Chemical and structural differences between low and high Ga ratio of polycrystalline $CuIn_{1-x}Ga_xSe_2$

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Thin-film solar cells based on a polycrystalline $CuIn_{1-x}Ga_xSe_2$ (CIGSe) absorber layer are among the most interesting photovoltaic devices as the optical band gap energy (E_g) of CIGSe can be optimized by tuning the Ga ratio, $x = \frac{[Ga]}{[Ga]+[In]}$. The highest labscale conversion efficiency (22%) has been reached for x^*~ 0.4, while from the theoretical point of view, CIGSe thin films could exhibit a better performance for x between 0.7 and 0.8 (E_g=1.4eV) [1].

A possible explanation of this suboptimal performance at large *x* is that large chemical and structural differences occur between low and high Ga ratio of polycrystalline CIGSe [2]. Our study aims at understanding the origin of these two regimes (below and above x*), by coupling a computational and an experimental approach.

We first focus on the bulk phase diagram of the $CuInSe_2$-$CuGaSe_2$ pseudo-binary system by combining *ab initio* density functional theory (DFT) calculations of Special Quasi-random Structures (SQSs) and thermodynamic modelling [3]. The resulting diagram of temperature versus x displays two different regions around *x**. We show that both the existence and dissymmetry of these regions may be associated with the radius difference between Ga and In[4,5], while keeping in mind that this approach requires a very specific care when handling ionic systems that will also be discussed [6].

Secondly, we carried out a structural and compositional analysis coupling XRD, EDX, and Raman techniques for different CIGSe stoichiometries. We illustrate quantitatively that crystallographic orientations differ for different Ga ratio, Cu composition and presence of Na diffusing from the soda-lime glass substrate. However, our first results show that the bulk properties remain invariant with x, which seems to contradict the computed phase diagram at the temperature under investigation. This apparent contradiction will be discussed considering the Cu composition, the temperature and the CIGSe layer growth process.

References

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