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MAGNETIC DECOUPLING IN SPUTTERED Fe/Si DE93 004204

SUPERLATTICES AND MULTILAYERS*

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Magnetic decoupling in sputtered Fe/Si superlattices and multilayers

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A series of sputtered Fe/Si superlattices was grown to study the magnetic coupling between ferromagnetic Fe layers (30Å thick) for Si spacer-layer thicknesses (t_{Si}) between 10 and 40Å. The material is ferromagnetically coupled for $t_{Si} < 13\text{Å}$ and antiferromagnetically coupled for $13\text{Å} < t_{Si} < 17\text{Å}$. For $t_{Si} > 17\text{Å}$ the magnetic Fe layers are uncoupled. X-ray analysis indicates that the system is well layered, but that the crystal structure remains coherent only for $t_{Si} < 17\text{Å}$. These results, along with our Mössbauer investigation, strongly suggest that the Si layer is crystalline for $t_{Si} < 17\text{Å}$, and is iron silicide in nature. For thicker spacers, Si becomes amorphous. We propose a model of the layering that is consistent with the known properties of Fe-silicide.

INTRODUCTION

Since the discovery of antiferromagnetic (AF) coupling of ferromagnetic layers across metallic, non-magnetic spacer layers,¹ there has been extensive experimental² and theoretical³ work in this field. Considerable effort has focused on determining the spacer layer materials which show AF coupling and the structural properties of the spacer layer that determine the strength and periodicity of the coupling. Since AF coupling has been observed for a wide range of metallic element spacer layers, recent work has turned to more exotic spacer layer materials. Toscano *et al.*⁴ observed AF coupling in Fe/Si/Fe trilayers grown at 40K by ultrahigh vacuum evaporation. In this paper, we examine the interlayer coupling in sputtered Fe/Si superlattices. The major finding is that for Si thicknesses in which the superlattice is coupled antiferromagnetically, the non-magnetic spacer layer material is a metastable crystalline Fe-Si compound.

A series of Fe/Si superlattices was grown at ambient temperature by d.c. magnetron sputtering onto sapphire and Si substrates. The base pressure of the sputtering system was 5×10^{-8} Torr and the Ar pressure was 3 mTorr. The superlattices were grown with the Fe layer thicknesses fixed at $t_{\text{Fe}} = 15, 30, \text{ and } 45 \text{ \AA}$ and the nominal Si layer thickness t_{Si} varied from 10-40 $\text{\AA}$, with either 12 or 50 bilayers. Structural properties were characterized by x-ray diffraction using Cu K_{α} radiation. The magnetic properties of the samples were studied by SQUID magnetometry, longitudinal Kerr rotation, conversion-electron Mössbauer spectroscopy (CEMS), and polarized neutron diffraction.

STRUCTURAL CHARACTERIZATION

The structure was characterized by both low- and high-angle x-ray diffraction. The Bragg peaks which result from the compositional modulation are clearly observed in low-angle x-ray diffraction out to high order (>3 orders), indicative of a superlattice structure with smooth layers. The crystalline coherence length ξ , can be estimated from the full width at half maximum (FWHM) of the Fe [110] diffraction peak. It is expected that if the Si spacer layer is amorphous, the separate crystalline Fe layers would scatter incoherently, and ξ would be given by the individual Fe layer

thickness t_{Fe} .⁵ This is observed for $t_{\text{Si}} > 20\text{\AA}$ where there is a weakly textured Fe [110] peak with a FWHM indicative of $\xi \approx 30\text{\AA}$, which is equal to t_{Fe} . This implies that the Si spacer layer is either amorphous or sufficiently disordered to limit crystalline coherence between the Fe layers. For $t_{\text{Si}} < 20\text{\AA}$, ξ increases to $\approx 150\text{\AA}$ and the main Bragg peak is located near the expected Fe[110] position with small superlattice peaks at lower angles, as shown in Fig. 1. This result clearly demonstrates that for $t_{\text{Si}} < 20\text{\AA}$ the Si layer is crystalline. The coherent structure of the spacer layer is only stabilized for thicker Fe layers. For Fe(t_{Fe})/Si(15Å) superlattices with $t_{\text{Fe}} = 30$ and $45\text{\AA}$, a coherent structure is maintained, but for $t_{\text{Fe}} = 15\text{\AA}$, the films are amorphous. Similar structural results were observed for evaporated Fe/Si superlattices where a crystal structure is not observed for Fe thicknesses of less than $20\text{\AA}$.⁶

The crystal symmetry of the spacer layer can be explored by performing an asymmetric diffraction scan to access diffraction planes which are not parallel to the layers. An asymmetric x-ray-diffraction scan performed with the scattering vector 29.5° from the surface normal is shown as an inset of Fig. 1. For the bcc [110] oriented Fe layers, this is the expected position of the bcc [211] peak. Since the films are polycrystalline in the plane, only the angle from the surface normal determines the reflection being probed. A single [211] peak is observed which is coherent over distances greater than $70\text{\AA}$. This result shows that the Si interlayer, in addition to being crystalline, also has a bcc crystal structure. For a superlattice in which the layers have different crystal symmetry (e.g. bcc/fcc), the asymmetric x-ray diffraction spectra consist of diffraction peaks from the constituent layers with coherence lengths limited by the layer thicknesses.⁷

MÖSSBAUER RESULTS

CEMS studies were performed on [Fe(30Å)/Si(17Å)]₁₂ and [Fe(30Å)/Si(15Å)]₅₀ superlattices. The CEMS results for the [Fe(30Å)/Si(15Å)]₅₀ superlattice is shown in Fig. 2. The data consist of a superposition of a magnetically split, six-line spectrum and a non-magnetic central portion which constitutes $\approx 20\%$ of the total spectral area. The polarization of the spins determined from the relative peak heights confirms that the Fe moments are in-plane with no out-of-plane

components in zero applied field ($H=0$). The magnetically split part of the spectrum was fitted to a distribution of hyperfine fields. The results of the fitting procedure are shown by the solid line in Fig. 2. The resultant hyperfine-field distribution were related to the bulk alloy properties determined by Stearns.⁸ The spectrum consists of predominately two magnetic phases: α -Fe (16% total Fe layer) and $\text{Fe}_{0.85}\text{Si}_{0.15}$ (58% total Fe layer) with the remaining magnetic component ($\approx 6\%$) fitted by a >15 at.% Si phase. For the $t_{\text{Si}}=17\text{\AA}$ sample, which has the much shorter structural coherence length, a clear quadrupole split doublet is observed for the nonmagnetic phase. The isomer shift (0.25 mm/s) and quadrupole splitting (0.59 mm/s) are consistent with amorphous Fe-Si, but not with any known crystalline phase.⁶ The nonmagnetic component for the $t_{\text{Si}}=15\text{\AA}$ sample shows substantially less quadrupole splitting, indicating formation of a crystalline spacer layer. The Si concentration of the nonmagnetic phase for both samples is 50 at.% or greater, since they are not magnetically order at room temperature. In the discussion section we utilize the known behavior of the Fe-Si system to understand the presence of two silicide phases.

MAGNETIZATION RESULTS

The in-plane magnetic hysteresis loop and saturation magnetization were measured by means of both a SQUID magnetometer and the longitudinal Kerr effect for fields up to 10 kOe. The saturation magnetization M_S at room temperature in these films is ≈ 1200 emu/cc of Fe. This value is reduced from the bulk value by 30%, in qualitative agreement with the Mössbauer results. Shown in Fig. 3(a) are the in-plane saturation field H_S and the ratio of the remanent magnetization M_r to M_S for an $[\text{Fe}(30\text{\AA})/\text{Si}(t_{\text{Si}})]_{12}$ series grown on sapphire. There is a peak in H_S and a minimum in M_r/M_S for $t_{\text{Si}}=15\text{\AA}$ which is suggestive of AF coupling that is quite strong ($J_{\text{AF}}=H_S M_{\text{stFe}}/4 \approx 0.5$ ergs/cm²). There is no evidence of additional AF-coupling peaks for thicker values of t_{Si} .

Polarized-neutron-reflectivity measurements were performed on an $[\text{Fe}(30\text{\AA})/\text{Si}(17\text{\AA})]_{12}$ superlattice to confirm the AF alignment of the Fe layers. For these experiments, it is necessary to apply a small magnetic field to define the polarization of the neutrons. The room-temperature

neutron reflectivity is shown in Fig. 3(b) for neutrons polarized parallel and antiparallel to an in-plane applied field of 30 Oe. The inset shows the Kerr loop for the same sample. A Bragg reflection is observed at $q_z=0.078\text{\AA}^{-1}$, which corresponds to a periodicity of $2\text{\AA}$ resulting from the AF alignment of the Fe layers. The Bragg reflection disappears when a field of 2 kOe is applied to the sample and the Fe layers align ferromagnetically. By fitting the intensity of the AF Bragg peak (solid and dashed lines in Fig. 3(b)), we find the average Fe moment is $1.5\ \mu_B$ in agreement with the measured M_S value. The critical angles (and momenta) for the two spin states are nearly identical, which indicates that in the presence of a 30 Oe field, the overall ferromagnetic alignment of the Fe layers is limited to yield a net moment of only $\approx 0.37\ \mu_B$.

DISCUSSION

X-ray structural analysis indicates that the system is well layered, but that the coherency of the structure undergoes a transition at $t_{Si}\approx 17\text{\AA}$. Below that thickness a coherence length of $\approx 150\text{\AA}$ is observed. Above that thickness, the crystal coherence is restricted to a single iron layer. These results, along with our Mössbauer investigation, strongly suggest that for Si layer thicknesses $< 17\text{\AA}$ the spacer medium is crystalline, and is iron silicide in nature. For $t_{Si} > 17\text{\AA}$, the spacer layer becomes amorphous.

Since a $30\text{-\AA}$ Fe layer is required to maintain the crystal structure, this suggests that the spacer layer structure is stabilized by epitaxial constraints to the bcc Fe. There appears to be two separate Fe-Si phases, $\text{Fe}_{0.85}\text{Si}_{0.15}$ and a nonmagnetic Fe-Si phase, as determined by the Mössbauer spectroscopy for the $t_{Si}=15\text{\AA}$ sample. The first phase is approximately the upper limit of solubility of Si in bcc Fe.⁹ Studies of sputtered Fe-Si thin films have shown that the crystalline bcc structure is observed up to 15 at.% Si and above 20 at.%, only amorphous structures are observed.¹⁰ The crystal structure, lattice spacing, and Si concentration of the nonmagnetic phase observed in our $[\text{Fe}(30\text{\AA})/\text{Si}(15\text{\AA})]_{50}$ sample are consistent with recent studies that demonstrated the epitaxial growth of metallic thin films of FeSi_{1+x} ($0\leq x\leq 1$) in a CsCl structure on Si(111).¹¹ The authors of Ref. 11 calculated the equilibrium lattice constant for FeSi to be $2.717\text{\AA}$ using a

full-potential linear augmented-plane-wave method, although this approach tends to underestimate the lattice spacing by at most 3%. The Mössbauer results and out-of-plane x-ray-diffraction scans are also consistent with ϵ -FeSi oriented in the (210) direction.

An explanation of the growth of our sputtered Fe/Si superlattices is shown schematically in Fig. 4. It appears that Si interdiffusion into the α -Fe layer up to the solubility limited of about 15 at.% Si precedes formation of the new Fe-Si phase. [Note that in Ni/a-Si superlattices, Si interdiffusion into Ni up to the solubility limit is thought to significantly reduce the interface energy which drives the formation of silicide phases.^{12]} For thicker Si layers, a metastable Fe-Si phase with ≥ 50 at.% Si, which is closely lattice matched to Fe, is epitaxially stabilized and forms a coherent bcc structure. When the Si layer becomes too thick ($>17\text{\AA}$), the more common amorphous silicide phase forms.^{12,13}

The implication of our results is that conduction electrons in the spacer layer are essential for the strong AF-coupling that we observe. We see no evidence of higher-order oscillations in the magnetic coupling for amorphous spacer layers. The magnetic properties are thickness independent for $20\text{\AA} < t_{\text{Si}} < 40\text{\AA}$. We conclude that higher-order AF coupling is not observed because the amorphous semiconducting character of the spacer could not communicate spin information effectively, while the interdiffused silicide layer that forms for thicknesses below $\sim 20\text{\AA}$ *does provide* an effective coupling medium.

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FIGURE CAPTIONS

- Fig. 1: High-angle x-ray diffraction spectra for $[\text{Fe}(30\text{\AA})/\text{Si}(15\text{\AA})]_{50}$ superlattices. Inset shows asymmetric x-ray diffraction scan with the scattering vector at an angle of 30° from the film normal about the bcc (211) reflection.
- Fig. 2: Mössbauer spectrum of $[\text{Fe}(30\text{\AA})/\text{Si}(15\text{\AA})]_{50}$. Solid line is calculated using the model described in the text.
- Fig. 3: (a) Squareness ratio and saturation field determined at room temperature from in-plane magnetic hysteresis loops vs. t_{Si} , for $[\text{Fe}(30\text{\AA})/\text{Si}]_{12}$ superlattices. (b) Polarized neutron reflectivity for an $[\text{Fe}(30\text{\AA})/\text{Si}(17\text{\AA})]_{12}$ superlattice in an applied field of 30 G at room temperature. Solid dots (open circles) are reflectivities for neutron spins parallel (antiparallel) to the applied field. The continued and dashed lines are calculated reflectivities.
- Fig. 4: Proposed structure of $\text{Fe}(30\text{\AA})/\text{Si}(15\text{\AA})$ superlattice based on Mössbauer and x-ray-diffraction results. Only the total $\text{Fe}_{0.85}\text{Si}_{0.15}$ thickness is known and is assumed to be equally divided in this figure.

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Fig. 2

$\longleftrightarrow \Lambda \longleftrightarrow$			
FeSi_{1+x}	$ \text{Fe}_{0.85}\text{Si}_{0.15} $	$\alpha\text{-Fe}$	$ \text{Fe}_{0.85}\text{Si}_{0.15} $
$0 < x < 1$			
CsCl structure	bcc	bcc	bcc
$\approx 10\text{\AA}$	$\approx 9\text{\AA}$	$\approx 5\text{\AA}$	$\approx 9\text{\AA}$

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