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Freestanding single-crystalline magnetic structures fabricated by ion bombardment

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Starting from an ultrathin Fe film grown epitaxially on top of a GaAs(001) substrate, we show that freestanding structures can be created by ion-beam treatment. These structures are single-crystalline blisters and only a few nanometers thick. Anisotropic stress in the rim of a blister induces magnetic domain states magnetized in the direction normal to the blister edge. Experimental evidence is provided that the lateral size can be confined by starting from a nanostructured template. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4906428]

Ultrathin magnetic films and small magnetic structures are the prototypical geometries for exploring fundamental aspects of magnetism at the nanometer scale. Intrinsic magnetic properties, however, are elusive because ultrathin ferromagnets are in contact with a substrate in almost all experiments. The interaction between substrate and magnetic film affects magnetic moments, magnetic anisotropies, and Curie temperatures. Different routes are explored to minimize these effects. Insulating spacer layers or templates, for instance, are used to electronically and magnetically decouple the film from the substrate, but a complete "isolation" is hard to achieve. Freestanding thin magnetic structures would be advantageous for many purposes, yet their fabrication and characterization—both structural and magnetic—are challenging.

Freestanding films are typically fabricated by reducing the thickness by means of focused ion beams, as, for instance, needed in transmission electron microscopy measurements.[1] An alternative approach starts by evaporating the ferromagnetic material onto a template, which is later removed in a wet-chemical process step, leading to freestanding films of a few atomic layers that extend over several hundred micrometers. Such films are required for transmission experiments that explore spin precession and spin transfer.[2] Recently, superparamagnetic CoO films were synthesized in the form of nanosheets by a surfactant-free wet chemical process,[3] and freestanding magnetic nanopillars were produced by focused electron-beam-induced deposition of an $Fe(CO)_5$ precursor.[4]

Here, an alternative route to achieving freestanding ferromagnetic structures is described. Starting from a thin Fe film grown on top of a GaAs(001) substrate, we show that freestanding structures can be created by ion-beam treatment of a flat, unstructured film.

This study was performed on 7–8 nm thick epitaxial Fe films grown on GaAs(001) epilayers and capped with 2 nm Au. The GaAs wafers were either undoped or Si-doped (doping concentration $5 \times 10^{18}/\text{cm}^3$). The epilayers were protected by an As cap for handling under ambient conditions, which were removed by annealing at 620 K for 1 h, leading to a slightly As-rich surface. The substrates were held at 210–290 K during Fe film growth. The results presented here were found to be independent of doping and growth temperature. Films were also grown through shadow masks in a nanostencil setup[5] to create patterned samples on which part of the GaAs remains uncovered. Magneto-optical Kerr effect measurements verified that the easy magnetization axes lie along the in-plane ⟨100⟩ directions.[6] Further experimental details are given in Ref. 7.

The samples were bombarded with a Ne^+ ion beam of variable energy (0.5–3.5 keV) at normal incidence and a beam current of ~2 μA. The total dose of this sputtering process was chosen such that it completely removed the Au layer (and eventually also the top part of the Fe film, typically 1–2 nm), so that a clean ferromagnetic surface was exposed. Topographic and magnetic maps of these surfaces were then acquired by spin-polarized scanning electron microscopy[8] in the same ultrahigh-vacuum tool at a base pressure of 1.5×10^{-10} mbar.

Remarkable topographic changes to the initially smooth surface appear after Ne^+ ion bombardment at 3.5 keV. The surface exhibits rounded protrusions of variable size ranging from 10 nm to several micrometer, in some cases even several 10 μm, see Fig. 1(a). Such protrusions are only found in the area on which the Fe layer had been deposited, see Fig. 1(b).

These protrusions are reminiscent of blisters, reported long ago in erosion studies of materials upon high-energy ion bombardment.[9] Noble-gas injection leads to gas accumulation in the sample, forming subsurface bubbles.[10,11] Ne has been shown to form a liquid phase inside the bubbles.[12] These bubbles get over pressurized if a low vacancy mobility prevents them from balancing their internal pressure by acquiring vacancies, which is generally the case for metals at room temperature.[10] Near the surface, the lateral stress induced by these bubbles[13] can be partially released by deforming the film, leading to the appearance of dome-shaped blisters near the

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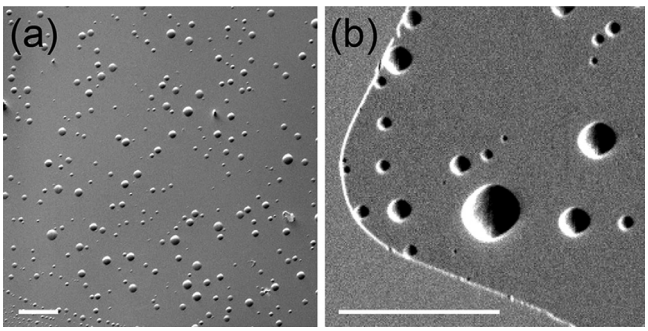

FIG. 1. Secondary electron micrograph of ~6 nm Fe on GaAs(001) after exposure to Ne^+ ions of 3.5 keV with a dose of $\sim 4 \times 10^{16}/cm^2$, as imaged by spin-SEM. (a) Extended film highlighting the large spread in blister size. (b) Structured film fabricated through a stencil mask, with the Fe film covering the top and the right part of the image and the bare GaAs substrate being exposed at the left and the bottom edge. No blisters are visible on the bare substrate. Scale bars correspond to 50 μm.

top surface.[14] These effects were reported for various noble-gas ions and in a wide ion-energy range, as well as for metals in bulk or foil form.[15–17] Blisters were also observed in semiconductors, for instance, in GaN nanowires[18] or in GaAs at elevated temperatures.[19] In our experiments, we employ an ion energy of 3.5 keV and an ion dose of $2 - 10 \times 10^{16}/cm^2$ for ion bombardment at room temperature. A key difference to earlier experiments is the use of an ultrathin epitaxial film rather than a bulk or foil substrate.

The in-plane crystallographic directions of the epitaxially grown Fe film are preserved in the blisters, see Fig. 2(a). The absorbed-current image taken by spin-SEM reveals a perfect fourfold symmetry, with lines and bands of different contrast along the ⟨100⟩ and ⟨110⟩ directions. These broad bands resemble the real-space analogon of Kikuchi lines, i.e., lattice fringe contrast originating from electron channeling of backscattered electrons.[20]

In the following, we substantiate our claim that the blisters we observe are single-crystalline curved layers that are only a few nanometers thick and have delaminated from the GaAs substrate. For this, we cut individual blisters in a focused ion-beam (FIB) system with a 30 keV Ga^+ beam. Most blisters collapse when they are cut locally, but in some cases they keep their shape. Depositing Pt or C onto a blister and its surroundings reinforces the mechanical stability enough so that blisters can survive the FIB cutting. Figure 3 proves that the blisters are indeed hollow and that the layer is freestanding.

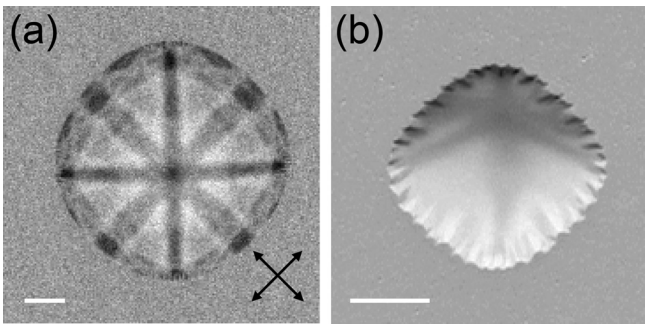

FIG. 2. (a) Absorbed-current micrograph of 6 nm Fe on GaAs(001) after Ne^+ bombardment at 3.5 keV with a dose of $\sim 5 \times 10^{16}/cm^2$. The blister exhibits broad lattice fringes (dark stripes) along the [100] directions and narrower fringes along the [110] directions, highlighting that it is single crystalline. A bright complex pattern with perfect fourfold symmetry is superimposed as well. Black arrows indicate the two equivalent [100] directions. (b) Secondary electron micrograph of a blister in a Fe/GaAs(001) sample that was post-annealed to 500 K after growth. Note the ripples at the edge of the blister. Scale bars correspond to 5 μm.

The depth at which delamination occurs cannot be derived from the FIB images alone, because the rim of the cut warps and the material starts melting, so that the intrinsic blister thickness is not accessible. We exclude delamination within the Fe film because in "exploded" blisters the area is free of Fe remnants. From Fig. 3(a), we also exclude delamination within the GaAs substrate: The exposed substrate is completely flat and free of fissures and fractures, which would not be the case if part from the substrate had delaminated with the film. Moreover, blistering is expected to start at a depth related to the ion implantation range,[15,21] which for Ne^+ at 3.5 keV is only 5 ± 3.3 nm,[22] i.e., within the Fe layer or at the interface. The fact that no blisters are observed in the uncovered GaAs substrate (see Fig. 1(b)) is further evidence that the GaAs substrate remains intact and the blister thickness is confined to the Fe film thickness.

We can influence the appearance of the blisters by modifying the Fe/GaAs interface by post-growth annealing the samples before ion-bombardment. Blistering is again observed, but now the blister edges have a rippled appearance and a less rounded shape, see Fig. 2(b). The edges of the blisters preferentially run along the ⟨100⟩ directions, reflecting the substrate's crystal symmetry. The blister size covers a similar range, but the average blister height is reduced by a factor of 2 compared with the non-annealed samples, as determined by atomic force microscopy. As annealing modifies the ferromagnet/semiconductor interface by creating an ordered interfacial layer,[24] we conclude that delamination occurs via cleavage at the interface, and hence, the blisters are indeed freestanding single-crystalline structures of only a few nanometer thickness.

We have shown that the Fe/GaAs interface is crucial for blister formation, i.e., the impinging ions induce stress in close proximity to this interface. The accumulation depth of the ions is determined by their mean free path, which depends on the ion species and their energy. We can suppress blister formation by reducing the Ne ion energy to 1 keV or less, or by using Xe^+ (3.5 keV) instead of Ne^+. Both these observations are consistent with the calculated ion ranges of 2.0 ± 1.3 nm and 2.1 ± 1.2 nm, respectively.[22]

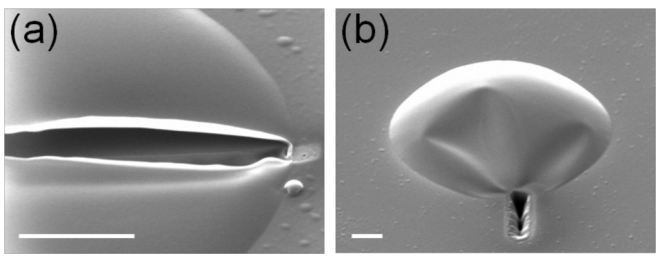

FIG. 3. Secondary electron micrographs of blisters cut open with a FIB tool. (a) Blister cut apart partially, displaying the void between the blister lid and the substrate, with a maximum clearance of more than 100 nm. (b) Blister that "deflated" upon cutting the lid from the edge. Scale bars correspond to 1 μm.

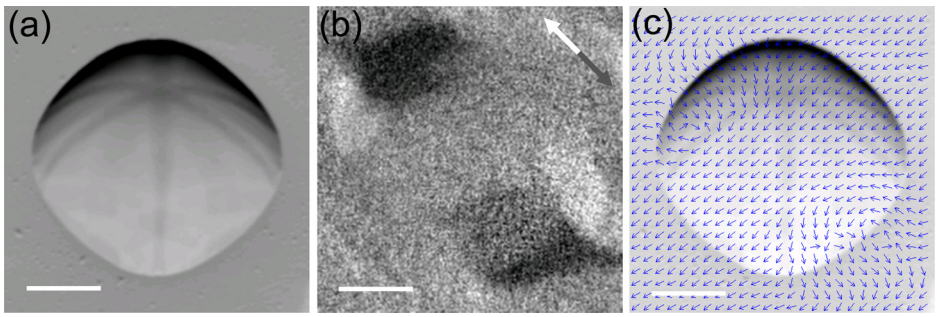

FIG. 4. The magnetic state of a Fe blister observed with spin-SEM: (a) Absorbed-current micrograph of the topography and (b) magnetization component along the diagonal given by the arrows ([010] direction). Black/white corresponds to the magnetization component along the black/white arrows, whereas gray means that the magnetization is orthogonal to this direction. (c) Superposition of the in-plane magnetization direction map with the blister topography to visualize the position of domains with respect to the blister rim. The arrow representation of the magnetization is calculated from magnetic images taken along two orthogonal directions within the plane. The exact same area is displayed in all three images. All scale bars correspond to 5 μm.

From the bulk lattice constants of bcc-Fe and GaAs of 2.87 Å and 5.65 Å, respectively, one estimates that an extended Fe film is under a compressive misfit strain of 1.6%. Strain measurements on Fe films deposited on As-rich GaAs substrates at low temperature revealed that indeed strain is compressive up to several nanometers.[23] The impinging ions lead to additional compressive strain in the film, which is relieved by blister formation.[13] The relief comes from both the deformation of the film as well as the release of ion gas from the subsurface bubbles into the void below the blister lid. Atomic force microscopy revealed that the blister height is about 10% of its diameter, i.e., blister formation leads to appreciable local bulging. The blister lid, once it has detached from the substrate, can radially relax towards the intrinsic Fe lattice constant, thereby reducing its strain. However, the compressive strain in the tangential direction can only be relaxed gradually from the edge towards the center of the blister causing anisotropic stress in its rim.

Qualitatively, a strain change in a ferromagnet is reflected in its magnetic properties such as in a change of magnetic anisotropy and/or the magnetic domain structure through a magnetoelastic energy contribution. Prior to ion-beam treatment, the Fe films were brought to a single-domain state with uniform magnetization along the [100] direction. After ion-beam treatment, we simultaneously imaged the topographic and the magnetic map by spin-SEM, see Fig. 4. Magnetic domains formed at the blister edge, typically with a pair of domains appearing along the rim, where the original magnetization direction was tangential to the edge, i.e., in Fig. 4(b) along the [010] direction. The magnetization in these newly formed domains preferentially points along the radial direction, constrained by the magnetocrystalline anisotropy which supports the ⟨100⟩ directions. In other words, the magnetization rotates from the [100] towards the macroscopically equivalent [010] direction in regions where [010] points along the blister radius. Thus, an additional magnetic energy term must exist to favor this direction. We conclude that the (partial) relaxation of the Fe lattice misfit in the freestanding lid leads to an appreciable magnetoelastic anisotropy, forcing the magnetization to align with the radial direction. Bulk Fe is known to react to stress in this way because of its negative magnetoelastic coupling coefficient,[25] whereas for Fe films remaining in contact with GaAs a positive value was reported.[26]

The energy is reduced further when domain pairs rather than single domains form—as in Fig. 4(b)—which result in a lowering of the magnetostatic energy. While magnetoelastic effects are abundant in magnetic materials, the direct imaging of the resulting domains in a freestanding membrane was not reported before. These domains are frozen in as long as no external magnetic field is applied, even when the blister starts to change appearance upon further ion bombardment.

Prolonged Ne^+ sputtering leads to blister collapse, which proceeds in a characteristic sequence, see Figs. 5(a)–5(c). The blister diameter gets reduced, and the outer rim area appears flattened. At the last stage, the blister implodes; leaving remnants of the Fe blister lid, see Fig. 5(c). Spin-SEM imaging proves that the flattened part of the original blister indeed consists of the originally warped blister lid: The magnetic contrast on the flat and on the curved part is the same.

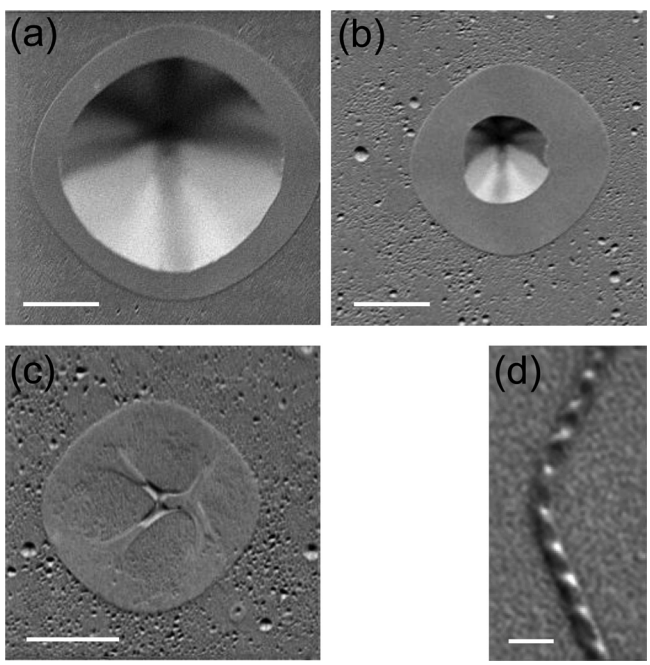

FIG. 5. (a)–(c) The "death" of three different blisters after prolonged Ne^+ bombardment at 3.5 keV, showing the reduction in blister size compared with the original blister and an outer smooth Fe film and finally the blister collapse. All scale bars correspond to 5 μm. (d) A curved ferromagnetic Fe strip of ≈250 nm width, fabricated by nanostenciling, after ion beam treatment. A sequence of blisters appears along the strip, with the blister diameter being limited by the wire width. Scale bar is 500 nm.

In some cases, the blisters spontaneously explode, uncovering a smooth surface as observed with FIB (see above).

In summary, we have shown a route to fabricating freestanding ferromagnetic structures by ion-beam treatment of Fe films grown on GaAs. These structures are single-crystalline blisters and only a few nanometers thick. Stress at the blister edge creates magnetic domains by the magnetoelastic effect. These domains, pinned at the rim, extend both into the blister and the adjacent area of the flat film. The creation of such freestanding structures should not be limited to the Fe/GaAs system reported here: Various materials and different film thicknesses should exhibit similar behavior. A decisive factor is that the ion range, which can be tuned by the ion energy, needs to correspond to approximately the film thickness. Blisters can also be induced in laterally confined nanostructures, as shown in Fig. 5(d). Preliminary experiments on nanostenciled magnetic nanostrips show that a series of blisters is induced that are limited in lateral extension by the 250-nm width of the strip. Magnetic blisters could be of interest for applications in magnetomechanical devices, in which magnetization and position are coupled, possibly as transducers in nanomagnetic logic concepts.[4]

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