Microstructure characterization of CIGS/GaP/Si tandem solar cells

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Background incl. aims

The conversion efficiencies of single-junction solar cells have reached values very close to the theoretical Schockley-Queisser limit (~33%). One strategy for exceeding this value is to use tandem solar cells [1] combining a top cell optimized to absorb solar radiation in the UV/visible range with a bottom cell optimized for IR absorption. As part of the ANR EPCIS project [2], we have developed this type of cell using a Cu(In,Ga)S2 (CIGS) layer (with a band gap of 1.65-1.70 eV) deposited at low cost by co-evaporation on a Si substrate (band gap of 1.1 eV). However, the adhesion of a CIGS layer to Si is very poor, so an intermediate layer is needed to improve adhesion. The GaP material was chosen for 3 main reasons : 1) its cubic symmetry with a lattice parameter close to that of Si and also to that of CIGS, for a GGI close to 20% (chalcopyrite material with pseudo-cubic quadratic symmetry with c~2a) can promote epitaxy growth of the CIGS film, 2) such layer can act as a diffusion barrier to prevent diffusion of species between layers during the growth process, and 3) its low absorption coefficient (2.26 eV indirect bandgap) coupled with an electron affinity around 3.8 eV make it an ideal selective contact for holes. Despite the above-mentioned advantages, electrical performances of our synthesized tandem solar cells are still very low. We combined different characterization techniques including S/TEM to try understanding the relation between microstructure and optoelectonic performances.

Methods

Growth parameters including temperature, GGI=[Ga]/([Ga]+[In]) and CGI=[Cu]/([Ga]+[In]) can be adjusted during the co-evaporation process to deposit the CIGS layer on the GaP/Si substrate. When CGI is greater (less) than 1, the film is described as "Cu-rich" ("Cu-poor"). Various S/TEM techniques (ACOM-TEM, probe-corrected S/TEM imaging, spectroscopies) were used to study the 'Cu-rich' and 'Cu-poor' CIGS films: the epitaxial growth of the chalcopyrite CIGS layer, the presence of other crystalline phases and their orientation, and the identification of certain intragranular defects. The interdiffusion of elements and the possible presence of an additional layer between the CIGS and the GaP were characterised by atom probe tomography (APT).

Results

In the Cu-poor sample, the existence of a CuIn5S8 thiospinelle phase (TS) was demonstrated in addition to the CIGS chalcopyrite phase (CH) with this epitaxial relationship: CH[100](001)//TS[100](001)//GaP[100](001)//Si[100](001). In the case of Cu-rich sample, the epitaxial relationship is the same, except that there is no TS phase (Figure). However, the interface between GaP and CIGS layer is much more abrupt in the Cu-poor case than in the Cu-rich case where a few nanometers thick layer made of Cu and P is detected.
Despite the presence of TS phase and interface layers, the desired chalcopyrite phase is the most present. Unexplained contrast in HAADF-STEM images for both kind of samples led us to consider defects such as cation antiphase boundaries. On the basis of these contrasts, three types of CAPB were conceptualized.

Conclusion

Combined with other more macroscopic techniques, S/TEM and APT have enabled us to show that the growth of a CIGS layer on a GaP/Si pseudo-substrate for the application of tandem solar cells is far from ideal. Differences in Cu stoichiometry in the CIGS film were also highlighted.

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Graphic:

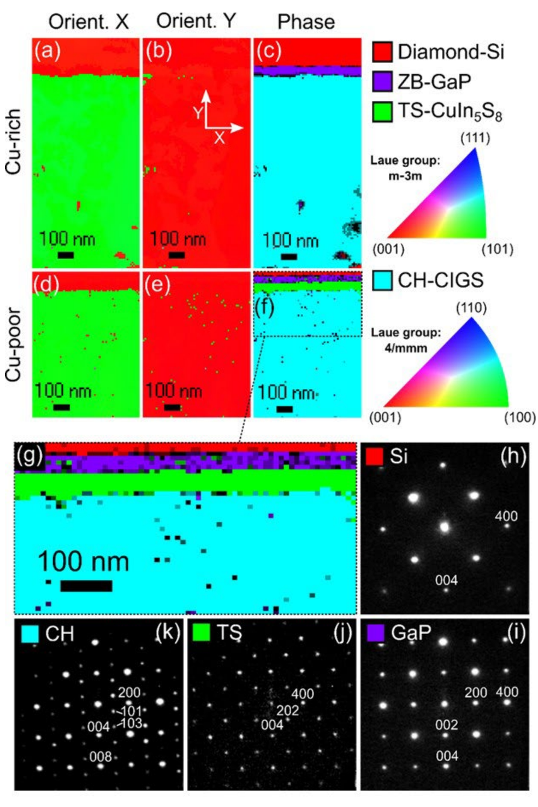

Figure : Orientation map a,d) along X, b,e) along Y, and c,f) phase map for the d–f) Cu-poor and a–c) Cu-rich sample. g) Zoom around the GaP layer for the Cu-poor sample. Electron diffraction patterns of h) the Si, i) the GaP, j) the TS, and k) the CH phases.

Keywords:

Tandem-solar-cell, epitaxy, secondary phases, interdiffusion

Reference:

[1] F. Martinho, Challenges for the future of tandem photovoltaics on the path to terawatt levels: A technology review, (2021).
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