Determination of Island Layer Relaxations in Pb/Si(111)7×7 by Using Surface X-ray Diffraction

P. Czoschke,[1] M. Holt,[1] H. Hong,[2] T.-C. Chiang[1]
[1]University of Illinois at Urbana-Champaign, Urbana, IL, U.S.A.
[2]Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.

Introduction

The Pb/Si interface is considered to be a prototypical metal-semiconductor interface and has therefore received a fair amount of attention. Recently, it has been shown that epitaxial growth at low temperatures (120-250K) of Pb on a Si(111)7×7 substrate produces uniform steep, flat-topped islands of preferred thicknesses [1]. This behavior has been attributed to quantum size effects (QSE). If this is the case, the quantized electronic states should also result in structural effects. Indeed, bilayer oscillations in island height have been observed by using scanning tunneling microscopy (STM) [2]. However, STM is primarily a surface-sensitive technique that does not give direct information about the layer thicknesses within the islands. Therefore, it cannot be inferred that this bilayer relaxation phenomenon penetrates through the whole film. With its longer penetration depth, surface x-ray diffraction (SXRD) has the ability to probe all the layers of the island with more or less the same scattering amplitude, so it should thus allow us to determine the individual layer thicknesses within the islands.

Methods and Materials

The surface diffraction station at beamline 34-ID of the University-National Laboratory-Industry Collaborative Access Team (UNI-CAT) was used for all of the experiments. The experimental setup consists of an ultrahigh vacuum (UHV) chamber mounted on a six-circle diffractometer. The chamber is equipped with reflection high-energy electron diffraction (RHEED), an Omicron electron beam evaporator, a crystal thickness monitor, and a 180° beryllium window to allow for incident and exiting beam travel. The sample can be cryogenically cooled with liquid nitrogen to a base temperature of 150K and/or can be annealed by direct resistive heating.

The Si substrates were cut from commercial wafers and degreased in methanol and acetone before being mounted in the chamber. The sample was cleaned in UHV by flashing at 1100°C for approximately 7 s. Surface cleanliness was checked by examining the 7×7 reconstruction by using RHEED. The Pb film was grown by evaporation at a rate of 0.8 Å/min (0.23 monolayer [ML]/min) at 150K. Throughout this report, coverages will be reported in terms of the atomic density of a (111) plane in bulk Si, 7.83×10^{14} cm^{-2}. When this notation is used, one full layer of bulk-like Pb would correspond to 1.2-ML coverage, since Pb has a smaller lattice constant than does Si.

Layer relaxations were determined by scanning the reflectivity of the surface after deposition and then fitting the profile to the model described below. Reflectivity measurements were taken by measuring the scattered intensity from the surface with a NaI scintillation counter positioned such that the exiting x-rays formed approximately the same angle with respect to the surface of the sample as that formed by the incoming x-rays. The sample was then "rocked" to collect all of the diffuse intensity due to the mosaicity of the crystalline surface. These "rocking curves" were then background-subtracted and numerically integrated to get the integrated intensity of that point on the reflectivity rod. The error for each point was estimated by a quadrature combination of the counting error and a 5% systematic error. An example of such a reflectivity profile is shown in Fig. 1(a). By using this method, accurate values for the integrated intensity can be measured to relatively large angles while

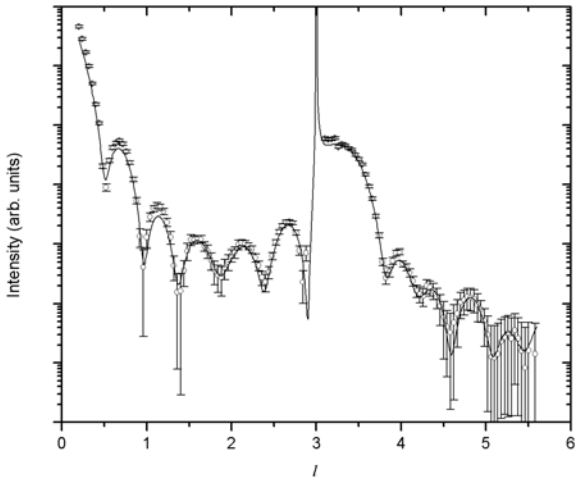

FIG. 1. Reflectivity of a sample with 4.5 ML of Pb. The five bumps in between the origin and the first bulk-like peak indicate the presence of predominantly seven-layer islands. The solid line is a best fit to the data from using the model described in the text.

background effects, such as thermal diffuse scattering, are taken into account.

The model used to fit the data was similar to that used by Edwards et al. [3], as follows. A structure factor is constructed with the following three contributions:

$$F(\ell) = F_{substrate}(\ell) + F_{DAS}(\ell) + F_{Pb}(\ell)\,. \qquad (1)$$

The variable l = the momentum transfer parallel to the surface normal in units of $2\pi d^{-1}$, where $d = \sqrt{3}a_{Si}$ = the unit cell length for Si in the (111) direction. The substrate contribution is that from a semiinfinite truncated crystal:

$$F_{substrate}(\ell) = f_{Si}(\ell)\cdot e^{-B\left(\frac{\sin(\theta)}{\lambda}\right)^2}\,\frac{1+e^{-i\frac{\ell}{12}}}{1-e^{-i\frac{\ell}{3}}}, \qquad (2)$$

where $f_{Si}(l)$ = the atomic form factor for Si and B = the Debye-Waller parameter for Si. $F_{DAS}(l)$ = the form factor for the 7×7 reconstruction following the Dimer-Adatom-Stacking fault model with the adatoms removed:

$$F_{DAS}(\ell) = f_{Si}(\ell)\cdot e^{-B\left(\frac{xin(\theta)}{\lambda}\right)^2}\cdot\left[\tfrac{48}{49}e^{i\ell z_1} + \tfrac{42}{49}e^{i\ell z_2}\right]. \qquad (3)$$

The layer spacings z_1 and z_2, in units of d, are the vertical heights of the two layers of Si that participate in the reconstruction. However, the relative occupancies of the two layers were fixed to the values for the DAS model. Finally, the Pb overlayer structure factor is:

$$F_{Pb}(\ell) = f_{Pb}(\ell)\cdot e^{2\pi i\ell z_2}\sum_n A_n e^{i\ell\Delta z_n - \frac{1}{2}q^2\xi_n^2}\,, \qquad (4)$$

where the free parameters are as follows: A_n = the relative occupancies of each layer, Δz_n = the height of each layer from the substrate, and ξ_n = a Debye-Waller-like factor, similar to that used by Robinson and Vlieg to account for the finite "thickness" of each layer [4], which corresponds to the standard deviation of the height of the atoms in that layer from their average height. The scattered x-ray intensity is proportional to the structure factor quantity squared modified by the appropriate geometrical corrections [5] and an overall roughness factor [6].

Results

An example of a reflectivity scan and its best fit is shown in Fig. 1. The sample had 4.5 ML of Pb deposited on it. The sharp peak at l = 3 is the (111) bulk Si Bragg peak, and the large bump after it at l = 3.6 is the corresponding peak for the Pb overlayer. The interference fringes between l = 0 and l = 3.6 are the well-known interference effects due to a thin film. In this case, the number of fringes indicates that the Pb overlayer is dominated by islands of seven layers. The solid line in Fig. 1 is the best fit of the model described above to the data. The reduced χ^2 value for the fit is 2.8. The fitted results for the individual layer relaxations are shown in Fig. 2. With respect to the expected bulk values, the first two layers appear to be significantly expanded, and the top layer appears to be contracted. However, the interior layers appear to have the spacing of bulk Pb, more or less.

All the thickness parameters in Eq. (4) were found to be on the order of that expected for bulk Pb, except for the first two layers, which showed unusually high values. The Pb/Si(111)7×7 system forms two wetting layers before forming islands, and it has been shown that these wetting layers appear to be highly disordered [7]. The high ξ values obtained for the bottom two layers therefore confirm this assertion.

Discussion

The results presented here, although preliminary, do not support the notion that all of the layers of the island participate in a bilayer thickness oscillation. It appears that only a small number of the interfacial layers next to the substrate and the outermost layer of the islands have thicknesses that deviate significantly from bulk Pb(111). However, since the bilayer relaxations were previously observed by comparing islands of different heights, it cannot be ruled out that islands of a height other than the seven-layer islands of the sample in this study would show different layer relaxations.

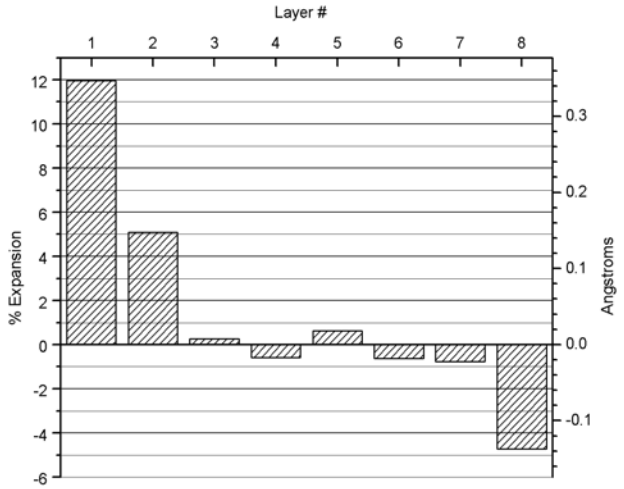

FIG. 2. The individual layer relaxations as found from the fit of the reflectivity curve in Fig. 1. Only the outermost layer and the wetting layers of the island appear to have significant structural relaxations.

Acknowledgments

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