Comment on “Carbon Atomic Chain Formation on the \(\beta\)-SiC(100) Surface by Controlled \(sp \rightarrow sp^3\) Transformation”

In a recent Letter, Derycke et al. [1] presented \(sp \rightarrow sp^3\) transformation on the carbon-terminated \(\beta\)-SiC(001) surface using scanning tunneling microscopy (STM). They claim that they have found evidence for the formation of carbon atomic one-dimensional (1D) chains consisting of alternatively buckled C-C atoms with a single-bonded \(sp^3\) structure (SSP3) and the structure has an extremely high thermal stability. However, no direct evidence for the \(sp^3\) bonding is provided in the Letter. In this Comment we show correct analyses of the STM images and we argue that the true stable structure on the C-terminated \(\beta\)-SiC(001) surface [2] is the triple-bonded C=C bridge dimer structure analyzed well in Refs. [3–5].

Figure 1(a) shows one of our high-resolution STM empty-states topographic images of the 1D structures formed at similar conditions as the Letter [1]. (i) We observe many triple lines of bright protrusions, as observed in Fig. 3 of [1]. This is inconsistent with the SSP3 model having a double line as its unit. The fraction of the total multiple line length over the total length of the double lines is \(~40\%\) for 1D coverage of 0.15 ML. (ii) The observed zigzag protrusions in the empty states are too wide to fit the location within each carbon atom, intervening extensively between the locations of C=C bridge atoms in the adjacent lines as shown in Fig. 1(a) [6]. In a topographic profile in Fig. 1(b), we observe that the peak of the empty-states protrusions on a SSP3 dimer is located 0.397 nm from the center of the SSP3 dimer with the FWHM of \(~0.49\) nm. The peak separation between the SSP3 dimer and the bridge dimer is only 0.121 nm. This indicates the carbon atoms on the 1D lines have hybridized orbitals with the adjacent bridge carbon atoms to form an extended empty state orbital. (iii) In contrast to the empty-states images, periodical lacks of protrusions are observed in the filled states images as shown in Fig. 1(c) and the image of the Letter. The typical periodicity is 0.92 nm, which is \(\frac{3}{2}\) times distance between the adjacent bridge dimer pairs. This corresponds to the defect density of \(~33\%\) on the 1D lines. Experimental features (i) to (iii) contradict the SSP3 model.

As observed in Fig. 1 of [1] and in our experiments, almost all 1D structures are found at step edges, with adsorbates [black arrows in Fig. 1(c)], or on broad large bright areas, whose origin is unknown. This suggests that the 1D chain formation is triggered not by the intrinsic surface stress [1] but by these extrinsic origins. Figure 1(d) shows an example of a very clean \(c(2 \times 2)\) surface perfectly covered by the bridge structure without the 1D structure with an extremely low adsorbate density of \(3 \times 10^{11} \text{cm}^{-2}\) [5]. The presence of such a clean bridge surface after a high temperature annealing is the strongest evidence of the stability of the C=C bridge structure. In our extensive experiments to improve the surface quality, we have found that the resultant perfectness in the bridge structure formation is improved drastically by reducing adsorbates and defects densities on initial Si-rich surfaces, the vacuum pressure in the annealing method, and impurity concentrations in the ethylene dose method. Further, no increase in the average length of the 1D structures has been found after the annealing, although the density of the 1D structure increases with increasing the density of the exotic origins. The limitation of the standing-alone length is the direct evidence of the higher thermal stability of the bridge structure than that of the 1D structure.

Last, it should be noticed that there is a consensus on the tripletility of the C-C bridge dimer bonding through the direct observation [3] and the theoretical calculations of the bonding length. So, the \(sp\) hybridization of the starting \(c(2 \times 2)\) surface is just a speculation at present, and further investigations are needed.

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[6] Details of the C=C structure will be published elsewhere.