ESRF	**Experiment title:** **Crystalline phase quantification of tin shells grown at cryogenic temperatures around InSb**	**Experiment number**: MA-4728
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Shifts: 18	**Local contact(s)**: Juan Rubio Zuazio	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

An-Hsi Chen* (Institut Neel CNRS Grenoble)

Moira Hocevar* (Institut Neel CNRS Grenoble)

Edith Bellet-Amalric* (CEA-IRIG Grenoble)

Joel Eymery* (CEA-IRIG Grenoble)

Report Summary

Superconducting tin (Sn) shells grown on indium antimonide (InSb) nanowires (NWs) at -190°C in ultra-high vacuum have recently shown great promise for topological quantum computing. Cryogenic grown thin films of metals have shown an improvement of the smoothness of the thin films. This is crucial for the homogeneity of different physical properties along the nanowires, in particular the superconducting gap. Crystalline Sn exhibits a phase transition at 11°C, with the high temperature phase being superconducting. Yet, depending on the geometry and nature of the substrate on which Sn is grown, the transition temperature can vary drastically. **To better understand this behaviour, we aimed at measuring in-situ the structural evolution of tin shells grown, at cryogenic temperatures, around InSb NWs.** The use of BM25 allows (1) for the control of the surface preparation (cleaning of the InSb surface using H_2 beam) prior to growth and analysis using XPS and (2) for the structural study of a growing Sn thin film cooled down to -190°C by GIXRD. Our study provides new structural data to understand topological hybrid materials.

Experimental procedure

One sample (substrate or nanowire substrate) is introduced in the experimental setup. Once the vacuum reaches 10^{-8} Torr, we heat the sample to 250°C below the decomposition temperature of InSb. The atomic hydrogen cell is switched on and heated to about 1000°C with a H_2 pressure of 10^{-6} Torr. The sample surface is then exposed to a flux of atomic hydrogen during 1h30. The surface is monitored before and after treatment by XPS and HAXPES. Then, the sample is cooled down to 80K and thermally stabilized. Once the alignment of the sample is done, GIXRD is performed to obtain the spectrum of reference. Next, Sn is deposited on the sample, either on the full surface of the substrate or on one side of the walls (facets) of the nanowire sample. After deposition , the sample is kept at 80K and Reflectivity and GIXRD is performed on the sample to study the crystalline structure of the Sn thin films and Sn shells. The sample is finally heated to room temperature, realigned and GIXRD is performed on the sample to study the phase and morphology transformations that occurred after warming up the sample.

Results

(1) Thin films

XPS is performed before and after atomic hydrogen cleaning at 300°C. We observe that 1h treatment leads to a decrease of the Sb3d(Ox) peaks by a factor 5, whereas the Sb3d peaks increase by a factor of 5. It means that the atomic hydrogen cleaning is efficient but the desoxydation of the InSb substrate is incomplete.

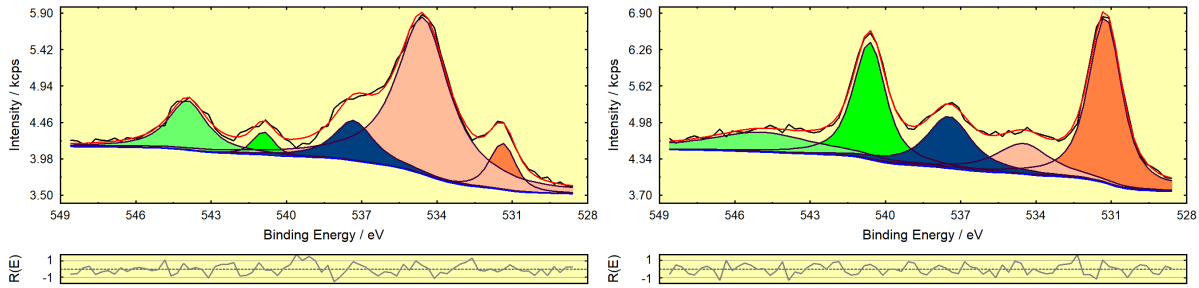
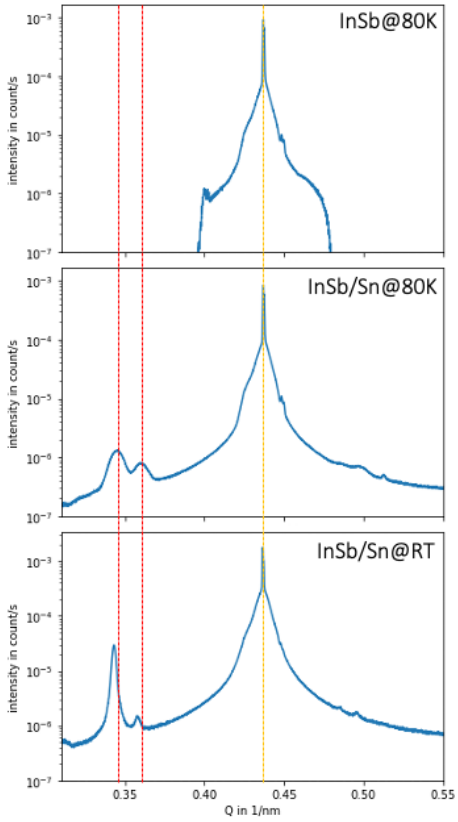


Figure 1: XPS analysis of Sb3d before and after atomic hydrogen cleaning

The thin film is then aligned with the X-ray beam along the (220)InSb Bragg peak which corresponds to 0.43668 nm^{-1}. We chose a photon energy of 12keV in order to study the sample crystalline structure in the diffraction mode (GIXRD) and the composition of the surface using the photoemission mode (HAXPES). The data presented on figure 2 shows the out-of plane XRD measurement performed on the InSb substrate at three different stages: (fig.2a) at 80K after atomic H cleaning, (fig.2b) at 80K after 12 nm of Sn deposition and (fig.2c) after warming up to 300K. Right after the deposition of the thin Sn layer at 80K, only two additional peaks corresponding to beta Sn (200) and beta Sn (101) appear on the spectrum. If present, the alpha Sn (220) reflection would appear at the same position as the (220)InSb reflection. We expected the presence of Pendellosung fringes around the (220)InSb reflexion, similarly to the ones observed in a laboratory setup that revealed the presence of the Sn thin layer. We also expected an broadening of the InSb(220) peak basis. Due to the absence of those two features, our preliminary analysis didn't provide a direct signature of the presence of epitaxial alpha Sn in the deposited thin film. We note that the other alpha Sn reflexions are absent and that the beta Sn reflexions shift towards smaller Q values due to the expansion of the crystal when heating the sample to room temperature.

Figure 2: Out of plane XRD of (110)InSb prior to Sn deposition and after Sn deposition at 80 K and 300K



The Sn layer thickness is measured using reflectivity: we record the intensity of the signal while sweeping the Gamma angle from 0° to 6°. From the period of the oscillations, we extract a deposited thickness of 6nm.

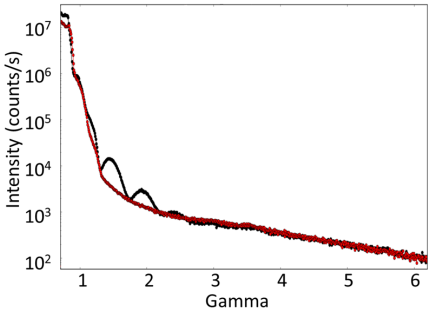


Figure 3: Reflectivity of the thin film of Sn before and after heating up to room temperature

After heating the sample to room temperature, the oscillations disappear, suggesting that the thin film dewetted, which was further confirmed by *ex-situ* SEM imaging. In conclusion, we deposited 6 nm of tin but did not observe the alpha Sn phase in this thin film.

(2) Core-shell nanowires

The nanowire sample consists in InSb nanowires grown on InP substrates through openings created in a SiNx amorphous layer (Figure 4A). Because the surface of interest is the (110) facet of the InSb nanowires, we use grazing incidence XRD to study the growth of Sn thin layers on the nanowires facets. First, we clean the nanowire sample using the same procedure as for the preparation the InSb substrate prior to Sn deposition. We didn't perform XPS due to the complicated 3D structure of the sample. Once the sample reached 80K, the Sn deposition started by opening the valve in front of the Sn source. After 6 nm of deposition (same time and temperature than on the 2D layer), the shutter was closed and the sample kept at low temperature for further measurement, and finally heated to room temperature to study the behavior of the Sn deposited on InSb after warming up.

An alignment matrix was defined with respect to the InP substrate prior to deposition, in order to align easily the sample before and after each process step. Figure 4 shows the XRD data obtained along the (h, -2k, l) projection at 80K before and after Sn deposition (Fig. 4A, 4B)) as well as at room temperature (Fig. 4C). We observe the presence of the (220)InSb peak at h=0.9 prior to Sn deposition. Once Sn has been deposited, two additional peaks appear. They correspond to beta Sn (200) and beta Sn (101) respectively, similarly to the results obtained on the thin films. Those beta Sn grains most probably nucleated on the SiNx amorphous layer. Interestingly, the (220)InSb peak shows a change in shape which could be a signature of the presence of alpha Sn on the nanowire facets. It is currently beeing investigated.

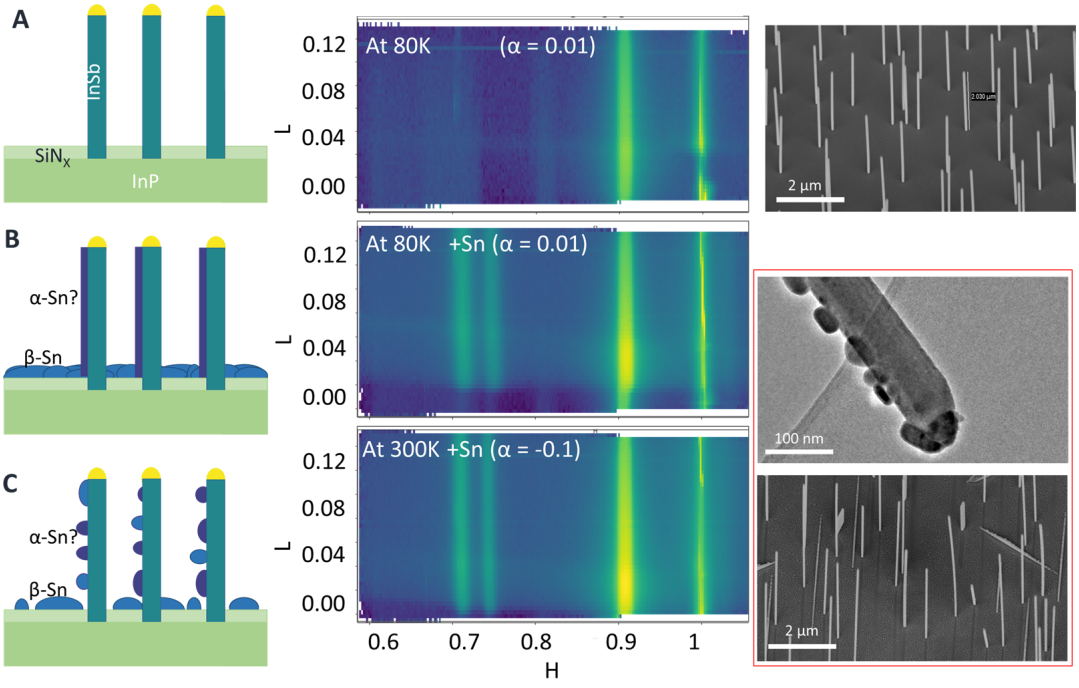


Fig. 4: **A.** *As grown InSb nanowires on SiNx patterns. Left: schematic. Middle: GIXRD along the (h, -2k, l) projection. Right: SEM image.* **B.** *InSb nanowires after Sn deposition at 80K. Left: schematic. Middle: GIXRD along the (h, -2k, l) projection.* **C.** *InSb nanowires after Sn deposition at room temperature. Left: schematic. Middle: GIXRD along the (h, -2k, l) projection. Right: TEM view of a single InSb with Sn grains. SEM image of the sample.*

Once at room temperature, we observe no change in the position of the nanowire and substrate peaks, whereas the beta Sn peaks slightly shift towards higher interplanar distances. This is due to the expansion of the grains due do the increase in temperature. After the experiment, we perform ex-situ SEM and TEM imaging of the nanowire sample. We observe that the Sn film dewetted both from the SiNx thin film and from the InSb nanowire facets. The preliminary TEM data do not show any presence of alpha Sn grains on the nanowire facets.

Conclusion and overview

The procedure to prepare Sn films at cryogenic temperatures (80K) on InSb substrates and nanowires was validated. The procedure of **atomic hydrogen cleaning requires an optimization** which could be performed eventually prior to an experiment at ESRF during an in-house experiment.

In the case of InSb(110) substrate, we couldn't confirm the presence of alpha Sn. Nevertheless, we observed the apparition of beta Sn grains during the Sn deposition at 80K. Those grains could have nucleated around residual oxide patches on the InSb substrate surface.

We were not able to resolve the alpha-Sn/InSb peaks because the lattice parameter between both materials is so close and because the Sn film adapted to the InSb lattice. We were still able to measure the thickness of the film by reflectometry at cryogenic temperature. When heating up the sample to room temperature, we also observed a fast dewetting of the thin film, leading to the conclusion that **capping by another layer is mandatory** prior to heating the sample to RT if we want to further study the phase transformation of the alpha Sn phase.

As a next step, we propose (1) to analyze carefully using X-ray photoemission spectrosocpy and HAXPES the surface preparation of InSb, in particular how to completely deoxidize InSb by atomic hydrogen without damaging the stoichiometry of the surface. We also propose (2) to cap the Sn thin film at cryogenic temperature with an Al thin film to prevent dewetting. They will enable the study of the alpha-to-beta phase transition above room temperature.

Finally, our research would clearly benefit from a second set of experiments mostly because of the difficulties encountered during the alignment of the samples and the creation of the orientation matrix. We suffered from instabilities in temperatures of the sample holder at cryogenic temperatures. The future experiment will also benefit from a larger 2D detector that will be installed in 2022. We will be thus able to observe simultaneously the evolution of the substrate and the nanowire peaks at the energy of interest.