Supporting Information for:

Defect characterisation in Cu$_2$ZnSnSe$_4$ kesterites via resonant Raman spectroscopy and impact onto optoelectronic solar cells properties

Mirjana Dimitrievska*1,2,3, Florian Oliva1, Maxim Guc1, Sergio Giraldo1, Edgardo Saucedo1, Alejandro Pérez-Rodríguez1,5, Victor Izquierdo-Roca*1

1 – Catalonia Institute for Energy Research (IREC), Jardins de les Dones de Negre 1, 08930 Sant Adrià de Besòs, Spain

2 – NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899-6102, United States

3 – National Renewable Energy Laboratory, Golden, CO 80401, United States

4 – Centres Científics i Tecnològics de la Universitat de Barcelona (CCiTUB), Lluís Solé i Sabarís 1-3, 08028 Barcelona, Spain

5 – IN²UB, Departament d’Electrònica, Universitat de Barcelona, C. Martí i Franquès 1, 08028 Barcelona, Spain

* Authors to whom correspondence should be addressed. E-mail: mirjana.dimitrievska@nist.gov, mirjana.dimitrievska@nrel.gov (M. Dimitrievska), vizquierdo@irec.cat (Victor Izquierdo)
**Figure S1.** Unprocessed Raman spectrum of the CZTSe material made under resonant Raman conditions with 1064 nm excitation wavelength. Clear contribution from the photoluminescence (PL) is observed, due to the bandgap electron excitations.
Figure S2. (top) Thin film (Sample 2) with lateral compositional gradients used for UV Raman measurements and defect identification. The surface of the samples has been etched to remove the presence of secondary phases. # Samples = 100. (Bottom) Compositional mapping of the Cu/(Zn+Sn) and Zn/Sn ratio of a 5 x 5 cm² graded kesterite film; each [column, row] corresponds to one cut sample cell. The compositional ratios of the Sample 2 are [Cu]/([Zn]+[Sn]) = 0.55-0.90, [Zn]/[Sn] = 1.1-2.0 based on the XRF measurements.
Figure S3. A comparison between the Raman spectra measured with 325 and 532 nm excitation, for CZTSe samples with different compositions. Strong enhancement in the Raman intensity of spectral regions around 176 and 250 cm\(^{-1}\) is observed for measurements made with 325 nm excitation. For clarity, spectra are normalized to 196 cm\(^{-1}\) peak intensity.
Figure S4. Intensity correlation between the Raman peaks located at 176 and 235 cm$^{-1}$ signalling similar origin effects responsible for the changes in the peak intensity.
Figure S5. Mappings of the optoelectronic properties with around 200 points showing device efficiency, open-circuit voltage ($V_{OC}$), short-circuit current ($J_{sc}$) and fill factor ($FF$).