Comment on "Long-Range Spatial Self-Organization in the Adsorbate-Induced Restructuring of Surfaces: Cu(110)-(2x1)O"

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In a recent letter, Kern et al. reported that anisotropic Cu–O islands form striped periodic supergratings on the Cu[110] surface. Kern et al. attribute the formation of these striped phases to long-range repulsive forces between stripes. We have shown using Monte Carlo simulations and a 1-D Ising model that short range adsorbate–adsorbate interactions are capable of producing striped phases.
Comment on “Long-Range Spatial Self-Organization in the Adsorbate-Induced Restructuring of Surfaces: Cu\{110\}-(2x1)O”

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In a recent letter [1], Kern et al. reported that anisotropic Cu–O islands form striped periodic supergratings on the Cu\{110\} surface. Kern et al. attribute the formation of these striped phases to long-range repulsive forces between stripes [1]. We have shown using Monte Carlo simulations and a 1-D Ising model that short range adsorbate–adsorbate interactions are capable of producing striped phases.

In the work of Kern et al., the observed spacings between Cu–O islands depended upon the coverage and temperature, and were studied using scanning tunneling microscopy (STM) and He diffraction [1]. This work differed from earlier STM studies of this surface [2–5] in that the surface was annealed. The surface without annealing displays disordered “strings” of adsorbates along the <001> direction which are one atom wide in the <110> direction [2–4], or islands of condensed strings [5]. Kern et al. found that annealed surfaces have “stripes” in the <001> direction which were several atoms wide and of quite uniform width and spacing.

Our sole purpose here is to demonstrate that short range adsorbate–adsorbate interactions are sufficient to reproduce the qualitative features observed for this interesting surface. We do not seek to extract specific interaction energies. Starting from randomly adsorbed O atoms on a Cu\{110\} surface at elevated temperature, we allow each adsorbate to move typically 10^6 times while cooling at a constant rate to room temperature. The main difference in the interaction energies in these simulations and those used in Ref. [4] is that we have added a weak adsorbate–adsorbate attraction for the next nearest neighbor site along the <1\bar{1}0> direction as suggested in Ref. [5].

We find for our simulations that upon cooling from 640K to 293K we form fairly evenly spaced stripes. The numbers and spacings of these stripes depend upon the coverage, but are the same for multiple runs at a given coverage. We find similar results for lattices of 30x30 and 60x60 Cu atoms, as well as for asymmetric lattices, all with periodic boundary conditions. We find that from ca. 8x10^4 up to ca. 10^7 moves per adatom, we obtain identical results. Fig. 1 shows the simulated stripe
spacing (D) vs. coverage which is in qualitative agreement with the experimental data — Fig. 2a of Ref. [1]. The vertical scale of this curve varies with the interaction energies chosen.

Randomly adsorbing O atoms at 294K, then allowing the same number of steps as for high temperature adsorption, results in disordered strings of adatoms as for some of the previous STM data [2–4] and as for the previous simulations [4], and also results in some two-dimensional growth as seen by STM in Ref. [5]. However, starting with one perfect stripe several atoms wide, we find that at 294K in $10^7$ moves per adatom this island breaks up into the number of stripes found for the annealed surface at this coverage. We take this as evidence pointing toward the striped phases being equilibrium structures in our simulations.

By considering the strong adatom attraction in the <001> direction as the determining interaction in string formation, and the weaker adatom attraction in the <110> direction as determining the string-string interactions, we have modeled the problem as a 1-D Ising model. Even in this simple model equally spaced bands are obtained with spacings and widths vs. coverage and temperature [6], in qualitative agreement with Fig. 2 of Ref. [1].

The essential qualitative features observed in the measurements of Kern et al. [1] on the O/Cu{110} surface have been reproduced using simulations which include only short range interactions including a weak adatom attraction in the <110> direction. This is in contrast to the suggestion that long range forces need be invoked in order to explain this data [1]. Details of the calculations summarized above will be published shortly [6].

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REFERENCES


FIGURE CAPTION

FIG. 1. Stripe spacing (D) vs. O atom coverage on the Cu{110} surface after cooling from 640K to 294K while allowing ca. $10^6$ moves per adatom.