Electronic Supplementary Information:

Prospects for defect engineering in Cu_2ZnSnS_4 solar absorber films

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Additional Maps used for secondary phase boundary analysis from Raman spectroscopy:

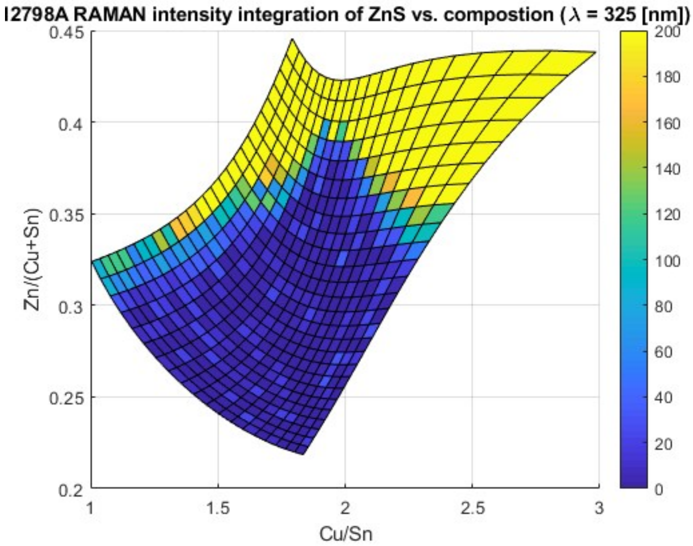

Figure S1 Intensity of Raman ZnS mode for sample A.

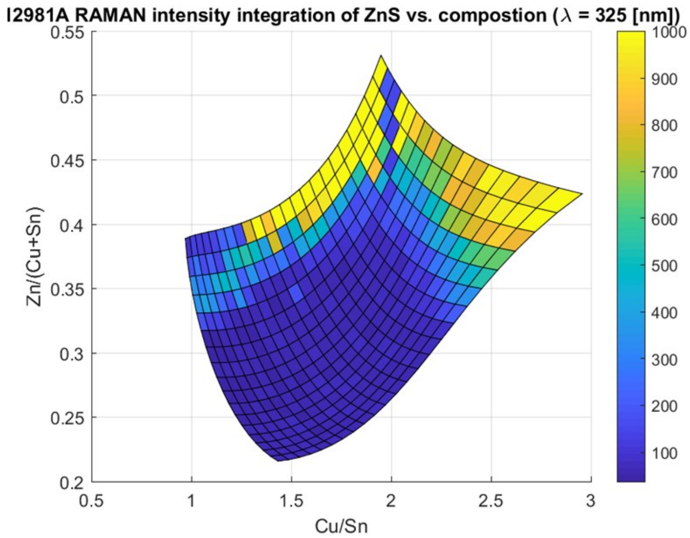

Figure S2 Intensity of Raman ZnS mode for sample B.

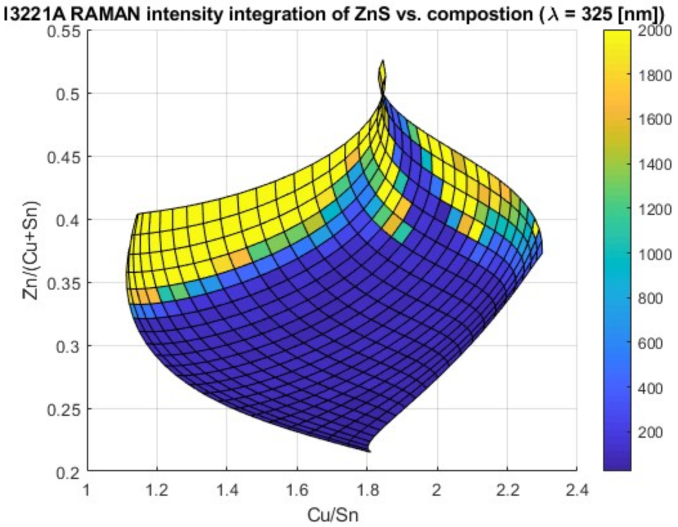

Figure S3 Intensity of Raman ZnS mode for sample C.

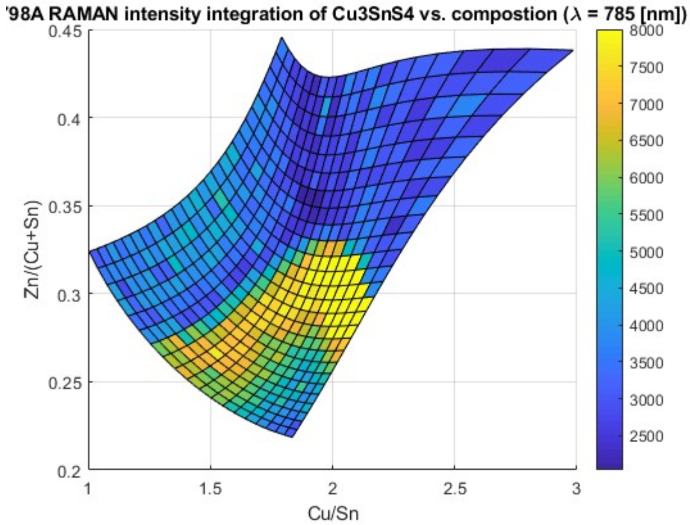

Figure S4 Intensity of Cu_3SnS_4 Raman mode of sample A.

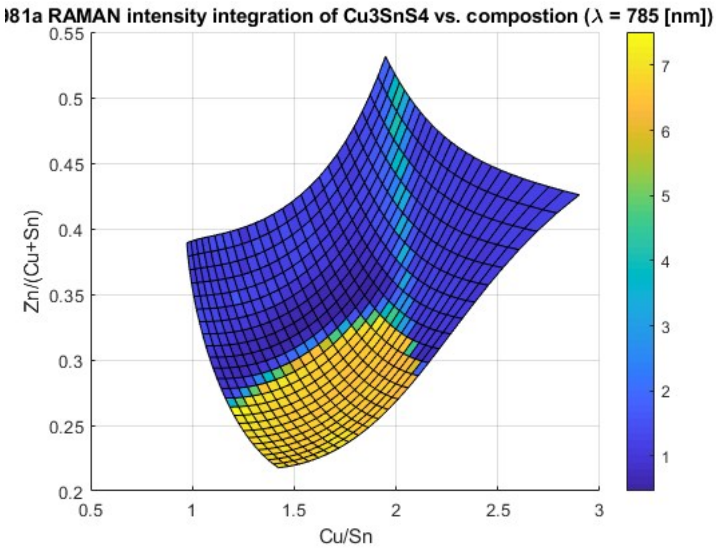

Figure S5 Intensity of Cu_3SnS_4 Raman mode of Sample B.

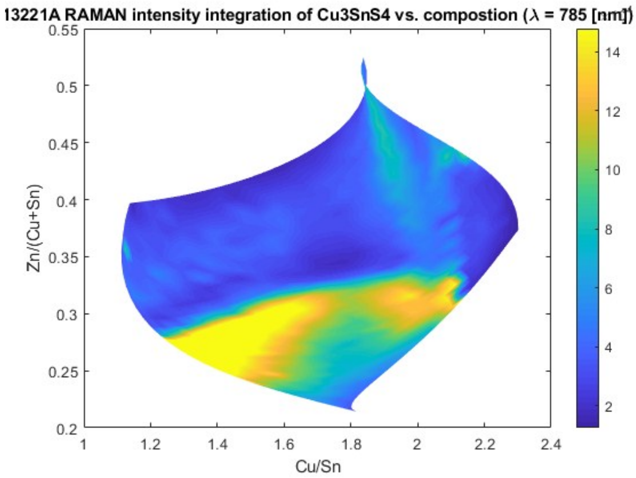

Figure S6 Intensity of Raman Cu_3SnS_4 mode for sample C.

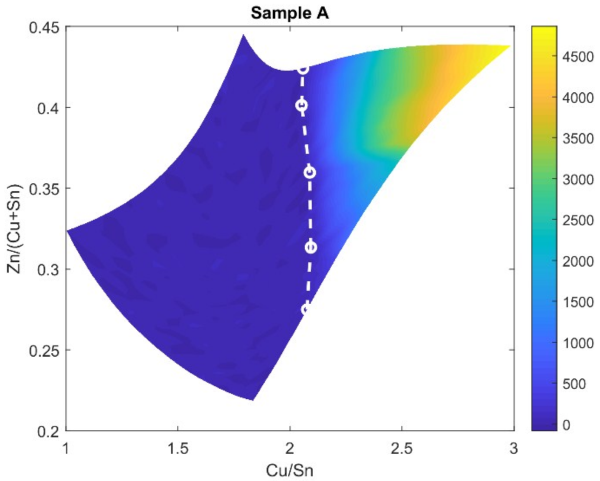

Figure S7 Intensity of Raman CuS mode for sample A.

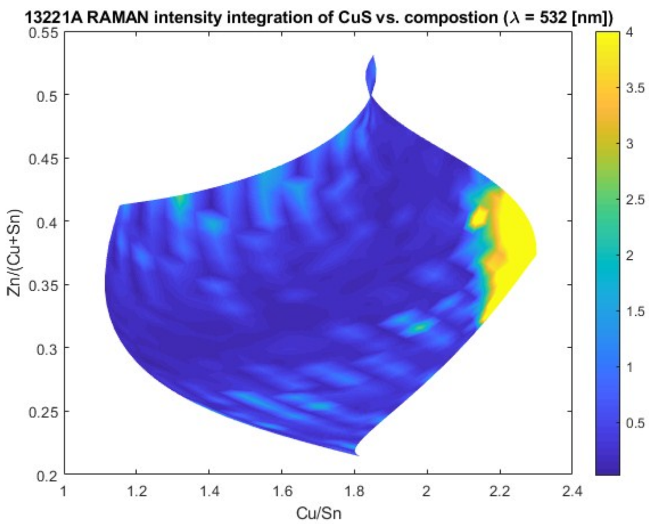

Figure S8 Intensity of Raman CuS mode for sample C.

Sketch illustrating GIXRD line scans

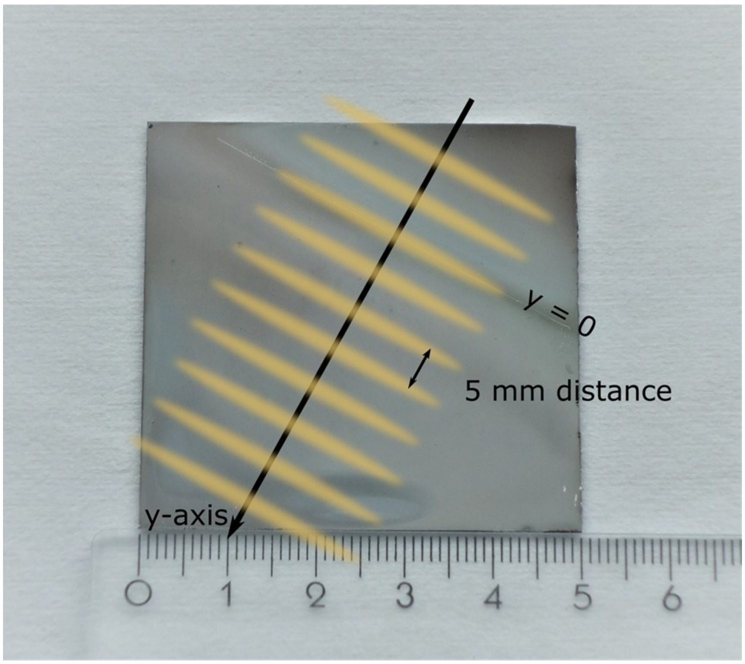

Figure S9 Sketch of the GIXRD line scan performed for SnS_x phase boundary analysis on CS samples.The Cu/Sn=2 line is along the line denoted with y=0. Each GIXRD scan covered an elongated area of the SC samples as indicated by the yellow areas.

STEM-EDX maps of Samples A-C

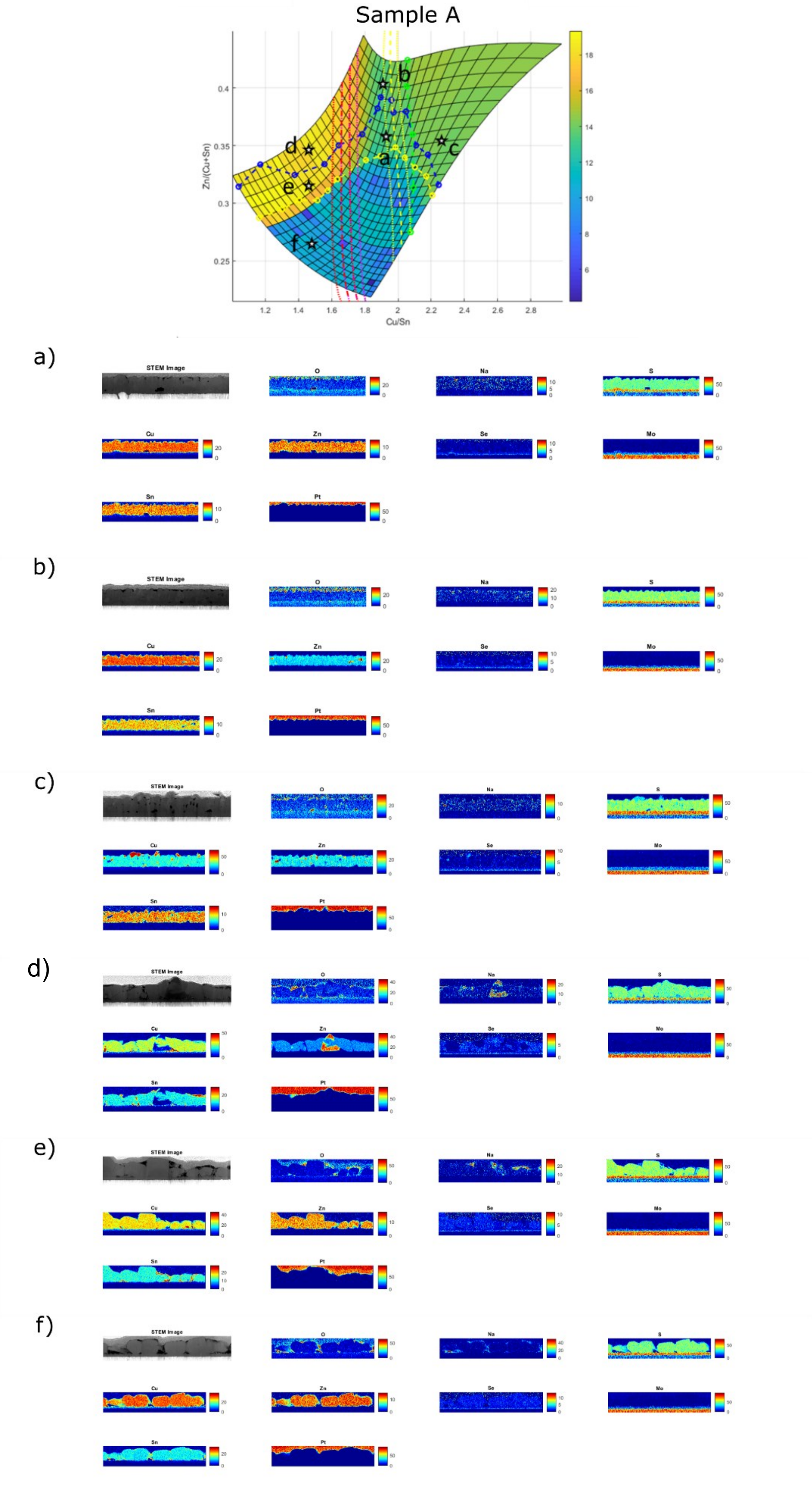

Figure S10 STEM-EDX maps for sample A at positions a – f.

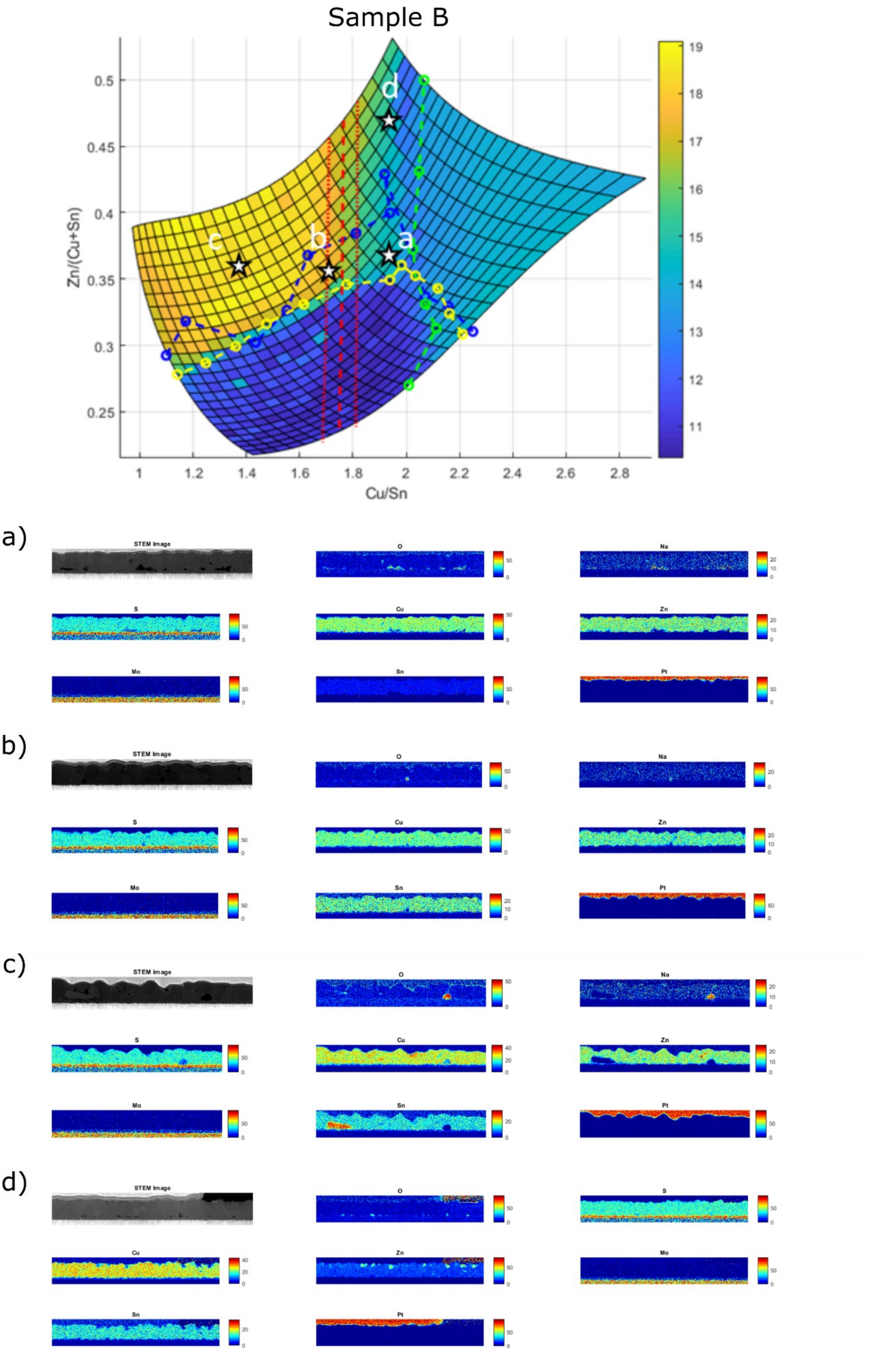

Figure S11 STEM-EDX maps of sample B for positions a – d.

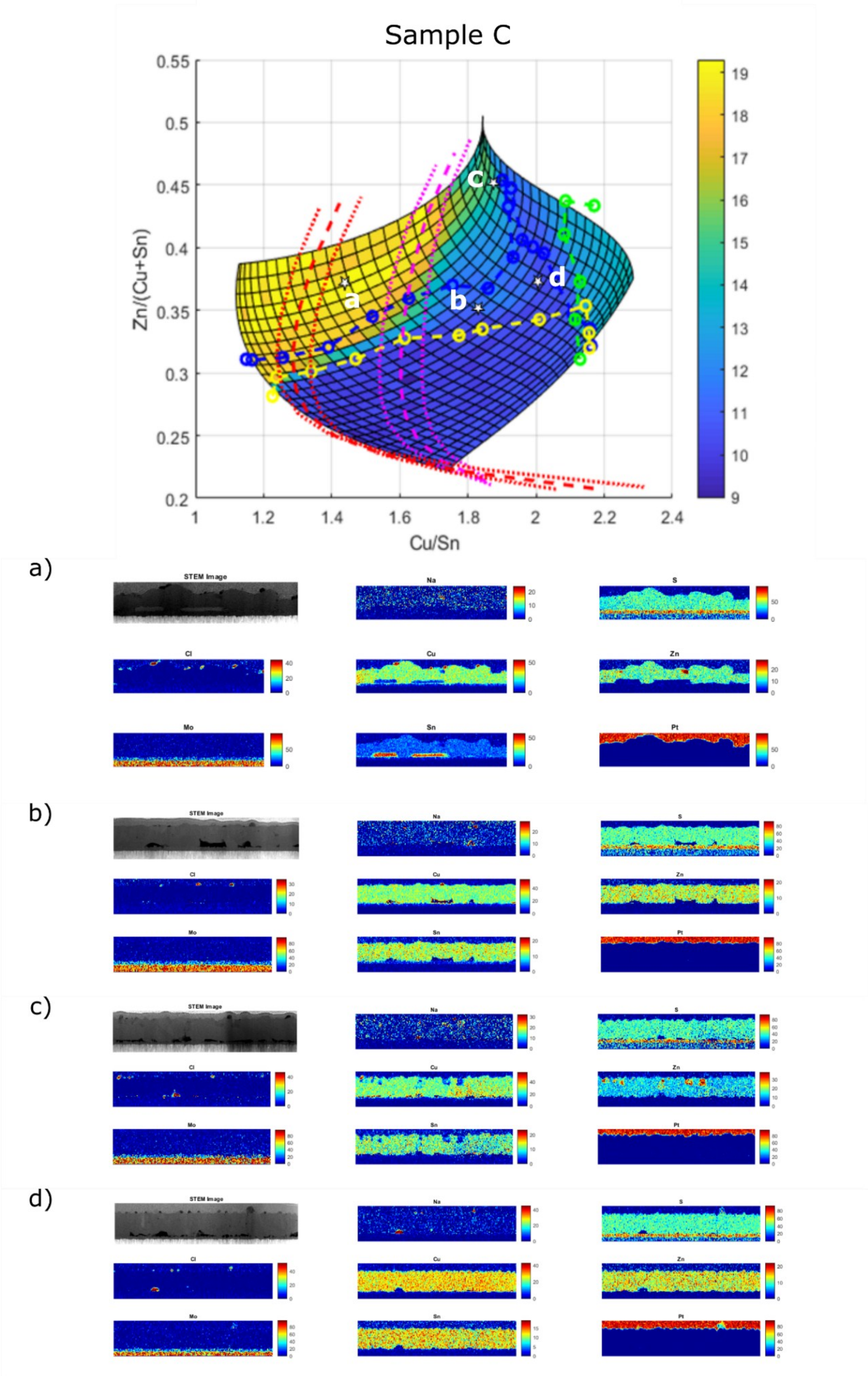

Figure S12 STEM-EDX maps of sample C for positions a – d.

STEM bright field images of Samples B and C

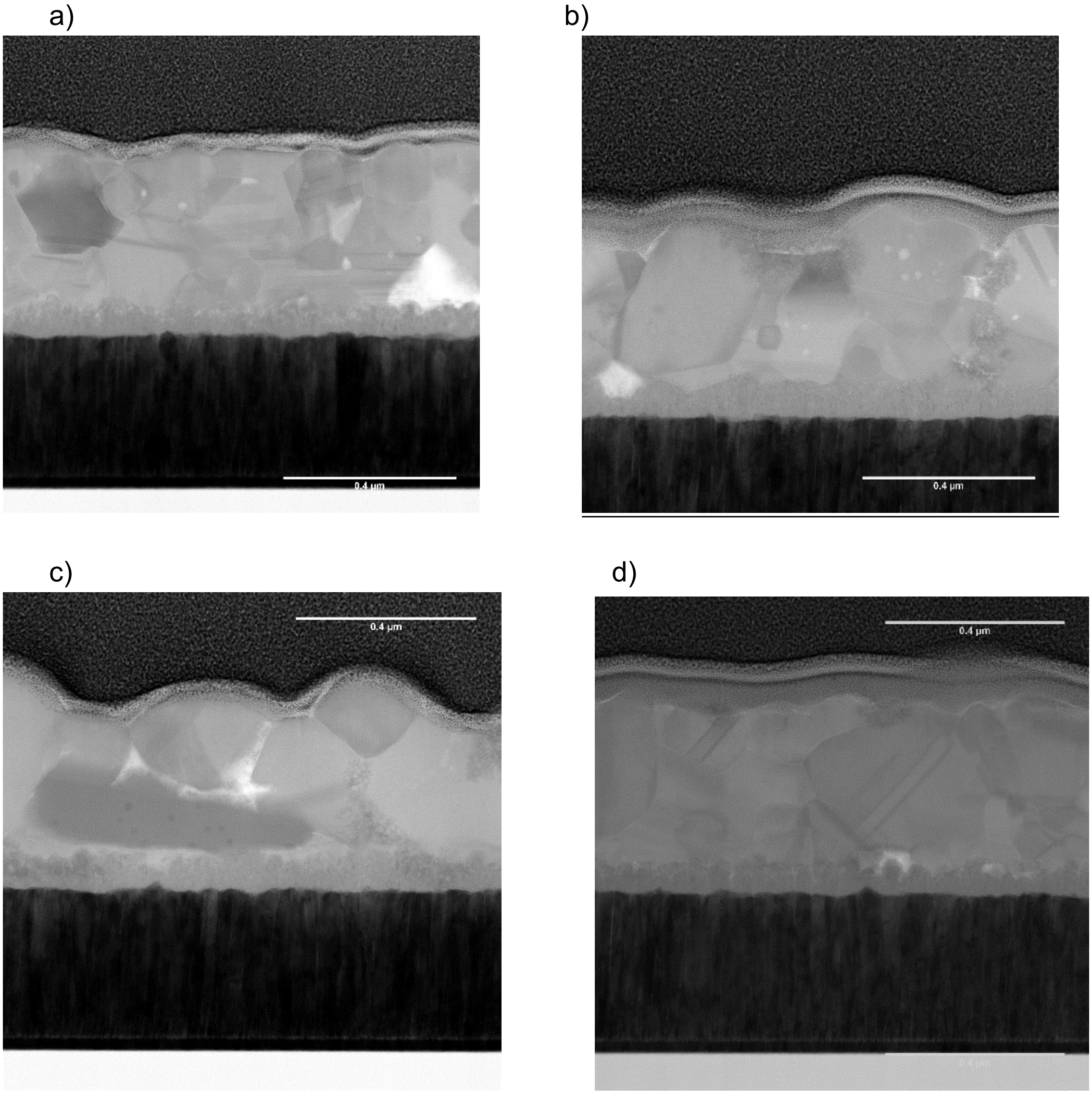

Figure S13 STEM bright field images of Sample B at positions a – d (see Figure S10 for sample overview).

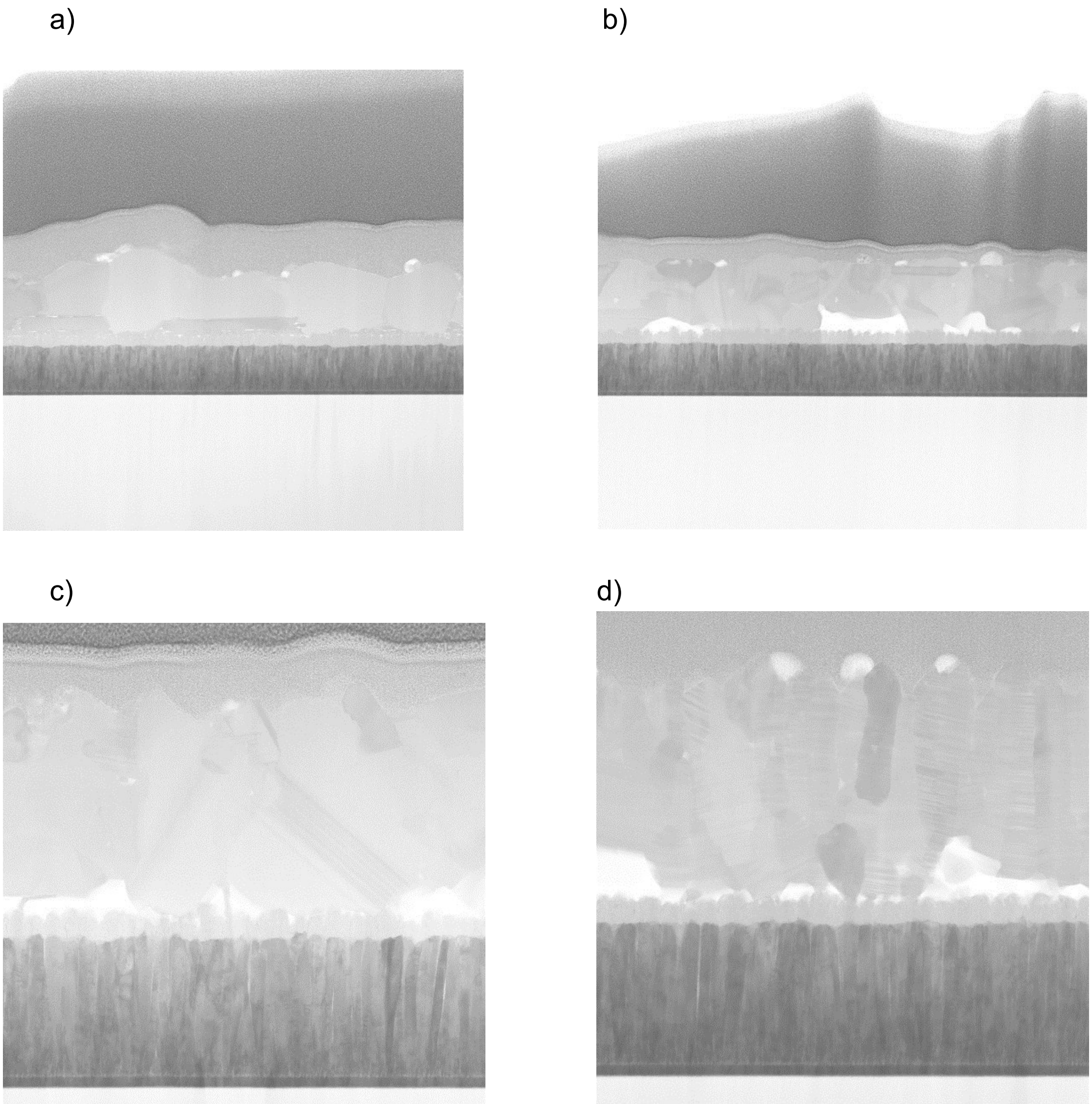

Figure S14 STEM bright field images of Sample C at positions a – d (see Figure S11 for sample overview).

Analysis of FWHM of peak at 337cm^-1 from Raman spectra

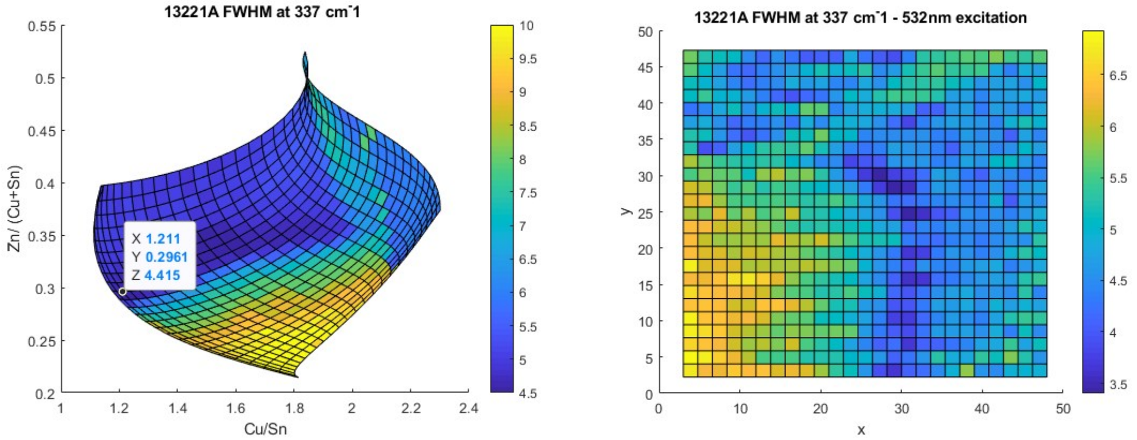

Figure 15 The FWHM for the CZTS peak at 337cm^-1 from Raman spectra measured with excitation wavelength of 785nm (left) and 532nm (right).

The full width half maximum (FWHM) can be used as a measure of the quality of the crystal structure. In the case of cation disorder we expect that the FWHM is smallest when cation disorder is less and the Q parameter is high. The FWHM of the CZTS peak at 337cm^{-1} shows the expected trends for Raman spectra measured under 785nm and under 532nm illumination. This confirms, that the trends observed from the Q parameter for different compositions reflect changes in the crystal quality.