

## Designing Superior Cu(In,Ga)Se<sub>2</sub> Solar Cells Through Understanding and Controlling Growth

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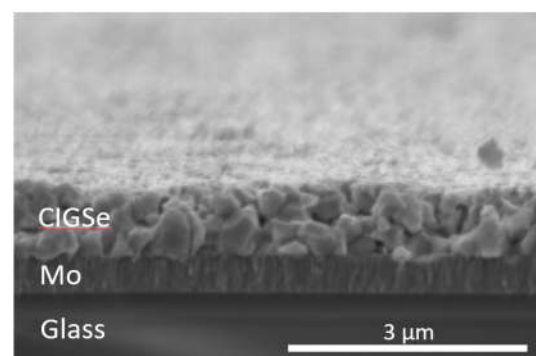
Cu(In,Ga)Se<sub>2</sub> (CIGSe) solar cells stand out as a stable and cost-effective alternative to mainstream Si photovoltaics, offering cost-effectiveness, application versatility, efficient manufacturing, and a reduced CO<sub>2</sub> footprint [1]. The best performing CIGSe solar cells and industry-scale modules are typically deposited using complex multi-stage processes throughout which the composition and temperature of the material is varied according to empirical optimization procedures [2]. To further increase power conversion efficiency, a detailed understanding of material properties during deposition and operation is required. Current studies on material formation primarily rely on methods examining the material before and after growth or quench the reaction and look at snapshots of the growth process. In recent advancements, in-situ X-ray diffraction experiments have been instrumental in tracking phase changes during the three-stage deposition process and defect dynamics during deposition [3]. However, these studies are limited as they are mostly based on diffraction data. To address these limitations and enhance the comprehension of materials processing, in-situ experiments were performed following several key areas in the chain to form ideal CIGSe solar cells. Special attention is given to phase transformations during the deposition process, focusing on defect formation and annihilation, segregation of elements between the different components and how this affects the crystallinity and electronic properties in-situ. Samples were prepared on glass/Mo substrates by sputtering of a Cu-In-Ga alloy at room temperature with simultaneous supply of Se by evaporation, forming an amorphous layer. In-situ Raman spectroscopy performed during crystallization shows that the first phase that is formed is the Cu-poor ordered vacancy compound (OVC), followed by the formation of Cu<sub>2-x</sub>Se at 350 °C. At 400 °C, the A<sub>1</sub> mode of the α-phase of CIGSe appears at ~166 cm<sup>-1</sup> along with the vanishing of the OVC phase. When cooling down, the Cu<sub>2-x</sub>Se phase

disappears and the OVC phase reappears. After crystallization, the A<sub>1</sub> mode shifts to the expected value of ~173 cm<sup>-1</sup> [4]. This red shift in wavenumber with increasing temperature is ascribed to effects of thermal expansion and changes in phonon occupation numbers. In-situ XRD shows that CIGSe starts to crystallize at 200 °C with preferential orientation along [112]. After 350 °C, the crystallite size increases to a maximum of 50 nm at 600 °C. Adding a Se layer on top of the a-CIGSe layer favours the formation of the α-phase during crystallization. After 15 minutes at the 500 °C stage, the A<sub>1</sub> vibrational mode of CIGSe reaches maximum intensity, starting to decrease after that. Moreover, the crystallite size starts to increase just after 200 °C reaching a maximum at a lower temperature of 400 °C.

## References

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## Figures



**Figure 1.** Cross-sectional SEM image of SLG/Mo/CIGSe after crystallization.

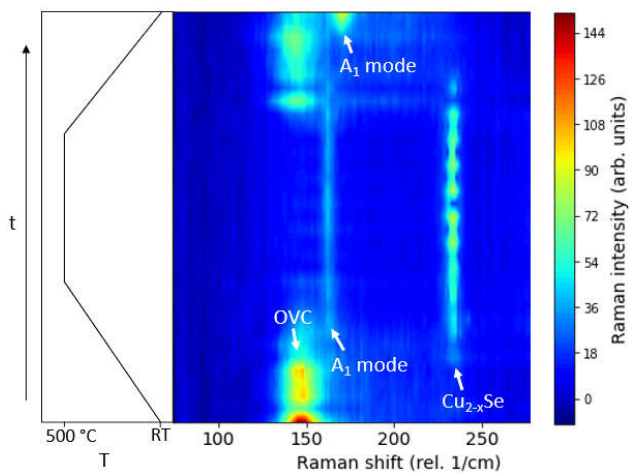


Figure 2. Raman spectra evolution with temperature of CIGSe.

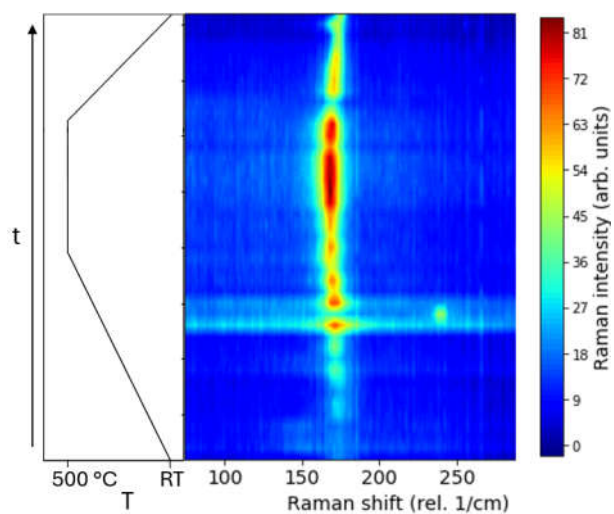


Figure 3. Raman spectra evolution with temperature of CIGSe with Se capping layer.

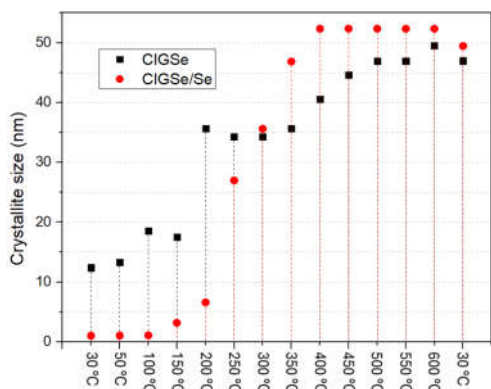


Figure 4. Crystallite sizes evolution with temperature for CIGSe and CIGSe/Se films.