ESRF	**Experiment title:** **In-situ wetting behaviour study of an undersaturated solution at a solid wall**	**Experiment number:** SI1977
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Report:

Wetting phenomena at solid-liquid interfaces play an important role in processes like lubrication, friction, melting and crystal growth. Of particular importance is obtaining an atomic scale understanding of the growth of crystals from a saturated solution, since many technological developments rely on the existence of perfect single crystals. With the advance of nanotechnology, there is a continuing search for relatively easy methods which allow growth to be controlled on the nanoscale. Liquid Phase Epitaxy (LPE) is such a thin film deposition technique, which is successfully used for numerous electronic and optoelectronic devices such as laser diodes and solar cells. However, although it has been observed for a long time that LPE-grown films have a superior structural quality compared to other growth techniques, the microscopic processes occurring during LPE remain completely unexplored for the reason that the process takes place at a deeply buried interface. With the HEMD set-up at ID15, it is now for the first time possible to reach these interfaces.

The principle of LPE relies on cooling a liquid metal solution below its saturation temperature. The supersaturation is determined by the temperature difference with respect to the liquidus. Starting in an undersaturated situation and cooling to a temperature below the liquidus, will give a supersaturated solution. The new equilibrium consists of, again, a solution and a newly formed solid (just as in solution growth). It has also been observed that thin films can be grown on substrates at temperatures above the liquidus. This phenomenon has been dubbed Interfacial Energy Epitaxy (IEE) [1], and has been shown to produce extremely thin (4 ML) Ge-Si films on Si(100), enabling the growth of nanometre thin films. The current understanding of this is that the surface energy γ is the driving force for crystallization on the Si(100) surface.

In our experiment, we have studied the formation of a Ge-Si heterostructure on a Si(100) substrate. The film was grown by LPE, using Indium as a solvent, in an ultra-high vacuum system. Due to technical problems with the existing portable UHV chamber we used, we were severely set-back in our preparation. Furthermore, due to a defective thermocouple, the substrate temperature could not be controlled accurately. The result of this was that the in-situ flash anneal of the substrate was not optimal, rendering a rough surface. Nevertheless, we could perform the envisaged experiments.

Results

Different kind of measurements were obtained, during different stages of the LPE process. Before contacting, X-ray reflectivity curves were obtained from the liquid metal (see Fig. 1). These curves show that the liquid metal surface is extremely smooth. Grazing incidence in-plane detector scans, shown in Fig. 2, clearly indicate that there is a very good measurable signal when shooting through the Silicon, of which the scattering also becomes visible.

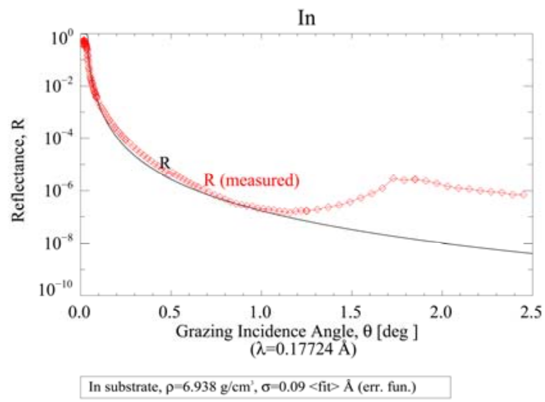

Figure 1 X-ray reflectivity from liquid Indium/vacuum interface. Shown are the data (symbols) and fit of a Fresnel-type curve, with very low roughness. The bump around θ=1.8° is the bulk liquid Indium scattering.

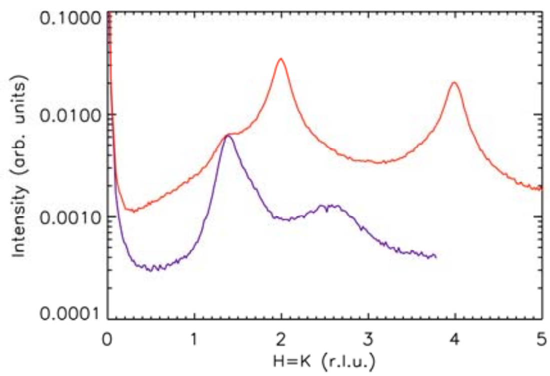

Figure 2. In-plane diffraction in grazing incidence geometry (L≈0). Shown are the liquid scattering from Indium before (below) and after contacting (top) with the Si substrate. After contacting, of which the curve is corrected for absorption of the Si, also the diffuse scattering of the Si Bragg peaks are visible at positions H=K=2, and H=K=4, where H and K are in Silicon units.

X-ray reflectivity curves taken after flashing of the Silicon and contacting with the liquid metal show two interesting features: 1) after contacting the reflected intensity increases 2) A thin film of several tens of nanometres grows during a period of 16 hours on the substrate. These results are understood qualitatively as follows: a rather rough Silicon surface is covered by the stronger scatterer Germanium. The overgrowth with Ge leads to a more strongly reflecting and possibly smoother interface. Since the system is not cooled down afer contacting, the growth is very slow. The details of these results are currently being worked out.

At temperatures above 350°C, we have also observed powderlike peaks on the GIXD scans of the liquid metal. After cool-down to 200°C, these peaks were absent. A search within a powder diffraction database gives rise to GeO_2 as the most probable structure. The exact nature and formation of this layer remains unclear and will be one of the focuses of a next beamtime.

Conclusions

The main conclusion is that in-situ x-ray diffraction during LPE of Ge on Si(100) using very high energies is absolutely feasible with the HEMD set-up at ID15. Furthermore, the Si-Ge system crystallizes very slowly, at these (undersaturated) thermodynamic conditions. These experiments, however, raise still many questions which should be answered in a next experiment. Special care has to be taken in order to have a better control over the UHV conditions and in-situ flash annealing of the substrate.

[1] P.O. Hansson, M. Albrecht, W. Dorsch, H.P. Strunk and E. Bauser, *Phys. Rev. Lett.* **73** (1994) 444