Comment on "Compressive Stress in Polycrystalline Volmer-Weber Films"

In a recent Letter [1] Koch *et al.* attempted to demonstrate that the reversible stress evolution observed during interruptions of Volmer-Weber growth cannot be explained by relaxation of kinetic roughening [2–4] or reversible nonequilibrium incorporation of adatoms at grain boundaries during growth [5]. We find the arguments made by Koch *et al.* fundamentally flawed.

First, the authors suggest that the observed reversible stress changes are associated with grain growth or recrystallization that occurs during growth interruptions [1]. As first pointed out by Shull and Spaepen [6,7], this cannot be the explanation since grain growth and recrystallization are not reversible.

In addition, Koch *et al.* suggest that the reversible tensile rise observed during growth interruptions occurs as "material moves away from the compressive surface regions between grains and then reattaches in the tensile grain boundary regions...". However, adatoms move to minimize chemical potential as defined by summing strain energy and curvature terms. Since the strain energy scales as the square of the stress, diffusion in general acts to minimize the magnitude of the stress (compressive or tensile). It has also been shown that the reversible stress changes observed during growth interruptions for polycrystalline and single crystalline (homoepitaxial) films of the same material are very similar in both magnitude and kinetic characteristics [3], so that an important role for grain boundaries seems unlikely.

Finally, an error was made in approximating the Laplace pressure acting on islands in the precoalescence regime. The authors incorrectly use the surface energy γ rather than the surface stress f [8,9]. Thus, the agreement between the capillary model discussed by Koch *et al.* and the experimental results (-1.2 GPa versus -1.3 GPa) would seem coincidental.

There are several competing models for the origins of growth interruption induced reversible stress changes during Volmer-Weber growth of polycrystalline [2–5], homoepitaxial [3], and heteroepitaxial [10] films. In variance with Koch *et al.*'s concluding remarks, this issue is not resolved.

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