

## Direct observation of elastic strain and relaxation at a metal-metal interface by Auger electron diffraction: Cu/Ni(001)

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We have used Auger electron diffraction with high angular resolution to measure elastic strain at a pseudomorphic metal-metal interface. Shifts in the position of the Cu  $L_3M_{4,5}M_{4,5}$  Auger intensity maximum along the [101] direction betray expansion of the Cu lattice normal to the Cu/Ni(001) interface resulting from the Cu-Ni lattice mismatch. In the pseudomorphic regime (up to 14 Å of Cu), the Cu lattice constant perpendicular to the interface has been determined to be  $3.71 \pm 0.03$  Å while the lattice constant is 3.52 Å in the plane of the interface (the lattice constant of Ni). Thus, the unit-cell volume of Cu is  $46.0 \pm 0.4$  Å<sup>3</sup> in the pseudomorphic overlayer, compared to a bulk value of 47.0 Å<sup>3</sup>. Above 14 Å, the lattice constant perpendicular to the interface drops as a result of dislocation generation and the relief of elastic strain. The critical coverage at which strain relief begins and the dependence of strain on coverage are in good agreement with simple classical models.

The assessment of atomic structure and order (or disorder) at solid-solid interfaces is of considerable importance for understanding the transition layer between two extended solids. This transition region is of great scientific interest and has significant ramifications in areas such as microelectronic and ultrathin magnetic materials development. Indeed, changes in lattice positions at the interface relative to those in bulk materials have marked influence on electrical transport phenomena and magnetic properties in the interfacial region. Although the importance of atomic structures at interfaces is well recognized, it is extremely difficult to determine atomic position with adequate precision.

In this paper, we describe the use of Auger electron diffraction with high angular resolution to determine atomic positions at an interface where pseudomorphic growth occurs—Cu/Ni(001). We have measured the Cu interlayer spacing *perpendicular* to the interface as a function of Cu coverage and have observed interfacial strain and subsequent lattice relaxation as dislocations are generated and propagate. Relaxation is observed to begin above a critical coverage of 14 Å. These results and analogous results which can be obtained with this technique for other interfaces will make it possible to map interfacial morphology with subangstrom accuracy, thereby setting the stage for precise modeling of other interfacial properties such as transport phenomena and magnetism.

The bulk lattice constants of Cu and Ni are 3.61 and 3.52 Å, respectively, leading to a lattice mismatch of 2.56%. For an interface with so much inherent mismatch, qualitative predictions based on simple energy-minimization ideas would indicate layer-by-layer growth at low coverage and the development of strain-relieving dislocations at higher coverage. In studies of these phenomena, Chambers and Jackson used transmission electron microscopy (TEM) and reflection high-energy electron diffraction (RHEED) to measure the Cu lattice constant *in* the plane of the interface and detect the presence and nature of dislocations as the overlayer thickness increased.<sup>1</sup> Matthews and Crawford<sup>2</sup> and Jesser and Kuhlmann-Wilsdorf<sup>3</sup> have developed models which predict this overlayer strain as a function of overlayer thick-

ness  $h$ . These models also predict a maximum critical thickness  $h_c$  for which pseudomorphic growth can be sustained without the introduction of dislocations and associated lattice relaxation. The value of  $h_c$  determined by TEM fell considerably short of the predicted values of 13–15 Å, however, although the behavior of strain versus coverage above  $h_c$  was in reasonable agreement with theory.

More recently, Egelhoff has shown that epitaxy at the Cu/Ni(001) interface can be easily detected by high-energy Auger electron diffraction.<sup>4,5</sup> He found that local maxima in the Cu  $L_3M_{4,5}M_{4,5}$  Auger intensity occur along low-index directions in polar intensity profiles. Subsequent single-scattering cluster calculations by Bullock and Fadley showed that zeroth-order forward scattering of outgoing electrons by atoms in the exit path is responsible for the strong maxima along the [101] and [001] directions, but that other peaks in the polar profile have a more complex origin.<sup>6</sup> Unfortunately, this work was performed at relatively low angular resolution so that, although the basic effect was observed, slight changes in bond distance as a result of elastic strain could not be detected. Our purpose here is therefore to build on the foundations established by Egelhoff and Bullock and Fadley and to use high angular resolution to look for effects brought about by elastic strain and relaxation.

Our measurements were performed with higher angular resolution ( $\Delta\theta\Delta\phi = 2^\circ \times 4^\circ$ ) than we used in previous studies of metal-metal<sup>7</sup> and metal-semiconductor interfaces.<sup>8,9</sup> A Ni single crystal, cut and polished to within 0.3° of (001), was chemically etched prior to insertion into the spectrometer (operating pressure  $< 5 \times 10^{-11}$  Torr). Light sputtering with 500-eV Ar<sup>+</sup> ions and annealing at 600°C produced a clean surface and a sharp low-energy electron diffraction pattern. Resistive evaporation of high-purity Cu from a W boat was monitored by a quartz-crystal oscillator. During evaporation, system pressures never exceeded  $2 \times 10^{-10}$  Torr. Polar profiles of Ni and Cu  $L_3M_{4,5}M_{4,5}$  Auger intensities were obtained from spectra collected in 1° steps for polar angles  $\theta$  of 10°–96°. Each spectrum was then smoothed, background subtracted, and area integrated.

In Fig. 1 we show polar profiles in the (010) azimuthal

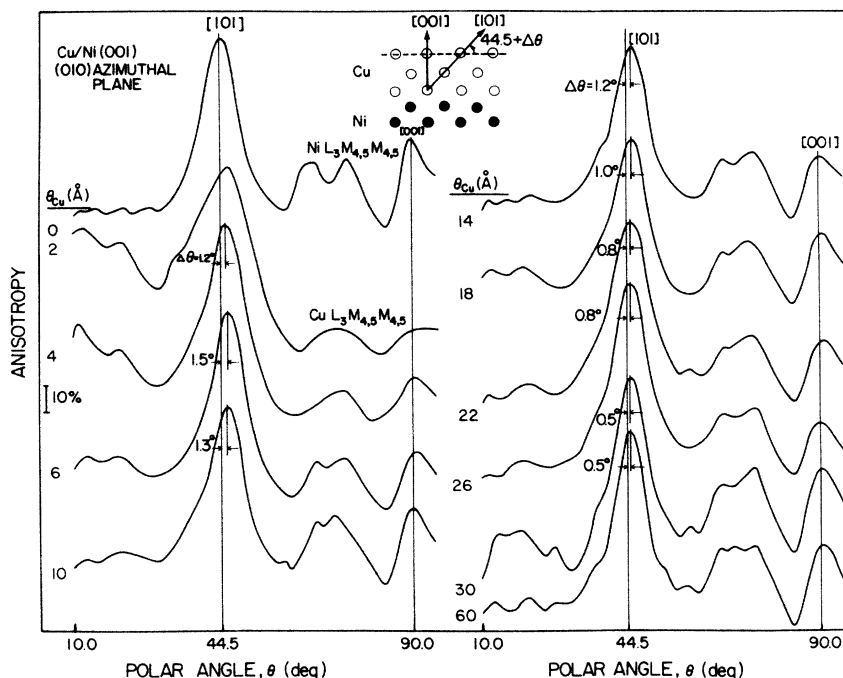


FIG. 1. Polar intensity profiles for Ni  $L_3M_{4.5}M_{4.5}$  (top, left-hand curve) and Cu  $L_3M_{4.5}M_{4.5}$  (all other curves) Auger emission from Ni(001) and the Cu/Ni(001) interface as a function of coverage, expressed in angstroms, where 1 Å = 0.525 ML. The uncertainty in  $\Delta\theta$  is estimated to be  $\pm 0.2^\circ$ . Incident electron beam energy was 5 keV.

plane for Ni  $L_3M_{4.5}M_{4.5}$  emission (at 844 eV) from Ni(001), and for Cu  $L_3M_{4.5}M_{4.5}$  emission (at 916 eV) from the Cu/Ni(001) interface at various stages of development. Maxima at near  $45^\circ$  and  $90^\circ$  result from zeroth-order forward scattering of the Auger electron waves by atoms in the exit path along the [101] and [001] low-index directions, respectively. The weaker features originate from a combination of zeroth-order forward scattering along other low-index directions (for example, [103] at  $71.6^\circ$  and [301] at  $18.4^\circ$ ), first-order diffraction associated with major zeroth-order forward scattering peaks, and interference phenomena resulting from the overlap of waves scattered from numerous atoms in the crystal. The peak along the [101] direction occurs at a polar angle of  $44.5^\circ$  for the substrate, rather than  $45.0^\circ$  as expected from the crystal structure, because of outgoing wave refraction at the solid-vacuum interface.

The results of Fig. 1 show that a broad peak develops at  $\theta \cong 46^\circ$  upon deposition of 2 Å [1.05 monolayer (ML)] of Cu. We interpret it as being due to the formation of limited amounts of second Cu layers on the base monolayer, consistent with the conversion from angstroms of Cu to monolayers on the Ni(001) surface. We can rule out the possibility of more than double-thick layers based on the absence of a peak at  $\theta = 90^\circ$ . Increasing the coverage to 4 Å (2.1 ML) of Cu results in the sharpening of the feature along [101] and the appearance of the maximum at  $\theta = 90^\circ$ , indicative of the presence of mixed two- and three-layer epitaxial Cu on Ni. The peak along [101] is as narrow for 4 Å as that seen for the clean substrate, but it is shifted to higher polar angle by an amount  $\Delta\theta = 1.2 \pm 0.2^\circ$ . This result suggests that by a coverage of approximately two monolayers, the interface consists of a structurally well-defined overlayer which has grown in a pseudomorphic fashion. Compression

of the Cu-Cu interatomic spacing in the plane of the interface, brought about by the Cu-Ni lattice mismatch, has been accompanied by lattice expansion perpendicular to the interface. This expansion, which is revealed by an increase in the polar angle at which the [101] feature is observed, continues to a coverage of 14 Å, above which  $\Delta\theta$  steadily drops to a constant value of  $0.5^\circ$  by 30 Å.  $\Delta\theta$  then remains unchanged until at least 100-Å coverage.

In addition to these polar profiles, we have also performed azimuthal intensity scans at fixed polar angles of  $7^\circ$  and  $45^\circ$  for interfaces with 6- and 100-Å coverage. The purpose of these studies was to see if such measurements reveal details related to structural differences accompanying lattice strain and relaxation. These angular distributions were, however, essentially identical for the two coverages and we conclude that the peak along [101] in the polar scans is the only feature which changes as relaxation occurs.

To determine the lattice constants of this system, we first note that in the studies by Chambers and Jackson, the absence of Moiré fringes in TEM images demonstrated that lattice misfit was taken up by elastic strain for a few ML of Cu on Ni(001).<sup>1</sup> Thus, for Cu lattice constant in the plane of the interface  $b$  can be taken to be the same as that of Ni for low coverages, namely, 3.52 Å. The lattice constant perpendicular to the interface  $c$  in the pseudomorphic overlayer can then be determined by comparing experimental polar profiles to those generated by single or kinematical scattering theory.<sup>10</sup> In Fig. 2, we present such a comparison for 6- and 10-Å coverages. We have employed free-atom, plane-wave scattering factors<sup>11</sup> and a cluster size of 81 atoms of Ni in each of four layers of substrate. Each layer of Cu in the overlayer also contained 81 atoms. Furthermore, we have made a 50% reduction in both the magni-

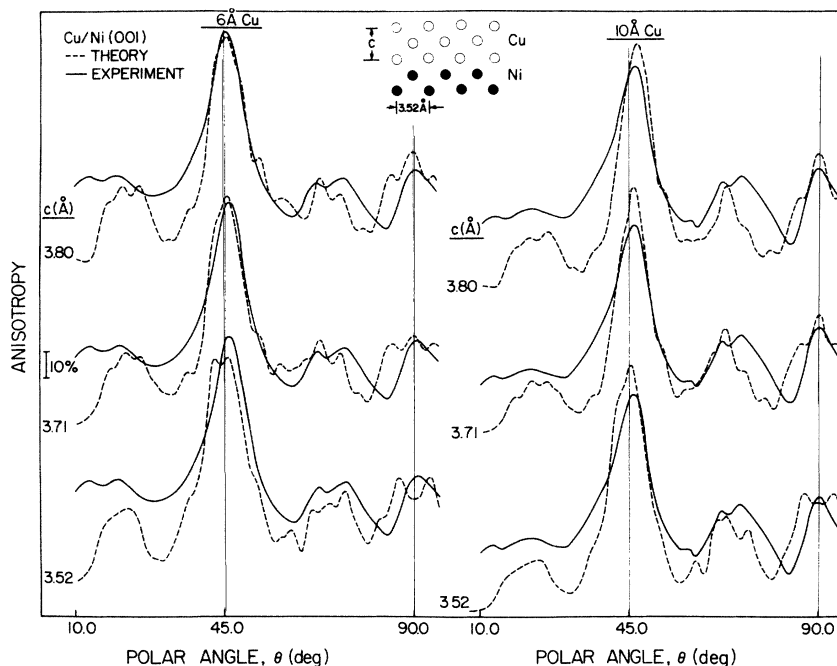


FIG. 2. Comparison of experiment and kinematical scattering theory for different choices of Cu-Cu interplanar spacing normal to the interface in pseudomorphic Cu/Ni(001).

tude of the scattering factors and the electron mean free path to compensate for neglect of multiple scattering and spherical wave effects.<sup>12-14</sup> The raw data (Fig. 1) show that  $\Delta\theta$  is  $1.5^\circ \pm 0.2^\circ$  and  $1.3^\circ \pm 0.2^\circ$  for 6- and 10-Å coverages, respectively. In the simplest picture,  $c$  can be estimated from the relationship  $c = b \tan(\theta + 0.5^\circ)$  where  $\theta$  is the observed polar angle for the peak along [101] and  $0.5^\circ$  is a correction for refraction at the solid-vacuum interface. This equation yields  $c$  values of  $3.71 \pm 0.03$  Å and  $3.68 \pm 0.03$  Å for  $\Delta\theta = 1.5^\circ$  and  $1.3^\circ$ , respectively. However, a detailed comparison of theory and experiment (Fig. 2) shows that optimal agreement in the position of the peak along [101] occurs at  $c = 3.80$  Å for 6-Å coverage and 3.71 Å for 10-Å. We therefore conclude that in the coverage regime where

pseudomorphic growth occurs (up to 14 Å), the average value of  $\Delta\theta$ ,  $1.3 \pm 0.2^\circ$ , leads to a perpendicular lattice constant of  $3.71 \pm 0.03$  Å. Since the lattice constant in the plane of the interface is the same as that of Ni, we arrive at a unit-cell volume of  $46.0 \pm 0.4$  Å<sup>3</sup> for pseudomorphic Cu on Ni(001) compared to a unit-cell volume of 47.0 Å<sup>3</sup> in bulk Cu.

Above a critical coverage of 13-15 Å, both of the aforementioned models, which are based on simple classical ideas about the forces and energies associated with strain buildup and dislocation generation, predict that it is no longer energetically favorable for the misfit to be accommodated entirely by elastic strain. Some of the strain could be relieved by the generation of dislocations. In Fig. 3 we plot

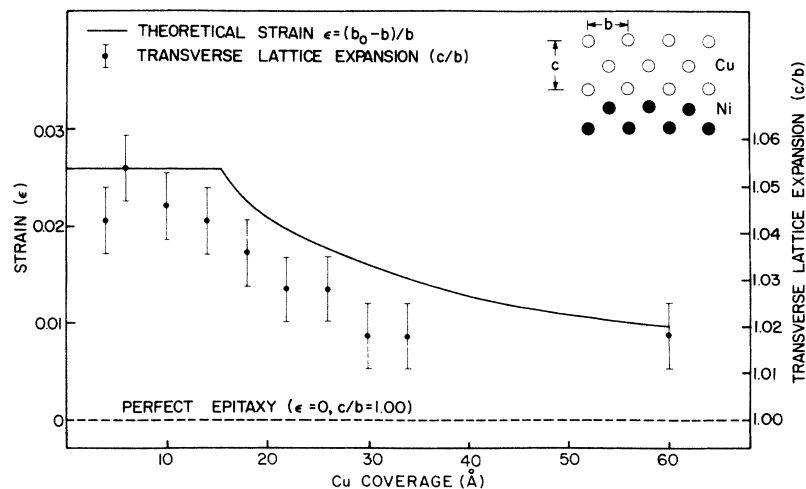


FIG. 3. Theoretical lattice strain (Ref. 2) and experimentally determined transverse lattice expansion vs Cu coverage.

the interfacial strain  $\epsilon$  versus coverage as predicted by the model of Matthews and Crawford.<sup>2</sup>  $\epsilon$  is defined as  $(b_0 - b)/b$ , and  $b_0$  and  $b$  are the lattice constants in bulk Cu and in pseudomorphic Cu on Ni(001) in the plane of the interface, respectively. Those authors then related strain to the overlayer thickness  $h$  for coverages above the maximum thickness for pseudomorphic growth  $h_c$  by equating the force on a dislocation which extends throughout the interface with the tension in the misfit dislocation line. The result is

$$\epsilon(h) = \left( \frac{G_i B (1 - \nu)}{20 G_0 h (1 + \nu) \cos \lambda} \right) \left[ 1 + \frac{10}{4\pi} \frac{(1 - \nu \cos^2 \alpha)}{(1 - \nu)} \ln \left( \frac{h}{B} \right) \right],$$

where  $G_i$  and  $G_0$  are shear moduli at the interface and in the overlayer,  $B$  is the magnitude of the Burgers vector for the dislocation line,  $\nu$  is Poisson's ratio for the overlayer material,  $\alpha$  is the angle between the misfit dislocation line and its Burgers vector, and  $\lambda$  is the angle between the slip direction and a vector in the plane of the interface which is perpendicular to the intersection line of the slip plane and the surface. In using this equation to generate the plot in Fig. 3, we have assumed that  $G_i \approx G_0$ , resulting in their mutual cancellation, and have used 0.32 for  $\nu$ , 2.55 Å for  $B$ , and 45° for  $\lambda$  and  $\alpha$ . The latter two numbers are taken from TEM results.<sup>1,2</sup> Moreover, by setting  $\epsilon$  equal to the natural misfit and solving the resulting transcendental equation for  $h$ , we arrived at a value for  $h_c$  of 14.8 Å. Accordingly, we show  $\epsilon(h)$  vs  $h$  for  $h > h_c$  in Fig. 3. Also shown in Fig. 3 is the experimentally determined ratio of  $c$  to  $b$ , which we will refer to hereafter as the transverse lattice expansion.

Once dislocations are generated and begin to propagate, the value of  $b$  is no longer unique along the interface. However, the value of  $c/b$  averaged over the area irradiated by the incident beam is a good measure of the extent of lattice relaxation. As shown in Fig. 3, the transverse lattice expansion is constant within experimental error up to 14 Å and averages 1.046 in this coverage region, but then drops monotonically to a final value of 1.018 by 30 Å. This value

then remains up to 100 Å (not shown). The experimentally derived value of  $h_c$  and the behavior of  $c/b$  vs  $h$  for  $h > h_c$  are in good agreement with the theoretical  $\epsilon(h)$  curve. The significance of this result is that we now have the necessary experimental and theoretical tools to accurately determine the structure of the strained overlayer and the maximum overlayer thickness for which coherence with the substrate can be maintained.

In contrast to the present work, TEM images of the Cu/Ni(001) interface showed the appearance of Moiré fringes at a coverage of  $\sim 8$  Å, indicating that dislocations had been generated and strain relaxation had begun.<sup>1</sup> One explanation of this discrepancy is that defects and impurities present on the TEM substrate caused the premature generation of misfit dislocations. The Ni surface was prepared for TEM by evaporation of 1500 Å onto a hot, cleaved NaCl surface and evaporation of Cu was carried out at  $\sim 10^{-8}$  Torr. It is then likely that the substrate had a high defect density and that impurities were incorporated into the interface as it was formed.

In this paper, we have shown that high-resolution Auger electron diffraction can be used to follow interfacial lattice strain and relaxation. Further, these results indicate that simple models such as those advanced by Matthews and Crawford<sup>2</sup> or Jesser and Kuhlmann-Wilsdorf<sup>3</sup> are useful in predicting the maximum overlayer thickness for which pseudomorphic growth can be sustained. Auger electron diffraction with high angular resolution, which allows accurate determination of the lattice spacing *perpendicular* to the interface, thus constitutes a powerful tool for interface structure determination. It is particularly valuable when used in conjunction with TEM, which can yield information on the lattice spacing *in the plane* of the interface.

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