

Interface structure and surface morphology of (Co, Fe, Ni)/Cu/Si(100) thin films

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(Received 28 February 1996; accepted for publication 25 July 1996)

We have examined bilayer Co/Cu, Fe/Cu, and Ni/Cu films deposited by molecular-beam epitaxy on hydrogen-terminated [100] silicon substrates. The magnetic metal/copper interface was examined by atomic resolution transmission electron microscopy and compared with the surface morphology as depicted by atomic force microscopy. The general orientation relationships across the magnetic metal/copper interfaces were found to be: $[001]_{\text{Co}}, \text{Ni} \parallel [001]_{\text{Cu}}$; $(010)_{\text{Co}}, \text{Ni} \parallel (010)_{\text{Cu}}$ and $[001]_{\text{Fe}} \parallel [001]_{\text{Cu}}$; $(110)_{\text{Fe}} \parallel (200)_{\text{Cu}}$. The latter system is equivalent to the $[\bar{1}\bar{1}\bar{1}]_{\text{Fe}} \parallel [011]_{\text{Cu}}$ and $(110)_{\text{Fe}} \parallel (100)_{\text{Cu}}$ Pitsch relationship, as has been reported earlier. Furthermore, there was a general correlation between interfacial and surface roughness, indicating that the initial interface character is propagated throughout the film during growth. © 1996 American Institute of Physics. [S0021-8979(96)03021-6]

INTRODUCTION

The magnetic anisotropy of thin films is strongly affected by their microstructure as well as interfacial effects, such as roughness, strain, and interdiffusion.^{1,2} Recently, the magnetic anisotropy of epitaxial nickel, cobalt, and iron films (2.5–50 nm) grown on Cu(100)/Si(100) was examined, using a ferromagnetic resonance technique.³ Epitaxial growth of the magnetic metals was confirmed using *in situ* reflection high-energy electron diffraction (RHEED), with Ni and Co growing with a face-centered-cubic (fcc) (100) structure and Fe growing with a body-centered-cubic (bcc) (110) structure with respect to the fcc Cu (100) lattice. RHEED patterns also revealed that the growth of the metals was predominantly three-dimensional, indicating a rough surface.

In this article we examine the copper/magnetic metal interface, utilizing cross-sectional transmission electron microscopy (TEM), and compare it with the surface morphology of the films as profiled by atomic force microscopy (AFM). We also present atomic resolution TEM micrographs to view the epitaxial growth and orientational relationships between the magnetic metal and the copper lattice and compare with earlier RHEED studies. Plan-view selected area transmission electron diffraction was undertaken on the Fe/Cu/Si(100) film to compare with earlier reported results on this system.

EXPERIMENT

The growth of Cu(100) on hydrogen-terminated Si(100) and of magnetic metals on Cu(100)/Si(100) is described elsewhere.^{3,4} Briefly, the films were grown in an ultrahigh-vacuum environment, using a molecular-beam-epitaxy

(MBE) deposition system with a base pressure better than 2×10^{-10} Torr. The evaporation rate was approximately 0.05 nm/s, based on a quartz-crystal thickness monitor, calibrated using a diamond stylus profilometer. RHEED patterns were continuously monitored during the deposition to gauge the quality and surface structure of the films. A magnetic metal (Co, Fe, Ni) film of 50 nm thickness was grown on a 150-nm-thick Cu(100) seed layer which was deposited on a Si(100) substrate. Si(100) substrates were etched with a 10% hydrofluoric acid solution prior to deposition for hydrogen termination.

The interface structures and orientation relationships of Co/Cu, Fe/Cu, and Ni/Cu bilayers were investigated by atomic resolution TEM. Section samples were fabricated by bonding two films face to face, mechanical thinning, and ion milling. Atomic resolution TEM was performed on a JEOL 4000EX transmission electron microscope, operating at 400 kV enabling a point-to-point resolution better than 0.18 nm. For analysis, electron micrographs were digitized with a Cohu series 4810 solid-state charge-coupled-device (CCD) camera into NIH IMAGE, version 1.35,⁵ modified to incorporate a fast Hartley transform [hereafter, fast Fourier transform (FFT)] routine.⁶ Interplanar spacings of selected regions were determined with reference to a Fourier power spectrum taken from a silicon standard, viewed down a $\{011\}$ direction. A plan-view Fe/Cu/Si(100) sample was also prepared and analyzed using a JEOL 2000FX TEM, operating at 200 kV. The surface morphology was examined by AFM using a Digital Instruments Nanoscope III Multimode AFM, operating in the tapping mode.

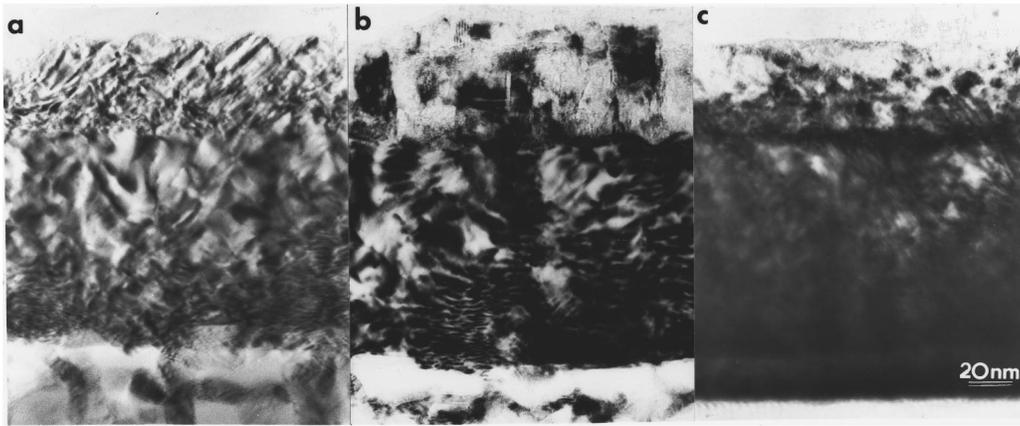


FIG. 1. Section view electron micrographs of (a) Co, (b) Fe, and (c) Ni thin films deposited on a Cu [100] seed layer. The substrate is [100] Si.

RESULTS AND DISCUSSION

Figures 1(a), 1(b), and 1(c) depict 50 nm Co, Fe, and Ni films deposited on 150 nm Cu layers [all on Si (001) substrates]. The Cu/Si(100) interface and details of the Cu layer have been described in detail elsewhere.⁴ A well-defined column structure is evident in the Fe film [Fig. 1(b)], but less so in the Co and Ni films [Figs. 1(a) and 1(c), respectively]. Wavy interfaces of Co/Cu, Fe/Cu, and Ni/Cu are clearly seen. We can estimate the interface roughness amplitude s from these images. In each case the observed lateral periodicity is $\sim 15\text{--}20$ nm. Table I lists these results. These interfaces are shown at atomic resolution in Figs. 2(a), 2(b), and 2(c) (interfaces indicated approximately by horizontal arrows). The interplanar spacings noted are those actually measured from corresponding Fourier power spectra taken from the region on either side of these interfaces (Fig. 3). These may reflect slight misorientations from the zone axis orientations. Nevertheless, they provide a self-consistent reference frame for the determination of lattice misfits, which are found to be -3.8% , -19.6% , and -4.5% , for cobalt, iron, and nickel, respectively. The following orientation relationships between the metal deposits and Cu can be clearly deduced from Fig. 3; $[001]\text{Co}$, $\text{Ni}||[001]\text{Cu}$; $(010)\text{Co}$, $\text{Ni}||(010)\text{Cu}$ and $[001]\text{Fe}||[001]\text{Cu}$; $(110)\text{Fe}||(200)\text{Cu}$. The Fe/Cu relation can be shown to be equivalent to the Pitsch relationship,^{7,8} $[\bar{1}\bar{1}\bar{1}]\text{Fe}||[011]\text{Cu}$ and $(110)\text{Fe}||(100)\text{Cu}$, as verified in earlier work by RHEED.³ As described by Dahman,⁹ this involves an approximately 10° rotation of the bcc lattice relative to the underlying fcc one, and promotes a good directional match in real space. However, this orientation relation is an approximate one, and holds exactly

TABLE I. Interface and surface roughness parameters. s is the interface roughness amplitude (cross-sectional transmission electron microscopy), $\pm 20\%$; R_a is the mean surface roughness amplitude (atomic force microscopy), $\pm 10\%$.

	s (nm)	R_a (nm)
Co	2.5	1.0
Fe	3.5	1.5
Ni	1.0	0.4

only along one direction. Along other directions, misorientations arise, as illustrated in the selected area transmission electron-diffraction pattern depicted in Fig. 4, which encompasses a number of Pitsch type “variants” of the form $\langle 1\bar{1}\bar{1} \rangle\text{Fe}||\langle 011 \rangle\text{Cu}$ and $\{110\}\text{Fe}||\{100\}\text{Cu}$.

Both the nickel and iron films retained their room-temperature equilibrium phases (fcc and bcc structures, re-

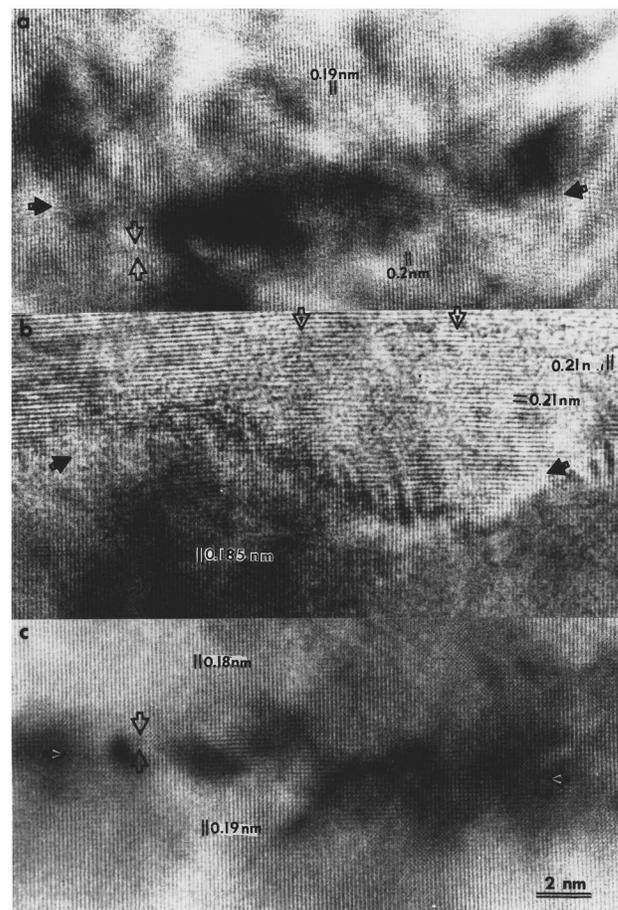


FIG. 2. Atomic resolution transmission electron section view micrographs of (a) Co, (b) Fe, and (c) Ni thin films deposited on [100] Cu. Interfaces are indicated approximately by horizontal arrows.

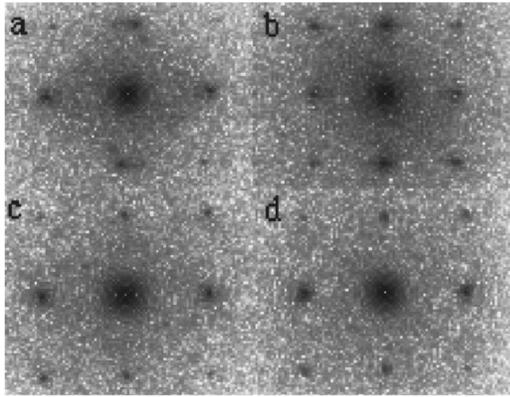


FIG. 3. Fourier power spectra of regions near the: (a) Co; (b) Fe; (c) Ni; (d) Cu interface.

spectively), while the cobalt grew with a fcc lattice (equilibrium structure is hexagonal close packed). Low-energy electron-diffraction studies¹⁰ have shown that iron deposited on (100) copper retains a fcc structure for thicknesses up to 10–14 monolayers after which it reverts to bcc.¹¹ Koike and Nastasi¹² also report initial fcc Fe layers grown on (001) Cu, as do Olsen and Jesser,¹³ who claim it to be pseudomorphic with the underlying copper for approximately the first 2 nm of growth. In the present work, the FFT power spectrum from the iron sample most nearly fits the bcc structure, even for the initial deposit [compare Figs. 3(b) and 3(d)].

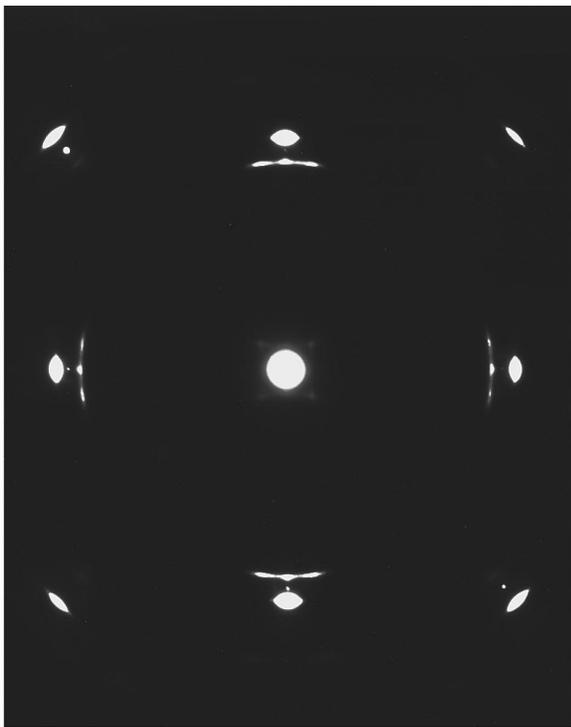


FIG. 4. Plan-view selected area transmission electron-diffraction pattern from a Fe/Cu/Si(100) sample. The innermost iron spots appearing in groups of three are due to Pitsch-type variants $\langle 1\bar{1}1 \rangle \text{Fe} \parallel (011) \text{Cu}$ and $\{110\} \text{Fe} \parallel \{100\} \text{Cu}$.

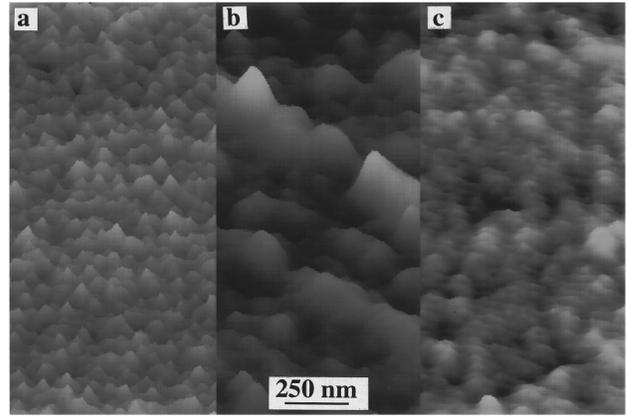


FIG. 5. Atomic force surface morphology images of (a) Co, (b) Fe, and (c) Ni thin films deposited on a [100] Cu. Full vertical scale: 25 nm.

As shown in Fig. 2(b), for the case of the iron deposit, the interface is rather abrupt (transition from one structure into another occurs within 5 nm), whereas for both the cobalt and nickel samples [Figs. 2(a) and 2(c)], it is best described as occurring within a band on the order of 10 nm in extent. In the latter two cases, bending of lattice fringes [vertical arrows in Figs. 2(a) and 2(c)] within a highly strained (as evidenced by the strain contrast present) transition regime is noted. The iron interface [Fig. 2(b)] suffers a more extensive deformation; however, little strain contrast is visible at the interface. No misfit dislocations are visible at the interface; unlike the case of Cu/Si (111),⁴ where the interface accommodates a large misfit by forming numerous dislocations. Instead, we find regions of slight orientational variation [indicated by vertical arrows in Fig. 2(b)] which may lead to the columnar structures referred to in Fig. 1(b).

The “waviness” of the iron/copper interface is not unexpected in light of the relative surface energies of the constituents involved. CO titration studies¹⁴ of Fe deposited onto (100) copper have shown that a substantial fraction of the surface remains covered with copper for thicknesses up to several monolayers. This is due to wetting of Fe by Cu. This wetting or creeping of Cu onto Fe surface may be the cause of the observed waviness in our Fe/Cu interface. From Zangwill¹⁵ we also find that the measured liquid surface tension values change in relative increasing order for Ni, Co, and Fe. Furthermore, Fe, Co, and Ni have much larger surface tension values than Cu. This means Cu wets or creeps more readily on Fe compared to Co or Ni. This suggests an increase in the interface waviness in the order Ni/Cu, Co/Cu, and Fe/Cu. This is indeed what is observed in Figs. 1(a), 1(b), and 1(c).

Figures 5(a), 5(b), and 5(c) show AFM images of Co, Fe, and Ni films deposited on Cu. These images are viewed at an angle of 45° to the page for clarity and are plotted with a 25 nm full scale vertical range (i.e., 100% white=25 nm). The general scale of the surface asperities can be correlated qualitatively with the interface waviness described above. The mean roughness R_a defined as the mean value of the surface relative to the center plane, was computed¹⁶ on 2.5 μm square areas selected as free from obvious surface arti-

facts. Results are tabulated in Table I. The lateral periodicity is $\sim 50\text{--}60$ nm. Both the order of magnitude and the general trends of R_a are comparable to the interfacial roughness amplitudes cited above from cross-sectional TEM. The observation that the magnitude of R_a is less than s is indicative of strain relaxation within the magnetic metal film layers (reduced driving force for surface roughening). This is also supported by the fact that the observed RHEED pattern changes from spotty (indicative of a three-dimensional growth) to streaky (indicative two-dimensional growth) as the thickness of the film increases. The noted correlation between s and R_a attests to the validity of relative surface roughness evaluations as characterizing the film/substrate interface—a point which has not, heretofore, been widely reported. It should also be stressed that care is necessary in interpreting such surface features as individual crystallographic regions. All of the films considered in this work are essentially single orientation layers and, as such, contain no grains, despite the presence of surface asperities resembling grains in the AFM surface morphology scans. These results indicate that the general profile of the initial (<10 nm) interface is propagated throughout the entire 50 nm film, and serves to underscore the importance of the substrate in promoting the resultant film morphology.

SUMMARY

The general orientation relationships across the metal/copper interfaces were found to be: $[001]\text{Co}, \text{Ni}||[001]\text{Cu}; (010)\text{Co}, \text{Ni}|| (010)\text{Cu}$ and $[001]\text{Fe}||[001]\text{Cu}; (110)\text{Fe}|| (200)\text{Cu}$, which is equivalent to $[111]\text{Fe}||011]\text{Cu}$ and $(110)\text{Fe}|| (100)\text{Cu}$, for example. There was general correlation between the surface roughness, profiled by AFM and the

interface “waviness,” as deduced by TEM. This was seen to adopt a configuration that minimizes the total surface energy of the interface, and is propagated throughout the film during growth.

ACKNOWLEDGMENTS

Transmission electron microscopy was carried out at the University of Michigan Electron Microbeam Analysis Laboratory. This work was supported by National Science Foundation Grant No. DMR-9321127.

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