Grazing-Incidence Diffuse Scattering from Vacancies in Homoepitaxially Grown Ag(001)

P.F. Miceli,[1] K. Li,[1] E. Conrad[2]
[1]Department of Physics and Astronomy, University of Missouri-Columbia, MO, U.S.A.
[2]School of Physics, Georgia Institute of Technology, Atlanta, GA, U.S.A.

Introduction

The extensive attention [1] given to understanding the evolving surface morphology of an epitaxially growing crystal has been focused on atomic-scale mechanisms confined to the surface. In large part, because of the fact that most experimental surface probes are sensitive only to the surface, little attention has been given to how subsurface defects originate at the surface or how such defects impact the subsequent surface morphology. Our recent x-ray scattering studies have shown that a substantial concentration of vacancies is incorporated during the low-temperature homoepitaxial growth of noble metals [2] and that their incorporation occurs in a temperature range where changes are observed in the surface morphology: reentrant smooth growth is observed for Ag(001) [3] and Cu(001) [4], whereas a steep pyramidal surface morphology is found for Ag(111) [5].

The present work represents an initial experiment to learn more about the nature of vacancies incorporated in the Ag/Ag(001) system. One recent study of a simulation [6] suggests that vacancies might originate from restricted "downward funneling" (DF) of adatoms near crystalline step edges, where the conventional DF mechanism is believed to be responsible for a surface smoothing that partially counteracts the step-edge diffusion barrier. These simulations predict large voids; thus, one important question to address experimentally is the physical size of the vacancy clusters.

Methods and Materials

X-ray scattering measurements were performed in a grazing incidence geometry, and scans were taken radially along the in-plane H direction through (H,0,L), where L = 0.03 reciprocal lattice unit (rlu) given perpendicular to the surface was held fixed. Using an incident x-ray energy of 16.2 keV, these scattering experiments were performed *in situ* in ultrahigh-vacuum (UHV). The newly constructed surface-scattering spectrometer located at MU-CAT (sector 6) at the APS was used. A Ag(001) crystal (10 mm in diameter by 3 mm in thickness) 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, acceleration voltage of 500 V, and current of ~10 μA. The sample was heated by electron bombardment, and the annealing was performed near 720°C. This procedure resulted in a surface correlation length (facet size) of ~500 nm. Low temperatures were achieved through the use of a closed-cycle refrigerator. The surface temperature of the sample was directly determined by measuring the thermal expansion. Ag was evaporated from a resistively heated crucible that had its evaporation rate calibrated by measuring the x-ray intensity oscillations that occur at the anti-Bragg position during layer-by-layer growth at 350°C. The experiments reported here had a deposition rate of 36 monolayers (ML) per minute and a total coverage of 690 ML that was deposited at a substrate temperature of 100K.

Results and Discussion

Measurements made through the (800) before and after deposition are shown in Fig. 1. An increase in diffuse scattering is observed after the film is deposited, and there is a noticeable asymmetry toward lower H. The direction of the asymmetry is consistent with vacancies [7], where we observe an average lattice contraction in the Bragg scattering but an expansion locally around the defect in the diffuse scattering.

The diffuse intensity averaged symmetrically about the high and low q side of the H = 8 position from the data in Fig. 1 is shown in Fig. 2, where q is the distance

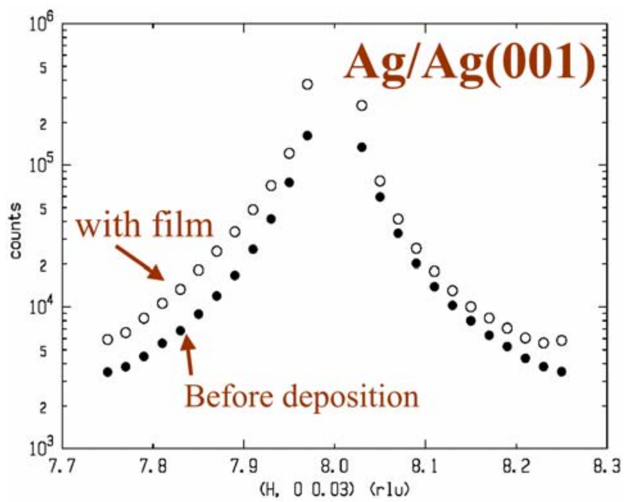

FIG. 1. Measurements before and after deposition.

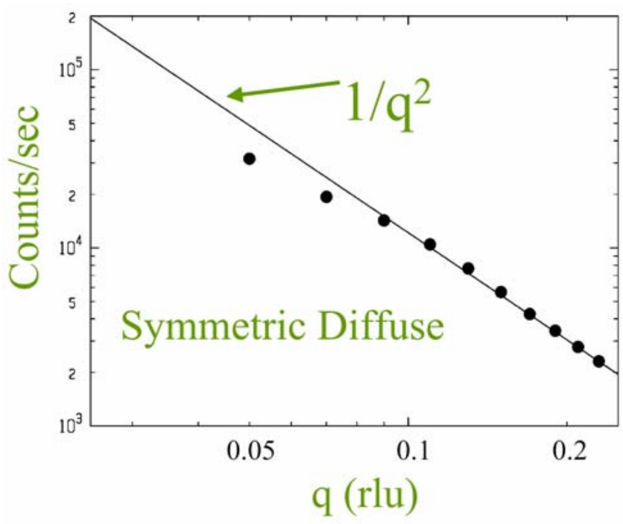

FIG. 2. Diffuse intensity averaged symmetrically about the high and low q side of the H = 8 position from the data in Fig. 1.

measured from H = 8. It can be seen that these data follow the $1/q^2$ Huang scattering predicted for point defects [7]. Although we do not presently understand the slight deviation at low q (and we do not believe this to be caused by the resolution), the fact that the scattering follows the point defect result to a large q suggests that the vacancy clusters are quite small.

We note that these measurements are for considerably thicker films than we studied previously in our Bragg scattering measurements of 5 to 25-ML films. We expect that there could be considerable differences as a function of the film thickness. However, these experiments demonstrate the ability to see the diffuse scattering due to vacancies and suggest that significantly thinner films can be studied in the future.

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