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Bottom-up approach for the nanopatterning of Si(001)

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The development of single-crystal semiconductor nanostructures, which – due to their atomic-like discrete energy levels – are promising candidates for improved laser applications, is a topic of intense research since the early 1990s.[1,2] A well-established procedure for the preparation of single-crystal quantum dots on semiconductor templates is film growth by the Stranski-Krastanow (SK) mode[3], where three-dimensional (3D) islands nucleate on top of a continuous wetting layer and relieve part of the misfit strain[4,5]. Prominent examples are Ge/Si(001)[1], SiGe/Si(001)[6], InAs/ GaAs(001)[7], PbSe/ PbEuTe[8], etc. Due to the statistical nature of the nucleation process the obtained quantum dot arrays typically are not very regular concerning their spatial arrangement and size distribution. For instance, in the case of the SiGe/Si(001) system pyramids, huts and domes are coexisting in a wide thickness range[9]. Therefore the spectral distribution is broad, although individual dots exhibit sharp resonances[10]. For the indirect band gap semiconductor Si the photoluminescence intensity is rather low. Therefore it is advantageous to develop small and uniformly sized SiGe quantum dots for increasing the island density.

Recently, the nanopatterning of substrates with a periodic superstructure has become a feasible alternative to SK growth as it promises a better control of the nucleation process, and thus improved spatial ordering and size uniformity for nanostructure growth. At present, top-down approaches are mostly employed for realizing patterned templates. For instance, a better spatial and size distribution has been obtained for SiGe quantum dots nucleating on the top plane of lithographically fabricated 1D and 2D mesa structures[11,12] due to the change in diffusion from a 2D to a 1D process. Furthermore, patterned templates have been prepared by intentionally generating a dislocation network[13] or by introducing regular arrays of defect centers via focused ion beam lithography[14]. However, whereas spatial and size distribution is improved by these preparation techniques, the achieved quantum dot density is very low (10^8–10^9/cm^2).

Here I report on a bottom-up approach for nanopatterning of the Si(001) surface. It is based on the selforganized intrinsic (2×*n*) superstructure of Si(001), which consists of extended stripes separated every few nanometers by a dimer-vacancy line (DVL) (Fig. 1). The Si(001)(2×*n*) structure is known for a long time[15] and was originally attributed to the effect of transition-metals, especially Ni contamination[16]. However, as convincingly shown by Men et al.[17] and others[18,19] the (2×*n*) exists also on the uncontaminated Si(001) surface and forms – although only occasionally – after repeated flashing of the sample to 1200 °C and subsequent quenching. A (2×*n*) structure has also been obtained, e. g., after ion-bombardment of Si(001)[20] or by etching with reactive gases[21].

Our combined scanning tunneling microscopy (STM) and Monte Carlo (MC) study of Si(001) reveals that the formation of the (2×*n*) superstructure depends strongly on the Si coverage in the surface layer. Both, experiments (Fig. 1) and simulations (Fig. 2), show that the (2×*n*) formation is restricted to a narrow coverage window. Whereas for Si-coverages lower than 0.5 monolayers (ML) small and randomly distributed 2D Si islands are observed, vacancy islands form at coverages above 0.85 that appear as randomly distributed nanodefects in otherwise (2×1) reconstructed terraces. The formation of the (2×*n*) takes place only in the coverage range of about 0.6–0.8 ML.

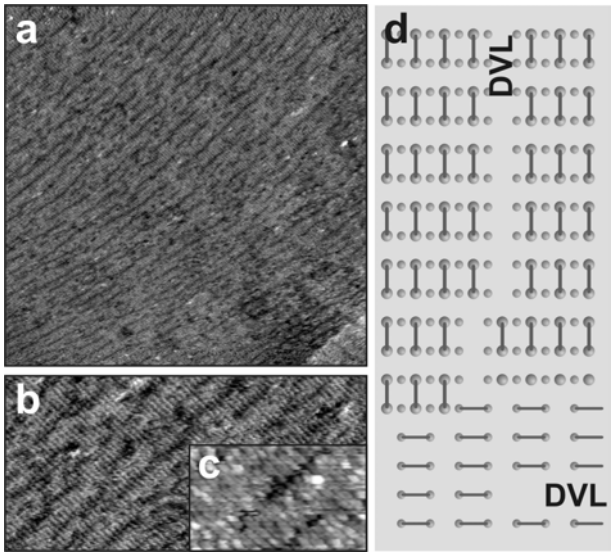

Fig. 1 (2×*n*) superstructure forming on the Si(001) surface after deoxidation at temperatures of 880–900 °C: a) 100× 100 nm^2 STM top view showing an extended (2×1)-reconstructed terrace decorated by a periodic pattern of elongated DVLs; U_{tip} = +2.0 V, I_t = 0.6 nA; b) and c) close-ups of a). d) Sphere model illustrating the (2×*n*) structure of Si(001).

Figure 1 a displays a typical STM image of a Si(001) sample prepared by a procedure described in detail in Ref. 22. The surface exhibits extended (2×*n*) reconstructed terraces containing a high density of DVLs. The DVLs run perpendicular to the dimer rows and are rotated by 90 ° on adjacent terraces in accordance with the geometry of the Si crystal lattice. The DVLs have organized to a quite regular stripe pattern with an average stripe distance of about 5 nm

and the length of the DVLs ranging between 5 and 20 nm. STM investigations at different sample positions indicate that the stripe pattern has formed uniformly over the entire sample. The long-range order of the stripe pattern is further corroborated by the LEED investigations which reveal relatively sharp (2×*n*) superstructure reflections in addition to the (2×1) pattern. From the LEED pattern a periodicity

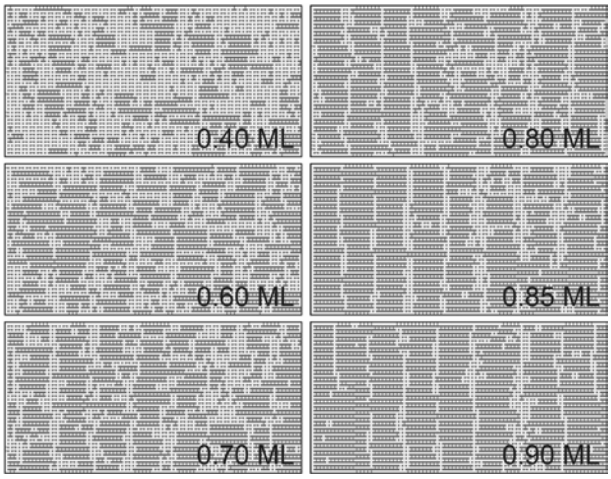

Fig. 1 Sequence of MC simulations, where a total of 1 ML was deposited atom-by-atom onto a completely filled (2×1) reconstructed surface of Si(001). After an atom was deposited each of the surface monomers and dimers had 1560 opportunities to hop to a free neighboring site; for details see Ref. 22.

of 5 nm is determined in good agreement with the average stripe distance observed by STM.

The annealing temperature of the Si)001) sample after oxygen desorption plays a decisive role for the development of the (2×*n*) reconstruction. If it is too low, ordering processes are suppressed. If it is too high, the dimer vacancies diffuse to the step edges and annihilate there. The experiments indicate that a temperature between 900–920 °C is required for oxide desorption, whereas the optimum temperature for a subsequent annealing is lower and lies at about 880 °C. For annealing at 1100–1200 °C – as used for oxygen desorption in the conventional preparation of Si(001) – the MC simulations in agreement with experiment yield (2×1)- reconstructed terraces with only a small number of missing dimers but without the formation of DVLs. The narrow vacancy concentration range explains the only occasional appearance of the (2×*n*) structure in previous quenching experiments, which obviously are not reproducible enough for establishing a defined vacancy concentration in the surface layer. In that respect sputtering and etching methods certainly are more reliable and moreover easier to control, but a detrimental surface contamination by impurities in the sputtering gas or the etching solution cannot be excluded.

As indicated by the MC simulations (Fig. 2) and recently confirmed experimentally, deposition of Si onto the oxygen-desorbed surface turns out to be a very promising method for adjusting the dimer vacancy concentration in the surface layer in a well-defined manner. It therefore seems to provide a feasible procedure for the nanopatterning of the Si(001) surface as a template for future quantum dot growth, as presently under investigation at our institute.

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