Growth and characterization of two- and three-dimensionally ordered quantum dots

Zhenyang Zhong, J Novak, J Stangl, T Fromherz, F Schäffler, and G Bauer
Institut für Halbleiter- and Festkörperphysik, Universität Linz, A-4040 Linz, Austria
E-mail: guenther.bauer@jku.at

Abstract Two dimensionally (2D) periodic pit structures are obtained by a lithograph technique on Si (001) substrate. Under proper growth conditions, the pit-pattern can be reserved after a thick Si buffer growth although the depth and the geometrical profiles of the pits are changed. During Ge deposition, GeSi islands preferentially grow at the pit bottoms, resulting in 2D ordered islands. In addition, the size homogeneity of these ordered islands is significantly improved. A kinetic model is proposed to qualitatively explain these results. By growing multilayer GeSi islands separated by thin Si spacers, three-dimensionally ordered islands or island crystal can be realized on pre-patterned substrates. These ordered islands helps to analyze the properties of a single island and island ensemble.

1. Introduction
Self-assembled islands during heterostructure growth have demonstrated a feasible routine to fabricate nanostructure materials or quantum dots. Enormous efforts have been devoted to the growth and the characterization [1] of those nanoscale islands for their promising device applications. However, the random positions and the broad size distribution of self-assembled islands on flat substrates without any pre-processing have not been significantly improved. Since different island size and/or different interplay between islands can change the properties of the islands [2], those disadvantages hamper the device applications based on a single island in integration and on island ensemble. Two- or even three-dimensionally ordered islands can be the solution to that problem.

Due to the simplicity and the sophisticated integration technique on Si, Ge/Si system has been an interesting subject of self-assembled islands [2-17]. On normal flat substrates, GeSi islands are always spatially random, and have a broad size distribution and often exhibit two types of shape, pyramid- and dome-like, simultaneously [9,13]. On vicinal substrates, quasi-periodic templates induced by step-bunching during buffer layer growth can lead to locally ordered islands[14,15]. By growing multilayer GeSi islands, locally ordered islands can be realized in the upper layers [5]. GeSi islands can also be aligned on the surface above a buried dislocation network [16]. In addition, local ordering can be realized in the case of densely arranged GeSi islands due to the elastic interaction among neighboring islands [17].

In order to obtain long-range ordered islands, substrates are always pre-processed. A pattern-assisted micro-step-network gives rise to the ordered islands growth [18]. Well-ordered islands arranged in a hexagonal lattice can be fabricated on latex nano-spheres masked Si substrates [19]. Recently, a combination of lithography technique and self-assembly is exploited to obtain long-range ordered islands. Two-dimensionally (2D) periodic windows in SiO\(_2\) masks fabricated by a lithography technique can result in 2D ordered islands due to selective epitaxy growth [20,21]. In the following,
we describe the fabrication of 2D pit-patterned pure Si (001) substrates without any mask. The islands preferentially grow in pits, favoring a long-range ordering. Meanwhile, these ordered islands manifest a much better size homogeneity than those on flat substrates. A simple kinetic model is proposed to qualitatively explain these phenomena. Moreover, multilayer GeSi islands grown on 2D pit-patterned substrates under proper conditions can yield island crystals, whose three-dimensional ordering was characterized by x-ray diffraction in reciprocal space and a corresponding simulation.

2. Experiments
Patterned Si (001) substrates were fabricated by holographic [22,23] or electron beam lithography. 2D pit-patterns with periodicities in the sub-micron range were generally obtained after reactive ion etching (RIE), as shown in figure 1a. After chemical cleaning and a thermal treatment at 900 °C, a Si buffer layer (>100nm) grows prior to Ge deposition by solid-source molecular epitaxy. These processes are crucial to preserve a pattern on the surface, and to eliminate surface roughness and/or damages induced RIE. In our cases, the Si growth rate is kept to be 0.5 Å/s. During buffer layer growth, the growth temperature starts at a low value, e.g. 450 °C, and is terminated at a high one, e.g. 650 °C. The pits become shallower, and their geometrical profiles will be changed after a buffer layer growth. To obtain desired pits for the subsequent GeSi island growth, it is necessary for different pit depth to adjust the growth conditions involving temperature, thickness and growth rate. For example, on the patterned substrate shown in figure 1a, a 130 nm Si was grown with ramping temperature from 450 °C to 550 °C, as shown in figure 1c. In the inset of figure 1c, a sharp ‘cross’ in surface orientation map (SOM) [24] clearly demonstrates the inverted-pyramid-like pits with dominant {1 1 n} facets. We also observe inverted-dome-like pits, as shown in figure 1d. The pit sidewalls are mainly composed of {1 1 3}, {15 3 23} and {1 1 9}, as denoted respectively by ‘1’, ‘2’ and ‘3’ in the SOM in the inset of figure 1d. In general, a higher growth temperature and/or a thicker buffer layer result in shallower pits and different shape of pits. We found that the island nucleation was affected by the geometrical profiles of pit as well as the growth conditions [25]. Therefore, it is necessary to investigate the actual dependence of the pit depths and their shapes on the growth conditions in further studies. Ge was then deposited on these pre-patterned substrates at an effective rate of ~0.04 Å/s at a temperature of more than 600 °C. For multilayer samples, GeSi islands
were separated by thin Si spacers [26]. In all these cases, although pure Ge was deposited, the islands are actually composed of GeSi alloy due to Ge-Si intermixing during growth at high temperature. The surface morphologies of samples are obtained by atomic force microscope (AFM). Multilayer GeSi islands were also characterized by transmission electron microscopy (TEM), x-ray-diffraction (XRD) and photoluminescence (PL) measurements.

3. Results and discussions

Figure 2a shows the surface morphology of one sample with 5 MLs Ge deposition at 650 °C on a patterned substrate with a 2D pit-pattern of periodicities 400 nm along <110> directions. Apparently, all islands are dome-like and are arranged in a square lattice, demonstrating a long-range ordering. The zoomed-in AFM image (amplitude) in figure 2b indicates that all islands locate at the bottom of pits. Around islands, ripple structures composed of \{105\} and \{001\} planes [25] are distinguishable. The reference islands grown on a flat substrate under identical conditions are shown in figure 2c. Some pyramid-like islands, as denoted by a white arrow in figure 2c, are observed as well as primarily...
The other interesting feature of islands on patterned substrates is their remarkably improved size homogeneity, as demonstrated by the distributions of island height in figure 2d (on the patterned substrate) and in figure 2e (on the flat substrate, the pyramid-like islands are not included.). These results are consistent with previous published ones [23].

The above features of ordered islands are associated with the preferential positioning of islands at pit bottoms. This has been confirmed by arrangements of islands in 2D lattices with non-orthogonal unit vectors, e.g. in a parallelogram lattice on patterned substrates [23]. Actually, the pit bottom is not the energetically favourable site for compressively strained GeSi islands. At too low temperatures the island formation is essentially not affected by the surface modulation [27]. In addition, at a high growth temperatures (> 600°C) the islands at pit bottoms always nucleate earlier and finally have larger volumes in comparison with those on flat substrates [23]. All these phenomena indicate that growth kinetics plays an important role in the island growth on patterned substrates.

Given that pit sidewalls are mainly composed of steps, and the activation barrier for ad-dimers or ad-atoms migrating downward is higher than upward over steps [27], a net flux of ad-dimers or ad-atoms from top terraces to pit sidewalls and at pit sidewalls downwards to the pit bottom can be formed during Ge deposition. As a result, aggregation of Ge at pit bottoms occurs, facilitating island nucleation and formation there. The alternative explanation is related to a possible minimum surface chemical potential at the pit bottoms [28], which drives the deposited Ge to aggregate there. Based on the this reasoning, it is reasonable to assume that only Ge deposited within a “pit unit cell”, as denoted by a white box in figure 2b, can possibly contribute to the islanding at the bottom of the corresponding pit. Accordingly, uniformed islands can be easily formed at pit bottoms within “cells” having the same areas, which collect the same amount Ge for each island. On the other hand, under proper growth conditions Ge is partially incorporated at pit sidewalls, particularly at the concaved corners between neighboring sidewalls [25]. Ripple structures can then evolve there to efficiently relax misfit strain. With sufficient Ge deposition, even secondary ordered islands might be formed on the top corners of inverted-pyramid like pits [25].

Multilayer GeSi islands separated by thin Si spacer layers have also been grown on 2D pit-patterned substrates [22, 26]. Based on the AFM images of uncapped islands in the top island layer [22, 26], we clearly observe that the lateral ordering and the size homogeneity of islands in each subsequent layer of multilayer islands samples are reserved on patterned substrates. Such lateral ordering in each layer, as well as the vertical ordering, of islands in one multilayer sample (13 periods, Si spacer layer thickness 25 nm, growth temperature for the Ge islands: 650°C, 6 ML; Si spacer layers grown at 550°C – 650°C, for further details see description of sample “A” in Ref. 26) has been confirmed by XRD reciprocal space maps. Such a scattered intensity distribution is shown in figure 3 around the (2 2 4) Bragg point in reciprocal space. The laterally periodic stripes along the vertical reciprocal space coordinate Q along the [001] direction are due to the small periodicity along growth direction in figure 3 and clearly demonstrate the rather perfect in-plane long-range ordering. The vertical ordering is demonstrated by the vertical satellites (VSn, n=+1, 0, -1, -2), whose vertical positions are indicated by white arrows. The (2 2 4) Bragg point of the Si substrate is also indicated by a white arrow. Simulations of the scattered intensities based on a kinematical approximation of diffuse scattering were done by Novak et al. [29] and gave information on the shape, the Ge content and the strain fields of the buried islands.

The vertical alignment of islands has also been confirmed by TEM. As an example such a cross-sectional TEM image is shown for a three-island layer sample (sample B, in Ref. 22), as shown in figure 4. Recently, such a vertical alignment was demonstrated by vertical pillar structures composed of periodic GeSi/Si, which were obtained by selective etching of Si between islands (mainly in-plane) from multilayer islands grown on a patterned substrate [30]. This vertical alignment has been explained in terms of local minimums of surface chemical potential just above buried islands. Accordingly, lateral 2D ordering of islands in the first layer on patterned substrates is transferred to the subsequent layers and 3D ordered islands can then be obtained. In addition, due to a periodic strain...
distribution on the surface of a Si spacer layer, we can argue that the surface of spacers is also composed of periodic “unit cells”. Furthermore, by optimizing the growth conditions and/or the amounts of deposited Ge [26] islands in different layers can also have the same size. Consequently, 3D-island crystals can indeed be formed. The size homogeneity of islands in sample in the 13-period 3D island crystal is demonstrated by well-separated narrow PL peaks from islands, as denoted by ‘islands’ in figure 5.

4. Summary
In summary, 2D and 3D ordered islands can be fabricated on periodically patterned substrates. The lateral ordering of the islands also helps to improve their size homogeneity. Our results also suggest that the sites of islands on patterned substrates are well defined and can be addressed even after Si capping. This facilitates the characterization of a single island and the device design based on single islands and their subsequent integration. Such ordered islands can also help to analyze the effect of interaction between islands on their properties, e.g. as a function of their distance.
Acknowledgement
This work was supported by the EU network NOE “SANDiE”, Brussels, and by the Austrian Science Fund, Vienna (SFB025).

References
[1] Stangl J, Holy V, and Bauer G 2004 Rev. Mod. Phys. 76 725
[2] Pchelyakov O P., Bolkhovityanov Yu B, Dvurechenski A V, Sokolov L V, Nikiforov A I, Yakimov A I, and Voigtländer B 2000 Semiconductors 34 1229
[3] Mo Y W, Savage D E, Swartzentruber B S, and Lagally M G 1990 Phys. Rev. Lett. 65 1020
[4] Schmidt O G and Eberl K 2000 Phys. Rev. B 61 13721
[5] Tersoff J, Teichert C, and Lagally M G 1996 Phys. Rev. Lett. 76 1675
[6] Vailionis A, Cho B, Glass G, Desjardins P, Cahill D G, and Greene J E 2000 Phys. Rev. Lett. 85 3672
[7] Sutter P, Zahl P, and Sutter E 2003 Appl. Phys. Lett. 82, 3454
[8] Ross F M, Tromp R M, and Reuter M C 1999 Science 286 1931
[9] Medeiros-Ribeiro G, Bratkovski A M, Kamins T I, Ohlberg D A A, Williams R S 1998 Science 279 353
[10] Zhang Y, Floyd M, Driver K P, Drucker J, Crozier P A, and Smith D J 2003 Appl. Phys. Lett. 80 3623
[11] Zhong Z, Stangl J, Schäffler F, and Bauer G 2003 Appl. Phys. Lett. 83 3695
[12] Kamins T I, Medeiros-Ribeiro G, Ohlberg D A A, and Williams R S 1999 J. Appl. Phys. 85 1159
[13] Rudd R E, Briggs G A D, Sutton A P, Medeiros-Ribeiro G, and Williams R S 2003 Phys. Rev. Lett. 90 146101
[14] Zhu J, Bruner K, and Abstreiter G 1998 Appl. Phys. Lett. 73, 620
[15] Lichtenberger H, Mühlberger M, and Schäffler F 2005 Appl. Phys. Lett. 86 131919
[16] Shiryaev S Y, Jensen F, Hansen J L, Petersen J W, and Larsen A N 1997 Phys. Rev. Lett. 78 503
[17] Floro J A, Chason E, Sinclair M B, Freund L B, and Lucadamo G A 1998 Appl. Phys. Lett. 73 951
[18] Ogino T 1997 Surf. Sci. 386 137
[19] Li N, and Zinke-Allmang M 2002 Jpn. J. Appl. Phys. 41 4626
[20] Jin G, Liu J L, and Wang K L 2000 Appl. Phys. Lett. 76 3591
[21] Kim E S, Usami N, and Shiraki Y 1998 Appl. Phys. Lett. 72 1617
[22] Zhong Z, Halilovic A, Fromherz T, Schäffler F, and Bauer G 2003 Appl. Phys. Lett. 82 4779
[23] Zhong Z, and Bauer G 2004 Appl. Phys. Lett. 84 1922
[24] Zhong Z, Halilovic A, Lichtenberger H, Schäffler F, and Bauer G 2004 Physical. E. 23 243
[25] Zhong Z, Schmidt O G, and Bauer G 2005 Appl. Phys. Lett. 87 133111
[26] Zhong Z, Chen G, Stangl J, Fromherz T, Schäffler F, and Bauer G 2004 Physica. E. 21 588
[27] Zhong Z, Halilovic A, Mühlberger M, Schäffler F, and Bauer G 2003 J. Appl. Phys. 93 6258
[28] Biasiol G, Gustafsson A, Leifer K, and Kapon E 2002 Phys. Rev. B 65 205306
[29] Novak J, Holy V, Stangl J, Zhong Z, Chen G, Bauer G, and Struth B 2005 J. Appl. Phys. 98 73517
[30] Zhong Z, Katsaros G, Stoffel M, Costantini G, Kern K, Schmidt O G, Jin-Phillipp N Y, and Bauer G Appl. Phys. Lett. in print