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Critical island size for a shape transition in strained Cu/Ni(100) islands

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ABSTRACT

We examine the shape instability of dislocation-free strained islands in heteroepitaxial growth, using continuum elasticity theory. Using the dipole interaction approximation for the strain-energy proposed by Pimpinelli and Villain, we have calculated the critical island size at which the shape instability may occur, and found that our expressions for the strain-energy and corresponding critical island size are very similar to those obtained by Li, Liu, and Lagally (Phys. Rev. Lett. 85 (2000) 1922). In addition, for the case of Cu/Ni(100) submonolayer islands we have carried out a direct comparison between the continuum elasticity predictions for the force monopole density and dipole interaction energy and atomistic calculations. Our results indicate that while the continuum elasticity expressions significantly underestimate both the force monopole density and the dipole interaction energy, the use of atomistic calculations leads to reasonable agreement between the two approaches. Our results also confirm that the experimentally observed ramified islands in Cu/Ni(100) submonolayer growth cannot be explained by equilibrium energetics arguments.

1. Introduction

The evolution of the island morphology in heteroepitaxial growth is a topic of significant experimental [1–11] and theoretical [12–18] interest due to its fundamental importance as well as its implications for nanostructure formation and stability. Unlike the case of homoepitaxial growth for which short-range interactions typically dominate, in the case of heteroepitaxial growth long-range elastic interactions may also play an important role. As a result, the equilibrium island-shape is expected to be determined by a competition between strain and surface and/or edge energies [1,4–7,10,11,13,19–21]. In particular, as shown by Tersoff and Tromp [12], strained 3D islands may undergo a spontaneous shape transition as a function of island-size [7,19,20].

More recently Li, Liu, and Lagally (LLL) [14] have used continuum elasticity theory to investigate the effect of strain on the stability of a square submonolayer island under biaxial isotropic stress. In particular, by expressing the finite-size correction $\delta E_{\rm strain}(s,t)$ to the strain energy of a rectangular submonolayer island of width s and length t (see Fig. 1) in terms of an integral involving the interactions between "force-monopoles" of magnitude F at the island step-edges, they obtained the result,

$$\delta E^{\rm LLL}_{\rm strain}(s,t)/E_u = 4(s+t) + 2(1-\nu_s)\Big[-4\sqrt{s^2+t^2} + s\ln\frac{\sqrt{s^2+t^2}+s}{\sqrt{s^2+t^2}-s} + t\ln\frac{\sqrt{s^2+t^2}+t}{\sqrt{s^2+t^2}-t} - 2s\ln\frac{s}{eb} - 2t\ln\frac{t}{eb}\Big] \quad (1)$$

where b is a cutoff length, ν_s and μ_s are the Poisson ratio and Young's modulus of the substrate, and E_u is the "unit strain energy" given by,

$$E_u = \frac{1+\nu_s}{2\pi\mu_s}F^2. \quad (2)$$

By combining this contribution to the island-energy with the contribution γP due to the step free-energy (where $P = 2(s+t)$ is the island perimeter and γ is the step free-energy per unit length) LLL obtained an expression for the critical island-width L_c above which there is a transition from a square to a rectangular island shape of the form,

$$L_c^{\rm LLL} = b\ exp\Big[\frac{\alpha+2}{2(1-\nu_s)} + 1.3\Big] \quad (3)$$

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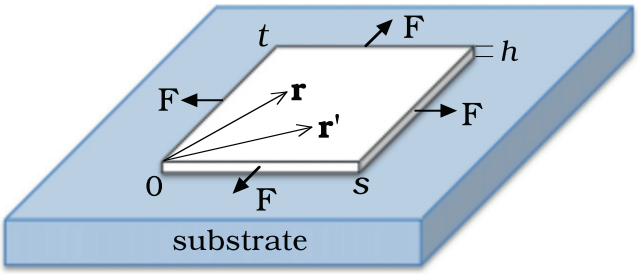

Fig. 1. Schematic diagram of 2D strained island of size $s \times t$ on a substrate. F represents the force monopole exerted along the island periphery due to the lattice mismatch. Here h is the height of the 2D island.

where $\alpha = \gamma/E_u$. These results have also been extended and applied to the case of anisotropic islands on an anisotropic or reconstructed (100) surface [5,17] for which both the step free energy γ and the sign of the force monopole F depend on the step orientation.

While Eq. (3) may be used to estimate the critical island-width for strained islands on isotropic (100) surfaces once the step free-energy γ and force-monopole density F are known, the analysis of LLL does not provide an estimate for F nor does it provide a prediction for the asymptotic strain-energy density. As a result, while this approach takes into account the step–step interaction, e.g. the interaction between the force monopole at a given position along the island step-edge and the substrate displacement due to the strain induced by the force monopoles at other points along the step, it does not take into account the contributions to the elastic energy away from the step-edge. In addition, it is based on the assumption that the force-monopole is constant along the island-edge and does not depend on the distance from the corner. Furthermore, while it has previously been suggested that the experimentally observed armwidth of the ramified islands observed in Cu/Ni(100) submonolayer growth might correspond to the critical island-width L_c for the equilibrium island-shape due to strain, this approach leads to a predicted value [22] which is several orders of magnitude larger than the experimentally observed armwidth. Accordingly, it is of interest to determine if this result can be confirmed using an alternative approach, in which the continuum expressions are directly "fit" to the asymptotic strain energy density (which can be directly calculated) rather than to the force monopole density.

We note that in previous work [23–25] it has been shown that for the case of biaxial (isotropic) strain the total strain energy of a submonolayer island may be approximately written in terms of a dipole interaction of the form,

$$E_{\text{strain}} = \frac{E_u'}{2}\int d^2\mathbf{r}\int d^2\mathbf{r}'\,\frac{1}{|\mathbf{r}-\mathbf{r}'|^3} \tag{4}$$

where the integrals are over the portion of the substrate below the island and the continuum elasticity prediction for the dipole interaction energy E_u' is,

$$E_u' = \frac{2\left(1-\nu_s^2\right)\mu_f^2\varepsilon^2h^2}{\pi\mu_s\left(1-\nu_f\right)^2} \tag{5}$$

where ν_f and μ_f are the Poisson ratio and Young's modulus of the film, h is the height of the island and the strain $\varepsilon = (a_s - a_f)/a_s$ where a_f (a_s) is the lattice constant of the film (substrate). While the dipole interaction is only exact for large length-scales [25], one of the advantages of this formulation is that it leads to an analytical expression for the asymptotic strain energy density corresponding to the strain energy per unit area for an infinite island. As a result, atomistic calculations of the dipole interaction strength E_u' may be used to determine the critical island-width for a particular system. We also note that while Eq. (4) has been previously used [16] to derive an expression for the critical island-radius of a circular island under isotropic stress, no direct comparison between the continuum expression (Eq. (5)) for E_u' and atomistic calculations has previously been carried out, while the corresponding calculation for a transition from a square to a rectangular island has also not previously been carried out.

Here we present the results of such a calculation which we have carried out in order to obtain explicit expressions for the asymptotic strain energy density as well as the finite-size corrections to the island strain-energy for a rectangular island of width s and length t. Somewhat surprisingly, we find that our results for the finite-size corrections to the island strain-energy are very similar to the LLL expression (Eq. (1)), and thus lead to similar expressions for the dependence of the critical island-width L_c on the ratio $\alpha' = \gamma/E_u'$.

We then consider the implications of our results on the stability of Cu/Ni(100) islands [1,2]. We find that if E_u' is calculated using the continuum elasticity expression (Eq. (5)) then a value of the critical island-width which is even larger than that previously obtained using Eqs. (2) and (3) is obtained. On the other hand, if the value of E_u' is directly and more accurately determined based on the asymptotic strain-energy density obtained from density-functional-theory (DFT) calculations [26], a significantly smaller value of L_c is obtained. However, in both cases we find that the critical island-width is significantly larger than the typical arm-width (approximately $22a_1$ where a_1 is the nearest-neighbor distance) observed in experiments on submonolayer Cu/Ni(100) growth. These results confirm our previous conclusion [22] that the experimentally observed shape transition cannot be explained by equilibrium energetics calculations based on continuum elasticity theory. However, they also indicate that since the critical width is very sensitive to the ratio $\alpha' = \gamma/E_u'$, care should be taken when determining this value, and if possible this should be done based on values calculated directly from atomistic calculations.

This paper is organized as follows. In Section 2 we first derive general expressions for the strain-energy of a rectangular submonolayer island using Eq. (4). In Section 3 these results are then used to calculate the critical island-width L_c for Cu/Ni(100) islands. Finally, in Section 4 we summarize and discuss our results.

2. Strain-energy for a rectangular island

In order to calculate the island strain energy for a rectangular island of width s and length t, we first note that Eq. (4) may be rewritten in the form, $E_{\text{strain}} = E_u' \times I(s,t)$ where,

$$I(s,t) = \frac{1}{2}\iint d^2\mathbf{r}\,d^2\mathbf{r}'\,\frac{1}{|\mathbf{r}-\mathbf{r}'|^3} \tag{6}$$

and where $\mathbf{r}$ and $\mathbf{r}'$ are two-dimensional vectors with $\mathbf{r} = x\hat{x} + y\hat{y}$ and $\mathbf{r}' = x'\hat{x} + y'\hat{y}$ and $0 \le x, x' \le s$ and $0 \le y, y' \le t$. In order to remove the factor of $\frac{1}{2}$ and avoid double-counting we then modify the integral so that it satisfies the restriction $x' \ge x$,

$$I(s,t) = \int_0^s dx\int_0^t dy\int_x^s dx'\int_0^t dy'\,\frac{1}{\left(|x'-x|^2 + |y'-y|^2\right)^{3/2}}. \tag{7}$$

We note that although not explicitly indicated, due to discreteness effects a cutoff b in the minimum distance $|\mathbf{r} - \mathbf{r}'|$ which is of the order of the lattice-spacing is assumed. As in Refs. [14] and [15] we assume a square cutoff which matches the lattice. In particular, due to the cutoff we exclude from integration over $\mathbf{r}'$ a $2b \times 2b$ square area centered on the point with coordinate vector $\mathbf{r}$, where b is a cutoff

length. Using this exclusion, the integral $I(s,t)$ may be written as the sum of 3 integrals corresponding to different "exclusion regions", e.g. $I(s,t) = I_1 + 2I_2 + 2I_3$. The first integral,

$$I_1 = \int_0^{s-b} dx \int_0^t dy \int_{x+b}^s dx' \int_0^t dy' \frac{1}{(u^2+v^2)^{3/2}} \tag{8}$$

(where $u = x' - x$ and $v = y' - y$) corresponds to "exclusion via x", e.g. to the contribution in which x ranges from 0 to $s - b$ while x' ranges from $x + b$ to s and y and y' range from 0 to t. Converting variables from x', y' to $u = x' - x$ and $v = y' - y$ this may be rewritten,

$$I_1 = \int_0^{s-b} dx \int_0^t dy \int_b^{s-x} du \int_{-y}^{t-y} \frac{dv}{(u^2+v^2)^{3/2}}. \tag{9}$$

In contrast, the 2nd integral,

$$I_2 = \int_0^{s-b} dx \int_b^t dy \int_x^{x+b} dx' \int_0^{y-b} dy' \frac{1}{(u^2+v^2)^{3/2}} \tag{10}$$

corresponds to "exclusion via y", e.g. to the contribution in which y' remains below $y - b$ while x' ranges between x and $x + b$ and x again ranges from 0 to $s - b$. There is an equivalent contribution in which y' ranges from $y + b$ to t while y ranges from 0 to $t - b$ and therefore this integral is multiplied by two. Again, converting variables from x', y' to $u = x' - x$ and $v = y' - y$ this may be rewritten,

$$I_2 = \int_0^{s-b} dx \int_b^t dy \int_0^b du \int_{-y}^{-b} \frac{dv}{(u^2+v^2)^{3/2}}. \tag{11}$$

Finally, the 3rd integral,

$$I_3 = \int_{s-b}^s dx \int_b^t dy \int_x^s dx' \int_0^{y-b} dy' \frac{1}{(u^2+v^2)^{3/2}} \tag{12}$$

is similar to I_2 except that it includes the previously neglected contribution in which x ranges from $s - b$ to s while x' ranges from x to s. Again, there is an equivalent contribution in which y' ranges from $y + b$ to t while y ranges from 0 to $t - b$ and therefore this integral is multiplied by two. Converting variables from x', y' to $u = x' - x$ and $v = y' - y$ this may be rewritten,

$$I_3 = \int_{s-b}^s dx \int_b^t dy \int_0^{s-x} du \int_{-y}^{-b} \frac{dv}{(u^2+v^2)^{3/2}}. \tag{13}$$

Carrying out the 3 integrals in Eqs. (9), (11) and (13) and combining we obtain,

$$\begin{aligned} I(s,t) = &-2\left(2-\sqrt{2}\right)b + \frac{2\sqrt{2}st}{b} + 2(s+t)\left[1 - ln\left(1+\sqrt{2}\right)\right] \\ &+2s\ln\frac{s+\sqrt{s^2+t^2}}{t} + 2t\ln\frac{t+\sqrt{s^2+t^2}}{s} \\ &-4\sqrt{s^2+t^2} - 2s\ln\frac{s}{b} - 2t\ln\frac{t}{b} \end{aligned} \tag{14}$$

Removing the first (constant) term and exchanging factors of s and t between the third term and the last two terms, and using the identity,

$$ln\frac{s+\sqrt{s^2+t^2}}{t} = \frac{1}{2} ln\frac{\sqrt{s^2+t^2}+s}{\sqrt{s^2+t^2}-s} \tag{15}$$

we obtain the dipole-interaction expression for the total island strain energy,

$$\begin{aligned} \frac{E_{\text{strain}}}{E_u'} = &\frac{2\sqrt{2}st}{b} + 4(s+t) - 4\sqrt{s^2+t^2} \\ &+ s\ln\frac{\sqrt{s^2+t^2}+s}{\sqrt{s^2+t^2}-s} + t\ln\frac{\sqrt{s^2+t^2}+t}{\sqrt{s^2+t^2}-t} \\ &- 2s\ln\frac{se^{1+\ln\left(1+\sqrt{2}\right)}}{b} - 2t\ln\frac{te^{1+\ln\left(1+\sqrt{2}\right)}}{b} \end{aligned} \tag{16}$$

We note that, ignoring the first area-dependent term, Eq. (16) is very similar to the LLL result (Eq. (1)). However, in the limit of large s and t the first term dominates. Multiplying this term by E_u' and dividing by the island area st we obtain the asymptotic strain energy density,

$$\rho_0(b) = 2\sqrt{2}E_u'/b. \tag{17}$$

In addition, multiplying Eq. (16) by E_u' and adding the perimeter energy $2\gamma(s + t)$ to obtain an expression for the total island energy $E_{\text{total}}(c,D)$ (where $D = \sqrt{st}$ is the island diameter and $c = \sqrt{s/t}$ is the aspect ratio) and setting the 2nd partial derivative with respect to c (at $c = 1$) equal to zero, the critical length for a transition from a square island to a rectangular island may be obtained,

$$L_c = b\,exp\left[\alpha' + 1.3 - ln\left(1+\sqrt{2}\right)\right] \simeq b\,exp\left[\alpha' + 0.42\right]. \tag{18}$$

3. Comparison with LLL result and application to Cu/Ni(100) islands

We first carry out a qualitative comparison between our results for the critical island-width L_c with the LLL result Eq. (3). In particular, we note that the predicted critical island-width L_c will exceed the LLL prediction if $\alpha' > \alpha_c'$, where $\alpha_c' = \frac{\alpha+2}{2(1-\nu_s)} + 0.88$. We now consider the case of Cu/Ni(100) which is of interest since previous experiments [1,2] over a temperature range from 250 to 345 K have indicated the formation of ramified submonolayer islands with a typical armwidth of approximately $22a_1$ (where a_1 is the nearest-neighbor distance). As noted in Section 1, while it has previously been assumed [1,2] that the experimentally observed island ramification may be explained by the use of equilibrium arguments [12], calculations which we have carried out [22] using Eq. (3) indicate that the corresponding critical island-width L_c is at least several orders of magnitude larger than the typical armwidth, thus suggesting that kinetic effects mediated by strain may play a role [22].

In particular, from DFT calculations [22] we have previously obtained $\gamma_{110}(T = 0) = 0.044$ eV/Å for this case. Similarly, from DFT calculations of the substrate and thin-film stresses σ_{xx}^s and σ_{xx}^f we have obtained using the expression [14],

$$F = \sigma_{xx}^f - \sigma_{xx}^s \tag{19}$$

the estimates $F = -0.134$ eV/Å^2, $E_u = \frac{1+\nu_s}{2\pi\mu_s}F^2 = 3.0 \times 10^{-3}$ eV/Å and $\alpha = \gamma/E_u = 14.7$. As discussed in Ref. [22], using Eq. (3) with cutoff $b = a_1$ then leads to an estimate of the critical island-width $L_c = 6.5 \times 10^5 a_1$ which is significantly larger than the experimentally observed arm-width (see Table 1).

For comparison, we now consider the critical island-width obtained using the dipole interaction expression Eq. (4). If the dipole interaction E_u' is estimated using the continuum elasticity expression (Eq. (5)) then a significantly higher value for the critical island-width L_c/a_1 is obtained (see Table 1). This is perhaps not surprising since the continuum elasticity expression [12,15] for the monopole density,

$$F = \mu_s \varepsilon h \tag{20}$$

Table 1
Comparison of results for the critical island width L_c for Cu/Ni(100) submonolayer islands calculated using Eq. (3) with results obtained using Eq. (18) for the case of a square cutoff. Values with an * in front are based on DFT calculations as discussed in the text, while those with a [2], [5], or [20] in front were calculated using the corresponding continuum elasticity equations.

	F (eV/Å^2)	E_u (eV/Å)	α	E_u' (eV/Å)	α'	L_c/a_1
Eq. (3)	*0.134	*[2]0.003	14.7	–	–	*6.5×10^5
Eq. (3)	[20]0. 062	[2]6.4×10^{-4}	68.7	–	–	6.8×10^{22}
Eq. (18)	–	–	–	*0. 0039	11.3	*1.2×10^5
Eq. (18)	–	–	–	[5]0. 0014	31.4	6.8×10^{13}

also leads to a significantly larger estimate for the critical island-width since it leads to an underestimate for the value of F compared to DFT calculations (see Table 1).

However, since our results imply a direct relationship between the asymptotic strain energy density ρ_0 and E_u', it is possible to obtain a more accurate value for L_c by using the asymptotic strain energy density to determine E_u'. In particular, Eq. (16) implies that,

$$E_u' = \rho_0 b/2\sqrt{2} \tag{21}$$

where b is a cutoff length. Accordingly, we have carried out DFT calculations of the asymptotic strain energy density ρ_0 for a complete monolayer of Cu on the Ni(100) surface, using the expression,

$$\rho_0 = \left(E_{ML,n} - E_{ML,m}\right)/A \tag{22}$$

where A is the unit-cell substrate area, and $E_{ML,n} = E_{\text{total},n} - E_{\text{substrate},n}$ is the energy (per unit cell) of a complete Cu monolayer (corresponding to the difference between the total unit cell energy with a complete Cu monolayer and without any Cu) for the case of a Ni(100) substrate with a "normal" bulk lattice constant ($a_{Ni} = 3.52$ Å). Similarly, $E_{ML,m} = E_{\text{total},m} - E_{\text{substrate},m}$ is the corresponding quantity for a "matched" Ni(100) substrate which has been expanded so that there is no mismatch at the interface.

Our DFT total-energy calculations were carried out using the Vienna ab initio simulation package (VASP) [27] with ultrasoft Vanderbilt pseudopotentials [28] and the generalized gradient approximation (GGA) using the Perdew–Wang functional (PW91) [29]. Bulk calculations resulted in lattice constants of 3.52 Å and 3.64 Å for Ni and Cu, respectively. Methfessel–Paxton [30] smearing with $\sigma = 0.2$ was used and for **k**-point sampling, the Monkhorst–Pack scheme [31] with a $8 \times 8 \times 1$ mesh was used along with a kinetic energy cutoff of 33.1 Ry. We used supercells of size 2×2 with a 20 ML thickness Ni substrate and a vacuum spacing of 12 Å. For the Cu/Ni case, an additional 1 ML Cu layer was added to both the top and bottom of the 20 ML Ni substrate. Full atomic relaxation of all atoms in the supercells was allowed. All geometries were optimized until the remaining forces were smaller than 10^{-4} eV/Å.

From our DFT calculations we obtain an asymptotic strain energy density $\rho_0 = 4.4 \times 10^{-3}$ eV/Å^2. Assuming a cutoff b equal to the nearest-neighbor distance a_1, we obtain $E_u' = 3.9 \times 10^{-3}$ eV/^{2}Å while using Eq. (18) this leads to an estimate for the critical island-width $L_c/a_1 \simeq 1.2 \times 10^5$ which is much closer to the LLL prediction and is also significantly larger than the typical arm-width observed in experiments on submonolayer Cu/Ni(100) growth. We have also obtained similarly good agreement between the LLL prediction and the dipole-interaction prediction for the critical island-width by carrying out atomistic calculations of the step free-energy γ and dipole-interaction strength E_u' for much larger systems using the Mishin, Voter, Bonney [32–34] (MVB) embedded-atom method [35] potential, although in this case the value of L_c is slightly smaller [36]. Thus our results confirm that the experimentally observed shape transition cannot be explained by equilibrium energetics arguments based on continuum elasticity theory. However, they also indicate that since the critical width is very sensitive to the ratio $\alpha' = \gamma/E_u'$, care should be taken when determining this value, and if possible this should be done based on values calculated directly from atomistic calculations.

4. Discussion

Using the dipole-interaction expression (Eq. (4)) proposed by Pimpinelli and Villain [24] we have derived expressions for the elastic strain energy of a rectangular island and critical island-width L_c as a function of the ratio $\alpha' = \gamma/E_u'$ of the step free-energy per unit length to the dipole interaction E_u'. Somewhat surprisingly we have found that the resulting expressions for the finite-size corrections to the strain energy, as well as for the corresponding critical island-width, are very similar to those previously obtained in Ref. [14] by Li, Liu, and Lagally [14] using a somewhat different 'force-monopole' approach. However, in contrast to this approach, our results also include the asymptotic strain energy thus allowing a direct connection between the value of the 'dipole interaction strength' E_u' and the asymptotic strain energy density.

In addition, we have applied our results to the case of Cu/Ni(100) submonolayer growth for which ramified islands have been observed. In particular, we find that using the continuum elasticity predictions for the dipole interaction strength E_u' and force monopole F leads to values of the critical island-width which are significantly larger than previously obtained using the value of F obtained from atomistic calculations [22]. In contrast, an estimate of the dipole interaction strength based on a DFT calculation of the asymptotic strain energy density, leads to an estimate for the critical island-width L_c which is close to—although somewhat smaller than—that obtained from DFT calculations of the force monopole F. However, all of these values are still significantly larger than the typical arm-width observed in submonolayer Cu/Ni(100) growth.

While our calculations were carried out at zero temperature, it is also of interest to estimate to what extent finite-temperature effects might affect our results. Previously we have calculated the finite-temperature corrections to the step free-energy for Cu/Ni(100) islands and have found [22] that this leads to a relatively small reduction in the step free energy and thus a relatively small reduction in the critical island-size. In addition, we have carried out atomistic simulations of rectangular islands with rough step-edges which indicate that roughness along the step-edge leads to island relaxation, thus slightly reducing the strain-energy and further increasing the critical island-width. Thus our results confirm that the experimentally observed shape transition in this case cannot be explained by equilibrium energetics arguments based on continuum elasticity theory. This is consistent with recent studies of mesoscopic relaxation in Cu/Cu(111) and Co/Cu(100) [37,38] as well as experiments on Co/N/Cu(001) [39] which indicate that for small islands, the island relaxation cannot be explained by continuum elasticity theory. This is also supported by kinetic Monte Carlo simulations [22] which indicate that kinetic effects (mediated by strain) may explain the ramified island shapes observed at $T = 250$ and 300 K.

In conclusion, we have used the dipole-interaction expression proposed by Pimpinelli and Villain [24] to obtain an expression for the strain energy of rectangular islands on an isotropic substrate. In addition, we have carried out a direct comparison between the continuum expressions for the force monopole density and dipole interaction energy and atomistic calculations. While our results indicate that the continuum elasticity expressions significantly underestimate both the force monopole density and the dipole interaction energy (thus leading to estimates for the critical island-width which differ by 9 orders of magnitude) our results also indicate that the use of atomistic calculations leads to reasonable agreement between the force-monopole and dipole interaction approaches. In addition, our results also confirm that the experimentally observed ramified islands in

Cu/Ni(100) submonolayer growth cannot be explained by equilibrium energetics arguments. In the future it would be of interest to carry out a more detailed comparison between atomistic predictions for the strain energy and the continuum predictions.

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