400-fold reduction in saturation field by interlayering

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The buildup of stress with increasing thickness of magnetic thin films is a common phenomenon that often induces undesirable anisotropies that can convert an otherwise magnetically soft film into a magnetically hard one. We found that by interlayering such a magnetic thin film with films that are either not lattice matched or have a different crystal structure, reductions in the saturation field as large as 400-fold can be achieved. Differences in grain size appear to be responsible.
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I. INTRODUCTION

The relationships among stress, magnetostriction, magnetic anisotropy, growth, and processing of thin magnetic films is a topic of long-standing interest in magnetism.[1–19] The impact on the saturation field of the sense layer is an important issue for magnetic sensors based on magnetoresistance effects. To detect small magnetic fields, these sensors need to incorporate a sense layer that is magnetically soft. Recently, we reported that an alloy known as conetic ($Ni_{77}Fe_{14}Cu_5Mo_4$) is particularly well suited for this purpose. The composition of $Ni_{77}Fe_{14}Cu_5Mo_4$ is designed to reduce the magnetostriction constant and the magnetocrystalline anisotropy as close to zero as possible. Films of the alloy are magnetically very soft at thicknesses of a few tens of nanometers.[20] In our work on implementing this alloy in magnetic tunnel junction (MTJ) sensors, it appears that such thicknesses are not likely to be adequate for reaching our goal of 1 $pT/Hz^{0.5}$ at 1 Hz.[21] Problems with $1/f$ noise and thermal magnetic noise appear to require a sense-layer thickness of hundreds of nanometers because such noise is predicted to scale inversely with sense-layer volume.[21,22] In the work presented here, we report a serious problem encountered with the saturation fields for such thicknesses and a practical solution to the problem. This work should lead to softer free layers in MTJs and to flux concentrators with higher gain.

II. EXPERIMENTAL

The samples were deposited on Si(100) wafers with 250 nm of thermal oxide. After bakeout, the deposition chamber has a base pressure of 7×10^{-8} Pa ($\approx5\times10^{-10}$ Torr), of which 90% is H_2. The $Ni_{77}Fe_{14}Cu_5Mo_4$ (at. %) films were deposited at room temperature by dc-magnetron sputtering in 0.3 Pa (2 mTorr) Ar at a typical rate of ~100 nm/min. The substrates were prepared for deposition by a cleaning process consisting of a few seconds in an ultrasonic bath containing a glassware cleaning solution, rinsing in a stream of ultrapure distilled water, and being blown dry in a filtered nitrogen gas jet. They were then loaded into the UHV sputtering chamber immediately and cleaned by neutralized-beam argon-ion etching to remove 2 nm of SiO_2. The estimated uncertainty in the reported measurements of the magnetic field strengths is ±3%.

In situ stress measurements were made on a HeNe optical bench using the wafer curvature method.[23] The substrate was a $60\times3\times0.1$ mm^3 wafer of borosilicate glass onto which a bonding layer of Ti and 250 nm of Au was evaporated. The samples were vapor deposited using the procedure outlined above. The curvature of the substrate was monitored while the films were electrochemically dissolved by reflecting the laser off of the glass/metal interface and onto a position-sensitive detector. The curvature is directly proportional to the cantilever force, which is the product of the film stress and the film thickness (in N/m). The films were dissolved in 0.1*M* H_2SO_4 at a potential of −0.31 V versus saturated sulfate electrode. The thickness of the material removed was calculated from the charge passed during dissolution. The average biaxial stress of the film was calculated from the change in force following dissolution, divided by the thickness of material dissolved. The estimated uncertainty in the reported stress values is ±5%.

III. RESULTS AND DISCUSSION

In our earlier work, we found that the $Ni_{77}Fe_{14}Cu_5Mo_4$ alloy in thin film form can have saturation fields as small as 0.005 mT (0.05 Oe).[20] While this sensitivity to small magnetic fields would be desirable in the sense layer of an MTJ sensor, we find that stray fields from the pinned layer set a practical lower limit of about 0.1 mT (1 Oe) on the saturation field of the sense layer. We have been able to achieve values in the range of 0.1–0.4 mT routinely in MTJs using $Ni_{77}Fe_{14}Cu_5Mo_4$ thickness of 20 nm. Recently, we began investigating layers a few hundred nanometer thick, as is needed for noise suppression.[21,22] The problem we encountered is illustrated in Fig. 1.

At the thickness of 400 nm, the saturation field is 20 mT (200 Oe), which is entirely unsuitable for the sense layer of

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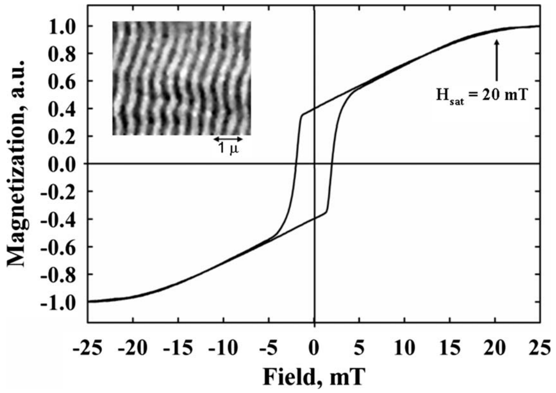

FIG. 1. The in-plane magnetic hysteresis loop of a 400 nm thick film of $Ni_{77}Fe_{14}Cu_5Mo_4$. The remanence is 40% as the film breaks up into stripe domains (the inset is a SEMPA image) with partial out-of-plane magnetization. The loop is almost azimuthally invariant (20 mT=200 Oe).

an ultralow magnetic field sensor or for a flux concentrator. A study by scanning electron microscopy with polarization analysis (SEMPA) indicates a partial out-of-plane magnetization with a break up into stripe domains at remanence, as is not unusual for thick magnetic films with in-plane stress.

It seemed likely that the best approach to stress relief in films of this thickness would be to use interlayers to interrupt growth and induce the renucleation of grains. Grain renucleation is a common effect when a film is deposited on an interlayer with a different crystal structure or a very different lattice constant. The effect is to induce grain renucleation by making epitaxy energetically prohibitive. Figure 2 illustrates the two structures.

Earlier work has shown that off-normal deposition of CoZrO results in the growth of partially isolated columns in that magnetic thin-film system. The columns can induce an out-of-plane anisotropy, which can be suppressed by interlayering with ZrO_2.[24] The result is a softer film in which the saturation field is reduced by a factor of 20.[24] The mechanism of softening in that case was reported to be different from the one reported here.

Figure 3 presents the hysteresis loop for a multilayer in which the 400 nm $Ni_{77}Fe_{14}Cu_5Mo_4$ is split into four segments interlayered with 5 nm Ag. The lattice mismatch is

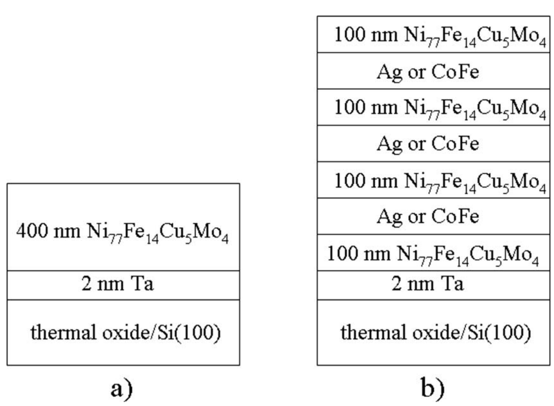

FIG. 2. An illustration of (a) the type of film that exhibits a large stress induced saturation field and (b) the interlayering of the film with a different material to induce grain renucleation and stress relief. The Ta is an adhesion layer needed to suppress stress-induced peeling in (a).

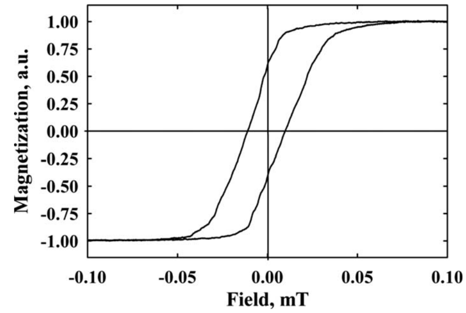

FIG. 3. The in-plane magnetic hysteresis loop of a multilayer film of four $Ni_{77}Fe_{14}Cu_5Mo_4$ segments 100 nm each thick with interlayers of Ag 5 nm thick, as illustrated in Fig. 2(b). The saturation field is reduced approximately 400-fold as compared to Fig. 1, the saturation magnetization is unchanged, and the loop is azimuthally invariant (0.1 mT=1 Oe).

16%. The saturation field is reduced approximately 400-fold as compared to Fig. 1. In contrast with the inset in Fig. 1, the magnetic domain image of the interlayered sample is featureless.

The transmission electron microscopy (TEM) image of the continuous 400 nm film in Fig. 4(a) shows a fine-grained film with little columnar growth. There is also no indication of magnetically isolated columns that might favor perpendicular shaped anisotropy.[24] The sample had a strong tendency to warp when it was being thinned down for TEM, indicating it was highly stressed. The multilayer film with the Ag interlayers did not exhibit such warping, indicating it had much less stress. Its TEM image in Fig. 4(b) shows frequent renucleation of grains on top the Ag layers and rather few examples of grain-to-grain epitaxy through the Ag layers.

The great difference in stress between these two films is likely to be related to the different average grain sizes. The continuous 400 nm film exhibits a high density of grains 10 nm or less in diameter. The multilayer exhibits a high density of columnar grains with a width of 20 nm and a height of 100 nm. Grain boundaries are commonly associated with

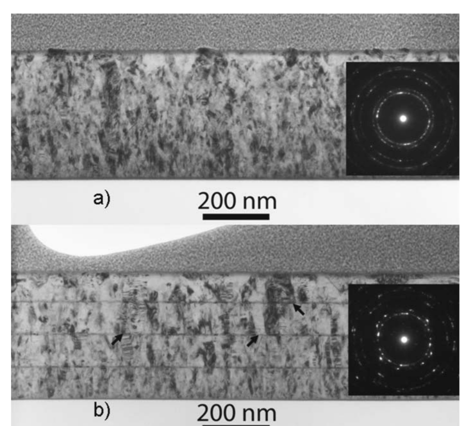

FIG. 4. TEM images of (a) the continuous 400 nm $Ni_{77}Fe_{14}Cu_5Mo_4$ film of Fig. 1, and (b) the multilayer of Fig. 3 consisting of four 100 nm $Ni_{77}Fe_{14}Cu_5Mo_4$ films separated by three 5 nm Ag films. The alternating light and dark bands seen in some grains along the growth direction are twin boundaries. The black arrows point to examples of locations at which renucleation of grains occurs. The grainy material on the top of each structure is glue.

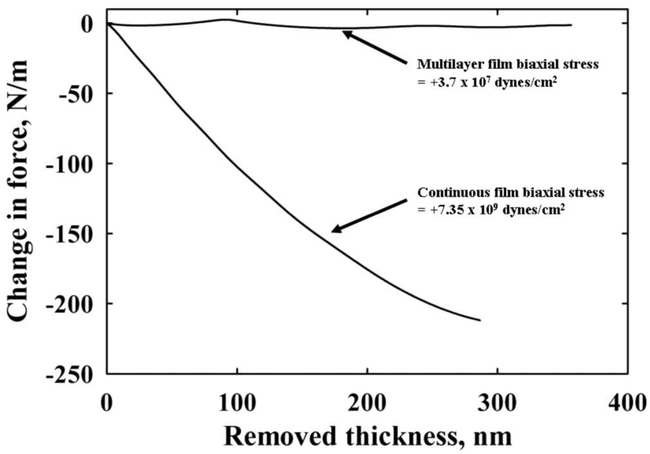

FIG. 5. Measurements (by electrochemical dissolution) of the biaxial stress in the continuous film of 400 nm $Ni_{77}Fe_{14}Cu_5Mo_4$ and in the multilayer consisting of four 100 nm $Ni_{77}Fe_{14}Cu_5Mo_4$ films separated by three 5 nm Ag films. The force change in the negative direction indicates the removal of material under tensile (positive) stress.

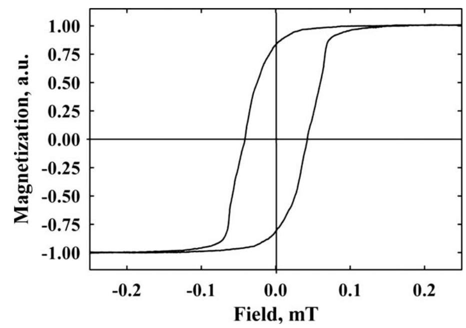

FIG. 6. The in-plane magnetic hysteresis loop of a multilayer film of four 100 nm $Ni_{77}Fe_{14}Cu_5Mo_4$ films separated by three 2 nm CoFe films, as illustrated in Fig. 2(b). The saturation field is reduced approximately 200-fold as compared to Fig. 1, the saturation magnetization increases very slightly, and the loop is azimuthally invariant (0.1 mT=1 Oe).

stress and smaller grains tend to produce larger stresses. Theoretical modeling of this effect may be found in Ref. 25.

Measurements of the biaxial stress in the two types of films, presented in Fig. 5, were made by electrochemical dissolution of the films deposited on thin glass cantilevers. The continuous film was found to have a biaxial stress 200 times larger than that of the multilayer with Ag.

Magnetic hysteresis loops for continuous $Ni_{77}Fe_{14}Cu_5Mo_4$ films showed that the onset of the partial out-of-plane magnetization occurred rather suddenly between 290 and 300 nm. Taking the critical thickness to be 295 nm, we can estimate the perpendicular anisotropy constant K_{perp} using the equation[26]

$$t_c = \left(\frac{1728}{\pi^4}\right)\frac{A^{1/2}M_s^2}{K_{\text{perp}}^{3/2}}, \tag{1}$$

where t_c is the critical thickness, A is the exchange constant [which is very likely close to the 1.1×10^{-6} ergs/cm of $Ni_{80}Fe_{20}$ (Ref. 27)], M_s is the saturation magnetization of $Ni_{77}Fe_{14}Cu_5Mo_4$, which is 616 emu/cm^3. Solving Eq. (1) for K_{perp} yields a value of 5.66×10^4 ergs/cm^3. Since this value will be $\approx 3/2\ \lambda\cdot\sigma$ (see Ref. 28) where λ is the magnetostriction constant and σ is the stress (7.35×10^9 dynes/cm^2 in Fig. 5), we can estimate $\lambda=5.1\times10^{-6}$. This value of λ is not particularly small and indicates that $Ni_{77}Fe_{14}Cu_5Mo_4$, which is very soft in its unstressed state due to the fine tuning of its composition, acquires a significant magnetostriction when it is under large stress.

It may be of interest that the reported tensile strength of $Ni_{77}Fe_{14}Cu_5Mo_4$ ranges from 5.3×10^9 to 9.0×10^9 dynes/cm^2.[29] Since our value for the continuous film is just in the middle of those values, at 7.35×10^9 dynes/cm^2 (or 735 MPa), it may be that the upper limit of the sustainable stress is set by the tensile strength of the film, and plastic deformation is occurring during growth.

While the 400-fold reduction in saturation field in Fig. 3 is encouraging, Ag has the undesirable effect of breaking the magnetic coupling between $Ni_{77}Fe_{14}Cu_5Mo_4$ layers, which defeats the purpose for ultralow-field sensors (but not for flux concentrators) and raises the question of whether a similar success could be achieved in a film with continuous magnetic coupling.

To test the idea, we used interlayers of 2 nm CoFe. Since CoFe is bcc and $Ni_{77}Fe_{14}Cu_5Mo_4$ is fcc, interruption of growth and grain renucleation is very likely while magnetic coupling throughout the structure will be preserved. Figure 6 presents the hysteresis loop for such a sample.

As in the case of Ag, the CoFe interlayers produce a sharp reduction in the saturation field, in this case to ≈ 0.1 mT (1 Oe). The saturation field is reduced approximately 200-fold. It may be of interest to point out that while 2 nm CoFe interlayers give the softest multilayer, even 0.5 nm CoFe interlayers reduce the saturation field to ≈ 0.2 mT!

In MTJs the stray fields from the pinned layer set a practical lower limit of about 0.1 mT (1 Oe) on the saturation field of the sense layer. Consequently, the saturation field in Fig. 6 is near the practical lower limit for the purpose of ultralow-field MTJ sensors and is also suitable for flux concentrators.

IV. CONCLUSIONS

The major conclusions of this work are as follows.

(1) Films of $Ni_{77}Fe_{14}Cu_5Mo_4$ are magnetically quite soft for thicknesses <100 nm but can become quite hard for thicknesses >300 nm, and the magnetostriction constant rises from near zero to $\approx 5\times10^{-6}$.
(2) Stress build up is responsible, and it induces a small perpendicular anisotropy.
(3) The magnetic softness can be restored by interlayering the soft magnetic material with films of different lattice constants or crystal structures to interrupt growth and promote renucleation of grains.
(4) Reductions in stress as large as 200-fold have been achieved.
(5) Reductions in coercivity to as low as 0.01 mT (0.1 Oe) have been achieved.
(6) Reductions in saturation field as large as 400-fold have been achieved.
(7) The large stresses appear to be attributable to small grain size.

(8) This work should lead to softer free layers in MTJs and to flux concentrators with higher gain.

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