Thin Film Growth: the Effects of Electronics and Kinetics

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Preliminary Examination
In partial fulfillment of the requirements of the

Preliminary Examination
Outline

Motivation

Experimental Apparatus

Surface X-ray Diffraction (SXRD)

Quantum Size Effects (QSE) and Quantum Confinement

I. Layer Relaxations in Pb/Si(111)

II. Temperature-Dependent Growth Studies

Summary
Preferred thicknesses
↑
Quantum confinement of electronic states

Layer-by-layer

(combination)

(Stranski-Krastanov)

(Vollmer-Weber)

Preferred thicknesses

3-D island formation

Quantum confinement of electronic states

X-rays
Quantum Size Effects in Pb/Si(111)

Island heights appear to be highly uniform. Preferred thicknesses are attributed to QSE and self-organization. Preferred thicknesses are preferred thicknesses.

Morphology depends on:
- Kinetic pathway
- Pb/Si interface
- Pb coverage
- Temperature

$\theta = \gamma$
\[ \alpha = \eta \]
Surface X-ray Diffraction (SXRD)

Since $q \cdot a_1 = q \cdot a_2 = 0$, the specular rod is insensitive to in-plane order.

Thin film overlayers will contribute an amplitude similar to the N-slit interference function,

$$ S_N(x) = \sum_{n=0}^{N-1} e^{i n x} $$

Bragg peaks

$$ 0 \leq x = 2\pi n $$

Specular reflectivity

$$ f_k = k \cdot f $$
Interlayer Relaxations in Pb/Si(111)

Previous STM study observed oscillations in step heights. Step heights correlated with electronic effects. But: step height ≠ layer thickness

- Magnitude of layer relaxations?
- Follow Friedel oscillations?
- Penetration into film?
- Preferred thicknesses?
- Samples grown on different Pb/Si interfaces exhibit different preferred thicknesses?

W. B. Su, et al., PRL 86 (2001) 5116
Conduction electrons in thin metal films take on particle-in-a-box-like states. Free-electron charge density exhibits Friedel oscillations in $z$-direction. Friedel oscillations have a wavelength 

$$\frac{2}{\pi} \sin \left( \frac{2\pi}{\lambda} z \right) = (z)$$

$$\frac{d\varphi}{\varphi} = \frac{d\psi}{\psi} \approx \frac{z}{\lambda}$$
\( q_p W \{ N \theta \}, \{ N \theta \} \) 

\[
(z) s \nabla - (p + z) s \nabla = (z) f \nabla
\]

where

\[
\left[ \left( p \left( \frac{z}{T} - u \right) + s \nabla \right) f \nabla + p \right] \sum_{T-u}^{T} + Tz = N^f z
\]

The atom \( z \)-positions are determined via the free-electron model:

The atom \( z \)-positions are determined via the free-electron model:

A range of different island heights is used with occupancies \( \{ N \} \) is used with occupancies \( \{ N \} \) is used with occupancies

\[
\sum_{N \theta}^{N \theta} \sum_{n_{\text{film}}}^{n_{\text{film}}} \sum_{\text{Si substrate}}^{\text{Si substrate}} \left( \sum_{N}^{N} \sum_{n_{\text{film}}}^{n_{\text{film}}} \sum_{\text{Si substrate}}^{\text{Si substrate}} \right) \left( \theta \right) \Omega \times \left( l \right) \Omega
\]

\[
\text{SXRD Reflectivity Model for Pb/Si(111)}
\]
Simulations are for 10ML Pb on Si(111)-7x7. For Pb(111), χ₉/2 ≈ 1.8d.

Used simplistic sinusoidal model for z₉. Half-order features appear with layer expansions. Simulations are for Si(111) - 7x7 10 ML Pb on for No layer expansions. Periodic layer expansions.
Layer Relaxations in Pb/Si(111)

Pb was deposited on both the 7$\times$7 and $\sqrt{3}$$\times$$\sqrt{3}$-$\sqrt{3}$ interfaces.

Profiles were fit with a range $\frac{4}{3}$ and $\frac{2}{3}$ of island heights to allow for a non-uniform distribution.

Profiles were fit with layer relaxations not reproduced without layer relaxations ($a = 0$) layer relaxations half-order features.

Profiles were fit with and without layer relaxations ($a = 0$) layer relaxations half-order features.

Probed at 115 K, Pb/Si(111)-7$\times$7

Probed at 180 K, Pb/Si(111)-$\sqrt{3}$$\times$$\sqrt{3}$-$\sqrt{3}$

($a$) 8.5 ML Pb on Si(111)-7$\times$7

($b$) 4.5 ML Pb on Pb/Si(111)

$N = 8$ islands

Annealed to 180 K

Deposited at 115 K

$N = 10$ islands

Deposited at 185 K

Probed at 180 K
Layer Relaxation Results

Oscillatory relaxations

More data needed for trends

<table>
<thead>
<tr>
<th>Parameter</th>
<th>N=10</th>
<th>N=8</th>
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<tr>
<td>$g$</td>
<td>$\sqrt{3}$</td>
<td>$7 \times \sqrt{2}$</td>
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Island Growth

**Growth curve model:**

\[
(\varepsilon v_z b)^N S^N \theta^{\{N\}} \underbar{\sum} = (v b)^N \overline{V}
\]

**Distribution of island heights**

**Monolayer vs Bilayer growth**
Island Growth Example — Pb/Si(111)

Application to other systems: •

Expect interface dependence •

As \( T \) increases, islands grow irreversibly •

bulk-like

Movement of Pb peak at \( l = 6 \) to \( l \approx 6.4 \) to •

Bilayer height selection even as film roughens •

Primary island height evolves as •

\[ \theta_{Pb/Si(111)}/^{3} \times \sqrt{3} - \theta_{Pb/Si(111)} \]

Started with 4.5 ML Pb on •
Unusual growth behavior has been observed in thin metal films attributed to QSE. Electronic confinement can lead to preferred thicknesses as well as characteristic structural effects. Both electronic (thermodynamic) and kinetic effects are important. Proposed experiments:

1. Layer relaxations in Pb/Si(111)
2. Studies of island growth with temperature

Need more data:

- Metal films attributed to QSE
- Unusual growth behavior has been observed in thin...
\[ \Theta \approx \frac{\pi}{2} \]  

**Resolution** •

Cross-section  

Ridge scan takes diagonal  

Cross-section of rod  

Rocking curve takes perpendicular

\[ |(g/ + \alpha)(g/ - \alpha)| \sim \frac{\lambda}{\|b\|} \]

\[ |g/ \cos - \alpha \cos| = \frac{\lambda}{\|b\|} \]

\[ g/ + \alpha \approx g/ \sin + \alpha \sin = \frac{\lambda}{\|b\|} \]
What we really want is the integral in reciprocal space,\footnote{\textit{Integrated Intensity (total energy measured by detector)}}

\[
 zb \psi(b) \int (-I) \frac{m}{2} \int \int \frac{m}{0} \int = \psi(b) \int \int \int
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Free-Electron Density in a Quantum Well (Expanded)
STM images taken with different biases: (a) -5 V (b) +5 V

- Electron fringes showing variable spillage of charge density into the vacuum
- "Real" island topology ↔ (a) "Island topology"
- Charge spillage into vacuum
Rocking Curves vs. "Ridge" Scans