Micromagnetism of Epitaxial Fe(001) Elements on the Mesoscale

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The size and orientation dependent micromagnetic structures of epitaxial Fe(001) thin film elements with in-plane anisotropy are reported. A transition from single domain to multidomain remanent states is observed upon reducing the element size beneath $\sim 50\ \mu$m, indicating that the *in-plane* dipolar field becomes competitive with the magnetocrystalline anisotropy at this size. Because of this competition, distinct micromagnetic structures arise according to the orientation of the element edges. The epitaxial elements are of high structural quality allowing the micromagnetic behavior to be controllably modified. [S0031-9007(97)02363-6]

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Epitaxial magnetic thin films with controllable magnetic anisotropies provide the experimentalist with an opportunity to study the interplay between dipolar (shape) and magnetocrystalline or interface anisotropies. In the case of ultrathin Fe/Cu(001) [1], Fe/Ag(001) [2], and Co/Au(111) [3,4] films, for example, the perpendicular spin orientation favored by the interface anisotropy is overwhelmed by the dipolar energy as the film thickness is increased, i.e., the spin reorientation transition (SRT) occurs. The question of how the micromagnetic structure evolves with thickness, i.e., how it is determined by the competition between interface anisotropy and dipolar interactions, is important in gaining an understanding of how this transition occurs, and the domain structure which forms in the vicinity of the SRT has therefore received much attention recently [1–6]. However, so far, the related question of whether *in-plane* dipolar fields can (and if so how) compete with strong magnetocrystalline anisotropy in determining the magnetic domain structure in epitaxial thin films has not been addressed. For a continuous epitaxial thin film with in-plane magnetization, a single domain state is predicted as the demagnetizing constant approaches zero [7,8]. Such a single domain state has been observed in epitaxial Fe/Ag(001) films [9], Co/Cu(001) films [10], and Fe/GaAs(001) films [11,12]. However, for an epitaxial thin film with in-plane anisotropy, can magnetic domains be created by decreasing the film in-plane dimensions? Also, and more importantly, *how* do the micromagnetic structures and microscopic reversal processes evolve with film in-plane dimensions and orientation, i.e., *how* are they determined by the competition between in-plane dipolar interactions and magnetocrystalline anisotropy? In analogy to the studies of the SRT, it is expected that novel micromagnetic phenomena may be observed by decreasing the film lateral dimensions, i.e., making mesoscopic epitaxial structures (elements). These epitaxial elements with controllable magnetic properties may also offer a range of applications [13].

Although a number of experimental investigations have been carried out on the micromagnetic structure of thin polycrystalline elements [14–18], to our knowledge no micromagnetic studies have yet been reported for epitaxial thin film elements. The micromagnetic structures of epitaxial elements are expected to be substantially different from the magnetic domain structures of polycrystalline elements in which the magnetocrystalline anisotropy is negligibly small. Furthermore, it has been found that the domain structure in polycrystalline elements is greatly influenced by defects [14]. Thus epitaxial single crystal thin elements with high structural quality are necessary for studying how the micromagnetic structures are determined by the interplay between in-plane inhomogeneous dipolar fields and magnetocrystalline anisotropy.

In this work the in-plane dimension and orientation dependence of domain structures and microscopic reversal processes in 150 Å thick epitaxial Fe(001) elements was studied for the first time by Lorentz transmission electron microscopy (TEM). The high spatial resolution of Lorentz TEM makes it a desirable technique for studying domain structures in small magnetic elements. However, due to the difficulties of preparing Lorentz TEM specimens of epitaxial elements, this technique has not been used so far to characterize magnetic epitaxial elements. In this work we used a selective chemical etching technique developed recently to prepare electron-transparent window specimens suitable for Lorentz TEM [11]. The procedure for epitaxial Fe film growth can be found in Ref. [11]. The film thickness was selected at 150 Å since for 150 Å thick epitaxial Fe films, the fourfold magnetocrystalline anisotropy is well developed and their micromagnetic structures are well understood from our earlier studies [11,12]. Before fabricating the elements, the magnetic anisotropies of the continuous Fe film were determined by magneto-optical Kerr effect (MOKE) vector magnetometry. MOKE

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measurements show that the original Fe(001) film has a predominant in-plane fourfold anisotropy with its easy axes parallel to the in-plane ⟨100⟩ directions as expected for an epitaxial bcc Fe film. Epitaxial square Fe elements with their edges parallel to the ⟨100⟩ or ⟨110⟩ directions and different size were fabricated using a combination of optical lithography, metallization, ion milling, and reactive ion etching techniques. Through optical lithography the design patterns were transferred from an optical mask to the resist layer, spun on the sample prior to the lithography. Then metallization and lift-off were performed. In these processes a Ti(500 Å)/Cr(20 Å) bilayer was deposited on the patterned resist layer, and then the resist was dissolved away, leaving only the patterns of Ti/Cr on the Fe film. We used ion milling to remove the Fe film surrounding the Ti/Cr masks to obtain square Ti/Cr/Fe elements. Finally, the Ti was removed by reactive ion etching using a combination of CF_4 and O_2 gases. A TEM bright field image of an epitaxial Fe element is shown in Fig. 1. The dark bands shown in this image are the bend contours arising from the single crystal GaAs membrane. It can be seen that the edges of the element are straight and smooth.

In this study both the Foucault and Fresnel modes of Lorentz electron microscopy were used [19]. The remanent domain states of Fe elements with their edges parallel to the ⟨100⟩ easy directions were investigated firstly. In the Lorentz electron microscope, an initial magnetic field H_i 120 Oe was applied along one of the ⟨100⟩ directions as determined from the diffraction pattern. In this field, all the elements of different size supported a single domain configuration. In the remanent state, it was found, however, that for elements of different size the domain structures are very different. Figure 2 shows remanent Fresnel domain images and corresponding magnetization schematics of three square Fe elements with side length $a = 55$, 30, and 12 μm. From Fig. 2 it can be seen that the 55×55 μm^2 element at remanence is almost in a single domain state much the same as for a continuous epitaxial Fe film [11]. However, fine spike domains are observed to form at the element edges perpendicular to the initial applied field. Figure 2(b) shows that as the size of the element decreases the edge domains extend. For the element with $a = 12$ μm the edge domains extend throughout the whole element forming a multidomain configuration as shown in Fig. 2(c). For these epitaxial Fe elements, the magnetic field has also been applied along the ⟨110⟩ hard directions to induce an initial single domain state. Lorentz TEM images show that at remanence, again a multidomain structure forms in the 12×12 μm^2 element and the 55×55 μm^2 element is almost in a single domain configuration.

The most remarkable new finding from these images is that the magnetic structure of an epitaxial thin film with in-plane anisotropy changes into a multidomain state at remanence upon reducing its in-plane size and that for the 150 Å thick film this transition occurs at a size of a few tens of microns. These results indicate that upon decreasing element size the in-plane demagnetizing field, which has been considered to be negligibly small for an epitaxial thin film [7,8], becomes important in determining the domain structures. In the case that the elements were initially magnetized along one of the ⟨100⟩ easy directions, due to the magnetocrystalline anisotropy, the magnetization vector tends to remain in that direction upon reducing the field. Hence "magnetic charges" would be uniformly distributed on the edges perpendicular to the magnetization vector. However, our previous work showed that the coercive field, determined by the nucleation and unpinning of 90° domain walls in an epitaxial Fe film of the same

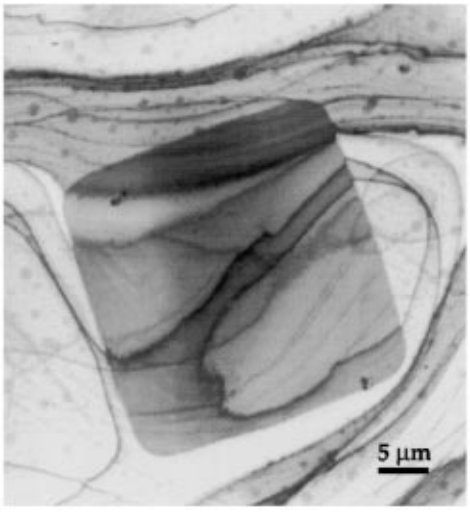

FIG. 1. Bright field TEM image of an epitaxial Fe(001) element supported on a GaAs single crystal membrane.

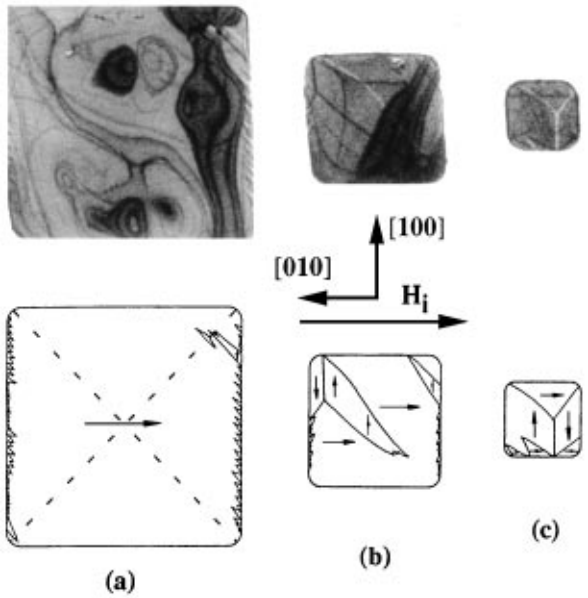

FIG. 2. Remanent domain images and corresponding magnetization schematics of the epitaxial Fe elements with edges parallel to the ⟨100⟩ directions. The field was initially applied along the $[0\bar{1}0]$ direction. The element sizes are (a) 55×55 μm^2, (b) 30×30 μm^2, and (c) 12×12 μm^2.

thickness, is about 8 Oe [11]. The in-plane demagnetizing field near the element edges is much larger than this nucleation field. Thus, upon reducing the applied field, domain nucleation occurs at the element edges as shown in Fig. 2(a). Since the magnetization within these spike edge domains is parallel to the edges, the magnetic charges at the edges and in turn the demagnetizing fields and magnetostatic energy arising from these charges are greatly reduced. However, since the normal components of the magnetization cannot be continuous across these spike domain walls, volume magnetic charges will develop at the walls and produce a demagnetizing field in the elements. The magnetic charges formed at the edge domain walls are oppositely charged for the two opposing edges, and therefore the demagnetizing field due to the charges at the domain walls along one edge acts to unpin the domains on the opposite edge. In the larger elements, such a demagnetizing field is smaller than the unpinning field since the element edges are well separated. The small edge domains thus cannot expand and the element is maintained almost in the original single domain state.

With decreasing element size, the charged edge domain walls approach each other. At a critical size, the demagnetizing fields arising from these charged domain walls become strong enough to cause the edge domains to unpin and expand throughout the whole element so that a multidomain remanent structure is formed. We have calculated the size at which the demagnetizing field approaches the unpinning field. The estimated size is about 15 μm and therefore is consistent with the experimental measurements. From an energy point of view, for the epitaxial elements with edges parallel to the $\langle 100 \rangle$ easy directions, the standard Kittel-type flux closure multidomain structure as shown by the dashed lines in Fig. 2(a) has a lower energy than the "single domain" structure with many small spike edge domains as shown in Fig. 2(a). The reasons for this are (i) the domain wall length in the Kittel multidomain structure is shorter than that in the single domain structure with small spike edge domains, hence the multidomain structure has a lower wall energy, (ii) whereas magnetic charges develop in the spike domain walls, in the multidomain structure, the domain walls are moved to positions where they are free of magnetic charges greatly decreasing the magnetostatic energy. However, for the continuous epitaxial films and larger elements the demagnetizing field is not strong enough to overcome the energy barrier arising from domain wall pinning to realize the low energy multidomain state.

It is found that the remanent multidomain configurations in the smaller epitaxial elements are strongly influenced by several factors such as initial magnetizing direction and element orientation. It can be seen that each remanent domain pattern shown in Fig. 2 attempts to achieve flux closure with the magnetizations lying parallel to the element edges. Since the edges of these elements are parallel to the $\langle 100 \rangle$ easy directions of bcc Fe, such domain configurations obviously minimize both the magnetostatic and anisotropy energies. For a square epitaxial Fe element with edges parallel to the $\langle 110 \rangle$ hard axes, the in-plane dipolar fields will compete with the magnetocrystalline anisotropy whose easy axes are now oriented along the diagonal directions of the elements. Such elements with a larger size are found again to be almost in a single domain state at remanence. Here, only smaller elements are considered. Again, a single domain state of a 12×12 μm^2 epitaxial element was first induced by applying a field along the $[0\bar{1}0]$ easy direction. Reducing the applied field results in domain nucleation and growth at the element corners as shown in the image [Fig. 3(a)] taken at $H_i = 6.0$ Oe. The image of the remanent domain structure and corresponding magnetization schematic is shown in Fig. 3(b).This remanent domain configuration is significantly different from those observed for the elements of the same size but with edges parallel to the $\langle 100 \rangle$ directions. As the field was initially applied along the $[0\bar{1}0]$ easy axis, the maximum demagnetizing field occurs at the two element corners [corners A and B in Fig. 3(a)]. Upon reducing the applied field, new domains nucleate and grow at these two corners. As the corners of the elements are always rounded off, the corner domains with the local magnetization oriented along the [100] easy direction [see Fig. 3(a)] reduce greatly the magnetic charges at the corners and in turn the magnetostatic energy. By further decreasing the applied field, these corner domains grow by domain wall displacement. For this element, whereas the magnetocrysalline anisotropy favors alignment of the magnetization parallel to the $\langle 100 \rangle$ easy axes, the in-plane dipolar field tends to form domain

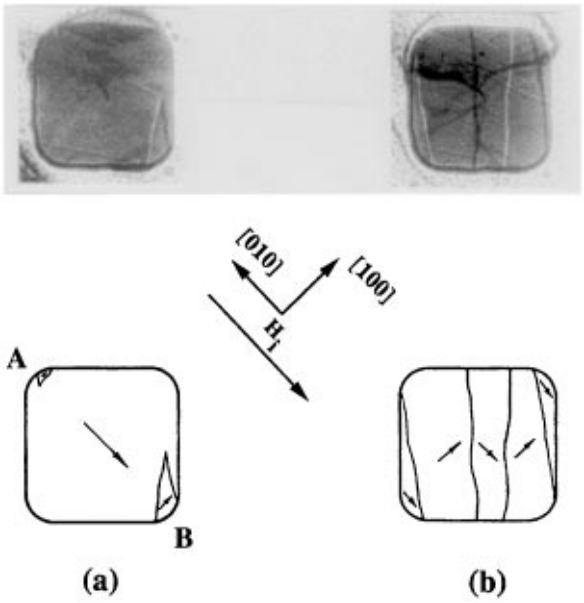

FIG. 3. Domain images and corresponding magnetization schematics of a 12×12 μm^2 epitaxial Fe element with edges parallel to the $\langle 110 \rangle$ directions. The field was initially applied along the $[0\bar{1}0]$ easy direction. (a) Corner domain nucleation and growth upon reducing the applied field ($H_i = 6.0$ Oe), (b) Remanent domain structure.

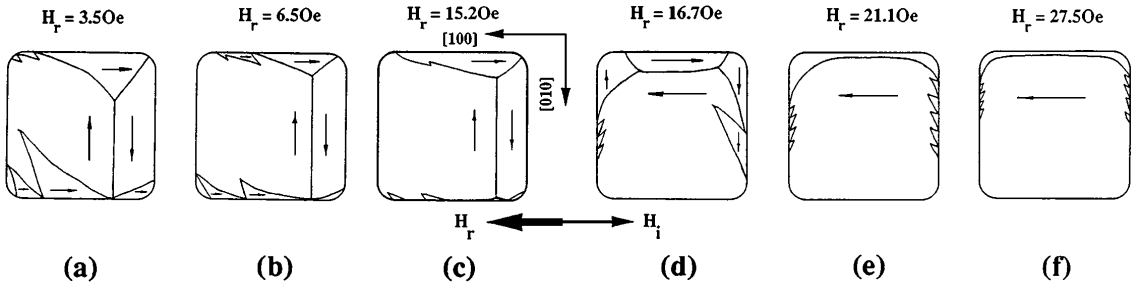

FIG. 4. Domain structures of the $12 \times 12\ \mu m^2$ epitaxial Fe element with edges parallel to the $\langle 100 \rangle$ directions during reversal along the [100] easy axis.

structures with magnetization vectors parallel to the element edges in order to reduce the magnetostatic energy. As a compromise, the element splits into an irregular stripe domain structure at remanence as shown in Fig. 3(b). In this structure, the magnetization vector in each domain remains parallel to the easy axes of the fourfold magnetocrysalline anisotropy at the cost of inducing magnetic charges at the element edges. However, the stripe domain structure brings the positive and negative magnetic charges closer, thus decreasing greatly the spatial extent of the stray field and in turn the magnetostatic energy in comparison with the single domain structure. A similar stripe remanent domain structure has also been observed when the element was initially magnetized along the $\langle 110 \rangle$ hard axes.

By recording domain images during field reversal, the microscopic reversal processes of these epitaxial elements have also been studied. Whereas the $55 \times 55\ \mu m^2$ elements are found to have reversal processes similar to those of continuous Fe films, the smaller elements show significantly different reversal behavior. For a $12 \times 12\ \mu m^2$ element with edges parallel to the $\langle 100 \rangle$ directions, the domain structures during reversal along the [100] direction are shown schematically in Fig. 4. On applying a reverse field, the domains with magnetization oriented opposite to the reverse field shrank. This continued until a critical field H_c was attained, whereupon an irreversible change took place. At this critical field, a domain with the magnetization oriented along the reverse field direction nucleated at the bottom edge and expanded rapidly [see Figs. 4(c) and 4(d)]. As this favorably oriented domain grew, the original vertical 180° domain wall became shorter and finally disappeared, and immediately a 180° domain wall parallel to the [100] direction appeared as shown in Fig. 4(d). A further increase in the reverse field caused the movement of the 180° domain wall towards the element upper edge. It was found that only by applying a strong reverse field (35 Oe) could the 180° wall be driven into the side of the element. The reversal processes shown in Fig. 4 are strikingly different from those of continuous films and of the $55 \times 55\ \mu m^2$ element where the magnetization reversal takes place by two 90° reorientations [11,20]. Strikingly different microscopic reversal processes in comparison with the continuous epitaxial Fe film have also been observed for reversal along the hard axes. The $12 \times 12\ \mu m^2$ element with edges parallel to the $\langle 110 \rangle$ directions also shows distinct reversal behavior. These results will be reported elsewhere [20].

In summary, this study shows that magnetic domains can be created in the epitaxial Fe thin films with a strong magnetocrysalline anisotropy by reducing their in-plane size beneath $\sim 50\ \mu m$. The drastic changes in the remanent state and microscopic reversal processes indicate that the *in-plane* dipolar field becomes competitive with the magnetocrystalline anisotropy at this size, and distinct micromagnetic structures arise controllably according to the orientation of the element edges.

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