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A sample chamber for in situ high-energy X-ray studies of crystal growth at deeply buried interfaces in harsh environments

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ABSTRACT

We introduce a high pressure high temperature chamber for in situ synchrotron X-ray studies. The chamber design allows for in situ studies of thin film growth from solution at deeply buried interfaces in harsh environments. The temperature can be controlled between room temperature and 1073 K while the pressure can be set as high as 50 bar using a variety of gases including N_2 and NH_3. The formation of GaN on the surface of a $Ga_{13}Na_7$ melt at 1073 K and 50 bar of N_2 is presented as a performance test.

1. Introduction

Interfaces are encountered on a daily basis. Examples are the formation of an interface between immiscible fluids, like oil and water; artificially bonded wafers, like microelectromechanical systems (MEMS); a solid in contact with its melt, like water on ice; a metal in contact with a semiconductor, like a Schottky contact; growth of single crystals from a liquid source, like gallium nitride (GaN) liquid phase epitaxy (LPE) growth. The 'buried' interfaces between two bulk systems, as described above, are often causing interesting phenomena. Examples are enhanced liquid density [1], liquid order [2] or liquid layering [1,3,4]. These phenomena are important factors with respect to the mass transport towards the growing interface and therefore growth speed and crystalline quality. Surface X-ray diffraction is capable of resolving a complete 3D structure of the interfaces described above. However, the structure at the interface is difficult to investigate with conventional X-ray energies ($E \approx 10$ keV) due to the attenuation by the material to be penetrated.

The last two decades, however, the creation of proper instrumentation [5,6] at high brilliance high-energy X-ray sources allowed scientists to start exploring these more realistic systems and more complex interfacial phenomena [7–10].

Many interfaces in crystal growth are formed in harsh conditions often combining high pressure, high temperature and dangerous gases or liquids to form the required material. A suitable sample environment is therefore key to investigate the buried interfaces formed during crystal growth in such environments.

This paper presents a reusable batch reactor specifically designed for in situ studies of growing, heated and pressurized buried interfaces in corrosive environments by means of high-energy X-ray reflectivity (XRR) and crystal truncation rod (CTR) measurements. The use will initially be limited to GaN growth using the Na-flux technique. However, this is only one of the experiments that can be envisioned.

To the best of our knowledge this chamber is unique in that it is the first large volume cell designed with a carbon fiber window that is capable of surface diffraction experiments at elevated pressure and temperature using corrosive gases and liquids. Previous pressure cells for XRR were not made for temperatures over 360 K nor corrosive materials ([9] and references therein). Pressure cells for CTR measurements are even more scarce [11], and to the best of our knowledge not available for the combination we present here.

2. Interface diffraction from deeply buried interfaces

Performing diffraction from deeply buried interfaces means that one of the materials forming the interface has to be penetrated. This implies the use of high energy X-rays, because the

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X-ray attenuation coefficient (μ) rapidly decreases with increasing X-ray energy: $\mu(E) \approx E^{-3}$.

An additional benefit of X-rays is that for increasing X-ray energy the scattering angle of a specific signal will decrease. This means that for high energy surface X-ray diffraction (SXRD) signals will be found relatively close to the (000) reflection which reduces the demands for X-ray windows in chambers.

A disadvantage of (high energy) X-rays is that they are not intrinsically surface sensitive and the bulk signal can drown the surface signal. By selecting surface sensitive reflections and additionally setting slits to suppress parasitic scattering and increase the signal to noise ratio this problem is largely overcome. Additionally one can increase the signal to noise ratio further by choosing small incident angles for CTR measurements, typically equal to or lower than the critical angle of reflectivity ensuring total reflection of the incident beam. The small resulting incident angle can lead to complications. Setting an incident angle lower than or equal to the critical angle has a consequence for the required surface quality. The use of high energy X-rays means that the critical angle of reflectivity is very small, typically around 0.03° at 70 keV. Even with a focussed X-ray beam, a small incident angle means illumination of a big surface (e.g. a vertically focussed beam of 8 μm at 0.03° incidence angle will illuminate approximately 15 mm of the sample surface). For this reason the sample surface has to be polished to an extremely smooth (surface roughness) and flat (wavyness, curvature) finish. A detailed description on SXRD is given elsewhere [12,13].

The chamber we describe in this paper is designed primarily for transmission SXRD at high energies. However, the available exit angle for scattered X-rays is chosen so that bulk powder diffraction and bulk liquid scattering, as presented in this paper, are optional alternative techniques. Additionally the chamber can be used with lower photon energies. Compared to 70 keV photons where the transmission through the chamber equals 85%, the transmission at 20 keV is still 68%. Note however that the incident and exit angles available might become an issue when utilizing X-rays of lower energy.

3. Experimental requirements and chamber design

3.1. The beamline

The chamber is developed for use on the high energy micro diffraction (HEMD) stage [5,6] at the high energy beamline (ID15) of the European Synchrotron Radiation Facility (ESRF). The HEMD setup is presented in Fig. 1.

To improve data acquisition the HEMD setup has been equipped with a 2D Maxipix detector with Cadmium Telluride (CdTe) direct detection sensor [14]. Such a detector allows for data collection in the so-called stationary mode [15]. Both the scattering signal and the background can be determined from a single image and there is no need for rocking the crystal. This speeds up data collection tremendously. Combining the use of a pixel detector, the possibility of XRR and CTR measurements, and the use of high energies ($E \geq 70$ keV) gives fast access to complete 3D structures of deeply buried interfaces.

3.2. Chamber requirements

The semiconductor Gallium Nitride (GaN) can be grown using several methods. One of the main techniques to grow

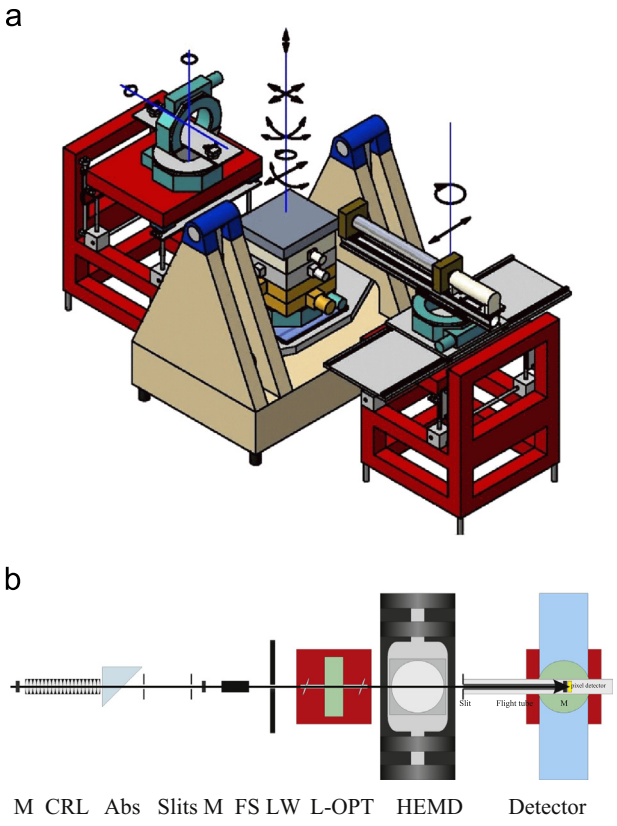

Fig. 1. (a) Schematic of the high energy micro diffraction (HEMD) setup at beamline ID15 of the ESRF showing from left to right the optics for liquid interface/surface studies (L-OPT in (b)), the HEMD and the detector table (Detector in (b)). (b) Top view of beamline setup with M, monitor diode; CRL, compound refractive lenses; Abs, PMMA Absorber; FS, fast shutter; LW, lead wall; L-OPT, optics for liquid interface/surface studies; HEMD, high energy micro diffractometer.