Helium atom scattering from an epitaxial nano-structure surface: Ni/Cu(100)

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Abstract. The growth of Ni thin film on the Cu(100) surface at 280 K has been studied by means of thermal-energy helium atom scattering. A near layer-by-layer growth is observed below ∼ 7 ML followed by a multilayer structure at higher coverages. Spot profile analysis shows Ni islands transfer from irregular shape to square with increasing Ni coverage. The mean island separation is measured as a function of coverage. The 0.5 ML Ni/Cu(100) surface turns out to be an ideal prototype, nano-structured surface for future interference measurements.

The most fundamental description of atoms at surfaces is through the atom-surface interaction potential, which has been studied intensely over the past century but, even today, remains difficult to determine accurately [1, 2]. In heterogeneous systems, one particular difficulty lies in separating geometric (or structural) contributions from electronic effects such as dispersion forces or atom-surface hybridisation. For example, recent helium atom scattering (HAS) studies demonstrated that measurements on a metallic thin-film depend upon the size of islands within that film [2, 3, 4]. Without appropriate structural characterisation, extracting fine details of the atom-surface interaction can therefore be difficult. In the present paper we characterise the structure of a nanotextured thin-film system and show it to form an ideal ‘reference’ for future study. We demonstrate that submonolayer epitaxial growth of nickel on Cu(100) can, under appropriate growth conditions, form remarkably well-defined and two-dimensional islands that are sufficiently large to isolate structural contributions to the helium-surface interaction.

Our choice of Ni/Cu(100) was guided by the expectation that the small lattice mismatch (\(a_{Ni}=3.52\) Å, \(a_{Cu}=3.62\) Å) would favour layer-by-layer deposition and predominately two-dimensional structures. However, the difference in surface energies (\(\gamma_{Ni}=1.94\) J/m\(^2\), \(\gamma_{Cu}=1.52\) J/m\(^2\)) could predict a tendency for Ni to wet the Cu(100) poorly and drive three-dimensional growth. Competition between these two factors is therefore expected to be critically dependent upon both the deposition rate and the substrate temperature. Previous low-energy electron diffraction (LEED) [5, 6] and scanning tunnelling microscopy (STM) [7, 8] studies at room temperature revealed a pseudomorphic, tetragonally-distorted Ni film. A recent medium-energy ion scattering (MEIS) study also indicated that alloying occurs above a surface temperature of 300 K [9]. Precise lateral characterisation of the overlayer structure and its dependence on growth conditions has not been performed previously but is especially accessible to HAS.

Our experiments were conducted using a helium atom diffractometer that allowed a wide range of scattering geometries [10] and had a base pressure of \(2 \times 10^{-10}\) mbar. The Cu substrate (1 mm thick, 10 mm diameter) was cut and polished in the (100) orientation (Surface Preparation Laboratory, The Netherlands). We used a He beam energy of 16.4 meV with a
Figure 1. Variation of normalised He specular intensity as a function of Ni coverage for Ni deposited on the Cu(100) surface at 280 K and under ‘antiphase’ scattering conditions in order to amplify the interference between terraces separated by the single Ni atomic step height.

velocity distribution of \( \sim 2\% \). Repeated cycles of Ar\(^+\) sputtering (30 min, 300 K, 800 eV, 5.0 \( \mu \)A/cm\(^2\)) and annealing (30 min, 800 K) were used to clean the surface until only a narrow, intense specular helium peak was observed (FWHM \( \sim 0.4^\circ \)). A lattice-rod scan \([11]\) of the surface indicated that the mean terrace width was in excess of 10 nm and hence the influence of steps on growth and morphology can be ignored here. Ni deposition was conducted at a substrate temperature of 280 K in order to avoid alloying \([9]\). Deposition was performed using a water-cooled electron-bombardment evaporator which typically increased the chamber pressure by less than \(1 \times 10^{-10}\) mbar. The deposition flux was \( \sim 1 \times 10^{-3}\) ML/s.

Figure 1 characterises the growth of Ni/Cu(100) at 280 K. The figure plots the variation of He reflectivity from the surface during nickel deposition and was taken under ‘antiphase’ scattering conditions, where destructive interference is expected between waves scattered from the top of growing islands and waves scattered from patches of ‘clean’ substrate. In addition to interference effects, the data of figure 1 is sensitive to diffuse scattering from aperiodic or ‘defect’ sites on the surface, most notably adatoms and from variations in the net step density. Thus, increasing surface roughness, for example through the nucleation of new islands, decreases the surface reflectivity. The data exhibit strong oscillations that are characteristic of a predominantly two-dimensional growth mode, implying that adatom migration across terraces and down step edges is facile at 280 K. We use the positions of intensity maxima and minima to indicate the integral and half-integral number of monolayers’ (ML) coverage respectively \([12]\). At half-integer coverage, the loss in reflected intensity is attributable to both increased surface roughness and interference effects; at integer monolayer coverage, little interference is expected and so the reflectivity serves as a direct measure of surface roughness. Figure 1 therefore indicates that well-ordered, layer-by-layer growth occurs over the first 1-6 monolayers, with a gradual increase in surface roughness that is consistent with a slow increase of step density with time. For the purposes of the present study, we are most interested in the structural details at the half-monolayer point, where the the surface is predominantly composed of single-layer, Ni islands, which have a compact shape.

Ni deposition curves have also been monitored in a wide range of Cu(100) surface temperatures (data not shown), which will be discussed elsewhere. Briefly, we find growth to be three-dimensional at 120 K, best described by a Volmer-Weber growth mode. Raising the substrate temperature to 200 K facilities wetting of the Cu(100) surface and growth switches to a Stranski-Krastanov mode (two-dimensional then three-dimensional after 1 ML). Around 360 K a clear alloying phase transition is observed, consistent with a previous MEIS study \([9]\). In the following discussion, therefore, we concentrate on the epitaxial structure formed at 280 K, a temperature chosen to ensure well-ordered pseudomorphic growth as well as precluding the
formation of surface alloys or substrate-overlayer exchange.

The lateral structure of the surface, during growth, can be inferred from measurements of the shape of diffraction peaks. Figure 2 illustrates ‘spot profiles’ of the specular peak at specific nickel coverages for growth at 280 K. The data was taken by raster-scanning the detector across the specular reflection in both in-plane and out-of-plane axes in order to plot the scattered intensity as a function of parallel momentum transfer. The plots all exhibit substantial broadening of the specular peak with respect to the sharp peak observed from the clean Cu(100) surface, indicating the presence of surface structures over length scales many times the dimension of the primitive Cu(100) unit cell. With increasing Ni coverage (0.5 ML, 4.5 ML and 8.3 ML from figure 2a to 2c respectively), the contours becomes more regular. Finally, a well-developed fourfold symmetry appears at the coverage of 8.3 ML, which indicates a four-fold rectangular (or square) distribution of Ni islands aligned along the close packed <110> directions.

More detailed, one-dimensional scans are plotted in figure 3a, which were performed by scanning the detector along the [100] direction (in-plane) whilst keeping the incident conditions fixed. Scans were taken at different Ni coverages, including those shown in figure 2. The broad contours observed in figure 2 are now revealed as comprising of a shoulder or distinct satellite peak. The separation of the two satellites is inversely proportional to the mean Ni island separation \( \langle L \rangle \) on the Cu(100) surface, while their shapes are related to the statistical distribution of Ni steps [13] and precise details of the islands’ form. Figure 3b shows the derived variation of \( \langle L \rangle \) with increasing Ni coverage, which implies that the repeat length between islands increases with coverage and hence islands become larger, on average, as the film thickens.

Combining the information from figures 1-3, we are now in a position to describe completely the thin-film growth of Ni/Cu(100) at 280 K. Below 7 ML, Ni growth proceeds by a layer-by-layer mechanism - i.e. two-dimensional Ni islands nucleate, grow and coalesce to form a new overlayer after the proceeding one is completed. We observe that layer-by-layer growth is not perfect, even at low coverages and despite the relatively slow deposition rate used here. The surface roughness increases continuously with coverage (figure 1). In an earlier STM work [8], a three-layer growth mode has been proposed starting from 3.5 ML. Our result, partially in agreement with this STM picture, suggests an even lower threshold for the onset of multilayer growth. During growth, the lateral topology of Ni islands continues to develop from irregular shapes to squares distributed along <110> directions on the Cu(100) surface. The mean island separation steadily increases from \( \sim 80 \) Å at 0.5 ML to \( \sim 110 \) Å at 6 ML. Since surface roughness increases simultaneously with island size, we surmise that, as the epitaxial strain is relieved, islands become larger but are
defined by more kinked and less regular edges. Above 7 ML, the oscillations of the deposition curve are lost. Previous STM work [7, 8] showed a pyramid-like island structure formed with more than four open Ni layers at this coverage. Our angular profiles in figure 3a agree with this model, since the the specular-satellite intensity ratio significantly decreases at 8.3 ML, compared to those at lower coverages.

Finally, we return to our original goal and identify the 0.5 ML Ni/Cu(100) system as a well-ordered, heterogeneous and nano-structured surface where it is possible to separate geometric contributions from the atom-surface interaction. We have demonstrated the growth to be predominantly two-dimensional and used diffraction profiles to indicate the formation of two-dimensional, rectangular Ni islands with a mean separation of 80 Å and hence mean dimension of 56 Å. This relatively large island size is particularly important as it allows us to exclude the modulation of the atom-surface potential via the small-island effects observed previously [3, 4]. With reference to the literature, we can also exclude alloying or overlayer-substrate exchange [9] in this temperature regime. Combined with the LEED results indicated a compressed Ni atomic step height [5, 6], the morphology on the 0.5 ML Ni/Cu(100) surface is well understood. In principle, an isolated study of He-Ni/Cu metal surface interaction, independent of structural effects, is now possible.

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