Temperature Dependence of Vacancy Formation during Homoepitaxial Growth on Ag(001)

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Introduction

There is considerable interest [1] in understanding the microscopic structure of surfaces, interfaces, and thin films from both a fundamental and technological perspective. Recently, kinetic Monte Carlo (KMC) simulations have made excellent progress in predicting many of the surface morphological features observed in homoepitaxial growth experiments [2-4]. However, for these simulations to accurately relate to real surfaces, one must include all of the microscopic kinetic mechanisms that are relevant.

Our recent x-ray scattering measurements have shown that a large concentration of vacancies occurs during low-temperature homoepitaxial growth on noble metal surfaces [5]. These vacancies, which are incorporated within the growing film and not observable by scanning probe microscopy methods, obviously must be intimately coupled to the surface kinetics. Reentrant *smooth* homoepitaxial growth at low temperature has been observed for the (001) surfaces of Ag [4] and Cu [6]; this behavior was explained [4] in terms of temperature-independent step-edge crossing mechanisms of the adatoms.

Here we report on x-ray reflectivity (XRR) measurements of the temperature dependence of vacancy formation for homoepitaxial films grown on Ag(001). An important finding is that the vacancies form at temperatures where reentrant smooth growth has been observed, suggesting that vacancy mechanisms must be fully considered for a thorough understanding of low-temperature epitaxial crystal growth.

Methods and Materials

These x-ray scattering experiments, which used an incident energy of 16.2 keV, were performed in an ultrahigh vacuum (UHV) by using the newly constructed surface scattering spectrometer located at APS Midwest Universities Collaborative Access Team (MU-CAT) sector 6. A Ag(001) crystal (10 mm in diameter by 3 mm thick) was mechanically polished to a miscut of ~0.1° and subsequently prepared in UHV by repeated sputtering and annealing cycles. Sputtering was performed by using an Ar pressure of 10^{-5} Torr, an acceleration voltage of 500 V, and a current of ~10 μA. The sample was heated by electron bombardment, and the annealing was performed near 720°C. Low temperatures were achieved through the use of a closed-cycle refrigerator that permitted cooling to 55K. The surface temperature of the sample was directly determined by measuring the thermal expansion. Ag was evaporated from a resistively heated crucible, and the deposition rate of 2 monolayers (ML)/min was determined by measuring the x-ray intensity oscillations at the anti-Bragg position that occur during layer-by-layer growth at 350°C.

Results

The XRR (Fig. 1) was measured in the vicinity of the (002) Bragg reflection for a nominal coverage of 20 ML deposited at the given substrate temperature. At the lowest temperatures of 108K and 65K, pronounced thin-film interference fringes and an asymmetry toward a higher angle appear. These features arise [5] from the compressive strain caused by the vacancies.

Discussion

Although these data will be analyzed to quantitatively determine the temperature dependence of the vacancy concentration, the results in Fig. 1 already demonstrate that the vacancies begin to form at rather high temperatures; at 204K, a clear asymmetry is present along with a slight shoulder. Recent scanning tunneling microscopy (STM) [4] studies of low-temperature Ag(001) homoepitaxy have shown that reentrant smooth growth begins at this temperature. Although simulations [4] suggest that the reentrant growth is consistent with "downward funneling," the present x-ray results indicate that vacancy mechanisms, which reflect dramatic changes in the microscopic surface kinetics, must also be included in a realistic description of epitaxial growth. Future experiments will be necessary to ascertain the specific configuration of the vacancies and the mechanisms by which they enter the growing film.

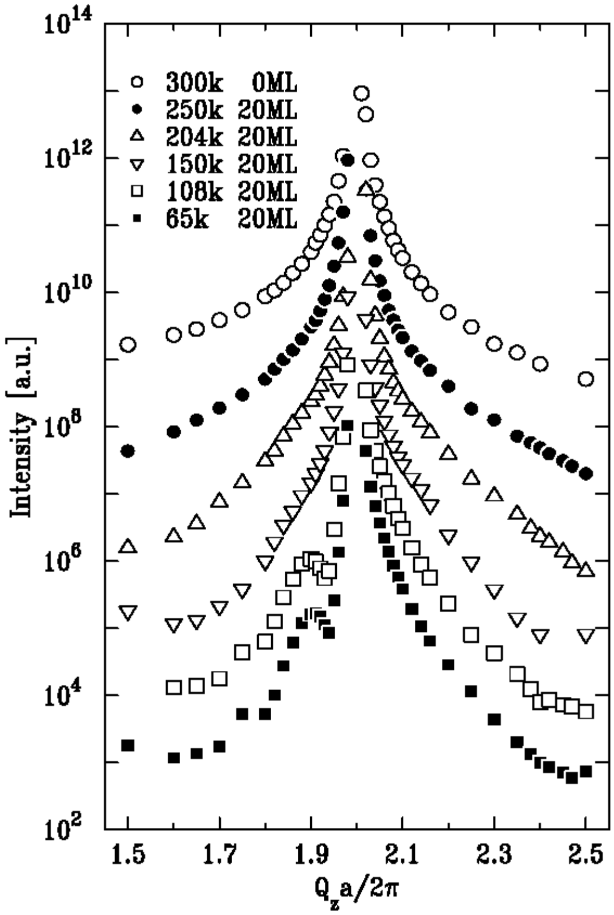

FIG. 1. XRR measured near the (002) Bragg reflection of Ag(001) after 20 ML was deposited at the given substrate temperature.

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