

### Comment on “Surfactant-Mediated Growth Revisited”

In a recent Letter [1], Meyerheim *et al.* claim that up to 0.15 monolayer of oxygen is trapped in subsurface octahedral sites in the growth of Ni on the missing row (MR) Cu(001)( $\sqrt{2} \times 2\sqrt{2}$ )R45°-O surface, on the basis of their surface x-ray diffraction (SXR) and stress measurements. They further question the validity of the traditional view that a surfactant floats on the top of the growing film. In this Comment, we show that all the evidences present are problematic so that the claim made is not supported.

It has been observed that [2], in the submonolayer growth of Co on the same surface, surface Cu atoms are displaced from MR structure by incoming Co atoms and aggregated into elongated islands with ( $\sqrt{2} \times 2\sqrt{2}$ )R45°-O reconstructed surface. The deposited Co atoms are immobile and randomly distributed in the oxygen adsorbed surface as individual Co atoms and clusters at low coverages ( $\leq 0.4$  ML). Then they aggregate into fcc Co patches with  $c(2 \times 2)$ -O on them at larger coverages ( $\geq 0.5$  ML). This special growth behavior not only causes both surface roughness and compositional inhomogeneity in the unique way in the early growth stage (0–2 ML), but also results in a specific interface structure. The published results [3,4] indicate that the same atomistic processes occur in the initial growth of Ni, although there is no explicit conclusion for that. Scanning tunneling microscopy (STM) studies show the same elongated islands for the growth of Ni on MR-reconstructed Cu(001) as observed for the Co growth. In Ref. [4], the ( $\sqrt{2} \times 2\sqrt{2}$ )R45°-O structure is actually resolved for the surface of the elongated islands. They are indeed Cu islands. Medium energy electron diffraction oscillations also show different period for the early three oscillations [5].

Based on the above analysis, we first question the validity of the SXR evidence in Ref. [1]. In SXR, the structure model is derived by best fitting the experimental data to it. The reliability directly depends on whether the real structure has been considered as a trying model. Clearly, the authors of Ref. [1] did not notice the special atomistic processes in the initial growth stage and the resulted specified surface roughening, compositional inhomogeneity, and the formed unique interface structure, though surface roughness induced by fractional occupancies and atomistic interface intermixing are considered in their structural model. The interface structure is always detected by SXR for ultrathin films, regardless of the investigated coverage range, the surface reconstruction of the growing film, and the growth mode in the investigated coverage range. If the interface structure in the structural model is different from the real one, the results obtained by best fitting the SXR data are not reliable. Thus the structure derived in Ref. [1] has not been verified.

Second, we would like to point out the stress measurement evidence is invalid. In Ref. [1], the authors calculated the overall stress change starting from clean Cu(001) and ending with  $c(2 \times 2)$ -O/8 ML Ni/Cu(001) to be 4.13 N/m. Then they compared this value with the 4.64 N/m of stress change measured during 8 ML Ni growth on the MR-reconstructed Cu(001) and ascribed the difference of 0.51 N/m to the insertion of oxygen atoms in the subsurface region. However, the above two stress changes are incomparable because the initial state is different. The stress change of 4.64 N/m is measured starting from the MR-reconstructed O/Cu(001). Therefore, to make a correct comparison, the stress change during the formation of the ( $\sqrt{2} \times 2\sqrt{2}$ )R45°-O structure on clean Cu(001) should be added to the latter case. It has been measured to be  $-0.6$  N/m by the same group [6]. After this value is added, the stress change shows no difference. Therefore, the stress measurement evidence is invalid. It even becomes a negative one since considerable stress change is expected if subsurface oxygen exists as Meyerheim *et al.* stated [1].

Additionally, if one third of oxygen stay in the subsurface region, STM should detect the obvious loss of the  $c(2 \times 2)$ -O surface. However, the existed STM image of 5.5 ML Ni on the MR-reconstructed Cu(001) [5] shows that the whole surface is covered by  $c(2 \times 2)$ -O. No loss of oxygen has been observed in our STM study of Co on the same surface.

In summary, the claim made in Ref. [1] on the oxygen position is not supported.

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