New $CIGS_n$ lamellar materials for photovoltaics applications

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Recently, our research group has identified new $CIGS_n$ lamellar phases in the Cu_2S-In_2S_3-Ga_2S_3 system [1]. $CIGS_4$ ($Cu_{0.32}In_{1.74}Ga_{0.84}S_4$), $CIGS_5$ ($Cu_{0.65}In_{1.75}Ga_{1,4}S_5$) and $CIGS_6$ ($Cu_{1.44}In_{2.77}Ga_{0.76}S_6$) are interesting compounds looking to their optical gaps (see Figure 1) that can be comparable to those of the chalcopyrite $CuIn_{0.7}Ga_{0.3}S_2$, studied as potential absorber in a tandem solar cell and well represented in the emerging thin-film photovoltaic [2]. As it is shown in Figure 2 these materials present a 2D structure with generic compositions $(M_{(Td)})_{n-2}(In_{(Oh)})S_n$ (M = Cu, In, Ga), with cations in tetrahedral (Td) and octahedral (Oh) sulphur environments. All compounds exhibit a van der Waals gap (~3.75 Å). We present in this work, the further study of these new materials in order to identify their potential for photovoltaic applications, by assembling a laboratory thin-film photovoltaic cell.

The synthesis of thin films was carried out by vacuum co-evaporation technique from elementary sources. Figure 3 shows an HAADF-STEM image of the scratched powder from a prepared thin film presented on the bottom of the figure. The measured distance between the octahedral In-S layers (observed as brighter lines) as well as chemical composition analysed by EDX, confirm that a $CIGS_5$ lamellar phase has been successfully deposited.

In order to optimise the heterojunction offsets, the flat band potentials of $CIGS_n$ compounds were measured, as a first step on bulk samples. Results were compared to that of the chalcopyrite $CuIn_{0.7}Ga_{0.3}S_2$ which was estimated around -5.3 eV.

A first SLG/Mo/$CIGS_5$/ZnOS/ZnO/ZnO:Al solar cell was formed without any interface optimization. The following photovoltaic parameters were obtained: V_{oc} = 400 mv and J_{SC} = 0.4 mA/cm^2. This first attempt shows there is still a serious need to optimize interfaces. Moreover, PL measurements are also in progress in order to better understand the absorber behaviour.

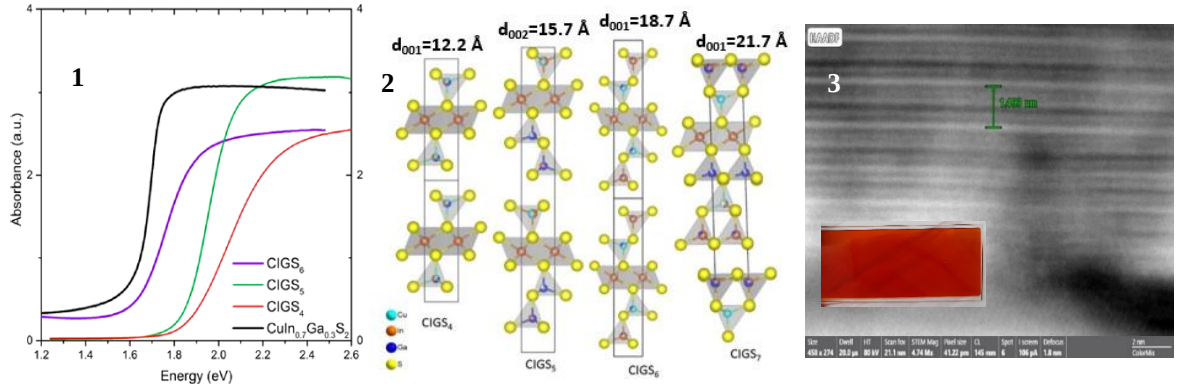

*Figure: **1)** Kubelka-Munk transformed reflectance spectra of $CIGS_n$ and $CuIn_{0.7}Ga_{0.3}S_2$ compounds.*
***2)** $CIGS_n$ structure types. **3)** HAADF-STEM image of $CIGS_5$ thin film.*

[1] Caldes, M., Guillot-Deudon, C., Thomere, A., Penicaud, M., Gautron, E., Boullay, P., Bujoli-Doeuff, M., Barreau, N., Jobic, S. and Lafond, A., 2020. Layered Quaternary Compounds in the Cu2S–In2S3–Ga2S3 system. *Inorganic Chemistry*, 59(7), pp.4546-4553.

[2] Thomere, A., Guillot-Deudon, C., Caldes, M., Bodeux, R., Barreau, N., Jobic, S. and Lafond, A., 2018. Chemical crystallographic investigation on Cu2S-In2S3-Ga2S3 ternary system. *Thin Solid Films*, 665, pp.46-50.