

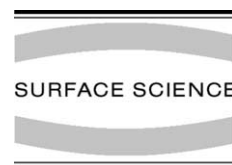


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Discussion

Comment on: “Formation of two dimension Ge cluster
superlattice on Si(1 1 1)-(7 × 7) surface”
[Surf. Sci. 506 (2002) L255] ☆

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In a recent Letter [1], Yan et al. reported on the adsorption of submonolayer Ge on Si(1 1 1)-(7 × 7), studied by in situ scanning tunneling microscopy (STM). The purpose of the present comment is to challenge the results presented in [1], and their interpretation. Indeed one of us (F. Rosei) has studied in detail the growth of Ge on Si(1 1 1) under various conditions [2–10], finding results that are in sharp contrast with what is reported in [1] by Yan et al.

In their introductory paragraphs, Yan et al. rightly point out that one way to achieve ordered arrays (for the self-organized growth of strained islands) is to use appropriate templates with periodic structures. In fact they then proceed to cite two relevant articles, which have demonstrated this possibility [11,12]. Actually, we believe it is quite inappropriate to consider the Si(1 1 1)-(7 × 7) reconstructed surface as a perfect template for the growth of Ge on Si, as they subsequently do. The Ge/Si(1 1 1) system has been studied quite extensively (for a recent review, see Ref. [13]) but to our knowledge only one recent work has demonstrated this concept [14], using *appropriate, specific* growth conditions.

On page 2 of their Letter, by discussing Fig. 1a and b Yan et al. claim that they observe Ge clusters on the 7 × 7 reconstructed Si(1 1 1) surface, and that, contrary to previous observations, their clusters have similar shape and size. First of all, we would like to point out that the quality and the overall resolution of the images reported in Fig. 1 from [1] is extremely poor, and therefore it is very hard to distinguish any significant feature in these two micrographs. Both images seem to display some kind of periodicity, but due to the low resolution we believe it is not possible to point (with reasonable certainty) to “clusters” in the images as the authors do, and that the periodicity in this case could very well be that of the 7 × 7 reconstruction or even of the 5 × 5 reconstruction, which is typical of a Ge/Si(1 1 1) wetting layer (even when incomplete) [5,15]. Consequently, we question their interpretation of these images, in which we are not able to identify the clusters with similar shape and size that Yan et al. claim to observe. A histogram reporting cluster dimensions (height and width) derived from analyzing many images could have demonstrated their presumed uniform size distribution, as was previously done by Masuda and Shigeta [14]. To better illustrate

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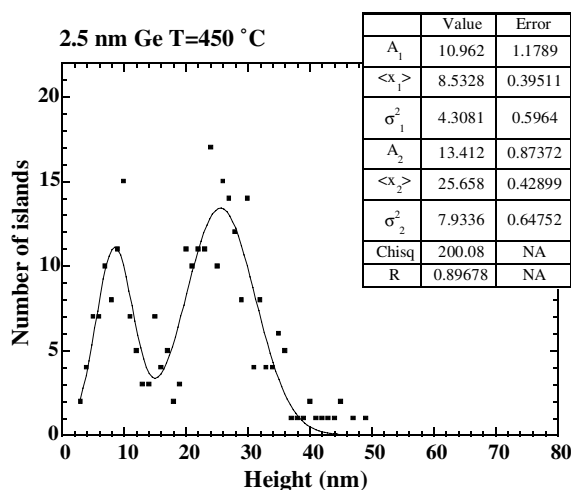


Fig. 1. Histogram of islands' height for $\theta = 2.5$ nm grown at $T = 450$ °C. Gaussian fits are also reported. The distribution is clearly bimodal, with values centered at $\langle x_1 \rangle = 8.5 \pm 0.4$ nm and $\langle x_2 \rangle = 25.7 \pm 0.4$ nm. From Ref. [2], with permission.

this point, we report in Fig. 1 below a histogram of the height distribution of 2.5 nm Ge deposited on Si(1 1 1) at $T = 450$ °C. Although the distribution is clearly bimodal, with two well-defined peaks centered at 8.5 and 25.6 nm, it is not possible to claim that the islands have a uniform size distribution. Actually the distribution may even be *trimodal*, with a third peak centered at about 43 nm, but unfortunately the statistics is not good enough to claim this with sufficient certainty. Generally speaking, we remark that self-assembled processes, including the present one, normally yield poor control over structure dimensions, unless growth parameters are appropriately optimized.

In an inset of the same Fig. 1 from [1], Yan et al. report a 2 D Fourier transform of the experimental image. They claim that the observed sixfold pattern confirms a macroscopic 2 D ordering of Ge clusters. Firstly, we would like to point out that the Fourier Transform (FT) is taken on Fig. 1a, the size of which is only 40×40 nm². Their claim that this confirms “macroscopic” ordering is therefore somewhat dubious, since the FT refers to such a small area of the sample surface. Secondly, in this context we expect that, in any case, the FT should exhibit a sixfold hexagonal pattern. This pattern is most likely induced by the substrate itself (which is a Si(1 1 1)-(7 × 7) surface), especially at such low coverages. To be conclusive, we believe that Yan et al. should report an average distance between clusters measured from STM images, and correlate it with the distances that can be deduced from the FT. At the same time, the presence of significant background noise which is clearly visible in the FT is an indication that the superlattice is probably not as ordered as claimed by Yan et al.

From previous studies [15] it is known that submonolayer deposition (0.45 ML) of Ge on Si(1 1 1) (albeit carried out at higher temperatures) yields a more intense reflection high energy electron diffraction (RHEED) 7×7 pattern with respect to the clean Si(1 1 1)- 7×7 . This is an indirect indication that (at least under these conditions) incoming Ge atoms initially fill vacancies and defects in the 7×7 reconstruction.

Further, on page 3 in [1] it is stated that the deposition of Ge atoms on Si(1 1 1)-(7 × 7) does not transform the Si surface until a few monolayers of Ge atoms have been deposited. We definitely do not understand what is meant by “transformed” in this context. We note however that at 0.75 ML Ge coverage (growth $T = 500$ °C), the RHEED pattern is still 7×7 , but STM images show 5×5 reconstructed “patches” [15]. This example shows that even below 1 ML, Ge deposition begins to “transform” locally the Si(1 1 1)- 7×7 surface. At about 3 ML ($T \geq 400$ °C) 3 D islands begin to form on the WL [15,16], whereas at lower temperatures (up to about 300 °C) this system is known to lead to amorphous structures

[17]. Crystalline 3 D islands may then be re-obtained by post-growth annealing at $T \geq 400$ °C for about 10 min.

On the same page, Yan et al. claim that, due to the low mobility of the incoming Ge atoms at this temperature (RT), the nucleation probability increases. This sentence is somewhat ambiguous. Both island nucleation and adatom mobility are activated processes so that the rate of island formation depends on a delicate balance between the activation energy for diffusion E_D , the Nucleation barrier E_N and the thermal energy kT . The nucleation probability should be roughly proportional to the square of coverage, and should decrease with decreasing temperature. Therefore at these low coverages (below 1 ML) we would expect that the nucleation probability would be very small. On the other hand, adatom mobility also decreases with decreasing temperature. It is therefore not obvious at all that a low mobility should increase nucleation probability, as stated in [1]. If the diffusivity is high, on the other hand, the probability per unit time that two atoms will meet on the surface is higher, and this will substantially increase the probability of nucleating clusters [7]. Perhaps what is meant here is that, when growing at low temperatures (as is observed for example on several metal on metal systems, albeit for growth at much lower temperatures), cluster nucleation is more likely to occur with respect to layer-by-layer growth, for example.

Further still, on page 4, Yan et al. state that their experiments strongly demonstrate that the initial growth of the amorphous Ge film on the Si(1 1 1)-(7 × 7) surface is not disordered as expected. We believe this sentence is quite contradictory. Firstly, if the film is amorphous, it obviously cannot be ordered. Secondly, given the fact that the film is incomplete (in the sense that it is less than one complete layer), it is quite hard to determine whether it is crystalline or amorphous. It only seems reasonable to say that it is disordered.

On the following page, Yan et al. now speculate that the Ge clusters may be amorphous, in contradiction with their previous statement on page 4. In the end we are not able to understand whether the authors believe their clusters to be amorphous or not. On the other hand, in the images presented on pages 4 and 5 (Figs. 3a, b and 5 in [1]), which are of slightly better quality and have a somewhat higher resolution than the ones reported in previous figures, it is not possible to see any ordered structure inside the clusters. Therefore our conclusion in relation to the images presented in [1] is that the clusters are indeed amorphous. This however is not particularly surprising, since it is consistent with what has been reported previously in the literature [17,18], i.e. that the growth of Ge on Si at room temperature leads to amorphous films.

Generally speaking, it is quite difficult to envisage that an ordered superlattice can form as a result of solid phase epitaxy. The claim that the clusters have similar shapes and sizes is not substantiated by the images presented. A statistical analysis of these clusters could have certainly helped in establishing this aspect. This type of analysis could have shown whether the clusters are indeed uniform in size, and it may have helped in understanding how many atoms the clusters are composed of.

Similarly, an X-ray diffraction experiment could help to establish the absence/presence of symmetry (i.e. order and uniform size) in this system. However, due to the very low coverage the overall signal may be too weak to provide a conclusive answer.

We would also like to draw attention to the scanning tunneling spectroscopy (STS) results reported by Yan and co-workers in Fig. 4 from [1]. These results are clearly inconsistent with the same measurements reported by the same authors (Fig. 4 from Ref. [19]) in a previous publication. In Fig. 4 from [19] STS spectra (dI/dV as a function of V) exhibit two peaks separated by a small gap of less than 1 eV, whereas in Fig. 4 from [1] the gap is more than 2 eV and the intensity of the first peak is much lower. This inconsistency however is not discussed or explained by Yan et al. in [1].

Finally, Yan et al. [1] stress the importance of their presumed ordered superlattice, for possible applications. Assuming that their observations (hopefully with better image quality and resolution) are reproducible, we do not believe that these preliminary results could lead to the development of any practical application. Any realistic device would require a much greater quantity of Ge to achieve the necessary active layer.

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