Supporting Information for:

Stoichiometry modulates the optoelectronic functionality of Zinc Phosphide ($Zn_{3-x}P_{2+x}$)

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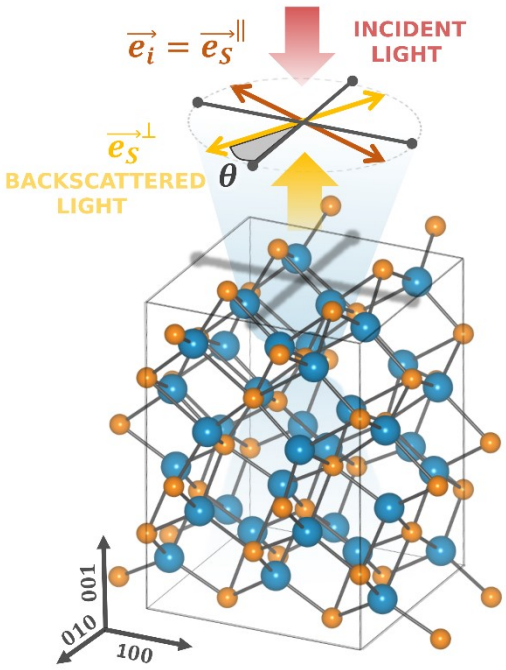

Figure S1. Schematic representation of the Raman scattering polarization measurements. Raman measurements are performed on the (001) basal plane with incident laser light (wide red arrow) projected along the -*z*-axis ([001] direction). The incident light polarization and the output polarization in perpendicular configuration are represented by the thin red and yellow arrows, respectively. The black arrows indicate the *x* and *y* axis of the crystal corresponding to [100] and [010] directions, respectively. The polarization angle $\theta = \pi/4$ was used for all three monocrystalline zinc phosphide thin films.

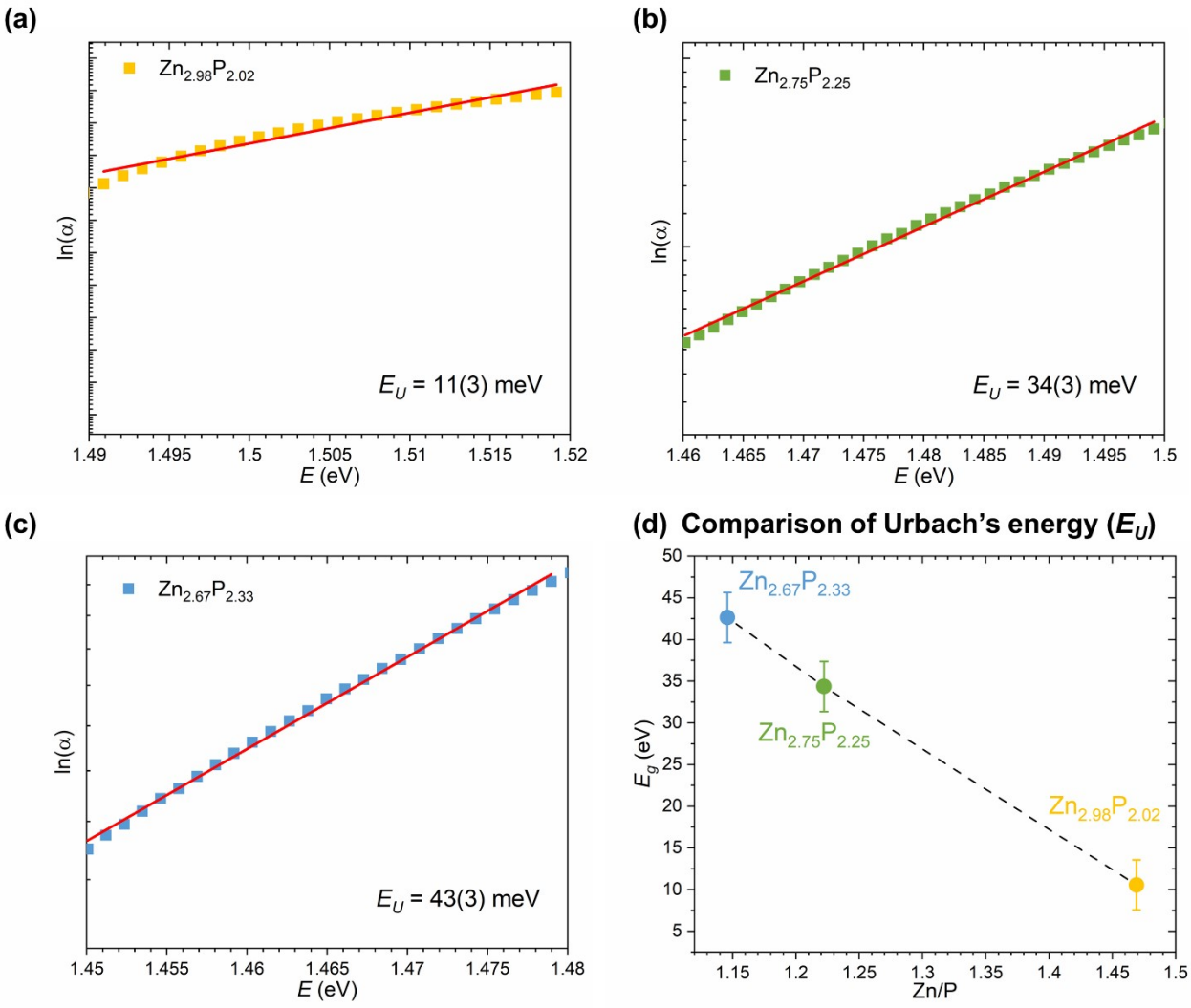

Figure S2. Urbach energy calculation from absorption measurements for samples with various compositions: (a) $Zn_{2.98}P_{2.02}$ (Zn/P = 1.47), (b) $Zn_{2.75}P_{2.25}$ (Zn/P = 1.22) and (c) $Zn_{2.67}P_{2.33}$ (Zn/P = 1.15). (d) Comparison of Urbach energy with composition.