and A2+DO_3), and x=25 (in the A2+DO_3 region) were selected for study. Combined with recent transmission electron microscopy[9,10] and neutron scattering[11] studies, we show that the keys to bridging the magnetic domain structure with the underlying microstructure are the perpendicular anisotropy energy and domain inhomogeneities/fluctuations during domain rotation, both of which are directly related to dispersed nanoprecipitates.[7–9]

Crystals used in this investigation were grown at Ames Laboratory by a Bridgman method as previously described.[12] The crystals were annealed at 1000 °C for 168 h, with heating and cooling rates of 10°/min, after which the crystals were considered to be in the as-grown state. All crystals were of dimensions $10\times10\times2$ mm^3. The (001)-oriented 10×10 mm^2 faces were polished down to a 0.25 μm finish. Careful investigations were performed by MFM using a Vecoo DI 3100a scanning probe microscope employing silicon cantilevers with commercial MESP tips.

Figure 1 shows the magnetic domain structures on the surfaces for various Fe-*x* at. %Ga crystals. Magnetic flux-closure domains can be seen for Fe-5 at. %Ga[13] in Part (a): which are similar to those of pure Fe whiskers[14] or Fe-3 wt %Si crystals.[15] The width of these domains was about 4 μm. Such closure domains minimize the demagne-

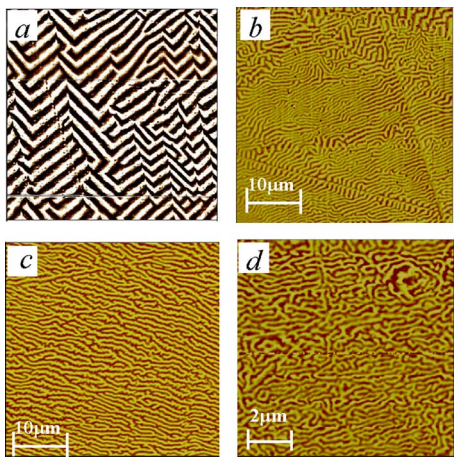

FIG. 1. (Color online) Magnetic force images of the domain structures of Fe-*x* at. %Ga, (a) x=5, cited from Ref. 13 (the image size is 100 $\times$100 μm^2), (b) x=12, (c) x=19, and (d) x=25.

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tization and magnetocrystalline anisotropy energies. With increasing Ga content to 12 at. %, two distinct features can be found in part (b): these are (i) stripelike magnetic domains became the dominant morphology and (ii) the domain width decreased dramatically from ~4 to ~0.7 μm. For Fe-19 at. %Ga, stripe domains remained the dominant morphology, however the stripes were shorter and less smooth than for $x=12$. Upon increasing the Ga content to 25 at. %, closure domains again became apparent, but they were much finer and more irregular than those in Fig. 1(a).

The formation of stripelike domains is generally attributed to a perpendicular magnetic anisotropy energy (K_u).[15] Such perpendicular anisotropy tends to align magnetic domains normal to the sample surface, which consequently increases the demagnetization energy (K_d). If $K_u < K_d$, stripelike domains form to minimize the total energy, which, for example, has been reported in amorphous ribbons with internal stress.[15] In addition, we note in the literature that stripelike domains have also been observed in NiMnGa shape memory alloys[16] and in Fe–Pd alloys.[17]

The perpendicular magnetic anisotropy we describe here is different from the magnetocrystalline anisotropy (K_1 and K_2) in a cubic structure. In fact, Rafique *et al.*[11] reported that K_1 decreased gradually with increasing Ga for $5<x<25$. Rather, K_u may be considered as a raised magnetoelastic energy barrier against domain rotation toward the direction of a magnetic field. Previous magnetization studies have shown higher saturation magnetic fields with increasing x.[18,19] In addition, by comparing the magnetostriction of quenched and slow-cooled samples, Clark *et al.*[19] concluded that quenching increased the magnetostriction at the expense of increased fields required for saturation. Such an energy barrier may be explained in two (or more) possible ways: (i) a localized strain associated with defects or microstructural inhomogeneities both of which can be described by the Kersten's theory[20] or (ii) a quenched random-field associated with an underlying strong heterogeneity that locally breaks symmetry following the Imry–Ma arguments,[21] which might then act as magnetic or magnetoelastic defects.[4] Following Kersten's theory, defects and microstructural inhomogeneities can serve as pinning forces to impede domain motion, and thus increase coercivity;[20] however, previous magnetization measurements of Fe-x at. %Ga have shown a small variation of coercivity Hc with increasing x.[18,19] Accordingly, Kersten's theory is not appropriate to explain the raised magnetoelastic energy barrier.

What could be a probable source, though, of a quenched random field? Possibly, clusters of short-range Ga-Ga pairs along the $\langle 100 \rangle$.[4] Or, the dispersion of nanoprecipitates with a local tetragonal symmetry coherently embedded in an A2 matrix,[9] which have been shown to have a c/a ratio of 1.04. The c-axis of said nanoprecipitates tends to align perpendicular to that of the remanent magnetization (i.e., the magnetic domain orientation),[7–9] and thus could be an important source for the perpendicular anisotropy. Furthermore, with increasing Ga content for $10<x<23$, the volume fraction of nanoprecipitates has recently been found to increase,[8] even though their size was nearly constant. Thus, this plausible source of random-field perpendicular anisotropy would increase with Ga content on approaching the boundary between A2 and A2+DO_3 phase fields near $x=19$: potentially explaining the evolution toward a stable stripelike domain state, over a closure one for $10<x<23$. However, for $x \geq 25$, a fully-ordered cubic DO_3 phase is known to form;[22] and thus, magnetic domains tend to form in a flux-closed patterns to minimize the demagnetization energy.

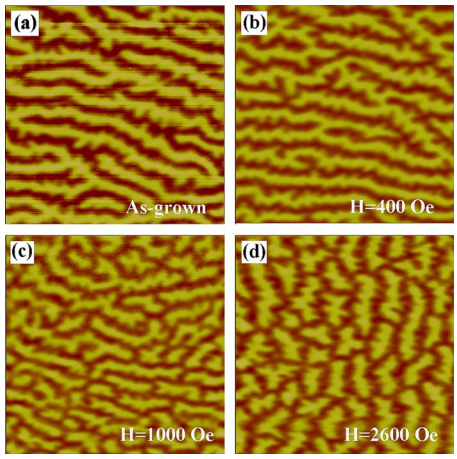

FIG. 2. (Color online) MFM images of Fe-19 at. %Ga upon applying a step-increased field normal to the sample surface, (a) as-grown state, (b) $H_\perp = 400$ Oe, (c) $H_\perp = 1000$ Oe, and (d) $H_\perp = 2600$ Oe.

Next, we studied the domain rotation upon applying a step-increased magnetic field normal ($H_\perp$) and parallel ($H_\parallel$) to the surface of a Fe-19 at. %Ga sample. It should be noted that the demagnetization field was not considered in either case. Figure 2 shows MFM images of 10×10 μm^2 for the sample upon applying $H_\perp$ to the surface. With increasing $H_\perp$, we can observe that (i) the size of the domains decreases and (ii) there are increased irregularities or fluctuations in the domain morphology. The averaged domain width decreased from about 500 to 400 nm upon applying $H_\perp = 400$ Oe. With further increase of magnetic field, the average domain length became increasingly smaller: about 10 μm for $H_\perp = 400$ Oe [see Fig. 2(b)], 3 μm for $H_\perp = 10^3$ Oe [see Fig. 2(c)], and <2 μm for $H_\perp = 2.6 \times 10^3$ Oe [see Fig. 2(d)]. The shrinkage of domain width can be explained by a classical closure-type domain argument. Upon applying $H_\perp$ to the sample surface, the elongation of closure domains is confined by two main domains, resulting in an increase of the magnetoelastic energy.

To better illustrate the increase of domain irregularities with $H_\perp$, we obtained two-dimensional fast Fourier transformations (2D-FFTs) of the four MFM images in Fig. 2. The log intensity plots of these FFT images are shown Fig. 3. As can be seen, the stripelike domains in Fig. 2(a) have a corresponding twofold symmetry in the 2D-FFT images of Fig. 3(a). Whereas, the domain irregularities resulted in a fourfold symmetry with increasing $H_\perp$. These domain irregularities also had a modulation length of ~210 nm, whose period did not noticeably change with increasing $H_\perp$, although the intensity did increase notably with $H_\perp$. Moreover, our analysis revealed that the distribution of domain irregularities in Fe–Ga crystals have preferred [100] and [010] orientations upon applying a magnetic field along the [001] direction, which is consistent with the reported tetragonal symmetry by Cao *et al.*[9] Please note that although the scale of the domain irregularities is larger than that of the dispersed nanoprecipitates,[7–9] it should be considered as the strain field scale around the nanoprecipitates. In addition, the resolution in the MFM images may be partially limited by the magnetic tip radius (~40 nm).

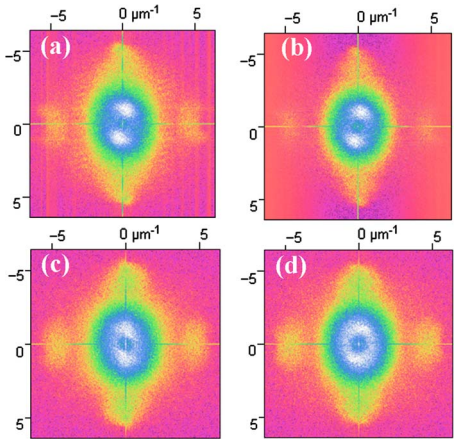

FIG. 3. (Color online) Log plots of MFM images of Fe-19 at. %Ga after 2D-FFT, (a) as-grown, (b) $H_{\perp}=400$ Oe, (c) $H_{\perp}=1000$ Oe, and (d) $H_{\perp}=2600$ Oe.

Finally, Fig. 4 shows the magnetic domain rotation process upon applying $H_{\parallel}$ to the surface of a Fe-19 at. %Ga crystal: right column with an image size of 40×40 μm^2, and left column with a higher resolution of 10×10 μm^2. These images with $H_{\parallel}$ were obtained just after those with $H_{\perp}$ in Fig. 2 where a large number of domain irregularities were found. With increasing $H_{\parallel}$, we observed that (i) the stripelike domain length increased and (ii) the domain irregularities were notably suppressed. The images clearly demonstrate that $H_{\parallel}$, following $H_{\perp}$, results in a near recovery of the original stripelike domain morphology.

Our findings of an inhomogeneous magnetization rotation, where very fine scale irregularities within conventional magnetic domain structures are nucleated under reverse or perpendicular bias, are conceptually similar to recent investigations of Fe-19 at. %Ga using small angle neutron scattering, where Wuttig *et al.*[23] have shown the presence of magnetic nanoregions with characteristic size of about 10–15 nm. These results, in conjunction with our MFM images, provide insight into an unorthodox magnetic domain rotation mechanism, where rotation occurs via domain nonuniformities, rather than by a homogenous rotation of the magnetization vector. Such a breaking of the translational invariant symmetry of the magnetic domains under field, and the creation/nucleation of magnetic nanoclusters with reverse or perpendicular orientations, are signatures of rotation in the presence of strong quenched disorder.[21]

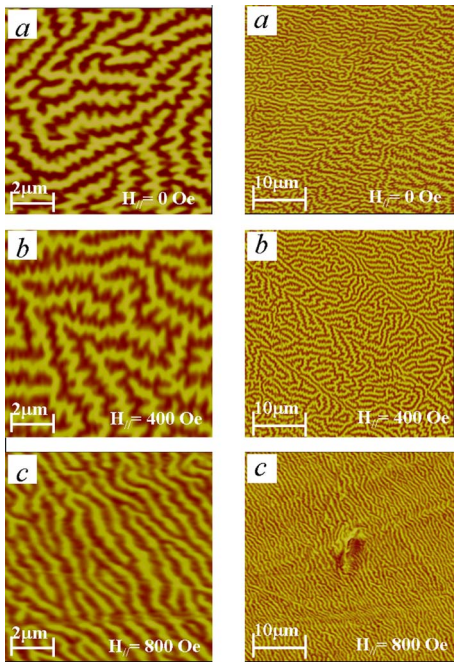

FIG. 4. (Color online) MFM images of Fe-19 at. %Ga upon applying a step-increased field parallel to the sample surface. The sample has been applied a field normal to the surface before MFM investigation.

In summary, the magnetic domain structures of Fe-x at. %Ga ($x=12$, 19, and 25) have been studied by MFM. It was shown that stripelike domains develop with increasing Ga content due to an increasing perpendicular anisotropy. During domain rotation under $H_{\perp}$, increased domain irregularities were observed, which indicate the presence of a quenched random-field, presumably from dispersed low-symmetry nanoprecipitates embedded in a cubic matrix.

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