ESRF	**Experiment title:** **Grazing-incidence three-dimensional DAFS mapping of free-standing Ge quantum dots**	**Experiment number**: 02-02-736 Block allocation with: 02-02-739
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Report:

Introduction: The aim of this experiment on BM02 was to study structural properties (local composition and strain) of GeSi islands grown on nominal Si(001) substrates, by combining **Multiwavelength Anomalous Diffraction** (MAD) and **Diffraction Anomalous Fine Structure** (DAFS) spectroscopy. The experiment was performed in grazing incidence geometry to reduce the scattering contribution of the substrate. Despite the wide knowledge acquired in the SiGe/Si system [1,2], there are still open questions concerning the interplay between the strain, the intermixing and their relations with the shape of the islands. In the following, we will especially show that grazing-incidence multiwavelength anomalous diffraction (GI-MAD) combined with GI-DAFS can be exploited to determine the mean Ge content even in the difficult region of the substrate-island interface and gather statistically significant information to correlate island composition with strain and morphology.

Experimental data and results: 5.5 MLs of Ge were deposited at a temperature of 650°C on the studied sample made of dome-shaped islands. MAD and DAFS measurements in Grazing Incidence were performed at beamline BM02 at the ESRF according to the experimental scheme described in a previous paper [3]. Figure 1 shows the square root of the scattered intensity ($\sqrt{I_{exp}}$) close to the in-plane Si(400) Bragg reflection, the modulus of the Ge and Si structure factors, as well as the Ge content, as a function of reciprocal unit h and island height z. Considering the Ge concentration for h lower than 3.97, the results clearly show a strong intermixing phenomenon leading to an island composition of about 60% Ge together with a slight linear increase of Ge concentration below h = 3.97.

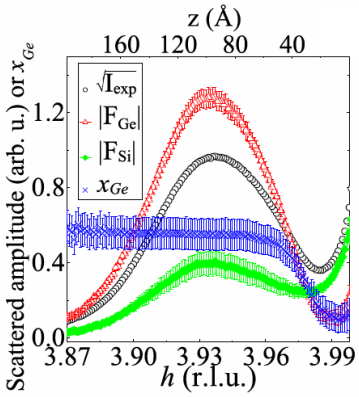
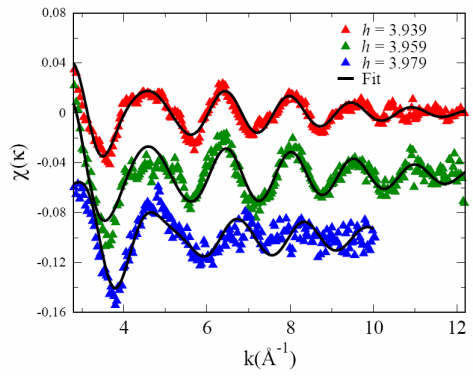


Figure 1. $\sqrt{I_{exp}}$ measured at 11.053keV (50eV below Ge K-edge) and at an incident angle α_i = 0.1°, $|F_{Ge}|$, $|F_{Si}|$ and Ge composition x_{Ge} plotted as a function of h around Si(400) and as a function of island height z.

Figure 2 displays background-subtracted Extended-DAFS spectra at three different h values (h = 3.939, h = 3.959, and h = 3.979), corresponding to iso-strain regions in the dots located at three different heights z (z = 9 nm, z = 6 nm, and z = 3 nm) above the Si surface. Slight changes are observed among the raw spectra, suggesting that compositional changes as a function of z must not be large. The best fit results are shown in table 1. As expected due to the overall spectra shape, the Ge content slightly increases from the base to the dome core, ranging from x_{Ge} = 0.54 to x_{Ge} = 0.6, showing that the composition change at the nano-islands/substrate interface is quite abrupt, and the Ge concentration stays almost constant as a function of z. This is in agreement with MAD results, except near the Si Bragg peak corresponding to substrate-island interface region (at h = 3.979 for instance). In this region, the Ge

content deduced from MAD is lower than the one deduced from EDAFS oscillations due to the scattering contribution of strained Si substrate beneath the QDs.



h	x_{Ge}	R_{Ge-Ge} (Å)	R_{Ge-Si} (Å)
3.979	0.54	2.41	*2.40*
3.959	0.57	2.42	*2.40*
3.939	*0.6*	*2.44*	*2.41*

Figure 2. Raw EDAFS spectra (triangles) and best fit curves (solid lines) recorded at three different *h* values (3.939, 3.959 and 3.979) from top to bottom. Curves are shifted for sake of clarity.

Table 1. EDAFS best fit esults. Error bars on interatomic distances range from ±0.01 to 0.03 Å. Error bars on x_{Ge} are of about 0.05.

To investigate the composition gradient issue, we have calculated the atomic structure for islands with two different chemical composition profiles C1 and C2, shown in Figure 3 (a). C2 is the profile commonly found in most MAD results, which consists of a Si-rich base with a stabilization of the composition at 40-50 Å above the substrate [4]. C1 represents instead an abrupt composition profile at the interface. Reciprocal space composition corresponds, according to the iso-strain method, to real space composition, as it is possible, in the case of free standing nano-islands, to associate Miller index values to *z* values. EDAFS spectra for models C1 and C2 are shown in Figure 3 (b), for several *h* values. We observe that the shape changes are much stronger for C2 due to the smooth variation of Si concentration. Looking at the EDAFS oscillations, model C2 describes much better the experiment.

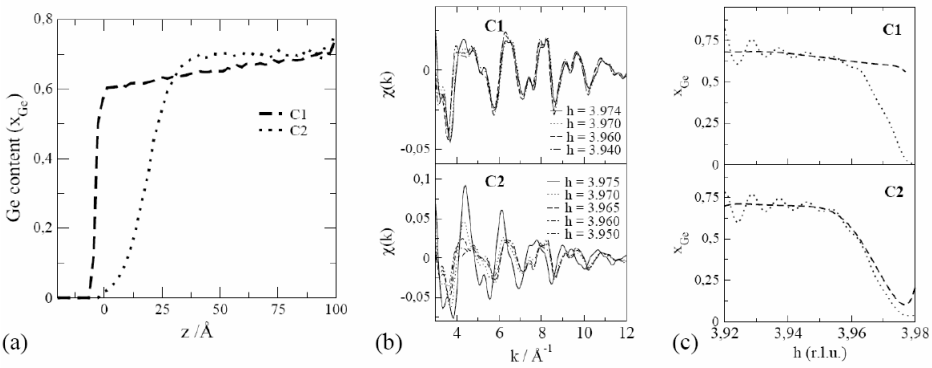


Fig. 3 (a) Chemical composition profiles for GeSi/Si(100) island models C1 and C2. (b) Extended DAFS calculated as a function of reciprocal unit *h* for models C1 and C2. (c) Ge concentration extracted back from the simulated MAD of models C1 and C2.

An important aspect that we wanted to quantify by atomistic simulations is how realistic is the raising, or gradient, of Ge content at the interface substrate-dome given by MAD. Once diffraction (MAD) is calculated for the cluster obtained by atomistic simulations, one can, applying MAD formalism, deduce the concentration profile corresponding to the theoretical MAD spectrum. The same model profile used to generate the cluster should be found if information is not lost on the way. As we can observe in Figure 3 (c), this is the case for model C2, characterized by a smooth concentration profile. For model C1 instead, which is more abrupt, information is lost, and an artificial smoother gradient is found. This shows that one must be careful about MAD results on concentration profiles that can look smoother than in reality.

Conclusion: These results show the great potential of combining MAD and DAFS spectroscopy together with Molecular Dynamics simulations to determine structural properties, as strain and composition, of nanostructures. In the case of sharp interfaces, MAD alone can not access to the mean Ge content in the region of the substrate-island interface and needs to be combined with DAFS measurements. The further step will be to compare the DAFS spectra with ab-initio simulations based on the Distorted-wave Born approximation (DWBA) simulation [5] of the diffracted intensity.

References:

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Reference related to this work:

- *Structural properties of Ge/Si(001) nano-islands and AlGaN nanowires by Diffraction Anomalous Fine Structure and Multiwavelength Anomalous Diffraction*
N.A. Katcho, M.I. Richard, O. Landré, G. Tourbot, M.G. Proietti, H. Renevier, V. Favre-Nicolin, B. Daudin, G. Chen, J.J. Zhang and G. Bauer, XAFS 14 Conference proceedings (2009), to be reviewed.