

- [3] M.J. Stowell and T.E. Hutchinson, *Thin Solid Films* 8 (1971) 41.  
 [4] G. Zinsmeister, *Thin Solid Films* 2 (1968) 497.  
 [5] E.Y. Jiang, M. Naoe and S. Yamanaka, *J. Tianjin Univ.* 4 (1984) 1.  
 [6] E.Y. Jiang and M. Naoe, *J. Tianjin Univ.* 3 (1985) 25.  
 [7] C.H. Lane and J.C. Anderson, *Thin Solid Films* 26 (1975) 5.  
 [8] M.J. Stowell, *Phil. Mag.* 26 (1972) 349.

## Effects of interfacial roughness on site-probed multilayers of Fe(100)/Ag(100)

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Mössbauer spectroscopy was performed on three  $[\text{Fe}_9(100)/\text{Ag}_{40}(100)]_{25}$  multilayers grown by molecular beam epitaxy (MBE). All of the Fe bilayer components in each sample were selectively loaded with 2 monolayers (ML) of  $^{57}\text{Fe}$ : at the Fe-on-Ag interfaces of one sample, at the Ag-on-Fe interfaces of another, and at the Fe bilayer centers of the third. The hyperfine fields, quadrupole splittings, and isomer shifts of these multilayers were measured at 16, 100 and 300 K. We observed widely different values and trends as the temperature increased, leading us to repeat an earlier claim that three rather than two distinct sites are needed to correctly describe the Fe bilayer components in Fe(100)/Ag(100) multilayered systems.

### 1. Introduction

Advances in ultrathin film growth technology in recent years have made it possible to fabricate high quality epitaxial thin films and multilayers of various crystalline orientations, which often depend sensitively on growth parameters. This group has already done extensive work on Fe(110)/Ag(111) multilayers and superlattices, and we find the Fe(100)/Ag(100) system fascinatingly different. Other groups have also found significant differences in these two systems [1–5]. In particular, our previous work on Fe(110)/Ag(111) [4] demonstrates that those Fe/Ag interfaces are equivalent: that is, there is little, if any, difference between Fe(110) grown on Ag(111) and Fe(110) which has Ag(111) grown upon it. The Fe(100)/Ag(100) system provides a striking contrast. Using transmission Mössbauer spectroscopy, an extremely local and site-specific probe, we have found evidence to corroborate an earlier claim we made that the Fe(100)-on-Ag(100) (“bottom”) interface behaves markedly different from the Ag(100)-on-Fe(100) (“top”) in-

terface, and both of course are distinct from the interior (“middle”) site of an Fe bilayer [6,7].

### 2. Experimental

Three multilayers of the form  $[\text{Fe}_9(100)/\text{Ag}_{40}(100)]_{25}$  were grown by molecular beam epitaxy (MBE) in a Perkin-Elmer 430B system equipped with in situ reflection high energy electron diffraction (RHEED), quadrupole residual gas analysis (RGA) and quartz crystal oscillator and electron impact emission spectroscopy (EIES) deposition rate monitors.

The films were grown on commercially available NaCl substrates, and the details of growth are described elsewhere [7]. The single difference between the previously described growth procedure and the one we employed for this investigation is that for the first time we have grown our multilayers at a constant, reduced substrate temperature ( $T_{\text{sub}} = 0^\circ\text{C}$ ). The work by Heinrich et al. suggests that reduced temperatures are required to obtain the best growth of Fe(100) and Ag(100) [8]. Substrate cooling was achieved by a cold-finger feedthrough cooled by liquid nitrogen. Epitaxial, single-orientation and surface flatness were verified by RHEED for each growth.

For the purpose of transmission Mössbauer spectroscopy, 2 monolayers (ML) of iron in each bilayer of each of the three films were composed

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of  $^{57}\text{Fe}$ , and the rest of the bilayer was composed of natural iron, which has a 2% abundance of  $^{57}\text{Fe}$  and is transparent to the Mössbauer effect. In one of the films the 2 ML of  $^{57}\text{Fe}$  were restricted to the iron-on-silver interface; in another they were restricted to the silver-on-iron interface, and in the last film they were located in the middle of the iron bilayer.

### 3. Results and discussion

All three films were magnetically characterized using transmission Mössbauer spectroscopy. The advantage of this technique lies in its capability to probe the magnetic environment in the immediate vicinity of the absorbing  $^{57}\text{Fe}$  nucleus. Thus by selectively depositing a small amount of  $^{57}\text{Fe}$  we can measure the nuclear hyperfine field of the iron layers as a function of distance from the Fe/Ag interface.

Mössbauer spectra were taken at three temperatures: 300, 100 and at approximately 16 K. The spectral lines were broadened to about twice their normal width, but in each case the data were well fit to a one-sextet and one-singlet model (the singlet arises from trace amounts of  $^{57}\text{Fe}$  in the windows of our cryogenic unit). Fig. 1 shows spectra from all the films at 16 K. The symbols denote data points, while the solid line shows the best fit to the data.

The values of the hyperfine field, quadrupole splitting and isomer shift at the various sites and temperatures are given in table 1. At low temperature all three sites show enhancement of the hyperfine field  $H$  over the bulk value. The 1 and 3% enhancements of  $H$  at the top and bottom interface sites, respectively, is to be expected; the 2% enhancement at the interior site is surprising, as one may instead expect bulk-like behavior in the middle of an Fe bilayer component of this thickness. At low temperature ( $T = 16\text{ K}$ ) a trend in  $H$  with distance from the Fe-on-Ag interface is evident: the hyperfine field is greatest for the Fe layers initially grown on Ag and decreases as the other surface of the Fe bilayer component is approached. This trend is interrupted at higher temperatures, however. At some temperature between 16 and 100 K the value of  $H$  at the interior

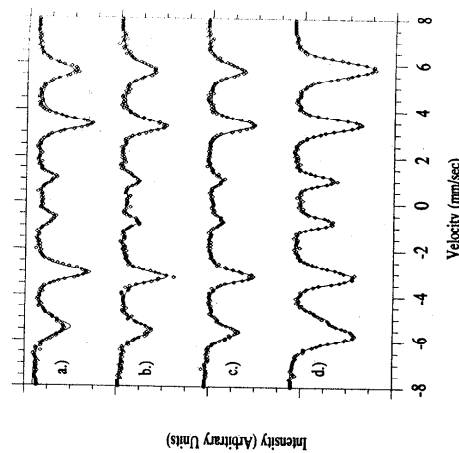


Fig. 1. Mössbauer spectra of each film at 16 K. (a) The top-probed film; (b) the middle-probed film; (c) the bottom-probed film; (d) the non-probed film.

site ( $H_{\text{mid}}$ ) drops below those of the top and bottom interfaces ( $H_{\text{top}}$  and  $H_{\text{bot}}$ , respectively). This is in contrast to our earlier work in which  $H_{\text{mid}}$  was enhanced relative to  $H_{\text{top}}$  and  $H_{\text{bot}}$  at low temperature but drops below  $H_{\text{bot}}$  at approxi-

Table 1

Values of the hyperfine field  $H$ , quadrupole splitting and isomer shift  $d$  at 16, 100 and 300 K for the top probed, middle probed and bottom probed multilayers. The quadrupole splitting values given are the single peak shifts ( $\epsilon$ )

Probed sample	Temperature (K)			
	16	100	300	
Hyperfine field (kOe)	top	345 (1.0)	334 (1.0)	299 (1.6)
	middle	349 (1.0)	331 (1.0)	290 (2.0)
	bottom	351 (0.6)	340 (0.6)	303 (1.0)
Quadrupole splitting (mm/s)	top	-0.03 (0.01)	-0.04 (0.01)	-0.04 (0.02)
	middle	0.0 (0.01)	0.0 (0.01)	0.02 (0.02)
	bottom	-0.03 (0.01)	-0.02 (0.01)	-0.02 (0.01)
Isomer shift (mm/s)	top	0.15 (0.01)	0.13 (0.01)	0.0 (0.02)
	middle	0.07 (0.01)	0.05 (0.01)	-0.06 (0.02)
	bottom	0.09 (0.01)	0.08 (0.01)	-0.05 (0.01)

mately 25 K [6,7]. Finally, even a superficial examination of the behavior of  $H$  at the three sites shows that the hyperfine fields at the two interfaces follow essentially the same temperature dependence, whereas the interior site shows a marked softening of the spin wave spectrum as evidenced by the more rapid decrease of  $H_{\text{mid}}$  as the temperature increases.

The quadrupole splittings and the isomer shifts present in the Mössbauer spectra of the three sites show interesting trends. Since the quadrupole splitting arises from the presence of an electric field gradient at the  $^{57}\text{Fe}$  nucleus, the relative size of the splitting correlates with the degree of asymmetry in the local electronic distribution. At  $T = 16$  and 100 K there is no distinguishable quadrupole splitting in the interior site, indicating a highly symmetric electronic environment. Moreover, at low temperature the splittings at the two interface sites are virtually identical. As the temperature is increased to 100 K the quadrupole splitting of the bottom interface increases (becomes less negative) while that of the top interface decreases (becomes more negative). As  $T$  is further increased to 300 K we detected no significant changes in the splittings of any of the sites in the probed films.

Of all the measurements we made, the behavior of the isomer shift,  $d$ , appeared most consistent. For all three sites and at all temperatures,  $d$  was strongly enhanced over the value for bulk  $^{57}\text{Fe}$  ( $d_{\text{B}} = -0.11\text{ mm/s}$ ). In addition, the smallest isomer shift was always seen in the interior site, and the top site consistently exhibited the largest shift. That is, for all three temperatures,  $d_{\text{top}} > d_{\text{bot}} > d_{\text{mid}}$ . Interestingly, the interface sites showed approximately the same change in  $d$  between 16 and 300 K, while the corresponding change in  $d$  for the interior site was slightly smaller.

Fe(100) grows on Ag(100) with a  $45^\circ$  rotation of its lattice in the film plane. This results in an in-plane lattice mismatch of only 0.77%, but an out-of-plane mismatch of 42.5%. Therefore, Fe that is deposited near Ag step-edges may experience an out-of-plane expansion, resulting in an enhancement in the quadrupole splitting. Such a tetragonal distortion has been seen by Egelhoff et

al. as well [5]. This tetragonal distortion could then be expected to relax over the thickness of the Fe layer. However, this model does not explain the large quadrupole splitting seen in the top interface.

In addition, the RHEED analysis showed that the Fe(100) surfaces were slightly rougher than the Ag(100) surfaces. This causes the top 2 ML of Fe to coordinate more Ag atoms than the other sites, which would suppress the hyperfine field relative to the bottom surface, which may explain the apparent dependence of hyperfine field on distance from the Ag underlayer.

### 4. Conclusions

Three  $[\text{Fe}_9(100)/\text{Ag}_{40}(100)]_{35}$  multilayers were fabricated by MBE. In each of the multilayers 2 ML of the Fe bilayer component consisted of the Mössbauer isotope  $^{57}\text{Fe}$ , located in turn at the bottom (Fe-on-Ag) interface, the top (Ag-on-Fe) interface and the center of the Fe bilayer. The hyperfine fields, isomer shifts and quadrupole splittings of the three films, and therefore of the three sites, were measured at  $T = 16, 100$  and 300 K using Mössbauer spectroscopy. We observed vastly different behavior between the three films, reinforcing the claim we made in an earlier work that three rather than two distinct sites are present within the Fe bilayer component of Fe(100)/Ag(100) multilayered systems. More work needs to be done to understand the mechanisms behind these differences.

### References

- [1] F.A. Volkening, B.T. Jonker, J.J. Krebs, N.C. Koon and G.A. Prinz, *J. Appl. Phys.* 63 (1988) 3869.
- [2] G. Lugert and G. Bayreuther, *Phys. Rev. B* 38 (1988) 11068.
- [3] G. Bayreuther and G. Lugert, *J. Magn. Mater.* 35 (1983) 50.
- [4] J. Korecki and U. Gradmann, *Phys. Rev. Lett.* 55 (1985) 2491.
- [5] C.J. Gutierrez, S.H. Mayer and J.C. Walker, *J. Magn. Mater.* 80 (1989) 299.
- [6] Z.Q. Qiu, S.H. Mayer, C.J. Gutierrez, H. Tang and J.C. Walker, *Phys. Rev. Lett.* 63 (1989) 1649.
- [7] C.J. Gutierrez, S.H. Mayer, Z.Q. Qiu, H. Tang and J.C. Walker, in: *Magnetic Properties of Low-Dimensional Sys-*

- tems II, eds. L.M. Falicov, F. Mejía-Lira and J.L. Morán-López (Springer-Verlag, Berlin, 1990) p. 139.
- C.J. Gutierrez, M.D. Wiecek, H. Tang, Z.Q. Oiu and J.C. Walker, *J. Magn. Mater.* 99 (1991) 215.
- [5] W.F. Egelhoff, Jr., I. Jacob, J.M. Rudd, J.F. Cochran and B. Heinrich, *J. Vac. Sci. Technol.* A8 (1990) 1582.
- [6] H. Tang, M.D. Wiecek, D.J. Keatney, D.F. Storm, C.J. Gutierrez, Z.Q. Oiu and J.C. Walker, in: *Magnetic Surfaces, Thin Films, and Multilayers*, MRS Symp. Proc. vol. 231, eds. S.S.P. Parkin, H. Hopster, J.-P. Renard, T. Shinjo and W. Zinn (1992) p. 405.
- H. Tang, M.D. Wiecek, D.J. Keatney, D.F. Storm and J.C. Walker, *J. Magn. Mater.* 104-107 (1992) 1705.
- [7] C.J. Gutierrez, M.D. Wiecek, H. Tang, Z.Q. Oiu and J.C. Walker, *J. Magn. Mater.* 99 (1991) 215.
- [8] B. Heinrich, K.B. Urquhart, J.R. Dutcher, S.T. Purcell, J.F. Cochran, A.S. Arrott, D.A. Steigerwald and W.F. Egelhoff, Jr., *J. Appl. Phys.* 63 (1988) 3863.

## Structure and magnetoresistance of Fe/Cu superlattices grown on Si(111)

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The Fe/Cu superlattices were grown on HF-cleaned Si(111) wafers at room temperature by electron beam evaporation in ultravacuum. X-ray diffraction and transmission electron microscopy indicate that epitaxial Fe/Cu superlattices are obtained for Cu layers thinner than 24 Å. This epitaxial growth is crucially dependent on which element is deposited first on the Si(111) surface. All superlattices show typically ferromagnetic hysteresis loops and have negative magnetoresistances between 0.2 and 0.3% at room temperature.

The research on metallic multilayered nanostructures has a growing scientific interest due to the remarkable physical properties and potential technological applications of these materials. Nevertheless, less attention has been given to the possibility of direct integration of these materials on silicon, which is the matrix element of most microelectronic devices.

Recently, the epitaxial growth of thick films of bcc-Fe (111) and fcc-Cu (111) on Si(111) surfaces were reported by Cheng et al. [1] and Chang [2], respectively. Since a wealth of information can be found in the literature on epitaxial growth of Fe on Cu and vice-versa we were attracted to fabricate Fe/Cu multilayered structures on Si(111) surfaces [3]. The Fe/Cu system is also very attractive from the magnetic viewpoint. For example, Petroff et al. [4] reported on the oscillatory interlayer coupling and a large magnetoresistance of the spin-valve type in Fe/Cu multilayers prepared by sputtering.

In the present paper, we report on the successful integration of Fe/Cu superlattices on Si(111) wafers. None of our samples showed any consis-

tent trends attributable to oscillatory antiferromagnetic coupling.

The samples were grown on HF-etched Si(111) wafers at room temperature in an ultravacuum system (Balzers UMS 500P) using two quartz crystal controlled electron beams. The base pressure was better than  $2 \times 10^{-8}$  mbar and the deposition rate was  $0.5 \text{ \AA/s}$  for both Cu(6N) and Fe(4N). Two series of multilayered films were prepared. In the first series, denoted by Fe/Cu, the growth of the multilayered films started with Fe on Si(111). In the second series, we start with Cu on Si(111) and we denote the set of films by Cu/Fe series. In both series the Cu thickness,  $t_{\text{Cu}}$ , is varied from 12 to 24 Å with a step of 3 Å. The Fe thickness and the number of bilayers are fixed at 15 Å and 10, respectively. All the multilayered films are finished with the layer of Cu.

We used Auger Electron Spectroscopy (AES) (PHI-SAM 590) to probe the possible contamination of our samples. In fig. 1 we show the AES spectrum of sample with  $t_{\text{Cu}} = 15 \text{ \AA}$  of the Fe/Cu series after sputtering with a 2 kV Ar<sup>+</sup> beam during 1 min. The contamination of the film by oxygen, carbon or nitrogen is below the detection limit of our experiment.

The crystalline structure and composition modulation of the samples was investigated by conventional  $\theta$ -2 $\theta$  X-ray diffraction (XRD) employing Cu K $\alpha$  radiation.

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