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Exploring new convergences between PV technologies for high efficiency tandem solar cells : Wide band gap epitaxial CIGS top cells on silicon bottom cells with III-V intermediate layers

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ABSTRACT: A new concept is proposed to develop efficient tandem cells based on silicon bottom cells and wide band gap $\text{Cu}(\text{In,Ga,Al})(\text{S,Se})_2$ (noted CIGS) top cells. It consists in introducing an epitaxial III-V buffer layer at the interface. The benefit of this configuration is discussed in terms of lattice and band offsets matching between CIGS and III-V's. Selected combinations are proposed, with AlGaAs and GaAlP buffer layers. Preliminary results with CuGaSe_2 on various substrates confirm the influence of the substrate on the growth of CGS and PV properties. Epitaxial growth on GaP and possible selective back contact effect with GaAlAs are evidenced. Preliminary results on using advanced characterization methods are presented ((quantum efficiency, x-ray diffraction, secondary ion mass spectroscopy, nanoAuger, cathodoluminescence).

Keywords: CIGS, epitaxy, III-V, silicon, tandem cells.

1 OBJECTIVES

In the new road map of PV towards increased efficiencies beyond the Shockley Queisser limit for single junctions, tandem junctions are taking a leading role, which will impose an hybridation between crystalline silicon and thin film technologies (III-V viewed as thin film junctions, perovskites, chalcogenides). Very important efforts are presently devoted to develop tandem cells by using perovskites or III-V junctions on the top of silicon bottom cells. We propose to also consider chalcogenide junctions as top cells, since they refer to well established commercial

technologies based on CdTe on one side and CIGS materials on the other side. Both technologies demonstrated record efficiencies beyond 22%, which would perfectly fit with more than 30% efficiency for tandems.

The aim of this work is to explore the potential of III-V/CIGS interfaces to fulfill the ideal back contact configuration shown in figure 1, which is lacking for present CIGS device structures (non selective Mo back contact)

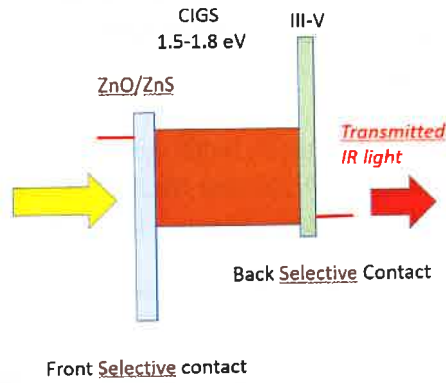


Figure 1. Scheme of an ideal CIGS solar cell, with front and IR transparent back selective contacts.

We will focus on the case of growing high efficiency wide band gap CIGS (1.6-1.8 eV) within the family of copper indium gallium selenosulfide ($\text{Cu}(\text{In,Ga,Al})(\text{S,Se})_2$) on top of

existing III-V buffer layers on crystalline silicon. The very low cost of silicon wafers and cells makes possible to consider now them for CIGS tandem solar cells with real industrial development perspectives. The key hypotheses for improving the efficiency of the wide gap CIGS cell beyond the actual level (>15%) are : (1) beneficial aspect of epitaxial growth which reduces the detrimental grain boundaries effects in wide band gap CIGS, (2) possibility of higher deposition temperatures (>550°) beneficial for life time increase in sulfide based CIGS, (3) optimization of band offsets matching between III-V and CIGS absorbers as compared to classical Mo/MoSe₂ interfaces, (4) new tandem architectures with 2 or 4 terminals.

The device concept for the 2 terminal configuration is given in figure 2.

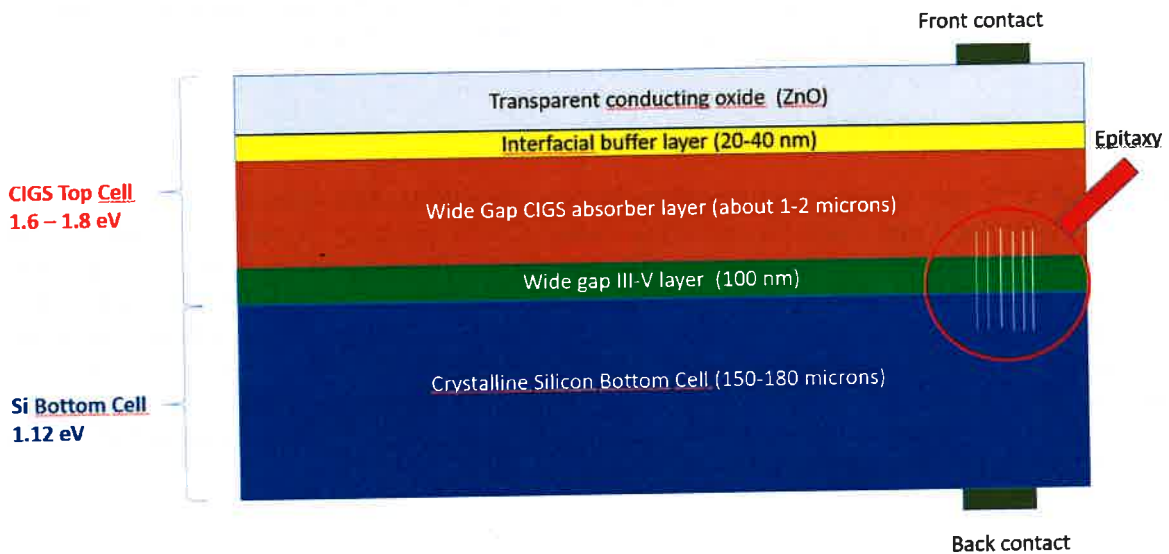


Figure 2. Scheme of the 2 terminal tandem Si-CIGS architecture concept with an interfacial III-V buffer layer.

2 FEASIBILITY ANALYSIS

CIGS solar cells are delivering record efficiencies (> 20%) in a limited zone of band gap values around 1.1 to 1.2 eV, corresponding to a Ga/(Ga+In) ratio (noted GGI) around 30% for $\text{CuIn}_{0.7}\text{Ga}_{0.3}\text{Se}_2$ pure selenides and $\text{CuIn}_{0.7}\text{Ga}_{0.3}(\text{S,Se})_2$ selenosulfides. Moving to lower and larger GGI leads to reduced efficiencies, in particular towards larger GGI associated to wide band gaps in the range of 1.6-1.8 eV, which are needed for top cells. A first systematic study was carried out at NREL in 2012 showing that increasing the temperature

allows to increasing significantly the efficiency up to 1.3-1.45 eV [1]. Recent breakthroughs occurred when using sulfides and selenosulfides, raising the record efficiencies up to 16% for band gaps about 1.55 eV and 11.9% for pure CuGaSe_2 at 1.7 eV [2] deposited on Mo/glass substrates. Very recently [3], a value of 15.7 % was obtained with epitaxial CIGSe cell on GaAs single crystal substrate, without Mo, with a GGI of 0.6 (E_g about 1.55). Figure 3 provides an overall view of possible combinations between CIGS absorbers and III-V's in terms of band gap and lattice matching. It appears that two domains exist which fulfill the required conditions. One concerns pure selenides on GaAlAs substrates (blue) and the

other one sulfoselenides on GaAlP substrates (red zone). III-V nitrides are not included since the lattice constants are significantly smaller.

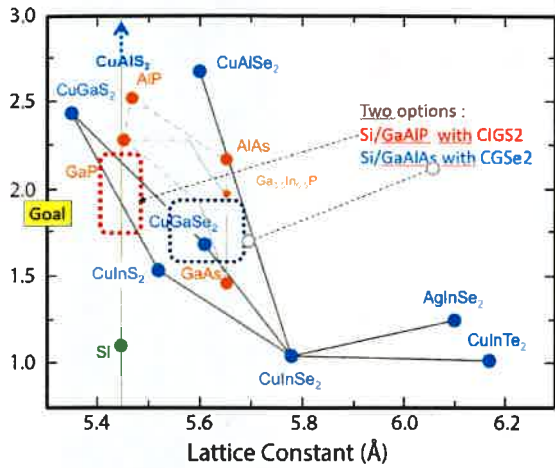


Figure 3. Energy gap (in eV) versus lattice constant for III-V's and chalcopyrites. The red and blue

rectangular zones are adapted for optimal lattice matching and band gap for CIGS top cells.

Considering now the selective contact criterion, the band offsets have to be considered. The conduction band edge energy of the III-V has to be higher than that of CIGS in order to act as an electron barrier/mirror, and the valence band has to be just higher (or slightly lower in case of a spike like configuration) than that of CIGS to provide an easy extraction of holes, giving the optimal open circuit voltage. The general figure given in figure 4 gives an overview of the band offsets for ternary CIGS sulfide and selenide compounds with Al, Ga and In, together with those of binary III-V compounds with N, P and Al. Silicon is included for comparison. The values are taken for the literature. It can be mentioned that some differences appear from one reference to the other as shown in the case of the selenide series (dotted line vs continuous line).

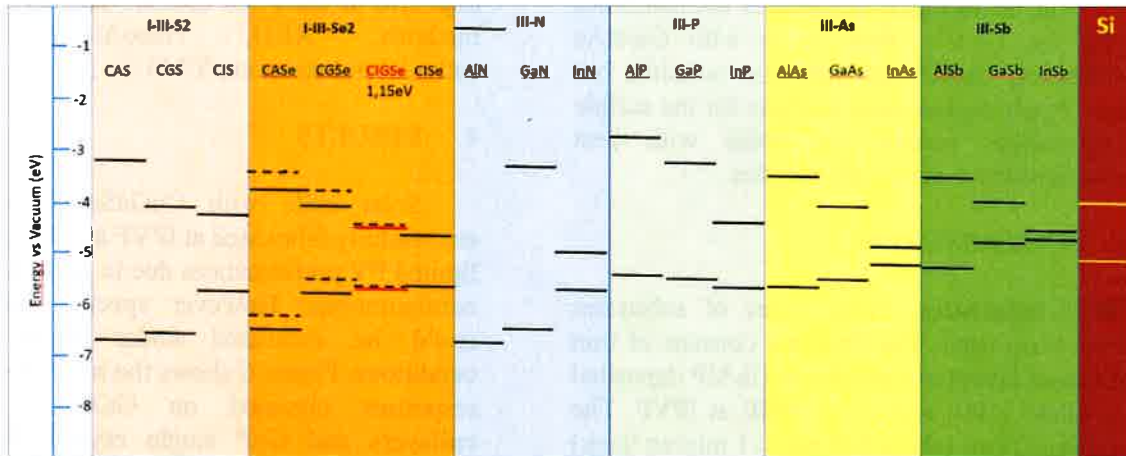


Figure 4. Band energy diagrams of ternary chalcogenides with selenium and sulfur, binary III-V's with nitrogen, phosphorus and arsenic, and silicon.

From both criteria, the combinations between a given CIGS absorber composition

and the III-V can be evaluated. Figure 5 gives the example of CuGaSe₂.

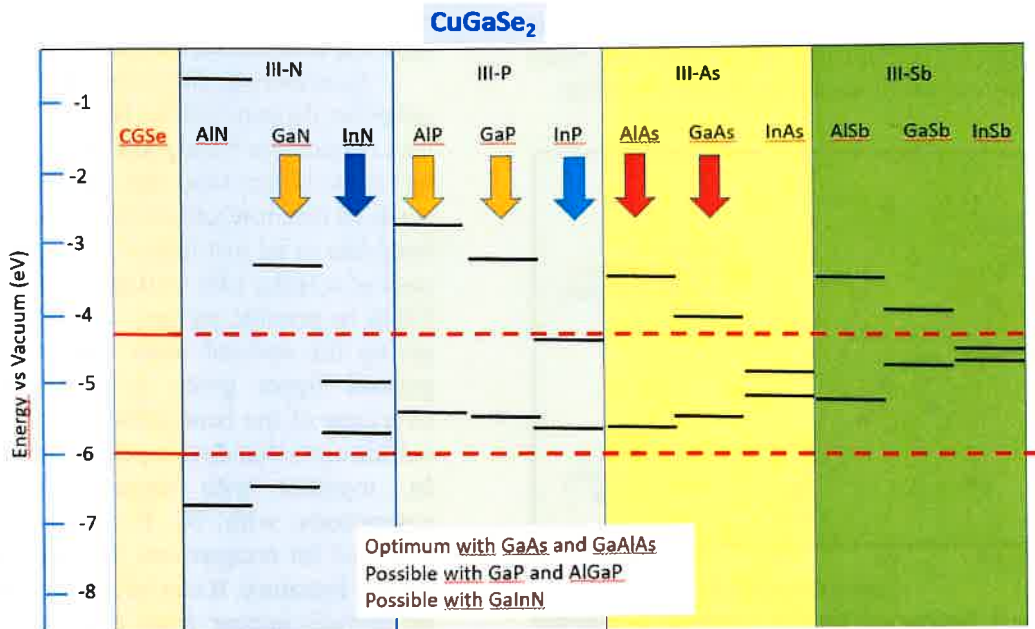


Figure 5. Prediction of the compatibility of III-V's buffer layers with CuGaSe_2 .

The arrows give the level of compatibility with the III-V's. Best fit is with GaAlAs substrates, GaAlP are then also suitable, but less. Applying the same analysis for the sulfide $\text{Cu}(\text{In,Ga})\text{S}_2$ reverse the order with best compatibility with the phosphides.

3 EXPERIMENTAL

III-V substrates. Three types of substrates have been used. The first one consists of thin epitaxial layers of GaAlAs or GaAlP deposited on GaAs (100) wafers by MBE at IPVF. The epitaxial films (about 300 nm to 1 micron thick) are p^+ doped (10^{18} cm^{-3}) and the GaAs is insulating. The second consists in thin GaP/GaPN epitaxial layers (230 nm) deposited on silicon (100) substrates by MOCVD at ISE Fraunhofer. The third one is a monocrystalline wafer of p type GaP (about 10^{17} cm^{-3}), with (111)B orientation. CIGSe layers have been deposited by single stage coevaporation at 570°C at IMN on these substrates. One reference sample with a standard three stage CIGSe absorber has been deposited on the GaP/Si(100) substrate. Cells have been completed for GaAs based substrates and the GaP single crystal with classical CdS/ZnO front contacts. In the case of GaAs substrate, back contact were taken on the III-V epilayers with silver dots. In the case of GaP it was simply done by contacting the back side of GaP by the silver dots. In other cases the CIGS surface was

kept free to carry out specific characterization methods: XRD, NanoAuger, XPS, cathodoluminescence, SIMS.

4 RESULTS

Solar cells with CuGaSe_2 have been successfully fabricated at IPVF at IMN but with limited PV performances due to non-optimized configurations. However spectral responses could be measured under short circuit conditions. Figure 6 shows the relative spectral responses obtained on GaAlAs, GaInP, epilayers and GaP single crystal. We can observe that the best result is obtained with $\text{Ga}_{0.51}\text{Al}_{0.49}\text{As}$ with a good collection towards larger wavelengths. This could be an indication for reduced back contact recombination as expected from the analysis. SEM analysis shows that large grains (micron size) are observed contrary to usually small grain structure of gallium rich layers deposited on Mo/glass substrates. This demonstrates that the use of single crystalline III-V substrate influence markedly the structural and PV properties of the CIGS layer as expected.

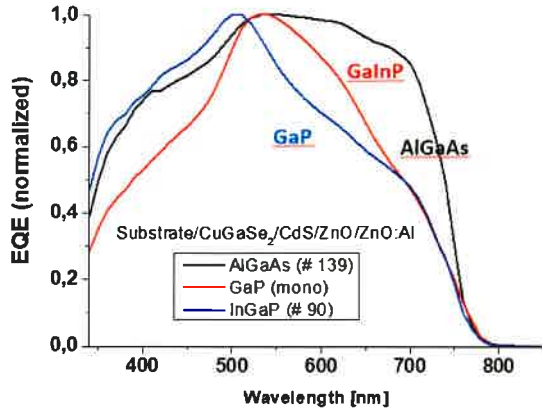


Figure 6. Relative spectral responses for CuGaSe₂ cells with different III-V's substrates.

XRD studies, both in $\theta-2\theta$ and ϕ scan configurations, have been carried out to analyze the structure of the films and the possibility of epitaxy (IPVF, FOTON). We observed that the film were generally not epitaxial, excepted for the GaP (111)B substrate where the epitaxial component was clearly visible in the ϕ scan configuration as shown in figure 7. The presence of 6 peaks can be attributed to a multipositioning effect (inset), as the (101) plane has only two possible orientations with respect to the (112) plane of CGS.

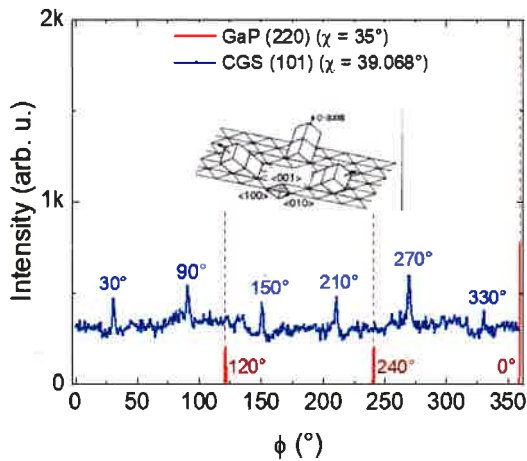


Figure 7. ϕ scans recorded on bare GaP (111) substrate and after deposition of a CuGaSe₂ layer.

Composition profiles have been addressed by SIMS (GEMAC) and NanoAuger (ILV) on Cross section. The interface appears sharp with no significant diffusion of copper across the interfaces. Figure 8 shows nanoAuger profiling on the cross section of the CIGS/GaP/silicon heterostructure.

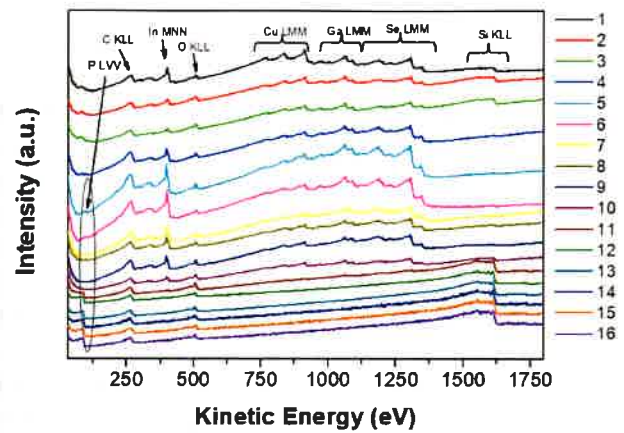
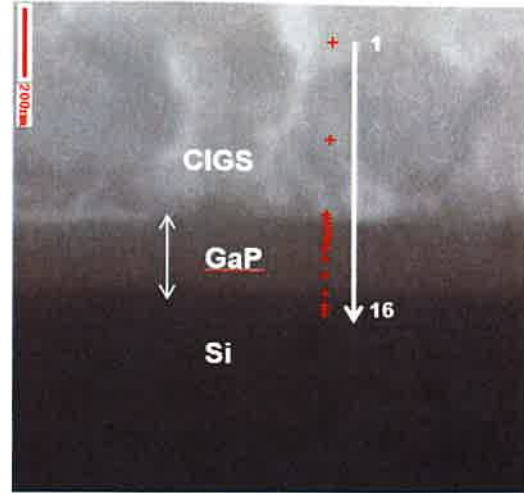


Figure 8. NanoAuger composition profiling across the cross section of a Si/GaP/CIGSe₂.

Cathodoluminescence experiments (C2N) show the presence of a sharp peak at about 1.68 eV on the CIGSe samples and high energies features related to possible high energy localized levels. As shown in figure 9, at room temperature the peak is sharper for the CGSe layer grown on the AlGaAs substrate, which is associated to the best spectral response. Some differences are thus observed for the different sample which may reflect different qualities of the layers.

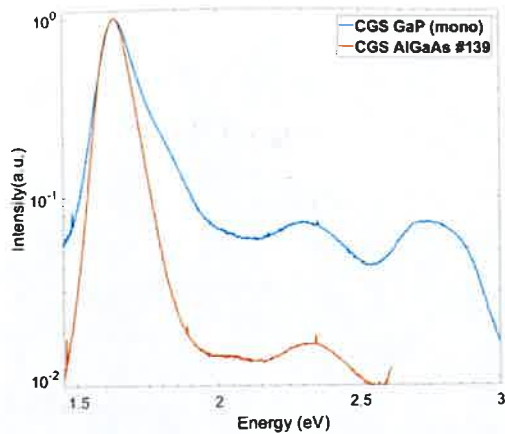


Figure 9. Cathodoluminescence spectra at room temperature for CGSe₂ layers on GaP and AlGaAs substrates.

5 CONCLUSION

A new concept for the realization of tandem cells between silicon and CIGS has been presented which consists in introducing a thin buffer layer of III-V layers between the two materials. First experiments seem to confirm the beneficial influence of this layer in the case of wide gap CGSe₂ absorber layers, in particular for GaAlAs buffers. Epitaxial growth has also been demonstrated. This is very promising for further development of this concept for efficient tandem cells combining silicon and CIGS. We can note that a recently published paper announced that an epitaxial cell of CuInGaSe₂ on GaAs (100) was able to deliver an efficiency of 20% [3]. This further supports the interest of developing III-V selective contact layers with CIGS and chalcogenides for high efficiency tandem solar cells.

6 ACKNOWLEDGEMENTS

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7 REFERENCES

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