Supplemental Information

## Reactive phosphine combinatorial co-sputtering of cation disordered ZnGeP<sub>2</sub> films

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Fig S1. Representative XRD data for the amorphous films reported in this work.



Fig S2. Representative Pawley refinement for this work.



S3. Defect formation energy plots corresponding to points 1 through 5 shown in the chemical potential stability phase diagram in the top left. The Fermi energy is referenced to the valence band maximum. The equilibrium Fermi energies were calculated at 400°C and are shown with the gray dashed line.



S4. Defect concentration diagram depicting the concentration of the primary point defects, calculated at 1000K, along the five points in the chemical potential stability diagram shown in S3. The trend in defect concentration matches that shown in Fig. 7 (b) of the main text.