Quantum Oscillations in the Layer Structure of Thin Metal Films

An x-ray study of smooth Pb/Si(111) films

P. Czoschke, L. Basile, Hawoong Hong, and T.-C. Chiang
Department of Physics, University of Illinois at Urbana-Champaign

Physics Illinois

Electronic Effects in Metal Films

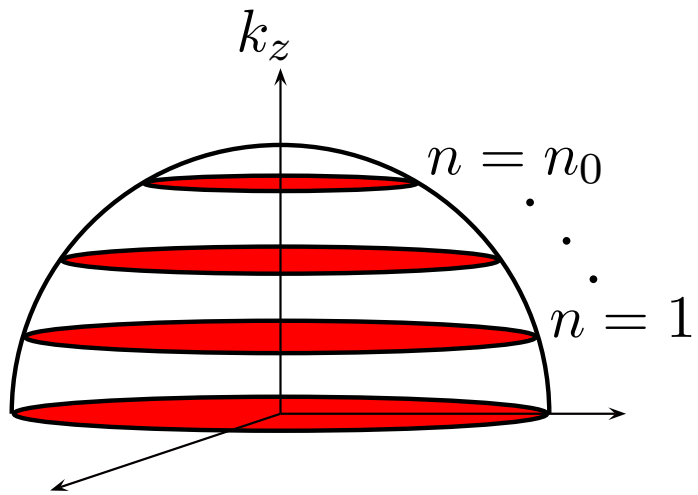
- For low temperatures, ultrathin metal films exhibit quantum size effects when approaching the atomic scale
- Significant electronic contributions to film energetics have been demonstrated
  - $\Rightarrow$ Self-organized “mesas” of preferred heights (Pb/Si, Ag/Si, Pb/Cu)
  - $\Rightarrow$ Smooth films (Ag/GaAs, Ag/Fe)
- Electron confinement may cause structural effects as well
  - $\Rightarrow$ Modulations in step height have been observed in STM and HAS studies
    - $\longrightarrow$ These techniques only sense the top surface morphology
    - $\longrightarrow$ X rays penetrate whole film, all the atomic layers of the film are probed as well as the film/substrate interface

What is the nature of any induced structural distortions of the atomic lattice?

Free Electron Model

Consider a 3-D free-electron gas confined to a 1-D quantum box:

$$\Psi_{\mathbf{k}}(\mathbf{r}) \propto e^{i(k_x x + k_y y)} \sin(k_z z)$$



Fermi sphere with subbands at $k_z = \frac{n\pi}{D}$

Charge density:

$$\rho(z) \propto \sum_{n=1}^{n_0} \pi \left(k_F^2 - k_z^2\right) \sin^2(k_z z)$$

Self-normalized density variations:

$$\delta\rho(z) \equiv \frac{\rho(z) - \langle\rho(z)\rangle_z}{\langle\rho(z)\rangle_z}$$

$$= -\frac{1}{C_D}\left(k_F^2 + \frac{1}{4}\frac{\partial^2}{\partial z^2}\right) \sum_{n=1}^{n_0} \cos\left(\frac{2\pi z}{D} n\right)$$

n_0-slit interference pattern with $\lambda \approx \pi/k_F = \lambda_F/2$.

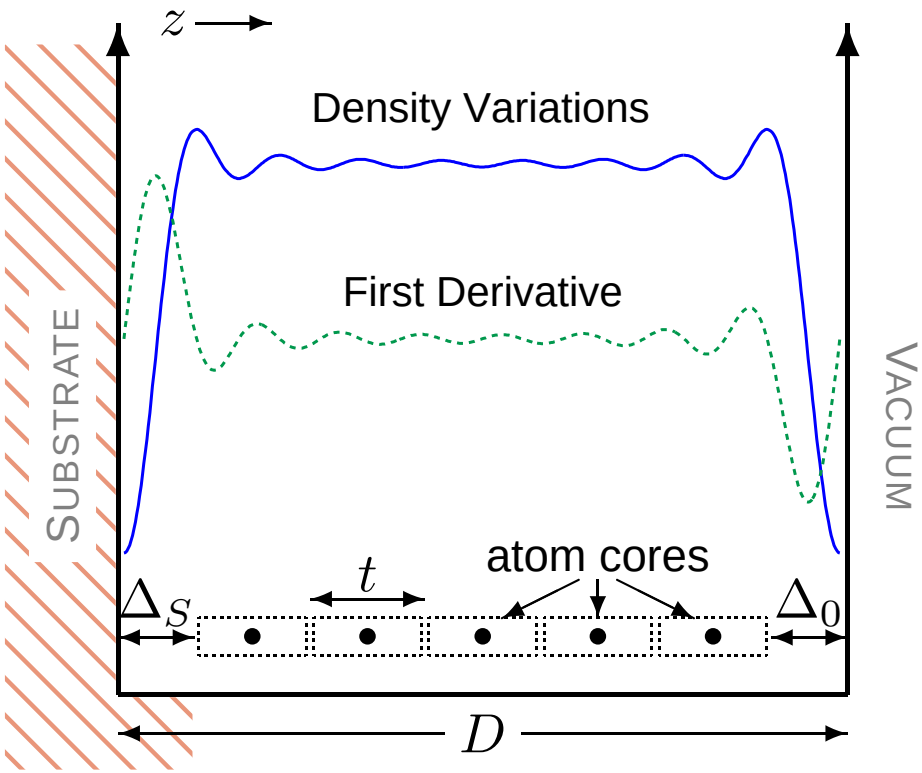
Charge Density in Pb/Si(111) Films

- Atoms shift from ideal positions $\Delta z \propto \frac{\partial}{\partial z}\delta\rho(z)$
- 4 parameter model for lattice distortions: Δ_S, Δ_0, t, A
- Layer-by-layer growth was found for ultrathin Pb films grown on the Pb/Si(111)-$\sqrt{3}\times\sqrt{3}$-β interface at low temperatures (110 K)

  $\Rightarrow$ 1-D Electron confinement

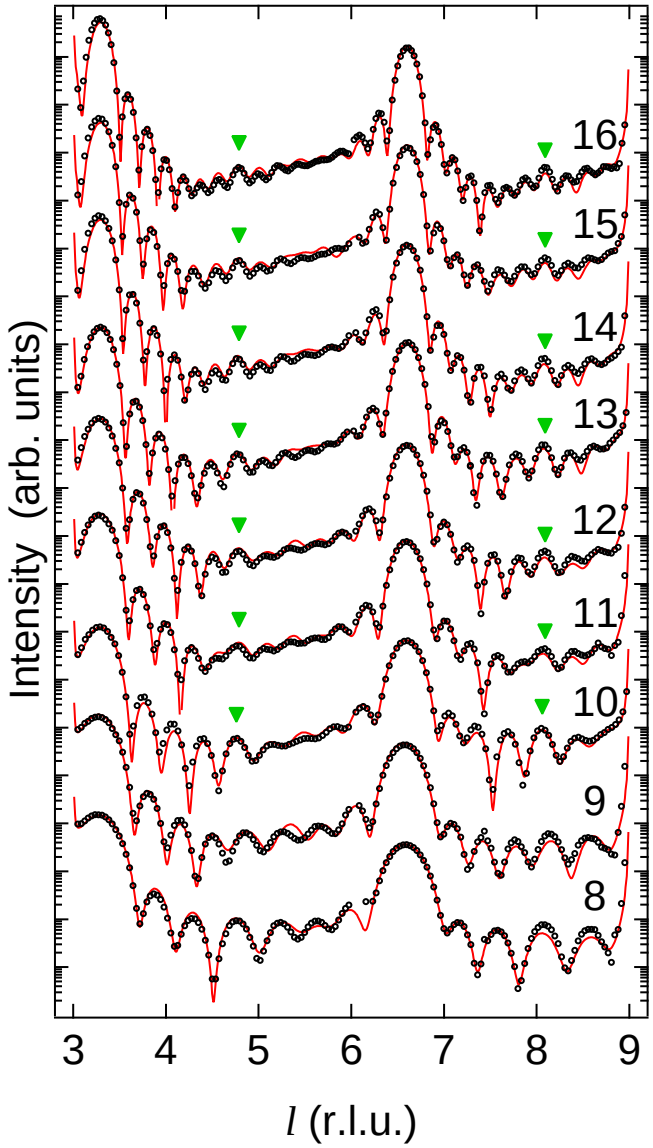
  $\Rightarrow$ Uniform quantum wells

  $\Rightarrow$ Δ_S, Δ_0, t, A may be thickness-dependent



Electrons are confined to a quantum box of width $D = \Delta_S + Nt + \Delta_0$

Extended X-ray Reflectivity Data



- Scattering from high l values gives sub-Å structural information
- Data was taken for $N = 6 - 18$ AL and $l = 0.2 - 12$ r.l.u.
- Spectra all have features at the $\frac{1}{2}$-order point for Pb(111)

  $\Rightarrow$ Analogous to fractional-order peaks for surface reconstructions

  $\Rightarrow$ Indicative of a quasibilayer superperiodicity in z direction

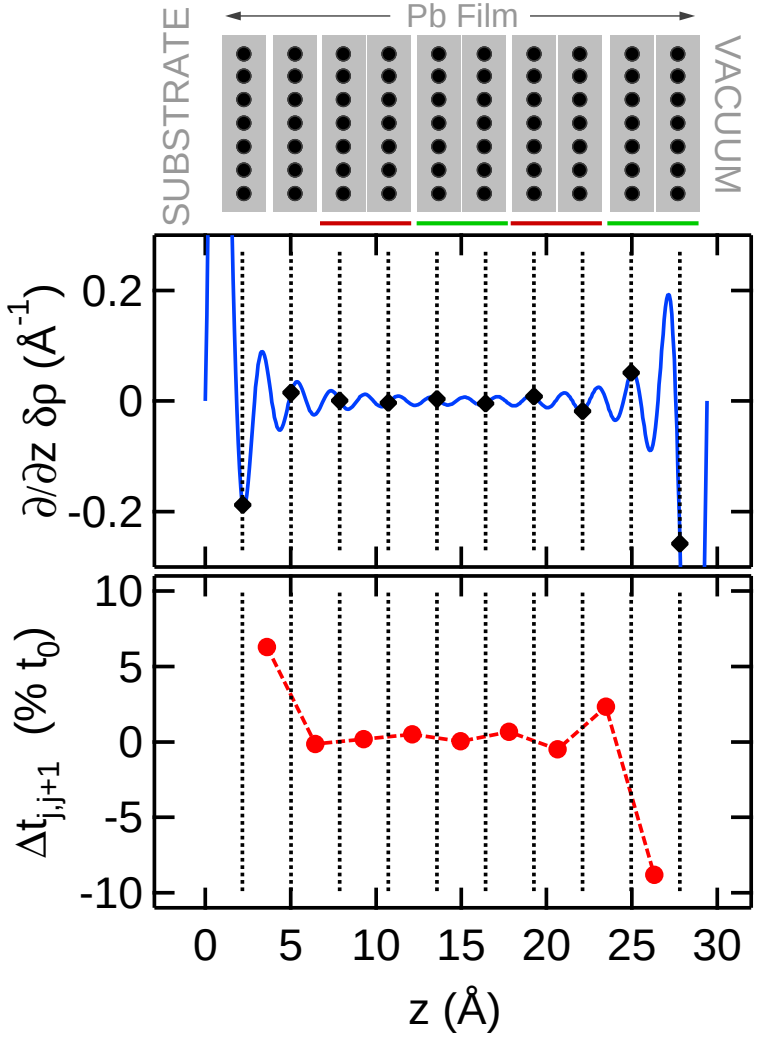
  $\rightarrow \lambda_F/2 \approx \frac{2}{3}$ AL

  $\Rightarrow$ Reproduced well by kinematic model using $\delta\rho$ lattice distortions

Structural Distortions of Atomic Layers

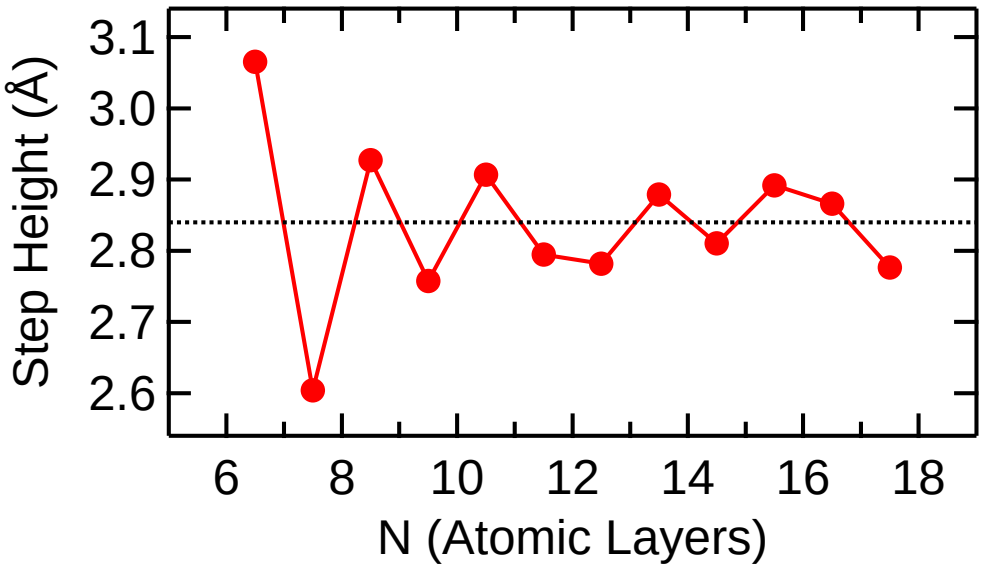
- Fits to data reveal variations in the interlayer spacings between individual atomic layers
- Quasibilayer oscillations are evident near film surface
- Significant distortion of film lattice near the substrate as well
- Distortions are qualitatively similar for the range of thicknesses

  $\Rightarrow$ Top interlayer spacing always compressed, etc.



Structural variations for N=10

Comparison to Previous Studies



- Step heights were deduced by taking the difference between net film thicknesses differing by 1 AL

  $\Rightarrow$ Net film thickness $\equiv \sum$ adjusted t's + Δ_0 (spillage into vacuum)

- Quasibilayer oscillations are present

- Similar to results from previous STM and HAS studies

Summary

- Interference of electron standing waves in metallic quantum well
  - $\Rightarrow$ Charge density oscillations
  - $\Rightarrow$ Structural distortions of the atomic lattice
- Lattice modulations are present at both film interfaces
- Step height modulations arise from subtle expansions and contractions due to different sized quantum wells
- Lattice distortions can be expected in metallic nanostructures

For more information see P. Czoschke et al. *PRL* **91** 226801 (2003)

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