Island Formation and Critical Thickness in Heteroepitaxy

In a recent Letter Chen and Washburn [1] proposed a mechanism for island nucleation in large-mismatch heteroepitaxy. The predicted coverage (Θ) dependence of the 3D island density $\rho_i(\Theta)$ reproduces the fast increase in the island density near the critical coverage $\Theta_c \approx 1.6$ ML [2]. Here we show that the critical coverage predicted by Ref. [1] depends strongly on the growth rate, thus contradicting, among others, the experimental results of Refs. [2,3].

The starting hypothesis of Ref. [1] is that during deposition submonolayer islands (platelets) form, and their size distribution follows $\rho(N,\Theta) = (\Theta/\overline{N}^2)\Phi(N/\overline{N})$ [4], where $\overline{N}$ is the average island size and Ref. [1] used the scaling function $\Phi(u) = 1.1u\exp(-0.27u^{3.7})$ [5]. Nucleation theory indicates that only platelets larger than a critical nucleus size N_c form 3D islands, the resulting island density being $\rho_i(\Theta) = \int_{N_c}^{\infty}\rho(N,\Theta)$. Using $\overline{N} = C\Theta$ with a proper choice of C, the density $\rho_i(\Theta)$ fits well the experimental results of Ref. [2]. However, this model assumes that C is constant, in contrast with the main results of submonolayer epitaxy, which predict that C *depends strongly on growth rate and temperature* [4]. Indeed, the average distance between islands is given by $l_d = (1/a)(D/F)^{\psi}$, where D is the diffusion constant of the adatoms, F is the growth rate, $\psi = 1/2$ [6], and a is a constant. Consequently, the complete expression for the average island size is $\overline{N} = \Theta l_d^2 = (1/a)(D/F)^{2\psi}\Theta$ which leads to

$$\rho_i(\Theta) = a\left(\frac{F}{D}\right)^{2\psi}\int_{l}^{\infty}\Phi(u)\,du\,, \qquad (1)$$

where $l = aN_c(F/D)^{2\psi}/\Theta$.

The island density given by (1) has a strong (F/D) dependence. To see this, in Fig. 1 we plot $\rho_i(\theta)$ as a function of θ (the plot is similar to Fig. 4 of Ref. [1]). Indeed, with a proper choice of (F/D), (1) provides a reasonable fit to the experimental results of Ref. [2]. However, a factor of 2 increase or decrease in the growth rate, while keeping the temperature constant, gives a rather different critical coverage, as shown in Fig. 1. This result contradicts numerous experimental observations that find that the critical coverage, within experimental errors, is independent of the growth rate. For example, Gerard [3] measured the photoluminescence spectra for three different growth rates, 0.125, 0.5, and 2 ML/s, and observed no change in the critical coverage. Under similar conditions the model of Ref. [1] predicts a *16-fold variation in* Θ_c, an effect that would easily be detected experimentally.

In fact the physical origin of the critical coverage is much studied for Ge/Si heteroepitaxy, and lies in the thermodynamic properties of the strained overlayer system [7], and not in the dynamics of the submonolayer islanding, as suggested by Ref. [1]. Indeed, while certainly dynamical effects play an important role, calculations based on equilibrium principles can reproduce many key aspects of quantum dot formation, including the critical coverage and the existence of several growth modes [8].

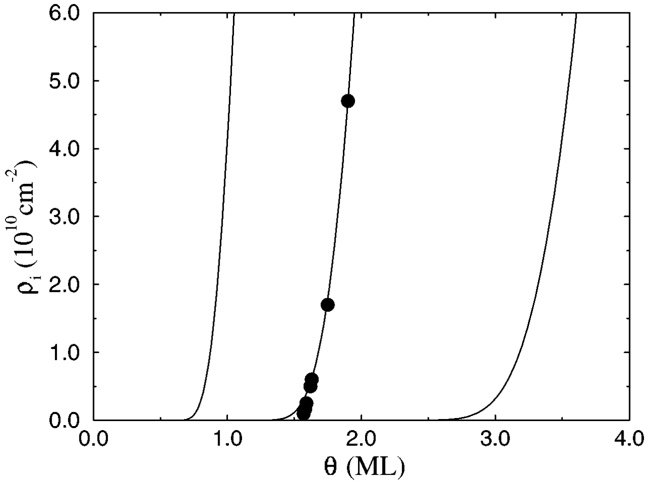

FIG. 1. Island density as a function of the coverage Θ, as predicted by (1). The growth rates corresponding to the three curves from left to right are F_1, $2F_1$, and $4F_1$, respectively. The dots correspond to the experimental results of Ref. [2].

I. Daruka and A.-L. Barabási
University of Notre Dame
Department of Physics
Notre Dame, Indiana 46556

Received 22 January 1997 [S0031-9007(97)02933-5]
PACS numbers: 68.55.Jk, 61.72.-y, 81.10.Aj

[1] Y. Chen and J. Washburn, Phys. Rev. Lett. **77**, 4046 (1996).
[2] D. Leonard, K. Pond, and P. M. Petroff, Phys. Rev. B **50**, 11687 (1994).
[3] J.-M. Gerard, *Confined Electrons and Photons*, edited by E. Burstein and C. Weisbuch (Plenum Press, New York, 1995); similar effects were documented by M. S. Miller *et al.*, Solid-State Electron. **40**, 609 (1996); M. S. Miller *et al.* (unpublished).
[4] M. C. Bartelt and J. W. Evans, Phys. Rev. B **46**, 12 675 (1992); J. G. Amar and F. Family, Phys. Rev. Lett. **74**, 2066 (1995); for a review, see A.-L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, 1995).
[5] One can also question the validity of $\rho(N,\Theta)$, derived for *submonolayer* epitaxy [4], for coverages as high as 2 ML s.
[6] Experiments on InP grown on GaAs indicate that $\psi = 1/2$ [W. Seifert (private communication)], different from the $\psi = 1/6$ prediction for nucleus free islanding [4].
[7] J. Tesoff, Phys. Rev. B **43**, 9377 (1991); C. Roland and G. H. Gilmer, Phys. Rev. B **47**, 16 286 (1993).
[8] I. Daruka and A.-L. Barabási (to be published).

0031-9007/97/78(15)/3027(1)$10.00