X-ray Diffuse Scattering Study of Vacancy Clusters in Ag Films Grown Homoepitaxially on Ag(001)

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Introduction

There is broad fundamental and technological interest in understanding the atomic-scale mechanisms that control epitaxial crystal growth. Most experimental studies of these systems have been performed using scanning surface probes which interrogate only the layer of atoms at the surface. Therefore, the combined surface and sub-surface sensitivity of x-ray scattering methods can provide both crucial and unique insight into the behavior of epitaxial crystal growth.

Recent x-ray reflectivity studies have shown, rather surprisingly, that vacancies are incorporated into films grown homoepitaxially on several noble metal surfaces at low temperatures [1]. Although the mechanism of vacancy incorporation is not presently understood, it is clear that the vacancies must incorporate at the surface using mechanisms that are also responsible for the evolving surface morphology. Therefore, understanding the vacancy incorporation mechanism is fundamental to understanding epitaxial crystal growth.

In the present study, we seek to determine the size of the vacancy clusters for a 100 monolayer-thick (ML) homoepitaxially grown film on Ag(001). Knowledge of this length scale is a first step in understanding the vacancy incorporation mechanism and it will provide an essential benchmark for theoretical simulations. The experiments measured the x-ray diffuse scattering near several Bragg reflections and the results were compared to simulations.

Methods and Materials

These measurements were performed at the Midwest Universities Collaborative Access Team (μCAT) beamline at the Advanced Photon Source using the UHV surface chamber and spectrometer at 6IDC. The Ag(001) surface was cleaned with multiple cycles of sputtering and annealing, yielding a surface free of impurities, as confirmed by Auger electron spectroscopy (AES).

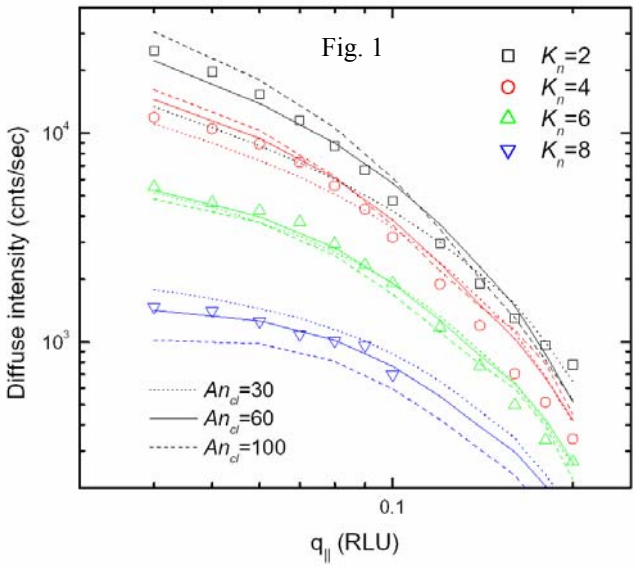
The scattering measurements were performed in grazing incidence geometry near four Bragg reflections (0, K+q, L) where K and L are Miller indices with L=0.1, K=2,4,6,8 and q was scanned in the measurements. 100 monolayers of Ag were deposited at a rate of 3.2 monolayers per minute from a thermal evaporation source onto the substrate, which was held at a temperature of 150K. Scans were performed before and after deposition so that the diffuse scattering intrinsic to the virgin crystal could be subtracted. Thus, the diffuse scattering discussed here will refer to the subtracted data.

Results and Discussion

Fig. 1 displays the symmetric diffuse intensity which is an average of the intensity on both sides of the Bragg reflection: [I(+q)+I(-q)]/2. The diffuse data also exhibit (not shown) a large asymmetry toward -q, consistent with diffuse scattering arising from vacancies.

Computer simulations of the diffuse intensity were performed in order to estimate a cluster size. To obtain analytic expressions from models [2,3,4] of defect scattering, simplifying approximations must be made such as assuming that the defects generate a small displacement field. In that case, $1/q^2$ behavior (Huang scattering) is expected at low q which evolves to $1/q^4$ (Stokes-Wilson) at higher q. We found that such an analysis could not reproduce the measured intensities self consistently at all orders of Bragg reflection. Instead, the rapid decay of the diffuse intensity with K in Fig. 1 suggests, after correcting for the atomic scattering factor and thermal Debye-Waller factor, the existence of large displacement fields arising from large vacancy clusters. Thus, our simulations [4] take full account of the large displacement fields using isotropic elastic theory.

As can be seen from the curves in Fig.1, good agreement for all orders of Bragg reflection is achieved



for a cluster size containing approximately 60 atoms, which corresponds to spherical diameter of approximately 12 Å. It was also determined that these clusters occur at a concentration of 0.05% (per atom), which yields a total number of missing atoms per volume that is consistent with previous estimates obtained from x-ray reflectivity. An important conclusion from this study is that an accurate interpretation of defect scattering requires measurements at multiple orders of Bragg reflections, which imposes mutual consistency of the intensities and constrains the model parameters.

The observed large vacancy cluster size is remarkable and suggests that once an atom resides at an overhanging site it provides a stable foundation to build a larger defect structure. Indeed, vacancies were found [1] to be stable upon annealing up to 300K. These results will provide a useful starting point to understand how the atomic-scale kinetics permit atoms to form such structures during epitaxial crystal growth.

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