Magnetic Properties of Epitaxial and Polycrystalline Fe/Si Multilayers

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Accepted to proceedings of 1995 MMM Conference; to appear in J. Appl. Phys. in April, 1996.

Abstract

Fe/Si multilayers with antiferromagnetic interlayer coupling have been grown via ion-beam sputtering on both glass and single-crystal substrates. High-angle x-ray diffraction measurements show that both sets of films have narrow Fe peaks, implying a large crystallite size and crystalline iron silicide spacer layers. Low-angle x-ray diffraction measurements show that films grown on glass have rougher interfaces than those grown on single-crystal substrates. The multilayers grown on glass have a larger remanent magnetization than the multilayers grown on single-crystal substrates. The observation of magnetocrystalline anisotropy in hysteresis loops and (hkl) peaks in x-ray diffraction demonstrates that the films grown on MgO and Ge are epitaxial.

The smaller remanent magnetization in Fe/Si multilayers with better layering suggests that the remanence is not an intrinsic property.
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The well-established picture of antiferromagnetic coupling in metal/metal multilayers would have to be extensively modified for coupling across insulating or semiconducting spacer layers, where the spacer does not possess a Fermi surface.[1,2] In particular the temperature dependence of the exchange coupling might be significantly different in ferromagnet/semiconductor multilayers where the exchange is mediated by thermally activated carriers.[1]

Unusual temperature-dependent magnetic properties have been reported for Fe/Si multilayers. For example, a large increase in the remanent magnetization has been observed at low-temperature.[3] If the interlayer antiferromagnetic (AF) coupling increases with decreasing temperature, as in multilayers with metal spacers, one might expect the remanence instead to *decrease* at low temperature. Proper interpretation of the remanent magnetization in Fe/Si multilayers may therefore be important to understanding the origin of the interlayer coupling in this system. One way to explore the origin of the remanence is to compare films of different crystalline quality.

I. EXPERIMENTAL METHODS

The films used in this study were grown using ion-beam sputtering (IBS) in a chamber with a base pressure of 2×10^{-8} torr. The deposition system is described in more detail elsewhere.[4] All samples used in this study were grown at a substrate temperature of 200°. All comparisons between films grown on glass and single-crystal substrates will be made on samples which were deposited *simultaneously* so as to eliminate any reproducibility issues.

The substrates used in this study were glass coverslips, MgO (001), Ge(001) and $Al_2O_3(11\bar{2}0)$. The MgO and Al_2O_3 substrates were cleaned according to a recipe reported by Farrow and coworkers.[5] The glass and Ge substrates were rinsed in solvents. All films are capped with a 200Å Ge oxidation barrier. The magnetic and structural properties of the films are stable for at least one year.

II. STRUCTURAL CHARACTERIZATION

Figure 1 shows high-angle x-ray diffraction spectra for a purely (001)-oriented (Fe40Å/Si14Å)x60 multilayer grown on MgO(001) and a purely (011)-oriented multilayer grown simultaneously on glass. While multiple superlattice satellites are observed around the Fe(002) peak for the film grown on MgO, the film on glass has only one peak corresponding to (011)-textured growth. A single superlattice satellite is typically observed in multilayers on glass on the low-angle side in keeping with previous observations.[6] An estimate of the crystallite sizes in these films can be derived using the Scherrer formula. This analysis gives a coherence length of 165Å for the film on glass and 188Å for the film on MgO.

Since the crystalline coherence lengths of these films are similar, the presence of high-angle satellites in the film on MgO must be due to better layering. The small-angle x-ray scattering data shown in Figure 2 confirm this hypothesis. The multilayer on MgO has 4 low-angle peaks, indicating a moderate degree of composition modulation. The multilayer on glass shows only two relatively broad peaks, indicating larger interfacial roughness and less order in the layering. The low-angle x-ray spectra are consistent with rocking curves which are only about 1° wide for films grown on MgO and Al_2O_3 but are typically 10° to 15° wide for films on glass. The bilayer periods determined from the low-angle peak positions are (41.0 ± 0.1)Å for the MgO film and (40.9 ± 0.2)Å for the glass film, the same within experimental error.

ϕ scans of the MgO and Fe [110] peaks for the film on the MgO substrate (not shown) demonstrate that it is oriented in-plane. While θ-2θ scans for (011)-oriented multilayers grown on Al_2O_3 substrates (not shown) also show multiple high-angle superlattice satellites, the ϕ scans for this film indicate only weak orientation in-plane.

The shape of the high-angle peaks and their superlattice satellites are described by a well-known theory.[7] Application of this theory to the Fe/Si multilayers is difficult because the iron silicide lattice constant, the thickness of the remaining pure Fe and the thickness of the iron silicide spacer can be estimated only roughly. A precise determination of the silicide lattice constant should make a quantitative analysis of these satellite features possible.

III. MAGNETIC CHARACTERIZATION

Figure 3a shows magnetization curves for 60-repeat (Fe40Å/Si14Å) multilayers grown simultaneously on glass and MgO(001). The saturation fields H_s appear to be similar for the two films. On the other hand, the remanent magnetization is 58% for the film on glass and 7% for the film grown on MgO. A remanence as low as 1% has been observed for other multilayers grown on MgO substrates. SQUID magnetometer data taken up to higher fields gives a saturation field of 9.75 kOe for the multilayer on MgO at room temperature. Assuming for a moment that the interlayer coupling is purely bilinear in nature, a well-known formula relates the saturation field to the AF coupling strength: $A_{12} = H_s M_s t_{Fe}/4$ where M_s is the saturation magnetization and t_{Fe} is the thickness of an individual Fe layer.[8] Use of this equation with $H_s = 9.75$ kOe and the measured magnetization $M_s = 1271$ emu/cm^3 gives $A_{12} = 1.2$ erg/cm^2. This AF coupling value is comparable in size to the coupling measured in metal/metal multilayers multilayers[9].

Figure 3b shows magnetization curves for Fe100Å/Si14Å/Fe100Åtrilayer films grown on Al_2O_3 $(11\bar{2}0)$, Ge(001), and MgO(001) substrates. All three of the magnetization curves in Fig. 3b were taken with the field applied along an Fe(100) easy direction. Significant in-plane anisotropy of the magnetization curves occurs for the films on the Ge and MgO substrates, similar to what has been observed for Fe/Cr/Fe trilayers.[8] The observation of magnetocrystalline anisotropy in the film on MgO but not in the film grown on Al_2O_3 is consistent with expectations from the ϕ scans, which show that the in-plane orientation of the film on MgO is much stronger.

Figure 3b once again demonstrates that the degree of remanence in Fe/Si multilayers is strongly related to the quality of layering. While the remanent magnetization of the epitaxial trilayers on Ge and MgO is only about 5% of the saturated value, the remanence of the polycrystalline trilayer on Al_2O_3 is close to 50%. The remanent magnetization of the trilayers on Ge and MgO is about 5% in the in-plane hard direction (H $\parallel$ Fe(110)) as well.

A SQUID magnetometer has been used to measure the magnetization curves of the IBS-grown Fe/Si multilayers at lower temperatures.[10] The temperature dependence of the remanent magnetization of these films is similar to that reported by other authors.[3]

IV. DISCUSSION

At the moment it is not possible to tell why the in-plane ordering of the films grown on $Al_2O_3(11\bar{2}0)$ is inferior to that grown on the (001) MgO and Ge substrates. The difficulty with the Al_2O_3 growth may have to do with the 6° miscut of the substrates, or it may be due to an intrinsic difficulty with (011) growth of the Fe/Si multilayers. Previous work has shown that AF coupling in Fe/Si multilayers is dependent upon formation of a metastable iron silicide spacer layer phase.[4] The possibility exists that the spacer silicide does not grow well on Fe in the (011) orientation. This question can be answered only by further growth studies on better (011) substrates and careful structural characterizations.

A related question is whether the larger remanent magnet moment in the (011)-textured films might be due to a fundamental difference in magnetic properties from the (001)-textured films. Because a 46-repeat Fe/Si multilayer grown on Al_2O_3 has a remanence of only about 10%, this is unlikely. Undoubtedly the trilayer on Al_2O_3 has a higher remanence than the multilayer because the thinner film is more greatly impacted by the poor substrate surface quality. The staircase morphology caused by the 6° miscut of this Al_2O_3 substrate may lead to wavy interfaces between the Fe and iron silicide films or to pinholes through the silicide layers. Wavy interfaces can cause increased magnetostatic coupling or even biquadratic coupling,[11] both of which would tend to increase the remanence. The large remanence of the multilayers grown on glass substrates is likely also due to pinholes or magnetostatic coupling.

Pinhole-induced may explain the unusual temperature dependence of the remanence.

(Magnetostatic coupling is expected to be approximately temperature-independent.) Fe atoms in bridges through the silicide spacer layers are expected have a reduced Curie temperature. A larger remanence at low temperature therefore makes sense if the remanence is derived from pinhole coupling and is not an intrinsic effect. Low Curie-temperature material may also be present in the iron silicide spacer layer or in at the iron/iron silicide interfaces.

By growing on a number of substrate materials and by using different deposition conditions, Fe/Si multilayers have been prepared with a varying degree of ordering. A large amount of accumulated evidence demonstrates that high remanence of the magnetization curves in Fe/Si multilayers is associated with interface roughness. The remanence is therefore not likely to be related to unusual exchange coupling but instead to originate from defects, perhaps pinholes through the silicide spacer layer. Since the remanent magnetization is caused by extrinsic effects, future studies should concentrate instead on measurements of the saturation field of the magnetization curves in order to learn more about the interlayer coupling.

We would like to thank E.E. Fullerton, J.A. Borchers, R.M. Osgood III and Y. Huai for helpful discussions, and B.H. O'Dell and S. Torres for technical assistance. Part of this work was performed under the auspices of the U.S. Department of Energy by LLNL under contract No. W-7405-ENG-48.

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FIGURES

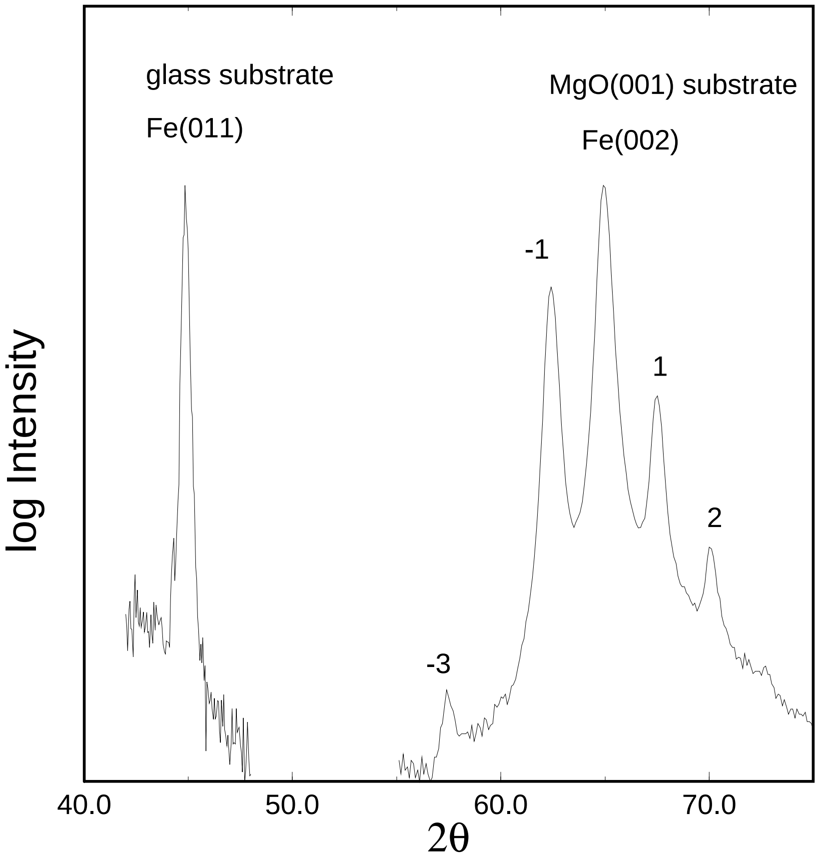


FIG. 1. High-angle x-ray diffraction spectra from two (Fe40Å/Si14Å)x60 multilayers grown on different substrates. High-angle x-ray peaks give information about lattice constants and stacking within the layers. The multilayer grown on MgO(001) has an Fe(002) peak with 4 satellites. The multilayer simultaneously grown on glass has only an Fe(011) peak.

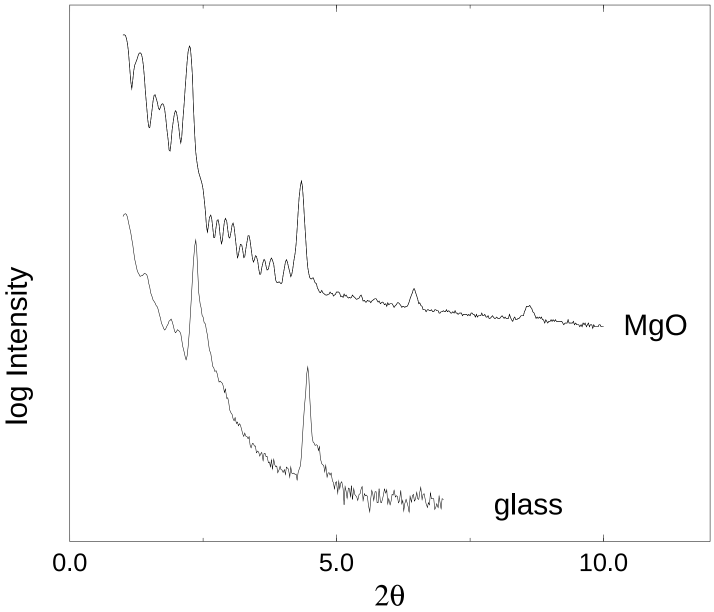
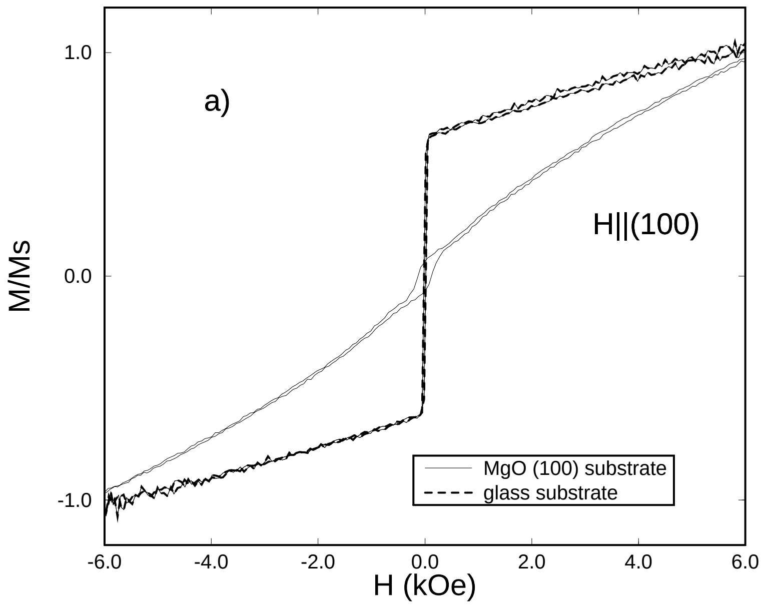


FIG. 2. Low-angle x-ray diffraction spectra for the same two (Fe40Å/Si14Å)x60 multilayers. Low-angle x-ray peaks give information about the bilayer period and layering of the multilayer. The multilayer grown on MgO has four relatively narrow peaks. The multilayer grown on glass has only two broad peaks, indicating a greater degree of interface roughness.


a)
H||(100)
MgO (100) substrate
glass substrate
M/Ms
1.0
0.0
-1.0
-6.0
-4.0
-2.0
0.0
2.0
4.0
6.0
H (kOe)

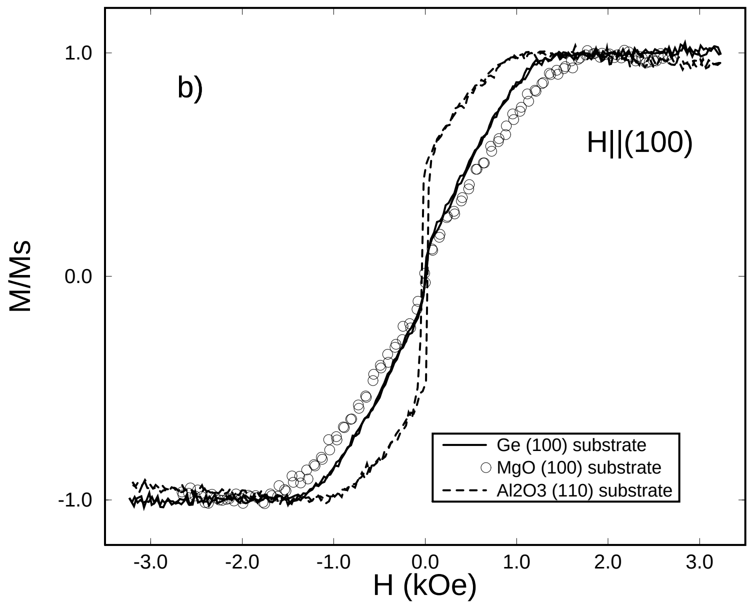

b)
H||(100)
Ge (100) substrate
MgO (100) substrate
Al2O3 (110) substrate
M/Ms
1.0
0.0
-1.0
-3.0
-2.0
-1.0
0.0
1.0
2.0
3.0
H (kOe)

FIG. 3. Magnetization curves for Fe/Si multilayers grown on various substrates. The magnetization data is normalized to the highest measured value in order to facilitate comparison of the shape of the two data sets. a) Hysteresis loops for the (Fe40Å/Si14Å)x60 multilayers simultaneously grown on MgO (001) and glass. The remanence is lower for the film grown on MgO. Magnetization curves for Fe/Si multilayers grown on various substrates. The magnetization data is normalized to the highest measured value in order to facilitate comparison of the shape of the two data sets. b) Hysteresis loops for (Fe100Å/Si14Å)x2 multilayers (trilayers) grown on Al_2O_3 $(11\bar{2}0)$, Ge(001), and MgO(001). All data are taken with the applied magnetic field along the Fe (100) easy direction.