Roughening and Fragmentation of Strained Ag Islands on Pt(111)

In a recent Letter [1] Blandin, Massobrio, and Ballone (BMB) have studied the thermal evolution of two-dimensional Ag islands on Pt(111) by molecular dynamics simulations using embedded atom potentials. The 4% lattice mismatch between Ag and Pt was shown to lower substantially the contour and kink energy of the pseudomorphic (1×1) Ag islands. Dynamic computations of the short time evolution (~ 10 ns) predicted a fragmentation of the strained Ag islands via kink formation and evaporation at $T \sim 600$ K.

BMB discuss their simulation results in relation with recent He-scattering experiments [2] which had detected an increase of diffuse scattering at coverages and temperatures similar to those found in their simulations. Röder et al. [3] have, however, definitely demonstrated that the irreversible decay of the specularly reflected He intensity upon heating above 550–600 K originates from the Ag-Pt intermixing in the topmost layer and has no relation to any fragmentation processes.

In this Comment we report on variable temperature scanning tunneling microscopy STM experiments which follow in situ the thermal evolution of strained Ag islands on Pt(111). The experiment provides direct evidence for the predicted island fragmentation via edge roughening and kink evaporation. The temperature for this morphology transition ($T = 300-350$ K) is, however, found to be substantially lower than predicted by the simulations.

The STM topographs in Fig. 1 show the thermal evolution of the morphology of compact 2D Ag islands on Pt(111). The islands in Fig. 1(a) have been grown by
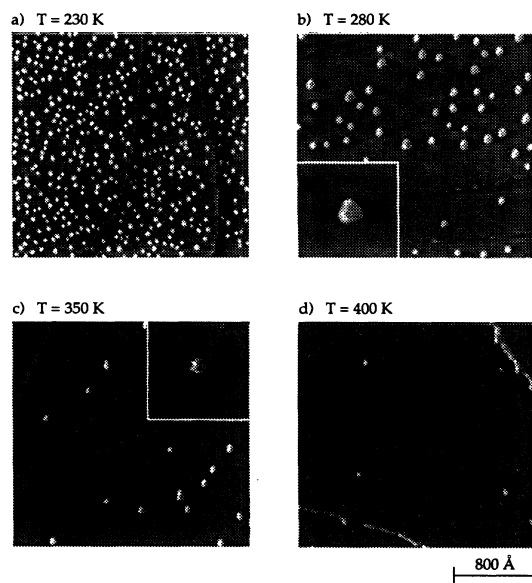
dimer nucleation at 50 K ($\Theta_{Ag} = 0.12$ monolayer) and subsequent annealing to 230 K giving a narrow island size distribution with an average cluster size of about 230 Ag atoms [4]. Further annealing to 280 K [Fig. 1(b)] leads to Ostwald ripening characterized by a broadened island size distribution and an increased average island size of about 1000 atoms. The large islands have developed a nice facetted shape [see inset in (b)] reflecting the anisotropy in step edge energies on fcc (111) substrates [4,5]. Around 350 K the morphology changes dramatically [Fig. 1(c)]; the large Ag islands are found to be in coexistence with a 2D gas phase [6]. The islands are already largely fragmented indicated by their substantially decreased average size. In the STM topographs the edges of the surviving larger Ag islands are imaged very rough [see inset in (c)], demonstrating that the fragmentation is driven via kink excitation (edge roughening) and subsequent evaporation of less coordinated edge atoms onto terraces. At 400 K [Fig. 1(d)] eventually all large Ag islands are disintegrated; the mobile 2D gas phase has now to a large extent been condensed at preexisting Pt steps.

The above experiments clearly prove the strain driven fragmentation of 2D islands via edge roughening and kink evaporation predicted by the static calculations of BMB. The transition temperature is, however, found to be a factor of 2 lower than predicted by the dynamic molecular dynamics (MD) simulations. This disagreement is of particular importance and uncovers the weak point of dynamic MD simulations. Even if the potentials would be correct, which is doubtful for the embedded atom model (EAM) potentials for Ag/Pt [7], simulations of thermodynamic configurations require "macroscopic" simulation times to ensure real equilibrium. As long as this time scale is inaccessible to computational power, caution should be taken in predicting thermodynamic equilibrium states.

FIG. 1. STM images of two-dimensional Ag islands on Pt(111) as a function of surface temperature; see text. The inset size in (b) and (c) is 450 Å $\times$ 450 Å.

Holger Röder, Harald Brune, and Klaus Kern
    Institut de Physique Expérimentale
    Ecole Polytechnique Fédérale de Lausanne
    CH-1015 Lausanne, Switzerland

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[7] The diffusion barrier for Ag adatoms on Pt(111) calculated with the EAM potential used by BMB [58 meV; Phys. Rev. B **47**, 13 687 (1993)] is a factor of about 3 too low compared to experimental data [(160 meV; H. Brune, H. Röder, and K. Kern, (to be published)].
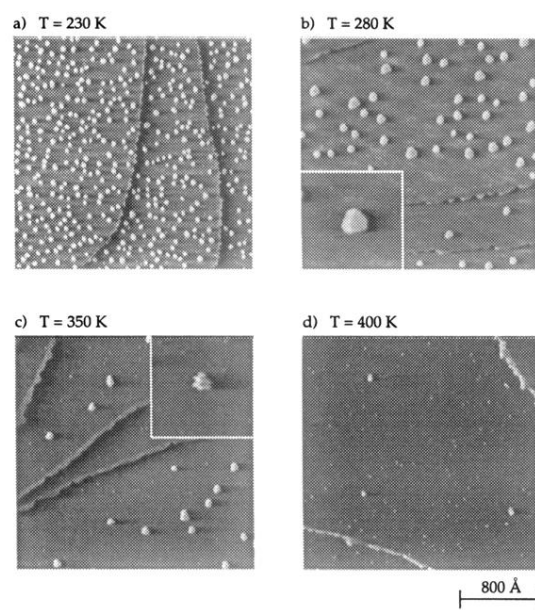
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