

Upton *et al.* Reply: The preceding Comment [1] states that morphological evolution during film growth and annealing depends on a number of factors including energetics, kinetics, nucleation, etc. We agree. Kinetic constraints can make global energy minima inaccessible. However, for the simple case discussed in our Letter concerning the thermal stability (initial breakup) of atomically uniform films, the surface energy is an excellent guide, and our analysis stands correct [2]. Our experiment was designed to avoid the complicating factors mentioned in the Comment.

A specific point of the Comment is the utility of STM for studies of complex structural evolution. We agree that photoemission is not well suited for studies of inhomogeneous systems; we did not attempt to study such systems [2]. Nevertheless, it is interesting to note that the STM and photoemission results agree qualitatively. Namely, the even film thicknesses of $N = 6$ and 8 monolayers (ML) are more stable than the odd ones of $N = 5, 7$, and 9.

A second point of the Comment is that films with $N = 4$ should be stable by straightforward extrapolation, but experiments indicate otherwise. In [2], the experimental results were compared to a first-principles calculation. Good agreement was found over the range of $N = 5-9$, but not for $N = 4$. As we explained [2], the discrepancy could be due to the inaccuracy of the calculation resulting from the lattice mismatch between Pb and Si.

This is confirmed by a recent x-ray diffraction study of Pb/Si [3]. Height distributions for thermally roughened films were analyzed to yield the surface energy. Figure 1 compares the second discrete derivative of a rough film's height population, p''_N , to the maximum temperature of film stability as reported in [2]. A negative p''_N suggests a height more stable than the adjacent heights, resistant to bifurcation, and should correspond to a higher maximum temperature of stability relative to the neighboring heights. In Fig. 1 the scale for p''_N is reversed to facilitate comparisons of the two measurements. There is a good correspondence in that the slope of each line segment between N and $N + 1$ has the same sign for both cases. In particular, $N = 4$ corresponds to an unstable configuration. The quantum oscillation period in Pb is 2.2 ML [2,3]. The even-odd oscillation phase can reverse over a sufficiently wide range in N . This explains why $N = 4$ appears to be an exceptional case.

In addition to the local stability against $N \rightarrow N \pm 1$ bifurcation, one may need to consider global stability. A spontaneous phase separation phenomenon was reported in [4]. There, Pb films were grown on Si(111)-(7 × 7) at a temperature where rapid Pb diffusion leads to the formation of $N = 6$ magic-height islands after the completion of

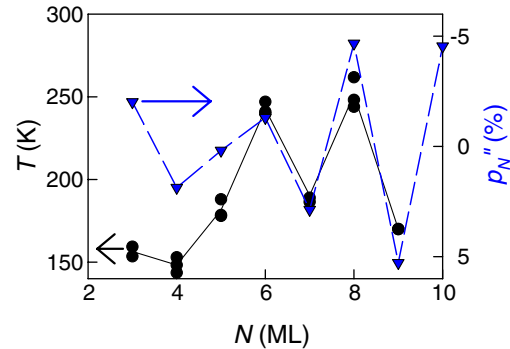


FIG. 1 (color online). Annealing temperatures at which the films of thickness N become unstable are indicated by circles with their averages connected by solid lines (scale to left) [2]. The second discrete derivative of the relative populations for a thermally roughened film are indicated by triangles connected by dashed lines (scale to right, reversed) [3].

a wetting layer. Thus, differences in growth conditions can indeed affect the final film morphology.

This work is supported by the U.S. National Science Foundation (Grant No. DMR-02-03003). We acknowledge the Petroleum Research Fund and the U.S. Department of Energy, Division of Materials Sciences (Grant No. DEFG02-91ER45439), for partial support of the synchrotron beam line operation and the facilities of the Frederick Seitz Materials Research Laboratory. The Synchrotron Radiation Center is supported by the U.S. National Science Foundation (Grant No. DMR-00-84402).

M. H. Upton, T. Miller, and T.-C. Chiang
Department of Physics
University of Illinois at Urbana-Champaign
1110 W. Green Street
Urbana, Illinois 61801-3080, USA
Frederick Seitz Materials Research Laboratory
University of Illinois at Urbana-Champaign
104 S. Goodwin Avenue
Urbana, Illinois 61801-2902, USA

Received 11 October 2004; published 23 February 2005

DOI: 10.1103/PhysRevLett.94.079702

PACS numbers: 73.21.Fg

- [1] M.C. Tringides and M. Hupalo, preceding Comment, Phys. Rev. Lett. **94**, 079701 (2005).
- [2] M. H. Upton, C. M. Wei, M. Y. Chou, T. Miller, and T.-C. Chiang, Phys. Rev. Lett. **93**, 026802 (2004).
- [3] P. Czoschke, H. Hong, L. Basile, and T.-C. Chiang, Phys. Rev. Lett. **93**, 036103 (2004).
- [4] H. Hong *et al.*, Phys. Rev. Lett. **90**, 076104 (2003).