

Using metallic interlayers to stabilize abrupt, epitaxial metal-metal interfaces

C. V. Ramana, P. Masse and R.J. Smith

Physics Department, Montana State University, Bozeman, MT 59717

Bum-Sik Choi

Physics Department, Jeonju University, Jeonju, 560-759, Korea

Abstract

An approach is described for stabilizing metal-metal epitaxial interfaces using a thin metallic interlayer. Rutherford backscattering and channeling techniques along with low-energy electron diffraction and keV He⁺ backscattering are used to demonstrate that an atomically thin layer of Ti metal deposited at the Fe-Al interface, a system well known for considerable intermixing at room temperature, prevents interdiffusion and enables the epitaxial growth of Fe films on the Al(100) surface. The resulting structure is observed to be stable for temperatures up to about 200 °C.

PACS: 68.35.Fx, 68.49.-h, 68.55.Jk, 61.18.Bn

Over the last few years the technology of thin film devices has evolved to the point where individual layers in a multilayer structure may be only a few nanometers thick [1]. Maintaining a well-defined architecture means that the films must be flat on an atomic scale and have abrupt interfaces. The interfaces should at least be thinner than the films making up the structure if the electrical and magnetic properties of the device are to be determined by the characteristics of the films rather than by those of the interface. Various schemes for promoting the growth of flat films have been considered, such as using a surfactant [2] or interfactant [3] to control the surface or interface energy of the growing film. While these techniques improve laminar growth, they are not specifically designed to prevent the interdiffusion that often occurs at the interface between metal layers.

A strategy to limit interdiffusion and to provide a stable template for abrupt, epitaxial metal-metal interfaces is reported here. We show that a single layer of Ti metal at the Fe-Al(100) interface, a system well-known for considerable intermixing at room temperature [4], prevents the interdiffusion of Fe and Al and enables the growth of an epitaxial Fe film. The idea is based on the fact that Ti films were observed to grow in a pseudomorphic fcc structure on Al(100) [5]. The concept is expected to be generally applicable for a large class of metal-metal interfaces.

Rutherford backscattering and channeling (RBS/c), along with low-energy electron diffraction (LEED) and low-energy ion scattering (LEIS) were the primary techniques used in the present work. In the channeling geometry a 1 MeV He^+ beam is incident along a low-index crystallographic direction. The energy spectrum of backscattered He exhibits a surface peak associated with ions backscattered from the topmost layers of the solid [6]. Incident ions missing

the target nuclei by more than a few tenths of an Angstrom are essentially undeflected and channel along the relatively open region between the rows of atoms. A surface atom displaced by a few tenths of an Angstrom uncovers the next atom along that row and the backscattered ion yield from substrate atoms increases. An adatom of a different element sitting directly above a substrate atom reduces the backscattering yield from substrate atoms. Low-energy (1 keV) He^+ scattering (LEIS) allows us to study the surface composition for the growing film and to investigate possible interdiffusion with monolayer depth resolution. Electron diffraction (LEED) is used to characterize the long-range order of the surface while high-energy ion channeling and shadowing provides quantitative information about local order for adatoms above substrate lattice sites.

The Al single crystals were prepared using standard techniques [5]. Metal films were deposited on the Al surfaces in vacuum using resistively heated wires. A deposition rate of about 0.3 monolayer/min was determined using ion backscattering. We use the Al(100) surface density of 1.22×10^{15} atoms/cm² to define one monolayer (ML) for Fe and Ti coverage. All metal depositions were performed with the Al sample at room temperature. The ultrahigh vacuum target chamber is connected to a 2-MV Van de Graaff accelerator through a differentially-pumped beam line [7].

In vacuum the crystals were cleaned by repeated cycles of Ar^+ ion bombardment at room temperature followed by annealing at 500 °C. The cleaning procedure was repeated until the LEIS spectrum showed only the Al backscattering peak with negligible peaks from any impurities such as oxygen. A total dose of 3×10^{14} ions/cm² was used to collect each channeling spectrum. No increase in ion yield attributed to beam damage during channeling was observed. Measurements with a non-channeling beam alignment were also made to avoid errors associated with the

shadowing of Ti or Fe atoms when measuring adatom coverage.

Figure 1 shows the high energy ion backscattering and channeling spectra obtained for 1 MeV He^+ ions incident on the Al(100) surface. The filled circles show a conventional backscattering spectrum with a non-channeling incident angle for the clean surface. The cutoff of the spectrum at 700 keV corresponds to those ions backscattered from the Al surface atoms at the set scattering angle of 105° . When the target crystal is rotated to align the [100] direction with the incoming collimated ion beam, the backscattered ion yield (open circles) is reduced due to channeling. Surface atoms now shadow bulk atoms. Those ions that scatter from the surface atoms give rise to the remaining peak in the ion yield at 700 keV. Calculations using the Rutherford cross section and the known experimental parameters allow us to convert the measured yield in counts to an effective number of target atoms (atoms/cm^2). The measured yield for the clean Al surface (not shown) was $15 \times 10^{15} \text{ at}/\text{cm}^2$, in reasonable agreement with the value of $14 \times 10^{15} \text{ at}/\text{cm}^2$ calculated using the computer simulation code VEGAS [8]. The channeling spectrum shown in Fig. 1 was obtained after the sequential deposition of 2 ML of Ti followed by 3 ML of Fe. He ions backscattered from the deposited atoms at the surface give rise to the two peaks at larger backscattering energies as indicated by the vertical arrows. It is straightforward to extract the areas of the two peaks as shown by the solid lines obtained using standard non-linear least squares methods. The experiments reported here consist of measurements of these three peaks in the ion yield as a function of Fe and Ti deposition on the surface.

The upper panel of Fig. 2 shows the variation of the Al surface peak area as a function of the Fe peak area for a series of Fe depositions with no Ti interlayer. The scattered ion yield from Al

atoms increases monotonically to a coverage of about 3 ML of Fe. These results are in agreement with previously published results for Fe deposition on Al(100) surfaces [4]. The interpretation is that Fe atoms alloy with the Al surface atoms, causing Al atoms to move off of substrate lattice sites, uncovering substrate Al atoms, and resulting in increased numbers of Al atoms visible to the incident ion beam. The reaction continues until the thickness of the alloy impedes the further interdiffusion of Fe and Al atoms at room temperature, at which point an additional 7 monolayers of Al atoms have become visible.

The lower panel of Fig. 2 shows the area of the Al surface peak as a function of Fe coverage with 1 ML of Ti deposited initially on the clean Al surface (circles), and in a later experiment for 2 ML of Ti deposited prior to Fe deposition (squares). The effect of the Ti interlayer is very remarkable. Now, the number of visible Al atoms *decreases* as Fe atoms are deposited on the surface. Rather than causing Al atoms to be displaced, the Fe atoms cover and shadow the substrate Al atoms, and must be growing in an epitaxial structure on the Al-Ti template. For such a reduction in ion yield to occur, the relatively narrow shadow cone created by the Fe atoms (0.11 Å cone radius at a distance of 4.05 Å behind the Fe atom) must partially overlap a substrate lattice site. That is, the Fe atoms are essentially within a horizontal distance of one shadow cone radius of the substrate lattice sites. From Fig. 2 it can be seen that using 2 ML of Ti prior to Fe deposition is even more effective in promoting the growth of an epitaxial Fe overlayer that shadows the Al. Adding a third monolayer of Ti (not shown) does not increase the shadowing significantly beyond that for 2 ML of Ti. We interpret this behavior as indicating that the Ti does not grow in a perfectly flat layer at room temperature, but instead grows with some second layer Ti atoms and

some uncovered regions of the Al substrate at the completion of 1 ML deposition. Adding the second Ti layer apparently completes the coverage of the Al surface atoms to stop the Fe-Al alloy formation. The broken line in Fig. 2 shows the behavior expected for the Al surface peak area assuming 2 ML of Ti atoms on Al lattice sites, followed by Fe deposition. The calculations were done using the VEGAS code, the interplanar distance of 1.43 Å for bcc Fe, and the Al vibration amplitude of 0.105 Å for both the Fe and Ti atoms. The simulated value for the clean surface (14×10^{15} at/cm²) is positioned at a coverage of -2 ML so that the Al surface with 2 ML of Ti is at zero Fe coverage. It can be seen that in the simulation the Ti atoms also shadow substrate Al atoms and decrease the scattered ion yield, unlike the measurements. For 7 ML of Fe the simulated and measured yields are quite close, suggesting that the order in the Fe overlayer is quite good. The lack of shadowing by the first Ti monolayer is consistent with results reported previously [5,9], and suggests that there may be some surface alloying occurring for the first layer of Ti atoms.

The Ti interlayer effectively stops the interdiffusion of Fe and Al atoms, leading to local ordering and shadowing of substrate atoms by Fe adatoms. It also results in improved long-range order for the Fe film that eventually grows on the surface, as shown by the low-energy electron diffraction (LEED) patterns of Fig. 3. The diffraction pattern for the clean Al(100) surface (Fig. 3a) quickly disappears with Fe deposition, but reappears at higher Fe coverage (Fig. 3b) with considerably broadened spots, in agreement with earlier reports [10]. The pattern is attributed to small Fe islands growing on an Fe-Al alloyed interface. Some degree of orientational order must be maintained by the small crystallites during alloying since the final pattern has the orientation of the clean surface. With the Ti interlayer a LEED pattern is obtained for much smaller Fe coverages,

and the individual spots are considerably sharper. The pattern for 5 ML Fe on an interlayer of 2 Ti ML is shown in Fig. 3c. Thus the interlayer not only stops interdiffusion at the interface, as shown by the shadowing in Fig. 2, but also promotes the growth of larger epitaxial islands. Patterns similar to that shown in Fig. 3c were visible for Fe coverages from 3 ML to the end of the experiments at about 8 Fe ML.

For device applications using the interlayer concept it is desirable to have thermal stability of the structure at the elevated temperatures required for film growth. We studied the Fe/Ti/Al(100) structure as a function of annealing temperature using low energy ion scattering to monitor the relative Fe, Ti, and Al surface compositions. Although the LEIS technique is not as quantitative as conventional RBS because of unknown scattering cross sections and neutralization rates at the surface, when used in the ion detection mode the technique does provide an extremely sensitive measurement of relative surface composition in many circumstances [11]. We performed LEIS measurements for the case of 2 ML Ti with an Fe deposition of 6 ML. Following Fe deposition, no Ti is visible in the LEIS spectrum, indicating that the Ti interlayer is buried at the interface. Up to a temperature of approximately 150 °C only slight changes in the Al and Fe peak areas were observed. After annealing the surface to 200 °C we observed increased Al yield and decreased Fe yield, suggesting that Al atoms were moving to the surface of the sample. After annealing to 300 °C the Fe atoms were clearly moving away from the surface, and, following an anneal at 400 °C, the Ti atoms had also moved into the Al substrate. The instability to diffusion for 5 ML of Ti on Al(110) at 450 °C was reported previously [12].

Finally, we present a simple model for understanding the effectiveness of the interlayer based on the interatomic distances for the intermetallic compounds having the least complex structures and the associated formation energies for these compounds [13]. First, we note that the (100) surface of bcc Fe has a lattice mismatch of only 0.3% compared to the Al(100) surface [14]. One might then expect ordered growth of Fe on Al(100) but interdiffusion occurs already at room temperature with the negative formation energy (-25 kJ/mole-at for FeAl) providing the driving force. The formation energy for Ti-Al is larger than that for Fe (-38 kJ/mole-at), but Ti remains on the Al surface up to 350 °C. This kinetic barrier to Ti diffusion exists presumably because of the larger metallic radius of Ti. Thus, Ti diffusion into the substrate probably occurs via an exchange reaction, or may require Al vacancy formation on the surface, with both mechanisms occurring more readily above room temperature. Now, with the Ti interlayer forming a surface alloy [9], the surface interatomic bonds have been strengthened relative to those of pure Al, and the energy required to break a bond and exchange an Fe and Al atom has increased. Apparently, the formation energy gained by Fe atoms moving into the lattice is no longer sufficient to drive the exchange reaction and diffusion is greatly reduced at room temperature. Heating the surface to 200 °C enables the Fe atoms to overcome even these stiffened surface bonds at the Ti-Al interface and the effects of the interlayer are compromised.

We have demonstrated the use of an atomically thin Ti interlayer to stabilize epitaxial growth for Fe films on Al(100) surfaces, a system which exhibits considerable interdiffusion at room temperature. Ongoing work will better define the structure of the Fe overlayer and its magnetic properties. These results suggest that other interlayer materials should exist, and we have

suggested a simple model based on atomic size and formation energies that should be useful for identifying other beneficial interlayer systems. Using interlayers for stabilizing metal-metal interfaces and promoting epitaxial growth should significantly advance the development of metallic thin film devices.

The authors are pleased to acknowledge the technical support of Norm Williams. This work was supported by the National Science Foundation under Grant DMR-007534.

Figure Captions

FIG. 1. Rutherford backscattering spectrum for 1 MeV He⁺ incident on clean Al(100) with a non-channeling incident angle (filled circles). Channeling spectrum (open circles) obtained with the ion beam directed along the [100] direction perpendicular to the surface. For this spectrum 2 ML of Ti were deposited on the surface prior to deposition of 3 ML of Fe. Vertical arrows indicate the energy position for ions backscattered from surface Fe and Ti atoms. The small triangle represents the background area subtracted from the Al peak area.

FIG. 2. Visible Al atoms for 1 MeV incident He⁺ ions, as a function of Fe coverage on Al(100). Upper panel shows the increase of Al surface peak area for Fe deposition with no Ti interlayer. The increase is attributed to alloy formation and disorder at the interface. Lower panel shows the decrease of the Al surface peak area as a function of Fe coverage with one (circles) and two (squares) ML of Ti deposited on the clean Al surface prior to Fe deposition. The decrease is attributed to shadowing of Al by Fe atoms. The broken line shows the calculated behavior expected for an epitaxial Fe overlayer.

FIG. 3. Low-energy electron diffraction (LEED) patterns for Fe deposition on Al(100). (a) Clean Al(100), 52 eV. (b) 9 ML Fe on Al(100) with no Ti interlayer, 52 eV. (c) 5 ML Fe on Al(100) with 2 ML Ti interlayer, 40 eV.

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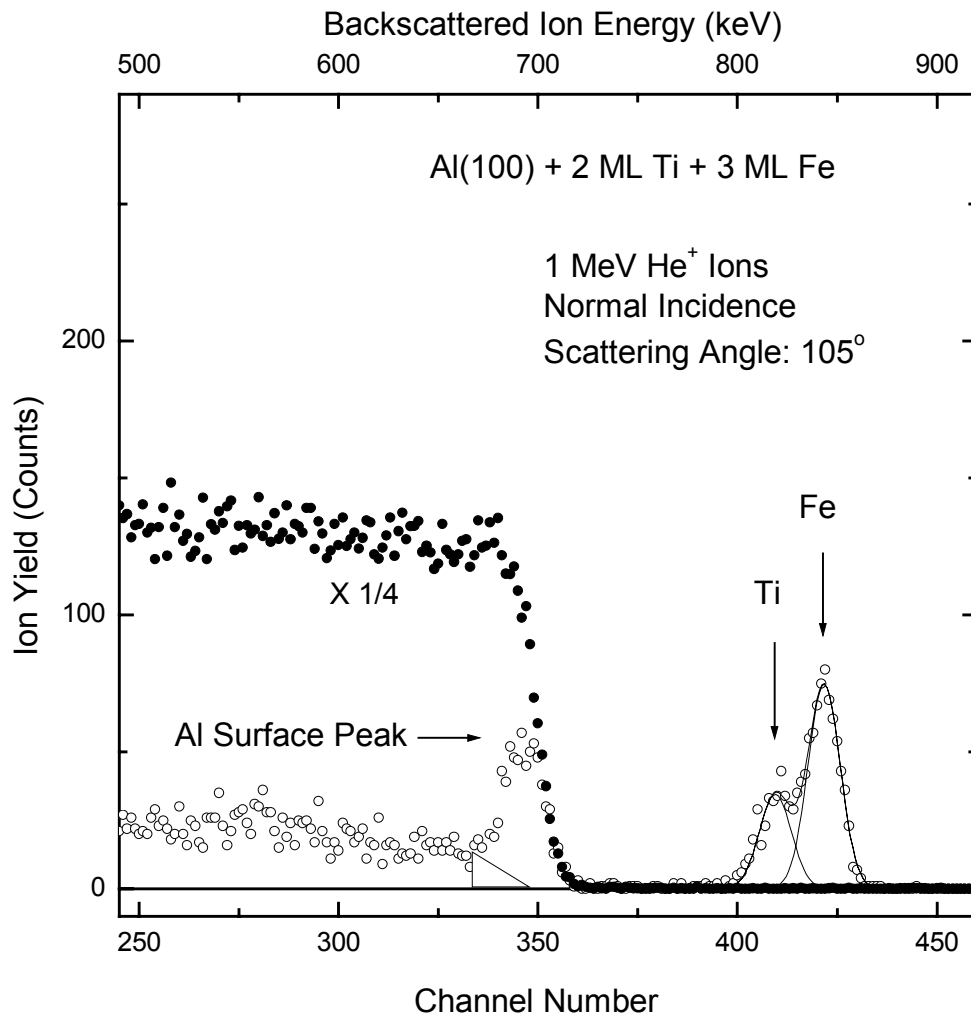


Figure 1

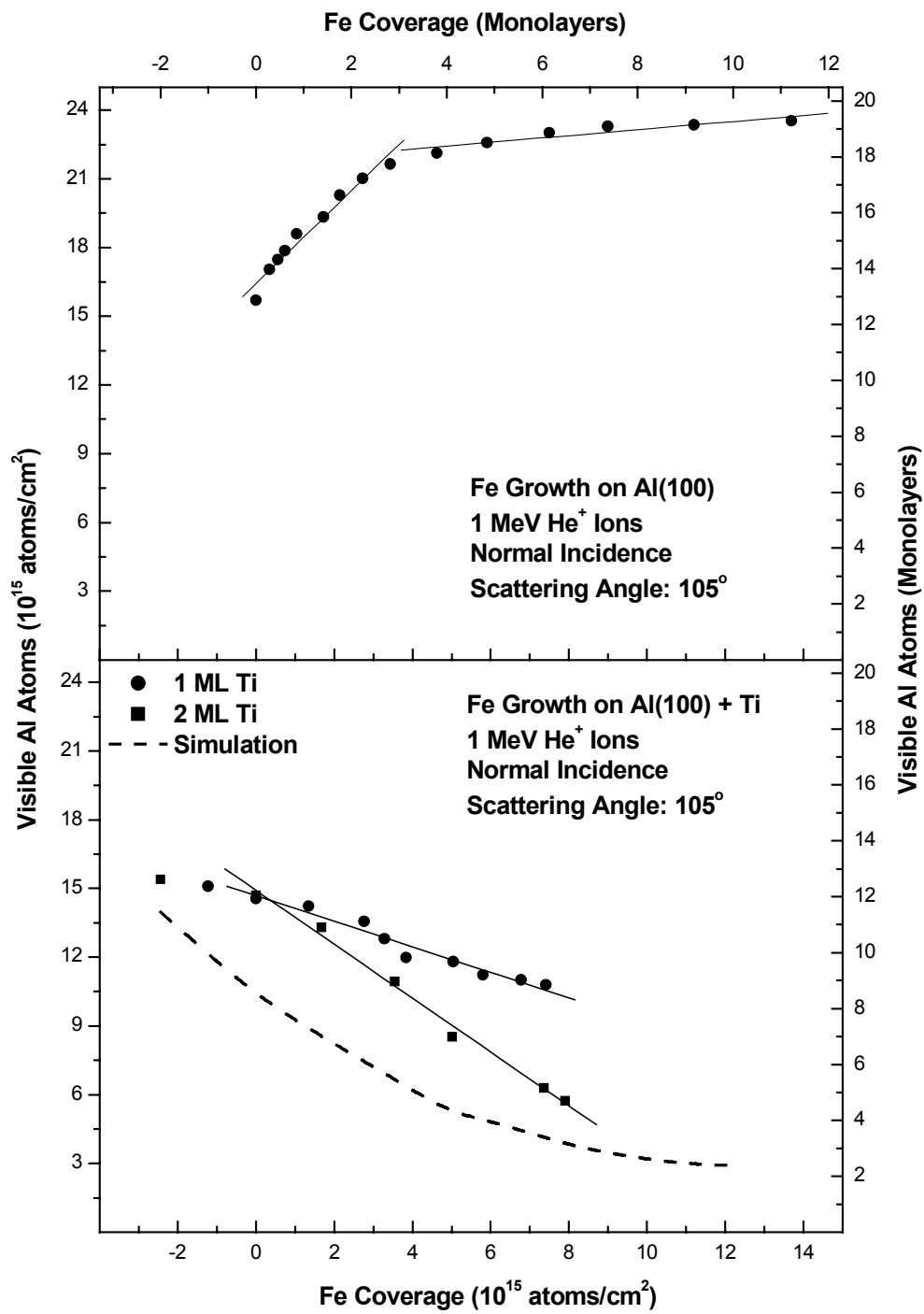


Figure 2

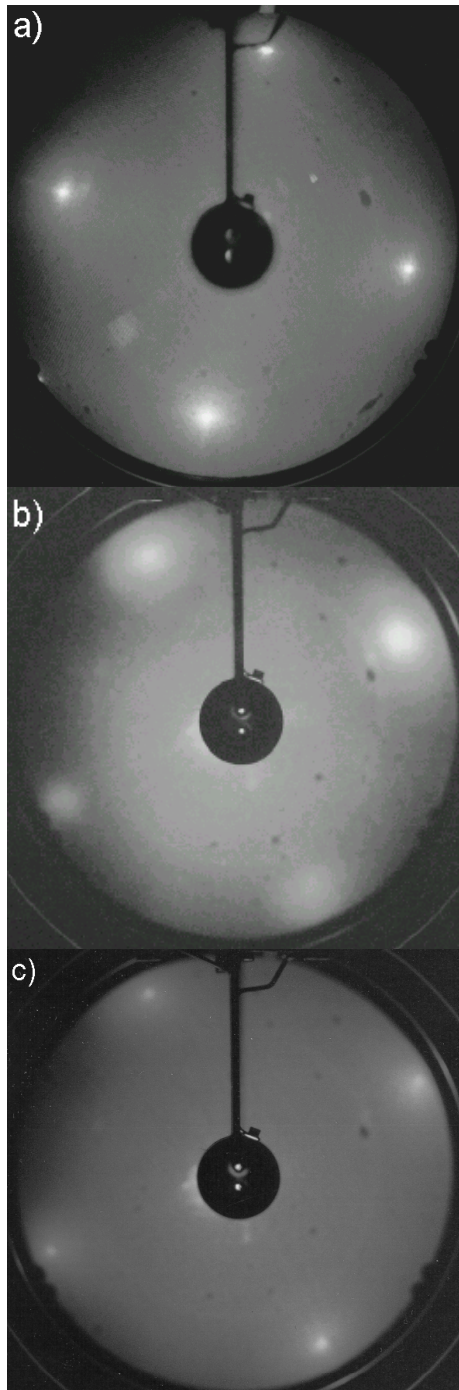


Figure 3