

## Supporting Information for:

### **Defect characterisation in $\text{Cu}_2\text{ZnSnSe}_4$ kesterites via resonant Raman spectroscopy and impact onto optoelectronic solar cells properties**

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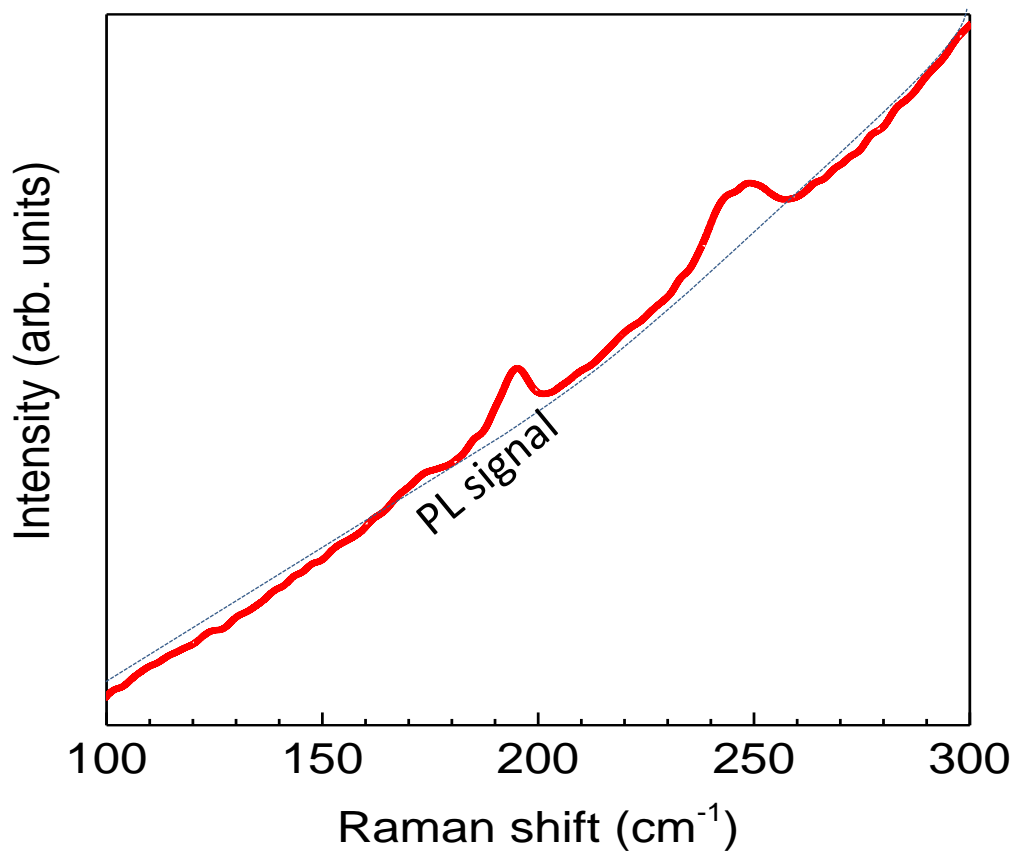
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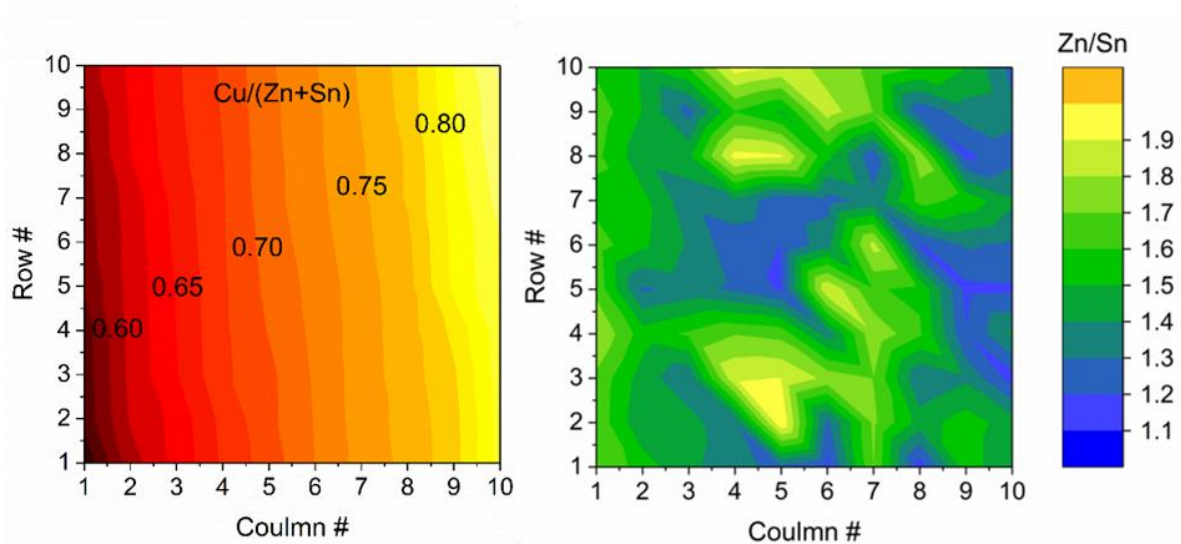
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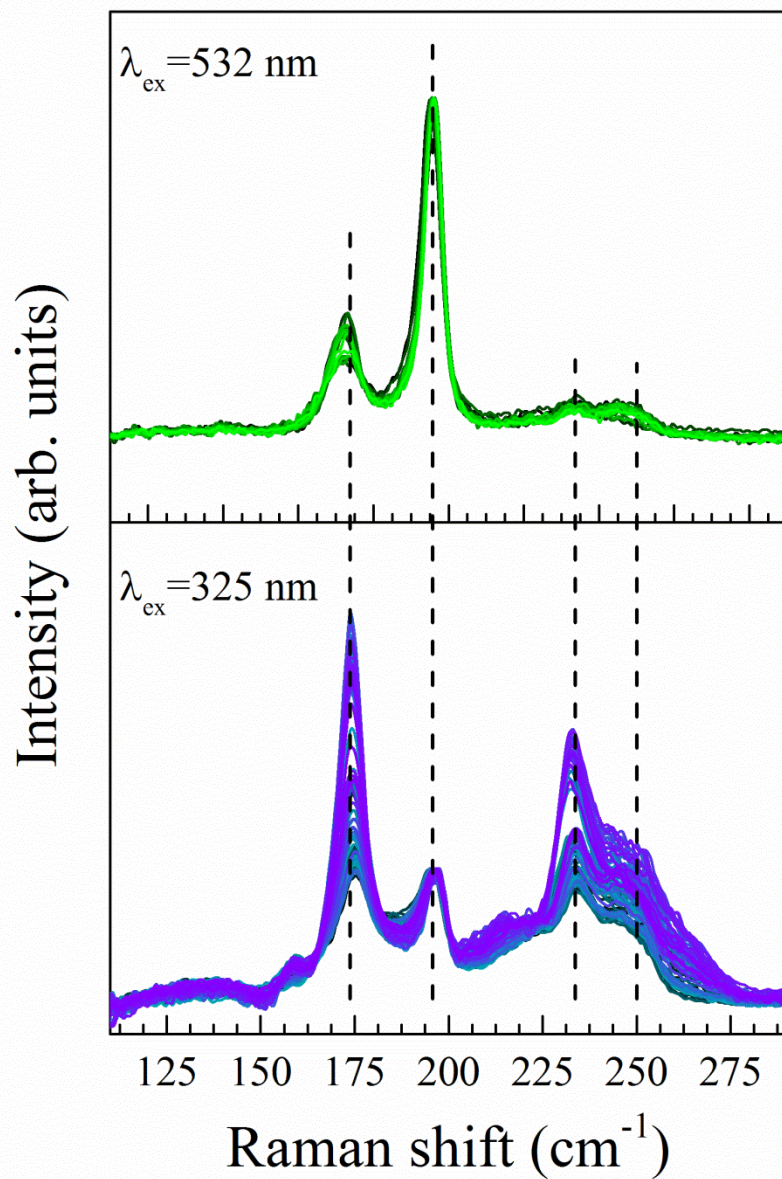
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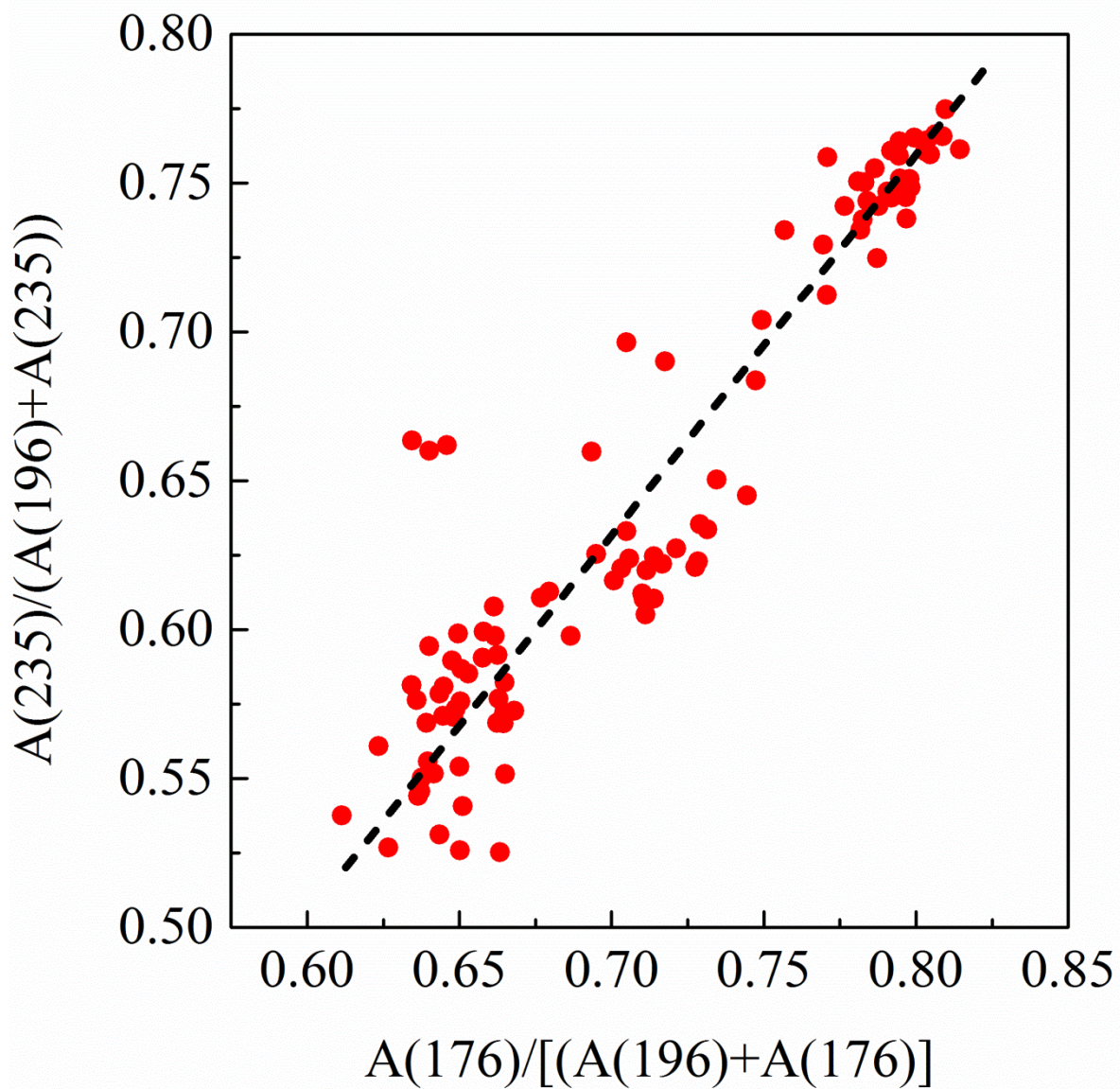
**Figure S1.** Unprocessed Raman spectrum of the CZTSe material made under resonant Raman conditions with 1064 nm excitation wavelength. Clear contribution from the photoluminescence (PL) is observed, due to the bandgap electron excitations.



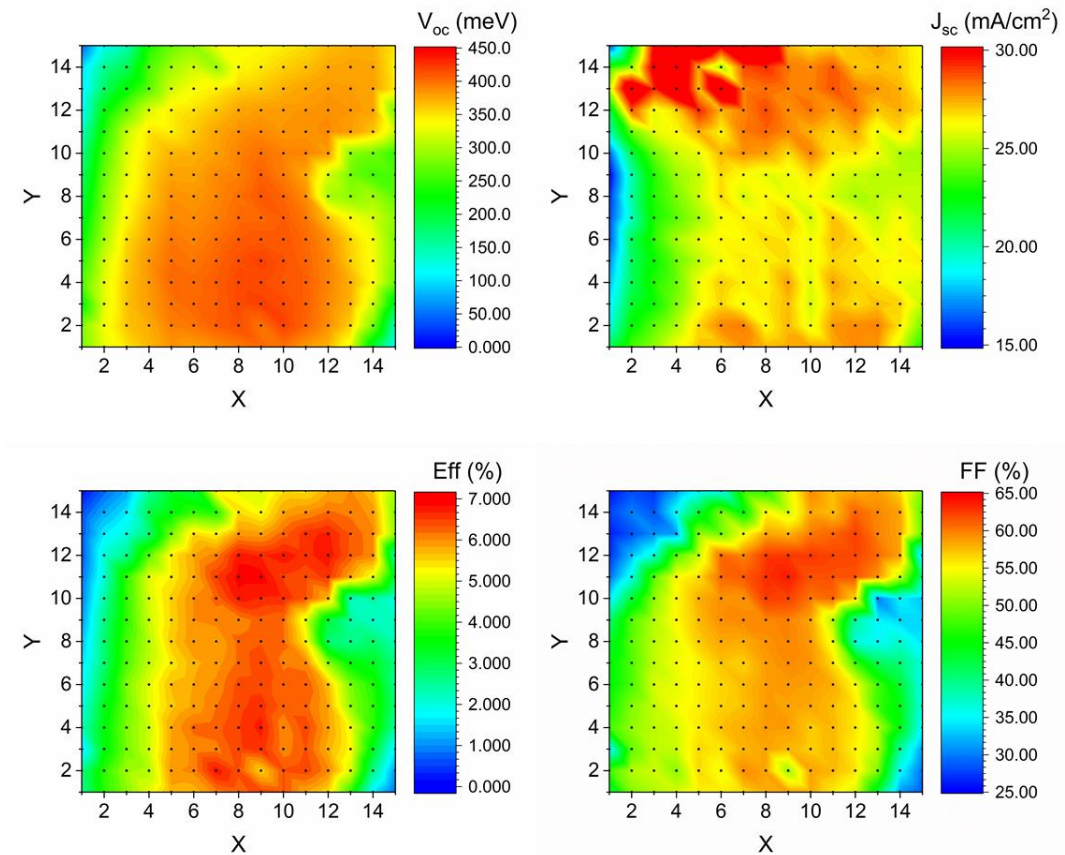
**Figure S2.** (top) Thin film (Sample 2) with lateral compositional gradients used for UV Raman measurements and defect identification. The surface of the samples has been etched to remove the presence of secondary phases. # Samples = 100. (Bottom) Compositional mapping of the Cu/(Zn+Sn) and Zn/Sn ratio of a 5 x 5 cm<sup>2</sup> 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  $\text{cm}^{-1}$  is observed for measurements made with 325 nm excitation. For clarity, spectra are normalized to 196  $\text{cm}^{-1}$  peak intensity.



**Figure S4.** Intensity correlation between the Raman peaks located at 176 and 235  $\text{cm}^{-1}$  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 ( $FF$ ).