Introduction of the Ge nanostructures in a Si matrix via nanosphere lithography deposition

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Experimental studies on patterning hexagonal Ge nanostructures have been conducted on Si substrates through deposition of Ge with polystyrene spheres as a mask. The size distribution of the patterned Ge nanostructures is narrow with the full width at half maximum being less than 10% of the dot size. The two-dimensional patterned Ge nanostructures were further introduced in a Si matrix. Cross-section transmission electron microscopy reveals periodic dark stripes representing the deposited Ge dots in an a-Si matrix.

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1. Introduction

The growth of germanium (Ge) thin films and structures on silicon (Si) surfaces has been the subject of extensive study due to the prospective device applications [1] and the fundamental research concerning the understanding of growth processes [2-3]. In the quest to expand integrated silicon technology, in particular to applications in optoelectronics, Ge/Si nanoheterostructures [4] with engineered band structures have come under intense investigation as important candidates for light-emitting quantum dot (QD) based devices. Research efforts have been dedicated to the exploration of “dotlike” structures obtained via Stranski-Krastanov (SK) growth mode, which comprises the formation of a wetting layer (WL) followed by three-dimensional island (“dot”) formation that relaxes the strain induced by the 4.2% lattice mismatch between Ge and Si. To control the size, shape and density, but mostly the spatial positioning of Ge dots, many strategies including combinations of lithography-based (top down) and spontaneous self-organization approaches (bottom up) have been pursued [5-8]. Work has focused on the assisted organization of Ge dots grown on prepatterned Si or SiO2 substrates either by chemical vapor deposition (CVD) [9-10], molecular beam epitaxy (MBE) [11-13], electron-beam evaporation with nanosphere lithography [14].

In this paper, we describe the patterning of Ge nanostructures via electron-beam evaporation using a colloidal particles mask. The intent of this approach is to demonstrate the capability to create a 2D Ge nanostructures that it will be introduce in a Si matrix.

2. Experimental methods

Initially, a monolayer of colloidal particles is self-assembled on a Si(111) substrate (purchased from Crystal GmbH) in a hexagonally close-packed arrangement [15]. Although areas without self-assembled nanospheres were observed throughout the surface, domains of closepacked monolayers of the polystyrene (PS) nanospheres extended over several hundred square micrometers and free of defects were observed by microscopy images. A metallic material is deposited onto the surface through the interstices of a colloidal template. The lateral dimensions of the nanopatterned features are determined by the interstitial spacings between the close-packed colloidal particles, while the height is controlled by the amount of material deposited [16]. Ge (99.999%) and Si (99.999%) purchased from Umicore was used for deposition to form the nanostructure patterns and, respectively, to introduce the 2D Ge nanostructures in a Si matrix. The materials were evaporated from an e-beam source ultra-high vacuum evaporator (BOC Edwards Auto 500) with a base pressure of 10^{-9} Torr, while the substrate was kept at room temperature. A microbalance was used to monitor the evaporation rate and the thickness of the Ge and Si thin films, with the microbalance calibrated by profilometry measurements. The deposition rate was ~ 0.1 nm/min at a working pressure of 5×10^{-6} Torr. The resulting Ge nanostructures were visualized by Scanning Electron Microscopy (SEM). The introduction of the Ge nanostructures in a Si matrix was characterized by cross-section transmission electron microscopy (XTEM).

3. Results and discussion

3.1. Ge nanostructures

When equally-sized spherical particles are closely packed on a surface in a single layer, the spheres form a periodical hexagonal pattern. With the monolayer as a mask, the deposited pattern due to the Ge flax passing through the space between the spheres will also form a hexagonal pattern. In our experiment, such patterning was

achieved in the Ge/Si system, where a monolayer of polystyrene nanospheres (latex particles) was used as deposition mask. A typical nanosphere lithography (NSL) fabrication is shown in the SEM images in Fig. 1, where an ordered hexagonal pattern of Ge nano-structures is obtained with 12 nm Ge deposition on the Si surface that was masked with 1500 nm latex nanospheres (a), respectively 6 nm Ge deposition on the Si surface with 300 nm latex nanospheres (b). A cluster-size distribution measurement from the AFM image reveals that the size dispersion of the Ge nanostructures in the pattern is very narrow, with a peak width at the half maximum (FWHM) less than 10% in respect to the measured size peak. This demonstrates that the NSL is a technique capable to fabricate ordered arrays of semiconductor nanostructures with a good size control.

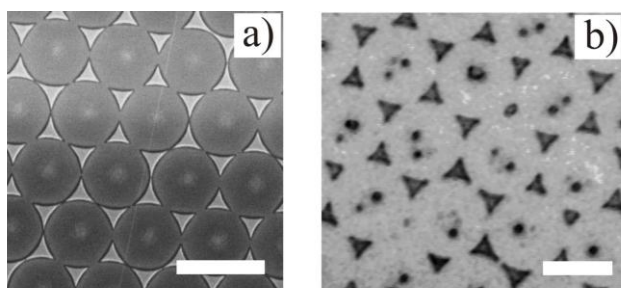

Fig. 1. SEM images of the Ge nanostructures obtained for a) colloidal particle mask of 1500 nm diameter (scale bar 1500 nm) and b) colloidal particle mask of 300 nm diameter (scale bar 300 nm).

3.2. Introduction of the Ge nanostructures in a Si matrix

The obtained Ge nanostructures were introduced in a Si matrix by depositing on top of them a 50 nm Si layer. A cross-section specimen for TEM observation has been prepared in the classical way, by mechanical polishing followed by ion milling on a Gatan PIPS machine. The TEM observations have been performed on a JEOL 200 CX electron microscope operated at 200 kV. The pattern of the deposited Ge islands, as revealed by the SEM image, suggests that the sectioning direction when preparing the cross-section TEM (XTEM) sample has important implications regarding the morphological information contained in the XTEM images. Real non-negligible chances are that the sectioning direction passes between the triangular Ge islands crossing no Ge island. The maximum length of a Ge island observable on a XTEM image equals the length of a triangle edge, which is around 250 nm, according to the SEM image (for a 700 nm colloidal particle mask). On the other hand, arbitrary sectioning directions may result on the XTEM images either in periodic Ge islands or, on the contrary, in a lack of periodicity. Therefore, the length, spacing and periodicity of the Ge islands observable on a XTEM image depend on the direction and position of the section line. A low-magnification TEM image of the sample that we prepared in cross-section is presented in Fig. 2. One can notice the 50 nm thick layer of amorphous Si (*a*-Si) uniformly covering the Si wafer. At a careful observation of the *a*-Si – Si interface, one can observe narrow dark stripes appearing periodically along the interface. White arrows are used in the image in order to facilitate their observation. These periodic dark stripes represent the deposited Ge islands. The noticed dark contrast is a mass contrast due to the difference between the atomic numbers Z of Si and Ge which influences the cross section of the incoherent Rutherford electron scattering. The separation distance between the Ge islands is around 750 nm, which corresponds to the expected periodic spacing along certain sectioning directions.

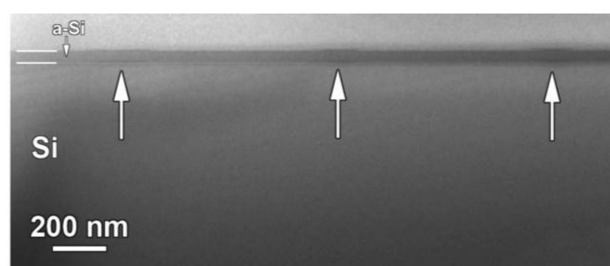

Fig. 2. Low-magnification TEM image revealing the Ge islands covered by the a-Si capping layer.

An individual Ge island is shown in the XTEM image taken at a higher magnification, in Fig. 3. The amorphous Ge island is recognized by the limiting dark contrast line at the interface with the amorphous Si capping layer. Although the Ge island does not show net borders, its section measures around 210 nm in length. This dimension corresponds to the expected range of values from the SEM image and it should be less or equal to 250 nm, the average edge length of the Ge triangular islands. The 50 nm thick covering *a*-Si layer follows the surface profile imposed by the Ge island, resulting in a variation of the overall thickness of 5.6 nm. White arrows on the righth side of the image in Fig. 3 are used to indicate the level difference. In Fig. 4, a high-magnification XTEM image from a region between the Ge islands is presented. The image shows the 50 nm thick deposited *a*-Si layer and, also, a 2 nm thick amorphous layer corresponding to the native amorphous SiO_2 covering the Si wafer.

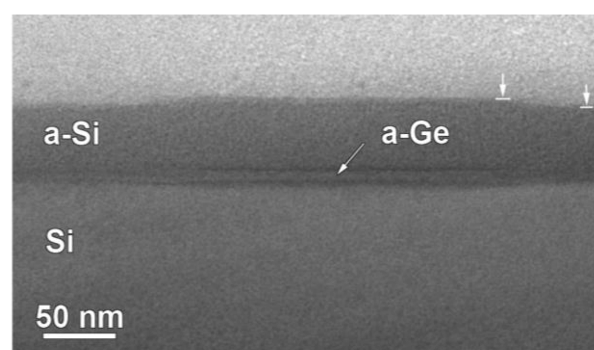

Fig. 3. High-magnificatoion TEM image showing a single a-Ge island embedded in the a-Si layer on the Si substrate.

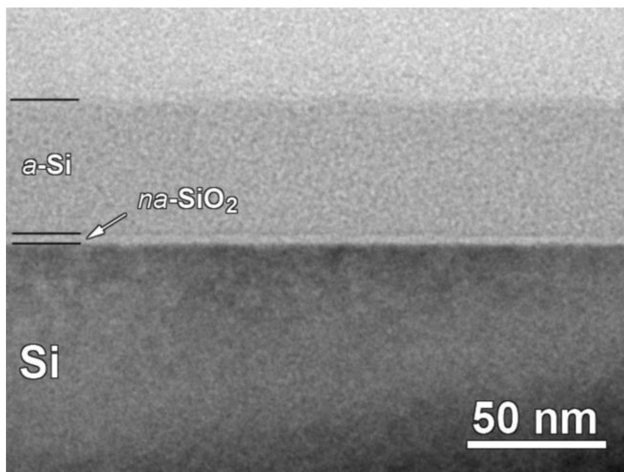

Fig. 4. High-magnification TEM image showing the interface between the a-Si layer and the Si substrate in the space between Ge islands.

3. Conclusions

Ge nanostructures, with lateral sizes from 150 to 400 nm and height from 6 to 12 nm, have been obtained using the combination of NSL and the e-beam evaporation. The 2D ordered structures on Si (111) substrates have been evidenced by SEM images. The 50 nm thick covering *a*-Si layer follows the surface profile imposed by the Ge nanostructures in the attempt to introduce them in a dielectric matrix.

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References

[1] J. Konle, H. Presting, H. Kibbel, K. Thonke, R. Sauer, Solid-State Electron. **45**(11), 1921 (2001).
[2] G. Medeiros-Ribeiro, A. M. Bratkovski, T. I. Kamins, D. A. A. Ohlberg, R. S. Williams, Science **279**, 353 (1998).
[3] J. Stangl, V. Holy, G. Bauer, Rev. Mod. Phys. **76**, 725 (2004).
[4] J. M. Baribeau, X. Wu, N. L. Rowell, D. J. Lockwood, J. Phys.: Condens. Matter **18**, R139 (2006).
[5] T. I. Kamins, R. S. Williams, Appl. Phys. Lett. **71**, 1201 (1997).
[6] G. Capellini, M. de Seta, C. Spinella, F. Evangelisti, Appl. Phys. Lett. **82**, 1772 (2003).
[7] A. Karmous, A. Cuenat, A. Ronda, I. Berbezier, S. Atha, R. Hull, Appl. Phys. Lett. **85**, 6401 (2004).
[8] A. Bernardi, M. I. Alonso, A. R. Goni, J. O. Osso, M. Garriga, Appl. Phys. Lett. **89**, 101921 (2006).
[9] T. I. Kamins, D. A. A. Ohlberg, R. S. Williams, W. Zhang, S. Y. Chou, Appl. Phys. Lett. **74**, 1773 (1999).
[10] L. Vescan, Mater. Sci. Eng., A **302**, 6 (2001).
[11] O. G. Schmidt, N. Y. Jin-Phillipp, C. Lange, U. Denker, K. Eberl, R. Schreiner, H. Grabeldinger, H. Schweizer, Appl. Phys. Lett. **77**, 4139 (2000).
[12] G. Jin, J. L. Liu, K. L. Wang, Appl. Phys. Lett. **76**, 3591 (2000).
[13] Z. Zhong, A. Halilovic, M. Muhlberger, F. Schäffler, G. Bauer, J. Appl. Phys. **93**, 6258 (2003).
[14] N. Li, M. Zinke-Allmang, Jpn. J. Appl. Phys. **41**, 4626 (2002).
[15] M. Ulmeanu, M. Filipescu. R. V. Medianu, Phys. Stat. Sol. (c) **12**, 3584 (2008).
[16] F. Burmeister, C. Schafle, T. Matthes, M. Bohmisch, J. Boneberg, P. Leiderer, Langmuir **13**, 2983 (1997).

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