INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON



# **Experiment Report Form**

# The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application**:

http://193.49.43.2:8080/smis/servlet/UserUtils?start

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Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

#### Reports on experiments relating to long term projects

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All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

### **Deadlines for submission of Experimental Reports**

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

#### **Instructions for preparing your Report**

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

<b>ESRF</b>	Experiment title: Investigation of Ge quantum dots ordering in Ge+SiO2 multilayers	Experiment number: SI9180
Beamline: BM20	Date of experiment:   from: 09.12.2010 to: 14.12.2010	Date of report: 17.02.2010
<b>Shifts:</b> 18	Local contact(s): Nicole Jeuter	Received at ESRF:
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## **Report:**

In our previous works [1,2] we have investigated the nucleation, growth and crystallization of Ge nanoclusters in amorphous  $SiO_2$  matrix. We have demonstrated that originally amorphous Ge clusters crystallize during post growth annealing creating crystalline quantum dots. Moreover, in Ge+SiO<sub>2</sub>/SiO<sub>2</sub> multilayers we found that the positions of the quantum dots are correlated creating a well developed rhombohedral lattice. We have explained this self-organization process by the influence of the morphology of the growing surface on the diffusion of Ge adatoms during the growth.

In order to prove this idea, we have investigated in-situ the nucleation and growth of Ge nanoclusters in  $Ge+SiO_2/SiO_2$  multilayers deposited on quasiperiodically corrugated surfaces. The substrates were produced by the ion beam erosion of Si(100) wafers creating quasiperiodic ripples, the AFM image of the ripples is shown in Fig. 1. We have chosen the parameters of the ion erosion so that the resulting ripple period (19±2) nm was close to the "intrinsic" distance of Ge nanoclusters on a flat substrate.

The rippled Si surface was used as a substrate for the deposition of the Ge+SiO<sub>2</sub>/SiO<sub>2</sub> multilayer. We deposited 5 bilayers Ge+SiO<sub>2</sub>/SiO<sub>2</sub> using dual (AC/DC) magnetron sputtering, the substrate temperature was  $\frac{1007 \text{ mm} 1560 \text{ mm}}{1007 \text{ mm} 1560 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ mm} 1000 \text{ mm}} = \frac{1000 \text{ mm} 1000 \text{ mm} 1000 \text{ mm}}{1000 \text{ m$ 



400°C and the Ge:SiO<sub>2</sub> molar ratio was approximately 75:25 in the mixed layers. After the deposition of each bi-layer we measured a grazing incidence small angle X-ray scattering (GISAXS) intensity map. The probing x-ray beam was directed parallel to the ripples direction, so that the intensity distribution measured in the reciprocal plane perpendicular to the primary beam reflects a spatial correlation of the ripple positions and the influence of the periodicity of the ripples on the correlation of the positions of the growing Ge nanoclusters.

GISAXS maps were measured for two incidence angles of the probing beam, i.e. at the critical angle and slightly above it. In the former case we see mainly scattering from the surface, while in the latter the beam penetrates into the deposited film (Fig. 2). All maps exhibit strong lateral satellites stemming from the ripple periodicity, these satellite maxima are elongated vertically and their height along  $q_z$  is inversely proportional to the ripple depth. The strongest satellites are visible for the substrate and their periodicity corresponds to the ripple period of 19 nm. The GISAXS maps taken with the critical incidence angle after the growth of individual bilayers (Fig. 2(a)) are rather similar to that of a naked rippled substrate, which indicates that the

morphology of the rippled substrate is replicated during the growth. The surface of each newly deposited layer contains ripples with a very similar geometrical arrangement. However, the degree of spatial correlation between ripple positions decreases during the deposition, since the lateral satellites become less pronounced.

For the examination of the inner structure of the film, the probing beam with a higher incidence angle was used (Fig. 2(b)). The resulting GISAXS maps exhibit vertical satellite maxima indicating that the local morphologies of the interfaces of different layers are correlated. This correlation might be described as a roughness correlation using standard models, or as a correlation of the positions of Ge nanoclusters. A detailed numerical comparison of the measured intensity distributions with the intensities following from these two structural models is necessary in order to confirm unambiguously the presence of the correlation of the positions of the nanoclusters induced by the ripples underneath, this work is underway.



Fig.2. GISAXS maps measured in-situ during deposition of  $(Ge+SiO_2)/SiO_2$  multilayer. (a) The surface contribution, the incidence angle equals critical angle, (b) the contribution coming mainly from the film volume, the incidence angle of probing beam was slightly above the critical angle.

After the deposition the film was annealed in vacuum up to 800°C and the GISAXS maps were measured insitu after each 100°C step in the annealing sequence starting from 600°C (Fig. 3). All maps are very similar showing that the layers keep their morphology during the annealing. With increasing annealing temperature, the overall GISAXS intensity increases indicating that the contrast in the electron densities in the clusters and the surrounding increases.



Fig.3. GISAXS maps measured in-situ during annealing of the deposited multilayer.

- [1] M. Buljan et al., Phys. Rev. B **79**, 035310 (2009).
- [2] M. Buljan et al., Nanotechnology 20, 085612 (2009).
- [3] A. Keller, R. Cuerno, S. Facsko, W. Moller, Phys. Rev. B 79, 115347 (2009).