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CRYSTAL STRUCTURE DEPENDENCE OF ANTIFERROMAGNETIC COUPLING IN FE/SI MULTILAYERS

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ABSTRACT

Recent reports of temperature dependent antiferromagnetic coupling in Fe/Si multilayers have motivated the generalization of models describing magnetic coupling in metal/metal multilayers to metal/insulator and metal/semiconductor layered systems. Interesting dependence of the magnetic properties on layer thickness and temperature are predicted. We report measurements that show the antiferromagnetic (AF) coupling observed in Fe/Si multilayers is strongly dependent on the crystalline coherence of the silicide interlayer. Electron diffraction images show the silicide interlayer has a CsCl structure. It is not clear at this time whether the interlayer is a poor metallic conductor or a semiconductor so the relevance of generalized coupling theories is unclear.

INTRODUCTION

The magnetic coupling of adjacent ferromagnetic layers separated by a broad range of non-magnetic metal spacer layers oscillate from anti-ferromagnetic to ferromagnetic as the spacer layer thickness increases[1]. The variation of the coupling can result in oscillations in easily measured quantities such as the saturation field and the magneto-resistance as a function of interlayer thickness.

Most features of the oscillating exchange coupling have been successfully explained by applying RKKY type interactions to the layered geometry and using the Fermi surface characteristics of the interlayer metal[2]. Recent experimental observations of anti-ferromagnetic coupling in Fe/Si multilayers[3, 4] have motivated generalization of models of interlayer exchange to include systems without well defined fermi surfaces such as semiconductors and insulators[5, 6, 7]. Among the most pronounced predicted differences between metallic and non-metallic interlayer systems is the strong temperature dependence of the coupling in multilayers with a non-metallic interlayer due to the thermally activated nature of the carriers which carry the exchange.

In this paper we present data describing structural and magnetic characteristics of Fe/Si multilayers deposited using ion beam sputtering and discuss their significance to the theories of Bruno[6] and Zhang[5]. Consistent with previous studies we find that increasing the Si interlayer thickness from 14Å to 20Å, while keeping the Fe thickness fixed at about 30Å, has a dramatic effect on the magnetic properties and the morphology of the multilayer. We find that for Si layers around 14Å thick, the multilayer maintains crystalline coherence in the growth direction through more than one bilayer period and magnetically the Fe layers are anti-ferromagnetically coupled resulting in a high saturation field. For slightly thicker Si layers (around 20Å) the crystalline coherence in the growth direction is only as thick as a single bilayer, and the saturation field is small consistent with either ferromagnetically coupled or uncoupled Fe layers. Further, for Fe layers sufficiently thin, crystalline coherence is not achieved in the multilayer. We find that even for 14Å thick Si interlayers, disordered ferromagnetic Fe layers are either ferromagnetically coupled or uncoupled. Our TEM study reinforces the assertion of Fullerton et. al.[3] that the crystalline iron-silicide that forms in the interlayer may be the CsCl structure. This silicide structure is likely stabilized in the multilayer because it is closely lattice matched to BCC Fe. We discuss the possibility that the crystallinity of the interlayer is crucial to produce AF interlayer coupling.

EXPERIMENTAL DETAILS

Our films were grown in a ion beam sputtering(IBS) systems described in detail elsewhere[8]. Briefly, four targets can be rotated in front of the 3 cm ion gun which sputters material up through a circular aperture in a stationary liquid nitrogen (LN) cooled Cu tray. A rotating tray above the Cu tray has positions for four substrates. The substrate to target distance is approximately 30 cm. The target carousel and ion beam voltage are computer controlled, and the layers thicknesses are monitored by a calibrated quartz crystal oscillator. The base pressure of the system is $1\text{-}2\times10^{-8}$ torr. We sputter in 2.5×10^{-4} torr partial pressure of UHP Ar which is about an order of magnitude lower than typical magnetron sputtering. With a beam voltage of 1kV and a beam current of 20mA the deposition rates are around 0.2Å/sec. IBS is unique in that at a fixed deposition voltage, the deposition rate can be independently adjusted by changing the beam current. As a result, the energetics of ion beam sputtering deposition can be quite different than those of thermal evaporation or magnetron sputtering.

We use glass and Si substrates and find no dependence in the magnetic or structural properties of the Fe/Si multilayers (MLs). We deposit at nominal room temperature (RT) or, by bringing the substrate tray into contact with the cooled Cu tray, at nominal LN temperature. Thick Si films sputtered under typical conditions are amorphous, and thick Fe films are BCC, polycrystalline, and textured in the (110) close packed direction.

The structural properties of the MLs were probed using a Rigaku rotating anode x-ray machine with a reflected beam monochromator and CuK_α radiation. Low angle θ-2θ scans reveal properties of the ML in the growth direction, and high angle scans measure the crystalline coherence. In addition, selected films were studied using TEM. RT magnetic characteristics of the MLs were measured using a vibrating sample magnetometer, and low temperature magnetic measurements were performed on a SQUID magnetometer.

RESULTS AND DISCUSSION

Figure 1 illustrates the effect of changing the interlayer thickness on the magnetic properties of Fe/Si multilayers. Film A ([Fe30Å/Si14Å]x50) (30/14 ML) has a low remanent magnetic moment (M_r/M_s=0.4) and a high saturation field H_s=1.7 kOe (H_s is define to be the field at which M(H) reaches 90% of its saturated value: i.e. $M(H_s)=0.9M_s$.). When the Si layer thickness is increased to 20Å, the magnetic behavior changes dramatically. As seen in figure 1, film B ([Fe30Å/Si20Å]x50) (30/20 ML) behaves like a single thick film of iron with a high remanence and a low saturation field. The magnetization of all of the films (1100-1200 emu/cm^3) is reduced from that expected if each iron atom had its bulk magnetization (1710 emu/cm^3 at RT). One expects in a perfectly layered system that roughly one monolayer of Fe at each interface would have a reduced moment due to Si nearest neighbors. The reduction we observe indicates more extensive interdiffusion. Our results are consistent with those of Fullerton et al on films deposited using magnetron sputtering[3]. The high saturation field and low remanence indicate the Fe layers are antiferromagnetically coupled through the interlayer. We calculate an AF coupling energy density $A_{12}=M_sH_st_{Fe}/2=$ 0.25 erg/cm^2 at RT. In film B, the Fe layers are either ferromagnetically coupled or uncoupled and are thus easily aligned in a small applied field.

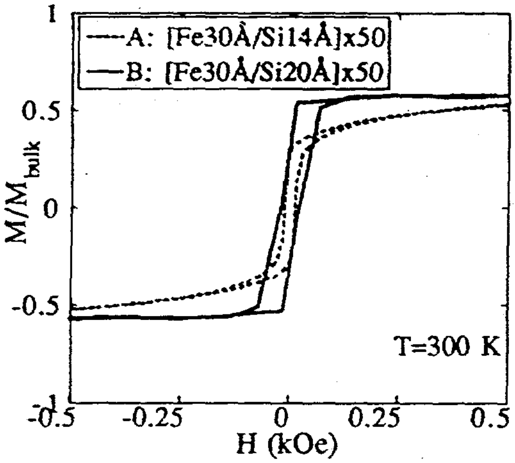

Fig. 1: Hysteresis loops for film A ([Fe30Å/Si14Å]x50) and film B ([Fe30Å/Si20Å]x50). Film A has a high saturation field and a low remanent moment indicating AF interlayer coupling. Film B has a low saturation field and high remanent magnetic moment characteristic of ferromagnetic interlayer coupling or no coupling.

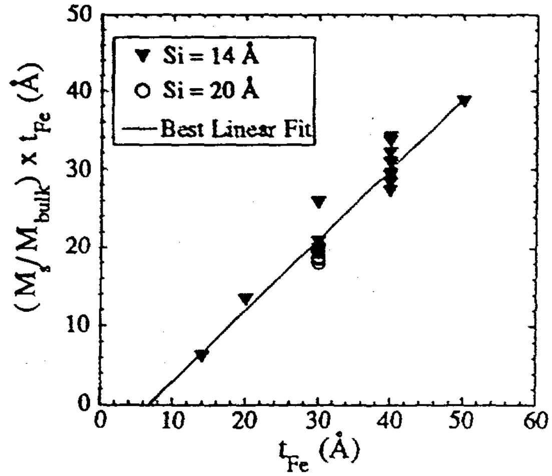

Fig. 2: Estimate from magnetization data of the fraction of Fe which is interdiffused into Si for [FexÅ/Si14Å] MLs. (M/MBulk) is the fraction of the expected bulk Fe magnetization which is experimentally observed. The best linear fit to the data gives $(M/M_{bulk}) \times t_{Fe}$ (Å) = $0.9t_{Fe}$ - 6Å, while an ideal multilayer would have $(M/M_{bulk}) \times t_{Fe}$ (Å) $= t_{Fe}$. An approximately constant thickness of Fe (6- 8Å) becomes non-magnetic due to interdiffusion into the Si layer.

The magnetic properties of Fe/Si MLs also show strong dependence on the Fe layer thickness. Figure 2 shows the saturation moment normalized to the bulk magnetization times the Fe layer thickness versus Fe layer thickness in MLs with 14Å thick Si interlayers. The linear dependence indicates the fraction of iron that is non-magnetic due to interdiffusion into the Si layer in independent of Fe thickness. Assuming Fe atoms either have the full bulk atomic moment or are non magnetic, the intercept shows 6-8Å of the Fe layer is lost into the Si layer. This is a low estimate of the total degree of interdiffusion because it has been shown that Fe atoms with 3-5 Si nearest neighbors retain a reduced but non-zero moment.[9] Notably we find the magnetic moment of MLs with 20Å of Si also shown in fig 2, are generally reduced from their 14Å counterparts. Thus it is reasonable to picture the entire Si interlayer interdiffused to some degree with Fe. The AF coupling energy is approximately constant for thick Fe layers but between 20Å and 15 Å A_{12} drops to zero. High angle x-ray scans indicate this drop may be a result of the thin Fe layer remaining amorphous. It may be that when the Fe layers are amorphous, the strain energy necessary to stabilize the crystalline silicide structure is not present.

Low angle θ-2θ x-ray scans of films A and B(fig.3) show 4 strong reflections indicating the ML are well layered. The ML peaks for the film B are narrower than those of film A indicating a reduced degree of roughness at the interfaces in the 30/20 ML. The bilayer periods derived from the positions of the ML peaks are reduced from the nominal periods by 4-8Å consistent with the reduced magnetization. Analysis of θ-2θ high angle x-ray scans reveals that most of the films are textured with Fe(110) perpendicular to the film plane. Using the well known Scherrer formula, the FWHM of the Fe(110) peaks can be used to approximate the range of the crystalline coherence in the growth direction. For the MLs that show square magnetic loops, the crystalline coherence extends over less than one bilayer period. On the other hand, in the AF coupled MLs, the crystalline coherence propagates typically through 2-3 bilayer periods. Thus the deposition of thinner Si layers allow crystalline coherence to reach from one Fe layer to adjacent layers and implies that the interlayer is itself crystalline. In addition, the AF coupled ML often show a strong Fe(200) reflection indicating a change in the preferred growth orientation in these films. This is the first report of a change in texture in Fe/Si MLs and may be

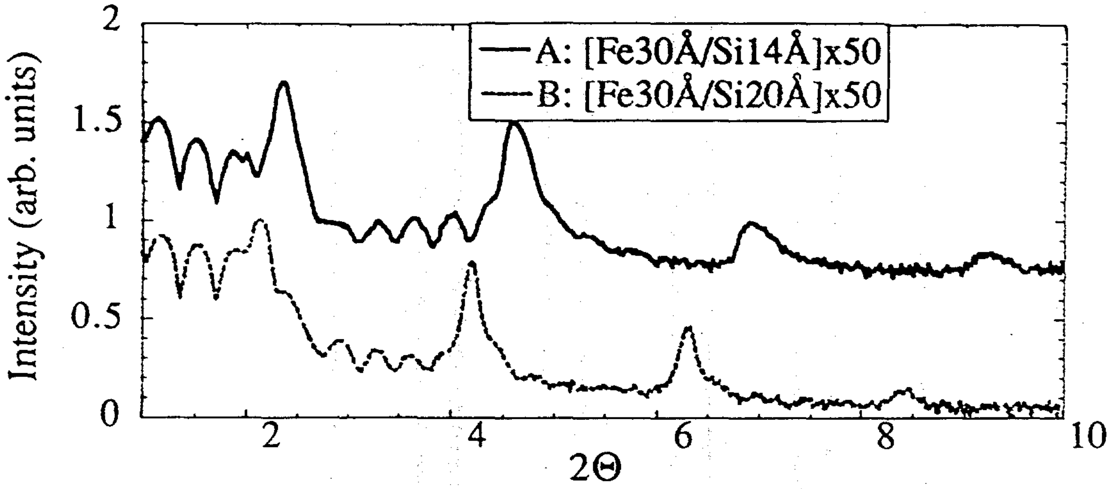

Fig 3: Low angle x-ray scans of the films shown in fig. 1. The ML peaks in film B are narrower than those of film A indicating better layering in the film that is not AF coupled. The interlayers in film B are amorphous while those in film A are crystalline.

unique to the energetics of IBS deposition. The change in texture is probably the result of an interaction between the Fe layer and the crystalline silicide interlayer that forms. A search in x-ray diffraction for Bragg reflections characteristic of the known iron rich silicide phases was unsuccessful.

In order to better correlate the magnetic characteristics of the Fe/Si multilayers to their structural properties and possibly identify the interlayer alloy phase, we carried out a detailed TEM comparison of two MLs. The first, [Fe40Å/Si14Å]x50, (40/14), showed AF interlayer coupling and the second, [Fe30Å/Si20Å]x50, (30/20), showed no evidence of interlayer coupling. Figure 4a,b show real space images of the two MLs. Both films are well layered, consistent with the low angle x-ray scattering results, but the grain structure in the growth direction of the two films is dramatically different. The 40/14 has grains that appear to reach from the substrate all the way through the ML stack. High resolution images confirm the crystalline coherence of Fe layers and the silicide interlayers in this ML. These results are consistent with the long coherence lengths derived from high angle x-ray scattering. In figure 4b no such extended crystalline coherence in observed. Instead the grain size is limited to

a

b

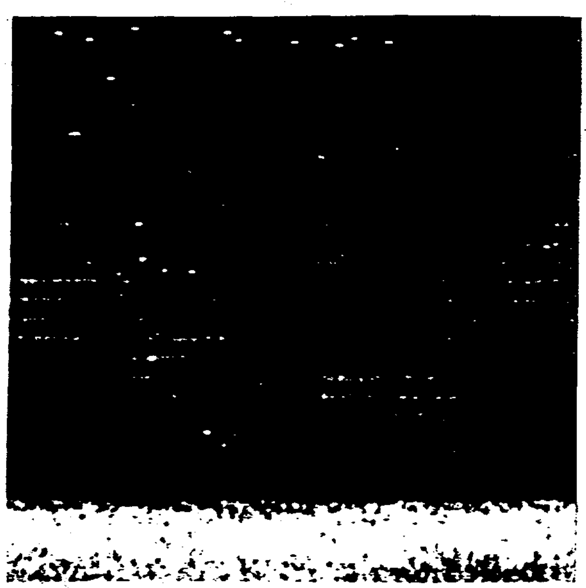

Fig. 4a,b: Real space TEM images of the ML shown in fig 1. Substrate is at bottom. Both ML are well layered. Film A which shows AF interlayer coupling has crystalline grains that reach through the entire stack. In contrast the grains in film B are limited to about the size of one bilayer thickness in the growth direction.

approximately one bilayer thickness. High resolution images confirm that the interlayer of the ML with 20Å of Si is amorphous. The long continuous layering observed in fig 4b are similar to Mo/Si MLs used for x-ray mirrors[10, 11].

Selected area electron diffraction images reinforce the marked difference in the structure of the two multilayers seen in the real space images. Figure 5b shows one nearly continuous ring consistent with the Fe(110) planes. Each Fe layer in this film consists of small grains with random in-plane orientation, textured in the (110) direction. On the other hand figure 5a shows more extensive crystalline order. The 6 bright spots on the (110) ring indicate much larger in plane grain size. The Fe(200) positions are the brightest reflections in the growth direction, indicating a (200) texture consistent with the x-ray scattering results. Significantly, intensity at the Fe (100) position is also evident. The (100) reflection is forbidden in the BCC structure of Fe and its presence in figure 5a is a clue to the interlayer crystalline phase. Both the CsCl and the Fe_3Si phases would produce (100) reflections in this position. The Fe_3Si phase is unlikely since it is ferromagnetic and would produce direct exchange coupling of the Fe layers. The CsCl structure is simple cubic with Fe at the corners and Si at the body center positions, and is closely lattice matched to BCC Fe. Fullerton et al have proposed that the spacer was either CsCl[3] or the epsilon phase[12]. The presence of a (100) reflection in figure 5a is direct evidence that the interlayer crystalline structure is CsCl. The equilibrium bulk binary phase diagram[13] shows the CsCl structure stable at RT for Si concentrations between 10 and 22 at. %. However, von Kanel has shown[14] that strain energy can stabilize the CsCl structure over much broader concentrations for silicide layers grown epitaxially.

In order to test the hypothesis that it is the loss of crystalline coherence and not the increased thickness of the silicide interlayer that results in the loss of AF interlayer coupling, we attempted to disrupt the crystalline coherence by growing the ML at a reduced substrate temperature. Two ML with the same nominal thicknesses were grown, the first on a nominal RT substrate and the second on a LN cooled substrate. [Fe40Å/Si14Å]x40 grown at RT consistently showed a high saturation field and low remanence. In contrast the LN cooled ML showed a square magnetic loop consistent with uncoupled Fe layers. Comparison of the low angle x-ray scans shows the LN cooled growth produces higher quality interfaces, and a bilayer period (52Å) much closer to the nominal period than the RT growth (49Å). Comparison of the FWHM of the high angle structural peaks in the two ML shows the crystalline coherence of the LN grown ML is limited to less than a single bilayer period, while that of the RT grown ML

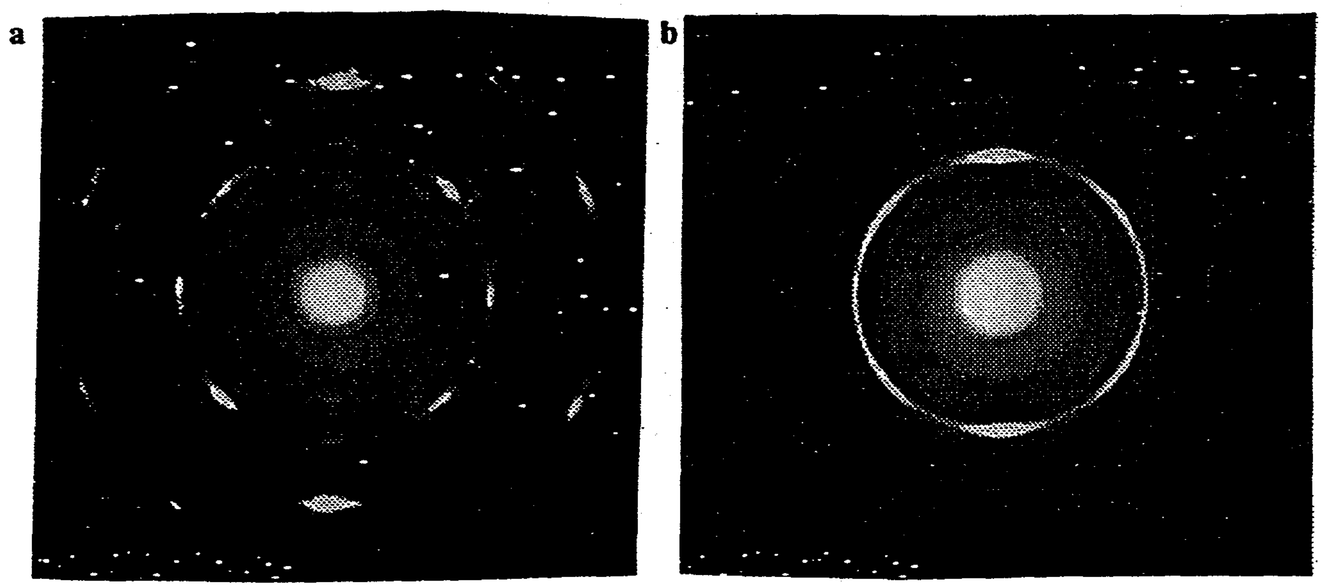

Fig 5a,b: Electron diffraction images of the MLs in fig 4 reinforce the conclusions drawn from the TEM images. (Growth direction along long axis of page.) The texture of film A is predominantly in the (200) direction in contrast to that of film B whose texture is in the usual (110) close packed direction. A faint (100) spot is visible in the image from film A indicating the crystalline silicide interlayer is in the CsCl crystal structure.

extends over 3 bilayer periods. The x-ray results indicate the low deposition temperature affects the crystal structure of the interlayer and that the loss of crystalline coherence through the interlayer eliminates the AF coupling.

CONCLUSIONS

Our results show that in the Fe/Si ML system, it is not straightforward to measure interlayer magnetic coupling as a function of the layer thickness. Unlike most metal/metal ML systems, the gross crystalline structure of Fe/Si MLs changes dramatically with changing layer thicknesses, and strongly affects the magnetic characteristics. We have shown that when the silicide interlayer is in the CsCl structure and is around 14Å thick, neighboring Fe layers are coupled anti-ferromagnetically. When the nominal Si layer thickness is increased to 20Å, the silicide interlayer is amorphous and the AF coupling disappears. It is possible, however, if the growth conditions can be adjusted to maintain the crystalline coherence of the silicide interlayer for larger layer thicknesses, then oscillations in interlayer coupling in Fe/silicide MLs may be measured that are similar to those observed in metal/metal MLs. We plan to try elevated deposition temperatures and post annealing of the MLs to explore this possibility. The exact stoichiometry of the silicide interlayer that produces AF coupling and whether it is a poorly conducting metal or a semiconductor is not clear at this point. The fact that when the Fe layers are amorphous, the AF coupling goes away is strong evidence that the AF coupling depends on the crystal structure of the silicide interlayer and not necessarily on its stoichiometry.

The relevance of the Bruno[6] and the Zhang[5] theories of magnetic interlayer coupling in ferromagnet/insulator or ferromagnet/semiconductor MLs to Fe/Si MLs is uncertain at this time. Future measurements of the conductivity of these MLs in the current-perpendicular-to-plane geometry may reveal whether the silicide interlayer is metallic or semiconducting.

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