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

Reports supporting requests for additional beam time

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

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

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.

ESRF	Experiment title: Spontaneous atomic ordering in SiGe nanocrystals in amorphous matrix induced by ion irradiation	Experiment number: SI1913
Beamline:	Date of experiment:	Date of report:
ID01	from: 23.09.2009 to: 28.09.2009	10.02.2010
Shifts:	Local contact(s):	Received at ESRF:
15	Thomas Cornelius	
Names and affiliations of applicants (* indicates experimentalists):		
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Report:

Semiconductor Ge nanocrystals embedded in amorphous SiO_2 matrix are intensively studied nowadays because of their prospective application as photonic structures and in effecient photovoltaic devices. The optical and electrooptic performace of these nanostructures substantially depends on the homgeneity of their sizes so that a reliable way of controlling their size is of a great technological importance. Simple kinetic simulations indicate that a narrow dispersion of sizes of Ge nanocrystals can be achieved by increasing the periodicity and regularity of their positions in the amorphous silica matrix. In our previous paper [1] we demonstrated that a quasiperiodic distribution of the positions of Ge nanocrystals can be achieved by a selforganization process during magnetron sputtering of Ge+SiO₂/ SiO₂ multilayers and subsequent annealing. This self-organization is caused by the influence of the nanocrystals buried below the growing surface on the surface morphology and consequently on the two-dimensional diffusion of Ge adatoms during the growth. However, this effect is rather weak and it can yield only a short-range ordering of the nanocrystal positions.

In the reported beamtime we investigated another way to improve the periodicity of the dot positions, based on the effect of a post-growth irradiation of a Ge+SiO₂/SiO₂ multilayer by highly energetic light ions. Preliminary considerations and experiments indicated that tracks of individual ions crossing the multilayer stacks create "weak points" in the individual Ge+SiO₂ layers, where Ge nanocrystals nucleate during the annealing. Consequently, such an irradiation gives rise to a three-dimensional array of Ge nanocrystals, in which the nanocrystal positions in different Ge+SiO₂ layers exhibit a long-range order [2,3].

We measured the positions of the nanocrystals before and after annealing of non-irradiated and irradiated multilayers by means of grazing-incidence small-angle x-ray scattering method (GISAXS) at the beamline ID01, using the wavelength of 1.127Å.

The scattered radiation was measured by a linear detector perpendicular to sample surface and a twodimensional distribution of scattered intensity in reciprocal space was reconstrusted from a series of the detector images taken at various in-plane azimuthal angles ψ of the detector arm. Compared to a standard experimental arrangement using a two-dimensional detector, the advantage of this experimental set-up consists in the fact that we did not have to use a beamstop shadowing out the specularly reflected beam, since we skipped a small angular region around $\psi=0$. We have simulated the intensity distribution using a novel structure model, combining an in-plane short-range order arrangement of the nanocrystals and a long-range order arrangement of the nanocrystals in different Ge+SiO₂ layers in the multilayer stack, for details see Ref. [3]. Example of measured and simulated intensity distributions is in Fig. 1, showing the results of non-irradiated and irradiated samples, the latter was irradiated by 3 MeV oxygen ions under the angle $\varphi = 30$ deg to the surface normal. In Fig. 1(a) showing the GISAXS map of a non-irradiated sample we can distinguish horizontal intensity sheets indicating a vertical correlation of the interface roughness. In the map of the irradiated sample (Fig. 1(b)) an inclined narrow sheet is visible, the direction of the sheet is nearly perpendicular to the irradiation direction. The intensity maps taken at various incidence angles were fitted to the structure model mentioned above and we vere able to distinguish the signal from the nanocrystals from the contribution of the interface roughness and to determine the parameters of the nanocrystals (mean sizes, size dispersion, dispersion of the distances between the nanocrystals etc.) [3].



Fig. 1 Measured (a,b) and simulated (c) GISAXS intensity maps of a non-irradiated (a) and irradiated (b,c) $Ge+SiO_2/SiO_2$ multilayer.

Additional measurements of x-ray diffraction from Ge nanocrystals in annealed samples demonstrated that there is no difference in the crystal structure and local chemical composition of Ge nanocrystals in non-irradiated and irradiated multilayers, so that the ion irradiation is a suitable way how to improve the periodicity of slef-assembling of Ge nanocrystals.

[1] M. Buljan, U.V. Desnica, G. Dražić, M. Ivanda, N. Radić, P. Dubček, K. Salamon, S. Bernstorf and V. Holý, Phys. Rev. B 79 035310 (2009).

[2] M. Buljan, I. Bogdanović-Radović, M. Karlušić, U. V. Desnica, G. Dražić, N. Radić, P. Dubček, K. Salamon, S. Bernstorff, and V. Holý, Appl. Phys. Lett. **95**, 063104 (2009).

[3] M. Buljan, I. Bogdanović-Radović, M. Karlušić, U. V. Desnica, N. Radić, N. Skukan, G. Dražić, M. Ivanda, O. Gamulin, Z. Matěj, V. Valeš, J. Grenzer, T. W. Cornelius, H. T. Metzger, and V. Holý, Phys. Rev. B, in print.