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He scattering study of the nucleation and growth of Cu(100) from its vapor

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The nucleation and growth of layers of Cu deposited in ultrahigh vacuum on Cu(100) has been studied by He scattering. The intensity of the specular beam shows oscillations as a function of coverage, with maxima occurring at the completion of successive monolayers. The amplitude of these oscillations depends on both temperature and angle of incidence. An analysis of the dependence with the incident angle at fixed coverage, combined with the Bragg conditions for interference, provides an accurate method for determining the height of the two-dimensional islands formed in the process of crystal growth. We find a value of 1.80 ± 0.06 Å in excellent agreement with x-ray determination of interlayer spacing of (100) planes. Measurements of the angular profile of the specular peak give an estimation of the average island size at different temperatures.

The understanding of the processes involved in epitaxial growth of a crystal from its vapor has been a topic of permanent interest for 40 years.[1] Many surface-sensitive techniques have been applied in the past years to characterize the series of atomic steps involved in growth phenomena, e.g., condensation, migration, and nucleation.[2] Whereas imaging techniques able to visualize individual adatoms, such as field ion microscopy (FIM), provide detailed information of small areas of the sample, diffraction techniques give the average over larger areas. As a typical example of the last type of techniques, the process of crystal growth has been recently studied by means of low-energy electron diffraction (LEED),[3,4] which, however, presents some problems due to the finite penetration of electrons in the solid. On the other hand, the scattering of atomic beams of He by surfaces is known to be sensitive only to the outer charge corrugation, thus, being especially suited to give average terrace size or step density of a growing crystal. Some of the advantages of using atomic beams for this purpose are their high sensitivity to detect disordered structures (adatoms, steps) at very low concentration,[5] and their capability to provide information on the state of aggregation of the adsorbates.[6]

In this Rapid Communication we report on the first results of a study of nucleation and growth of copper evaporated on the (100) surface of the same metal using atomic beam scattering.

The experiments were carried out in a UHV chamber equipped with standard surface-science techniques: Auger spectroscopy, low-energy electron diffraction, and Ar-ion sputtering. A supersonic He nozzle beam of 63 meV ($k = 11$ Å^{-1}) entered the chamber and impinged upon the crystal surface at a selected angle of incidence ϕ. The scattered He intensity was measured by a movable quadrupole mass spectrometer. A more detailed description of the experimental setup has been already published.[5,7] A copper wrapped tungsten filament placed near the crystal surface was used as a source of Cu vapor. The rate of Cu deposition was calibrated by measuring the peak heights of selected substrate and adsorbate Auger transitions in a cobalt foil mounted next to the crystal. The evaporation rate was typically 10^{12} atoms/cm^2 s. The Cu coverage θ was known to within 10% of a monolayer.

Prior to Cu deposition, the crystal was cleaned until no impurities were detected by Auger spectroscopy. The reflectivity of the clean and annealed crystal for He (between 20% and 60% at room temperature) was typically used as an indicator of the state of the surface, more sensitive than Auger spectroscopy. In the time scale of the experiments the maximum decrease of the intensity due to contamination was $\sim 1.5\%$.

Upon Cu deposition the LEED pattern was still (1×1) with sharp spots and only at the lowest temperature attainable, $T \sim 210$ K, some spot broadening (hard to assess quantitatively) was detected.

Information on the growth of the copper crystal was first obtained by measuring the reflected He intensity as a function of deposition time at a given angle of incidence. In Fig. 1 we show the relative intensity of the specular beam, $I(\theta)/I_0$, where I_0 represents the initial intensity at an angle of incidence $\phi = 67°$, as a function of the amount of Cu evaporated (in monolayers) for various substrate temperatures. A number of observations are relevant. (a) The data show an oscillatory behavior with a period of one monolayer similar to that observed in other systems by electron diffraction.[3,4,8,9] Maxima are observed at the completion of each deposited monolayer, while minima correspond to half-layer coverage. The oscillations in the reflected intensity are simply the result of the interference between waves reflected at two different levels of the surface. (b) The intensity oscillates in a limited range of temperature for the Cu flux used in these experiments, i.e., $235 \lesssim T \lesssim 365$ K. Below 235 K, $I(\theta)/I_0$ decreases continuously without oscillations. Above 365 K, the initial value stays constant independent of the amount of Cu evaporated onto the sample. These observations seem to indicate that nucleation of Cu islands on the (100) terraces of the substrate is taking place, the number and size of islands being a strong function of the (temperature-dependent) diffusion coefficient. At $T \gtrsim 365$ K the diffusion of Cu adatoms to step edges is fast enough to prevent nucleation at the center of the terraces, producing only a lateral displacement of the existing steps. Therefore, the step density within the transfer width of the experimental apparatus (500 Å) remains constant and so does the reflected intensity. On the other hand, below 235 K the adatom diffusion is slow enough to produce a continuous decrease of the order of the surface upon vapor deposition.

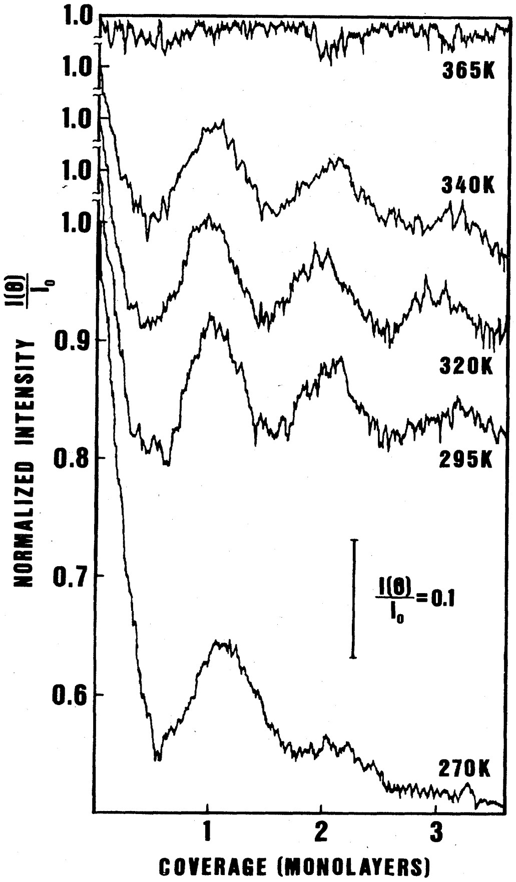

FIG. 1. Normalized intensity of the He-specular peak vs amount of deposited copper in monolayers for different substrate temperatures. Deposition rate was 13 min per monolayer. The angle of incidence was 76°. The curves are displaced vertically for clarity.

(c) *For a given temperature, the amplitude of the oscillations decreases with increasing coverage.* In this temperature range, a layer-by-layer growth is expected on a general thermodynamic basis[10] for a Cu crystal growing in equilibrium with its vapor. However, the referred observation indicates that the layer-by-layer growth is not perfect. That is, the successive ad-layers are not completed when the next layer starts to form. This disorder accumulates until after some monolayers, depending on crystal temperature, a steady-state rough structure is reached. These structures are not equilibrium structures; however, they are kinetically stable at these temperatures, as shown by the invariance of the reflected He intensity upon termination of Cu evaporation. The general features of this model are nicely reproduced by a Monte Carlo simulation.[11] (d) *The normalized intensity at half monolayer coverage, $I_{\frac{1}{2}}/I_0$, is smaller the lower the temperature* (see Fig. 1), suggesting that the number of islands is large at low temperatures (and their size small), whereas at high temperatures fewer and larger islands are formed.[4]

Quantitative information of the average size or distance of the islands can be obtained from measurements of the spot profile of the specular peak. The spot profile after deposition of half a monolayer of Cu has been measured for "in plane" scattering as a function of temperature. For a fixed incidence angle of 77°, the full width at half maximum

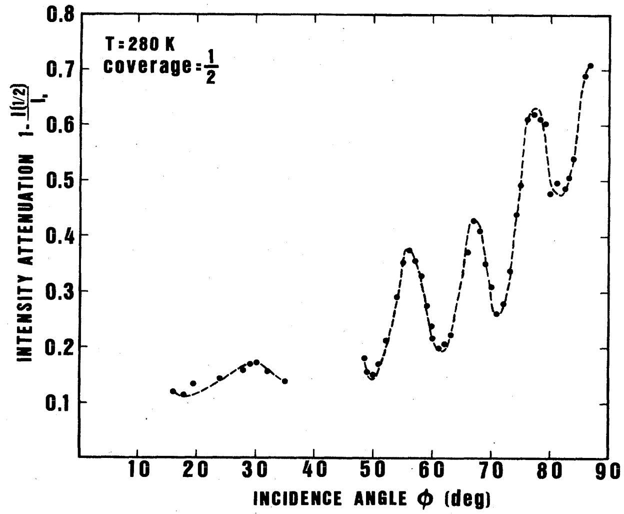

FIG. 2. Intensity attenuation at half monolayer as a function of incidence angle ϕ. The deposition rate was 5 min per monolayer and the substrate temperature 280 K. Maxima and minima correspond to destructive and constructive interference, respectively. Ranges of scattering angle (0°–15° and 35°–48°) were not accessible for measurement.

(FWHM) $\Delta\phi$ has an initial value of 0.62° at 320 K and zero coverage. After deposition of half monolayer at 210 K, the sample was kept at the chosen T and the spot profile measured. As expected, $\Delta\phi$ continuously decreases with the chosen increasing T. Some significant values are 2.98° at 240 K, 1.83° at 280 K, 1.30° at 295 K, 0.99° at 320 K, and 0.69° at 355 K. At 240 K two shoulders can also be observed separated by 2.37° that reflect random distribution of islands sizes and/or distances. A characteristic distance of 60 Å can be deduced from the angular separation of the shoulders at 240°. The characteristic correlation distance increases then with temperature until at 355 K it reaches a value of approximately 470 Å.

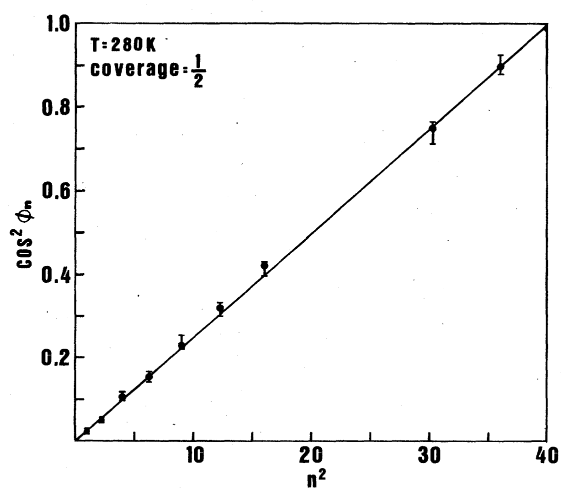

FIG. 3. Squared cosine of incidence angles satisfying Bragg condition for destructive ($n =$ half integer) and constructive ($n =$ integer) interference. These angles correspond to the maxima and minima shown in Fig. 2. In all cases the coverage was $\frac{1}{2}$.

When the angle of incidence ϕ varies from 0° to 90° the conditions of the interference between waves scattered at two levels of a surface change accordingly from constructive to destructive a number of times. It is straightforward to show that for islands of height H the interference condition is

$$H|k^2\cos^2\phi_i+(2m/\hbar^2)D|^{1/2}=\pi n \quad ,$$

where k is the He wave vector, m the mass of the helium atom, D the attractive potential well depth,[12] and n a half-integer number for destructive interference and an integer for constructive interference.

A measure of the normalized intensity as a function of the angle of incidence ϕ for fixed values of T and θ would then yield, in a simple and accurate way, the island height. The results of such a measurement at $\theta=\frac{1}{2}$ monolayer and $T=280$ K are shown in Fig. 2. The figure actually displays the *decrease* or attenuation of the normalized intensity, i.e., $1-(I\frac{1}{2}/I_0)$, rather than the intensity itself. The intensity attenuation shows again an oscillatory behavior. The number and position (angle of incidence, ϕ_n) of the maxima and minima depend critically on the value of H. By plotting $\cos^2\phi_n$ vs n^2 (n, integer for minima, half-integer for maxima), a straight line is obtained with a slope of π^2/k^2H^2 and ordinate $(2m/h^2k^2)D$. From the data depicted in Fig. 3 values of $H=1.80\pm0.06$ Å and $D=0$ are obtained in excellent agreement with both the bulk value of $a=1.8037$ Å given by x-ray diffraction for the (100) interlayer spacing of copper[13] and the depth well calculated by García, Barker, and Rieder[14] for the He-metal surface interaction potential.

In conclusion, we have shown in a simple case that He scattering is a powerful technique in studying crystal growth. The detailed information obtained on microscopic details of the processes involved contains a promise for the understanding of the epitaxial growth of more complex abosrbate/substrate metal systems.

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