Comment on "Missing-Row Asymmetric-Dimer Reconstruction of SiC(100)-$c(4 \times 2)$"

In a recent Letter [1] Lu, Krüger, and Pollman report *ab initio* total energy pseudopotential calculations using the local density approximation (LDA) for β-SiC(001)-$c(4 \times 2)$ suggesting a missing-row asymmetric-dimer (MRAD) model. The latter is inconsistent with different classes of experiments and other *ab initio* LDA studies [2–6]. Previous experimental data are misinterpreted, making comparison with calculations misleading. Reference [1] hinges on the assumption of a 0.5 Si adlayer in contradiction with experiments. Computationally, there are open issues concerning the accuracy of LDA in the present case.

Based on STM, we have proposed a model of rows made of alternating up and down dimers (AUDD) within the row, reducing the surface stress [2]. This model is supported further by STM image simulations taking into account tunneling current and tip-sample distance [2] and by recent *ab initio* LDA calculations [5,6]. The AUDD model fully agrees with a "bare" Si-terminated surface, with no adlayer as established by quantitative experiments using different probes [4]. The MRAD model, at 0.5 Si adlayer on this Si-terminated surface [1], is inconsistent with these results. Instead, Lu *et al.* base their calculations on a photoemission study tentatively exploring possible higher Si coverages [7].

In Ref. [1], the simulated STM images are compared with our experimental topographs [2]. In the latter, the spots represent dimers [as confirmed by electron scattering quantum chemistry (ESQC) STM simulations] [2]. In Lu *et al.* STM image simulations [1], bright spots correspond instead to individual atoms. Their calculations use the Tersoff-Hamann approach known to depend on the altitude selected to represent the surface density of states [8]. First, the altitude is not specified and does not appear in Ref. [1] leading to a wrong comparison with our experimental scans with a real z scale [2]. Second, the density of states does not correspond to a constant STM current simulation imaging.

The AUDD model [2] explains very well the temperature-induced $c(4 \times 2) \Leftrightarrow 2 \times 1$ reversible phase transition at $\approx$700 K that we have established experimentally [3]. Calculations in Ref. [1] are performed at 0 K and cannot account for surface behavior at $\approx$700 K. To make the MRAD model consistent with such a phase transition, dimer breaking has to be invoked [1]; this does not occur for Si surfaces at $\approx$700 K [9] and is highly unlikely for refractory β-SiC(001) at such low temperatures.

Ab initio calculations used so far in trying to explain the β-SiC(001)-$c(4 \times 2)$ surface reconstruction are based on LDA [1,5,6]. While this approach has been very successful for bulk solids, surfaces, or molecules, there is no guarantee that it is always adequate. In fact, LDA is known to underestimate semiconductor band gaps—e.g., Lu *et al.* [1] found 1.1 eV instead of 2.34 eV for β-SiC. Sometimes LDA wrongly predicts a material to be metallic while it is in fact semiconducting. Such an error could have consequences for surface structures, which may be the case for β-SiC(001).

Spontaneous AUDD formation was found in our LDA cluster study [5], in excellent agreement with STM [2]. Because of finite sizes, cluster calculations tend to increase band gaps, which may be at the origin for finding AUDD [5]. We also performed pseudopotential plane wave calculations. Without any external stress, no AUDD are formed. To probe possible pseudopotential effects, we have performed all-electron LDA calculations with the same periodic model and a numerical localized basis. Starting from an AUDD geometry, the periodic model computations show a very flat energy hypersurface between an AUDD array and a geometry with all Si dimers in the same plane. This flatness is indeed consistent with the reversible semiconducting $c(4 \times 2) \Leftrightarrow$ metallic 2×1 phase transition [3], except that LDA may incorrectly favor the metallic state. Surface stress could lead to band gap opening. A structure corresponding to a nonmetallic state, i.e., AUDD, could be favored in a stressed system as indeed found by Catellani *et al.* [6]. We suggest that LDA overemphasizes the metallic state and thus does not predict AUDD in unstressed slab models.

To summarize, calculations in Ref. [1] are inconclusive and provide no convincing argument supporting the MRAD model [1]. The latter is ruled out by experiment.

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