ESRF	**Experiment title: 1D and 2D ordering on vicinal surfaces probed by in-situ high-temperature GISAXS experiments**	**Experiment number**: 02-02-828
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Report:

Aim

This experiment is part of a large study devoted to the self-organization of vicinal surfaces of sapphire through thermal treatment. We have previously demonstrated that according to the duration of the treatment, 1D or 2D lattices can be obtained. This experimental work was mainly realized by ex-situ thermal treatments and observations through GISAXS and AFM. Although we have clearly observed these two types of ordering, the quantitative study of their kinetic is really difficult through ex-situ experiments. On the other hand, taking into account the temperatures under consideration (more than 1200 °C), in-situ measurements are very challenging, nevertheless it was the experimental objective of this proposal. The scientific aim of our high temperature **in-situ** measurement was twofold: the first aspect concerns the 1D ordering and the determination of the kinetic of the evolution of such ordered surface and the second point was to try to identify the process of the 1D to 2D transition.

All the measurements were realized using the new prototype furnace that we have developed through the QMAX research program funded by the ANR (ANR-09-NANO-031). This furnace was put on the top of the motorized goniometric head that we have implemented on the D2AM kappa diffractometer. Through a procedure that we have previously established, the out-of-plane and in-plane orientations of the vicinal surfaces respect to the goniometer axis and the X-ray primary beam were defined with an accuracy of few thousandths and few hundredths of degree respectively. The scattering signal close to the central point of the reciprocal space, i.e. the GISAXS signal, was recorded at 8 keV with an incidence angle close to 0.3° and using a 2D detector (XPAD) located at 5050 mm from the sample. The 3D reciprocal space maps were recorded by the acquisition of set of (q_y-q_z) maps as a function of the azimuthal angle φ. The samples that we have studied were elaborated with a miscut angle equal to 10° respect to the (006) sapphire planes and in such a way that the step edges are parallel to the $[1\bar{1}0]$ direction. The azimuthal orientation was determined by diffraction of the (1112) planes and in all the cases, the zero value of the φ rotation was defined to be the situation where these planes were under diffraction condition, i.e. the primary beam was orthogonal to the [110] direction. Because the $[1\bar{1}0]$ direction of sapphire is orthogonal to the [110] one, in this situation the step edges are parallel to the primary X-ray beam. After convenient alignment, we were able to record complete 3D maps with convenient signal-to-noise ratio in only 15 minutes. Because significant evolution of the surface took more than 10 hours, our study was really time-resolved.

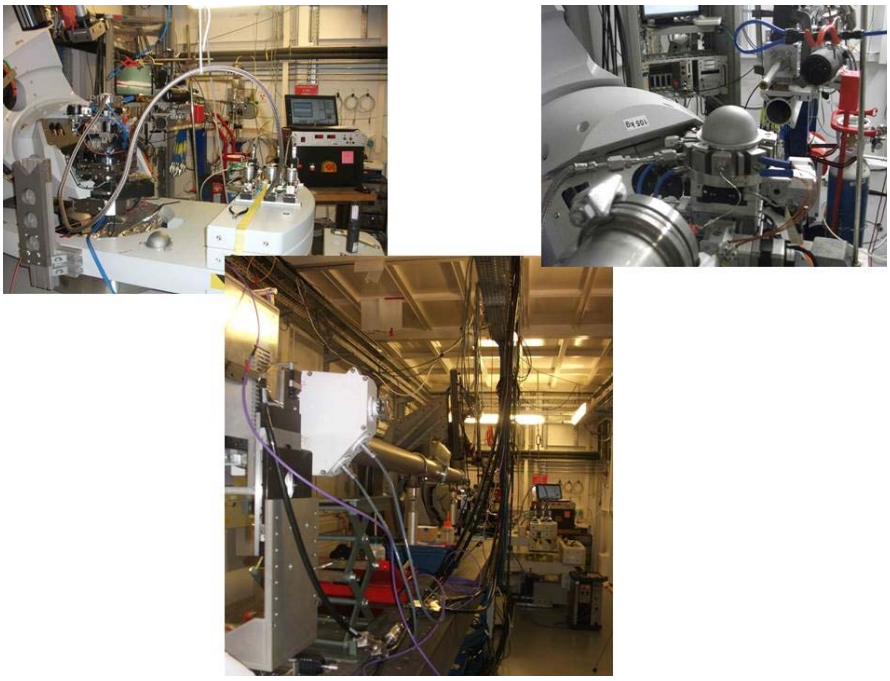


Fig.1. views of the experimental set-up

Experiment

The complete building of the experimental configuration (see fig. 1) took roughly one day and the convenient alignment of the sample in the working conditions (i.e. including electrical, gas and water cooling system connections of the furnace) was particularly tricky. Nevertheless, we were able to found a configuration allowing full 360° rotations of the sample around the φ axis.

After various unsuccessful experiments, we recorded at 1325 °C, during roughly 38 hours, the evolution of the diffuse scattering signal close to the central part of the reciprocal space. Projections of this signal respect to the q_z axis are reported in figure 2. These images are typical q_x,q_y GISAXS maps. Before any thermal treatment, the vicinal surface is made of primary step without any order. We observe an isotropic scattering signal (see fig. 2a). The thermal expansion of sapphire associated to the temperature increase has been compensated by a z-translation of the furnace and the sample has been realigned when the temperature reached the measurement value (i.e. 1325 °C in that case). Rotational realignment is also clearly mandatory in order to keep the normal of the vicinal surface parallel to the φ-axis and consequently the incidence angle of the X-ray beam constant during the rotation of the sample around this φ-axis. Such realignment took some time and the first GISAXS high-temperature measurement was realized after 1h45 on isothermal treatment (see fig. 2b). At this stage, the reciprocal space map exhibits very clearly a scattering line along the q_y direction which is due to the ordering of the step. All along the thermal treatment, this scattering line is observed. Scans along the q_y axis will allowed us to followed the evolution of the period of the steps.

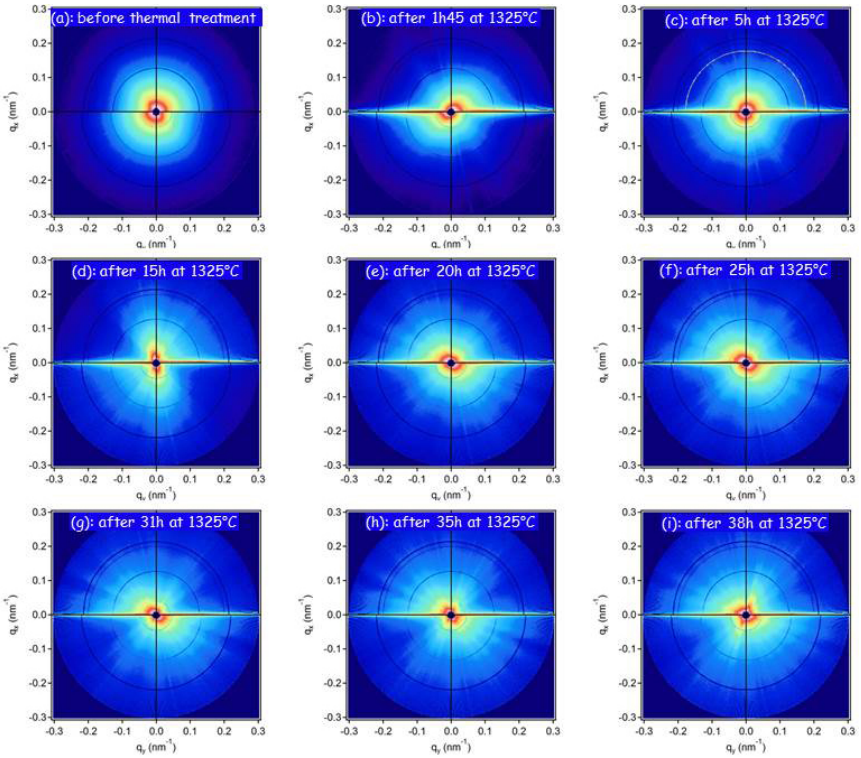
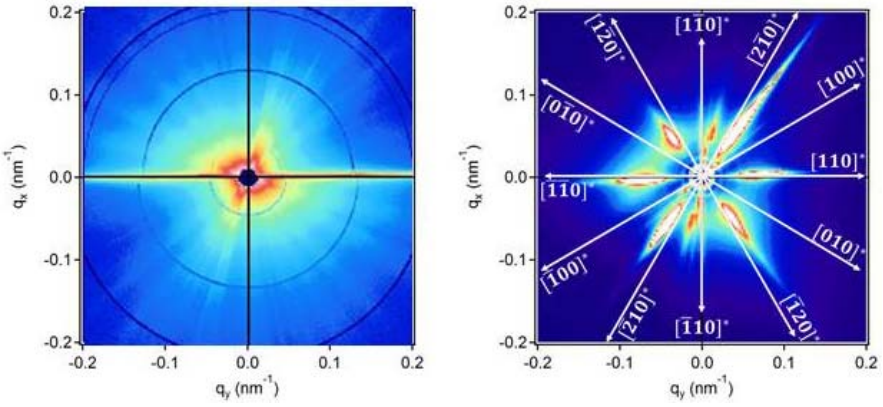


Fig.2. evolution of the diffuse scattering signal during isothermal treatment at 1325°C under oxygen.

Some structuration of the scattered signal is appearing onto the maps recorded after more than 5 hours, with specific diffuse scattering lines being observed. We reported in fig. 3 a comparison between the last map obtained during this in-situ measurement and the one obtained during a previous experiment by ex-situ measurement on a sapphire exhibiting clear 2D ordering. Specific directions underlined in fig. 3b are also observed in fig. 3a. The presence of these directions is due to the appearance of the 2D ordering. It is thus clear that we have observed during this in-situ experiment the transition between the 1D and the 2D self-organization of the vicinal surface. Interpretation of these results is in progress.



(a) *(b)*

Fig.3. appearance of the 2D ordering.

(a): map obtained by in-situ measurement after 38 hours

(b): typical map obtained by ex-situ measurement on a sample exhibiting 2D ordering and the corresponding indexation of the reciprocal space direction respect to the sapphire lattice

As shown fig. 2b, due to the complexity of the setting procedure, we were able to record the first map only after 1h45. In order to evidence the appearance of the 1D ordering we will have to realize new experiment in which the setting procedure will be done under neutral atmosphere. Moreover, the kinetic of the evolution that we observed seems to be slower than that one we evaluated through ex-situ measurements. A convenient probe allowing accurate evaluation of the partial pressure of oxygen must be implemented on the furnace.