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Strain-induced self-assembly of Ge nanodashes, nanodumbbells, and dot chains on Si(001)

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We investigate the growth of self-assembled Ge nanostructures on top of embedded Ge nanowires on Si(001) substrates. Ge nanostructures, such as nanodashes, nanodumbbells, and dot chains are observed simply by tuning the growth temperature and thickness of the Si spacer between the Ge layers. The self-assembly process is governed by the surface strain fields generated by the embedded Ge nanowires and is well-described by our theoretical calculations. The catalyst-free and horizontal growth of such Ge nanostructures directly on Si(001) is attractive for investigating exotic transport properties through Si/Ge-based quantum devices. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4824121]

The formation of defect-free three dimensional islands (quantum dots (QD)) via the Stranski-Krastanov (SK) growth mode, observed more than 20 years ago in lattice mismatched semiconductor heteroepitaxy, triggered overwhelming interest in fabricating OD electronic and optoelectronic devices.^{1–7} A particularity in strained epitaxial layers is the ability to fabricate QD layer stacks separated by wellcontrolled spacer layers. During deposition of such multilayers, the surface strain fields induced by the embedded QDs significantly influence the formation of the QDs at the growth front. For instance, vertical alignment of quantum dots was observed in systems of weak elastic anisotropy, including Ge/Si^{8,9} and InAs/GaAs,¹⁰ while anticorrelation and a face-centered cubic alignment of quantum dots were found in systems of strong elastic anisotropy, such as CdZnSe/ZnSe¹¹ and PbSe/PbEuTe,^{12,13} respectively. Fine tuning of the spacer layer thickness and growth temperature has led to the formation of quantum-dot molecules on top of embedded individual quantum dots in the Ge/Si system.¹⁴

The formation process of stacked nanostructures is crucially affected by the strain fields on the spacer surfaces induced by the embedded quantum dots.^{8,15,16} This was firstly demonstrated in weak anisotropic systems using a simplified model, where the anisotropy was neglected and the embedded three-dimensional island was modeled as a point force dipole of zero-dimension.⁸ In this model, the strain profile on the spacer surface is characterized by a single maximum of tensile strain above the center of the embedded island. The tensile strained areas are energetically favorable for nucleation and growth of surface islands. Thus, vertical alignment of stacked islands was observed, as demonstrated in many experiments.^{8–10} Later, the embedded island was modeled as a 3D rectangular cuboid and an analytical expression for the surface strain distribution was obtained by taking the integral of the point force dipole over the rectangular cuboid.¹⁶ Based on this model, calculations revealed that an embedded Ge island in Si generates four maxima of tensile strain on the (001) surface above its four corners, given that the base of the embedded island is much larger than its height and the thickness of the spacer layer.¹⁶ As a result, four islands formed in the regions of the four strain maxima over a certain range of relevant growth parameters.¹⁴

The surface strain profile depends on the shape and size of the embedded nanostructures and the spacer thickness.¹⁶ Different surface strain profiles result in different stacking and alignment phenomena leading for instance to the formation of lateral quantum dot molecules.¹⁴ Very recently, we have reported that under certain conditions, SK growth leads to the formation of one-dimensional (1D) Ge/Si nanowires (NWs) (or quantum wires) on standard Si(001) surfaces.¹⁷ Highly uniform {105} facetted Ge NWs with a height of only three unit cells and a length of up to two micrometers have been obtained simply by in-situ annealing of the deposited 2D Ge wetting layer (WL) under ultra-high vacuum conditions.¹⁷ The extension of SK growth from 0D quantum dots to 1D NWs provides possibilities for creating different strain profiles on spacer surfaces, which allow fabricating various types of coupled quantum structures.

In this letter, we report about the self-assembled growth of Ge nanodashes, dumbbells, dot chains, and matchsticks on top of embedded Ge NWs by varying the growth temperature and Si spacer thickness. As schematically outlined in Fig. 1, initially, Ge NWs are obtained by *in-situ* annealing the deposited Ge WL,¹⁷ followed by the Si spacer layer. The sample is completed by the second Ge nanostructure growth on top of the spacer layer.

Samples were grown by solid-source molecular beam epitaxy (MBE) at a base pressure of 5.0×10^{-11} mbar. The growth rates of Ge and Si are 0.056 and 1.0 Å/s, respectively. We initially deposit a 4.4 monolayer (ML) Ge WL at a substrate temperature of 570 °C and then keep the substrate temperature at 560 °C for 3 h *in-situ* annealing, as detailed in Ref. 17. After annealing, Ge NWs with a uniform height of 1.9 nm and a length up to 1 μ m are obtained. Their surface is bounded by four {105} facets, indicating that their base width is 10 times of the height, i.e., 19 nm.^{2,17} Such Ge NWs

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FIG. 1. Schematic growth sequence for obtaining capped Ge NW on Si(001) for subsequent Ge nanostructure growth on the top (the wetting layer is not plotted).

were then capped with Si spacer layers. The growth temperature for the Si spacer varied from 480°C for the first 2 nm to 530 °C for the rest of the layer. The lower growth temperature of 480 °C is used to minimize the Si-Ge intermixing during Si growth. As demonstrated in Ref. 18, the Ge {105} facets are preserved if the growth temperature of the Si spacer layer is kept at 500 °C or lower. In turn, the increased growth temperature of 530 °C is used to flatten the surface. After the Si spacer layer growth, top Ge layers of 3.5, 3.8, and 4.3 ML are deposited on the Si spacer thicknesses of 3.5, 6.0, and 10.0 nm, respectively. The reduced Ge coverage, compared with the initial 4.4 ML in the bottom layer, was chosen due to a reduced WL thickness caused by surface strain fields induced by the embedded Ge NWs-similar to what has previously been reported for Ge/Si island multilayers.¹⁹ After the top Ge layer growth, samples are immediately ramped down to room temperature without any further annealing. Two sets of samples are investigated. The first set consists of Ge layers grown at different temperatures from 540 to 620 °C on a 6 nm thick Si spacer layer. For the second set, we fix the growth temperature of the top Ge layer at 620°C but vary the Si spacer layer thickness from 3.5 to 10 nm.

Figures 2(a)–2(d) show atomic force microscopy (AFM) images of self-assembled nanostructures obtained after the deposition of 3.8 ML Ge on a 6 nm thick Si spacer layer at 540, 560, 580, and 620 °C, respectively. At a growth temperature of 540 °C, we observe strictly aligned and closely spaced nanodashes with lengths of tens of nanometers. They are orientated along either of the two $\langle 100 \rangle$ directions, identical to the orientation of the embedded NWs.¹⁷ At an

increased temperature of 560 °C, the dashes are less abundant and mostly substituted by broken nanowires (Fig. 2(b)). The space between the dashes and the broken NWs is only a few nanometers. For a growth temperature of 580 °C, the formation of complete NWs is retained (Fig. 2(c)). We note that, although pure Ge was deposited, the dashes do not consist of pure Ge, which we predominantly attribute to strainfield induced intermixing.²⁰ Furthermore, the height of the dashes/nanowires gradually increases from 1.8 nm to 2.6 nm with higher growth temperatures due to increased Si-Ge intermixing at elevated temperatures.²¹ When we further increase the growth temperature to 620 °C, dumbbells are observed, each of which consists of a NW with two additional islands positioned at its ends (Fig. 2(d)). There are two kinds of dumbbells. For one kind, the two islands are square or rectangular based huts with a height of about 6 nm and are connected with the wire, as marked by the red dashed rectangle. The other kind incorporates two dome islands²² which are separated from the NW by a small gap. The domes have a diameter of about 75 nm and a height of 15 nm, and the wires have a height of about 4.5 nm. To explain these results, it is useful to recall that for sufficiently thick spacer layers surface islands can generate compressive strain around their bases much larger than the tensile strain induced by the embedded NWs. Because the dome islands are larger than the huts, they produce higher compressive stress in their vicinity²³ and therefore separate from the NW easier than the smaller and shallower huts.

The tensile strain on the surface depends on the spacer layer thickness. It is therefore interesting to study to what



FIG. 2. AFM images showing Ge nanostructures on top of embedded Ge NWs. Nanodashes (a), broken wires (b), wires (c), and dumbbells (d) are obtained at different Ge growth temperatures of 540, 560, 580, and 620 °C, respectively. The Si spacer layer thickness is 6 nm. (e) and (f) show dumbbells and dot chains obtained at 620 °C with different Si spacer thicknesses of 3.5 and 10 nm, respectively. The dashed rectangles in (e) and (f) mark dumbbells where dots and wires are connected. Scale bar: 250 nm.

extent the spacer layer thickness influences island formation. We keep all parameters of the sample shown in Fig. 2(d) constant except for the spacer thickness. Figs. 2(e) and 2(f) show AFM images of the nanostructures after the deposition of 3.5 and 4.3 ML Ge at $620 \,^{\circ}$ C on a Si spacer layer with a thickness of 3.5 and 10.0 nm, respectively. In Fig. 2(e), we see that domes now tend to connect to the NWs, as marked by the dashed red rectangle, where on one end the NW is connected with a dome and on the other end with a hut. The connection of the dome with the NW is attributed to the increased tensile strain induced by the embedded NWs at a reduced spacer layer thickness. In turn, for a larger spacer thickness, a decreased tensile strain leads to the formation of 1D dot chains (Fig. 2(f)). We shall discuss the strain distributions for different spacer thicknesse later.

Figures 3(a)-3(f) show 3D AFM images of some of the intriguing nanostructure arrangements obtained by our growth procedure. In these configurations, the wires and islands are connected or closely spaced, as quantified by AFM linescans passing through the centers of the nanostructures (Figs. 3(g)-3(k)).

Let us now provide an explanation for the observation of the different nanostructure configurations based on surface strain field calculations. As discussed above, the NW is bounded by four {105} facets and has a triangular crosssection. There is still no analytical expression available for the strain field generated by such structures. We model the embedded NW as a summation of ultra-thin rectangular cuboids (sheet) along the growth direction. For an embedded rectangular cuboid with a volume $V = l \times w \times h$ (where *l*, *w*, and *h* are the length, width, and height of the rectangular cuboid, respectively) at a spacer layer thickness *d*, the strain on the spacer layer surface (*x*, *y*, 0) is given by¹⁶

$$\varepsilon = \frac{\varepsilon_0 (1+v)(1-2v)}{\pi (1-v)} \times \sum_{i,j,k=1}^2 (-1)^{i+j+k} \tan^{-1} \frac{x_i y_j}{z_k \sqrt{x_i^2 + y_j^2 + z_k^2}}$$

where $x_i = x \pm l/2$, $y_j = y \pm w/2$, $z_1 = d$, and $z_2 = d - h$. Parameters v and ε_0 are Poisson's ratio of the spacer layer material and the lattice misfit with $\varepsilon_0 = (a_e - a_s)/a_s$, respectively. For Ge/Si, $a_e = 0.5656$ nm, $a_s = 0.5431$ nm, and v = 0.218.

Figure 4(a) shows the strain profile on the spacer layer surface induced by an embedded Ge NW of l = 1000 nm and w = 20 nm (2 nm in height) with d = 6 nm. The NW induces tensile strain with two maxima on its two ends, as further shown by the line scans of the surface strain along the length and width directions passing through (0, 0, 0)(red lines in Figs. 4(c) and 4(d), respectively). The tensile strained regions are energetically favorable for the nucleation of Ge adatoms and eventually lead to the formation of Ge dumbbell structures. In Fig. 4(b), we schematically illustrate the occurrence of a dumbbell on top of an embedded Ge NW.

We further calculate the surface strain for different Si spacer thicknesses (Figs. 4(c) and 4(d)). For the smallest Si spacer of 3.5 nm, we calculate the largest tensile strain and reveal two clear maxima at both ends, leading to the formation of dumbbells even without any separation between the dome and the NW (Fig. 2(e)). For an increased Si spacer of 10 nm, the reduced and almost flat strain results in the formation of chains of homogeneously sized dome-shaped islands (Fig. 2(f)). Indeed, at a spacer thickness of 30 nm, we calculate a negligible surface strain, and, experimentally, observe randomly distributed dome islands (not shown). The surface



FIG. 3. (a)–(f) 3D AFM images showing different Ge nanostructures. (g)–(k) The corresponding AFM line scans passing through the centers of the nanostructures shown in (a)–(d) and (f), respectively. For structures shown in (a)–(e), the Si spacer layer thickness is 6 nm and the Ge growth temperature is 620 °C. For (f), the Si spacer layer thickness is 3.5 nm and the Ge growth temperature is 600 °C.



FIG. 4. (a) Surface strain profile on a 6 nm thick Si spacer layer induced by an embedded Ge NW with a width of 20 nm and a length of 1000 nm. (b) Schematically shows a Ge dumbbell on top of an embedded Ge NW. (c) and (d) Line scans of surface strain distribution induced by an embedded NW (width: 20 nm, length: 1000 nm) at different spacer layer thicknesses in the cross-section (c) and the central (d) plane of the embedded NW.

strain also depends on the lateral size of the embedded NWs: The larger the width (height) of the NW, the larger the induced surface strain. An embedded tapered NW¹⁷ therefore induces a tapered strain field on the spacer surface, which in turn leads to the formation of asymmetric nanostructures such as the matchsticks observed in Fig. 3(f).

Finally, we qualitatively explain why different types of structures are observed at different temperatures (Figs. 2(a)-2(d)). Reducing the growth temperature leads to a reduced surface diffusion length of the Ge adatoms and the energetically preferential area (on top of embedded NWs) cannot accumulate enough material. The lower the growth temperature, the smaller the surface diffusion length and the less Ge material is accumulated. This is why first small dashes, then larger broken wires and finally complete NWs and dumbbell structures are observed with increasing temperature (Figs. 2(a)-2(d)).

In summary, we have shown that Ge nanostructure arrangements, such as dashes, dumbbells, and dot chains, on Si(001) can be obtained simply by varying the growth temperature and the Si spacer layer thickness. Experiments were interpreted by calculating the strain profiles on the spacer layer surface induced by embedded NWs. In such Ge nanostructures, different components (huts, wires, and domes) are either connected or closely spaced, providing intriguing possibilities to investigate the fundamental physics, such as tunnel coupling and exotic quantum states. For instance, recently, it was suggested that the Majorana-fermion zero mode could be detected in a system consisting of a quantum dot coupled to the end of a semiconductor nanowire, as shown here in Fig. 3(c).²⁴ In addition, the closely spaced islands can induce

large compressive strains in the Si regions between the islands and tensile strain above the islands when they are overgrown with Si. The compressive and tensile strain in Si channels can significantly enhance the hole^{25,26} and electron mobility,²⁷ respectively. We therefore expect that the monolithic growth of such advanced Ge nanostructure configurations directly on Si(001) opens up possibilities for both fundamental experiments and fabrication of technologically relevant Sicompatible Ge nanodevices.

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