SUPPLEMENTARY MATERIAL

Physical properties of new ordered bimetallic phases $M_{0.25}Cd_{0.75}PS_3$ (M = Zn^{II}, Ni^{II}, Co^{II}, Mn^{II}).

P. Fuentealba*, C. Olea, H. Aguilar-Bolados, N. Audebrand, R. C. de Santana, C. Doerenkamp, H. Eckert, C. J. Magon, E. Spodine.

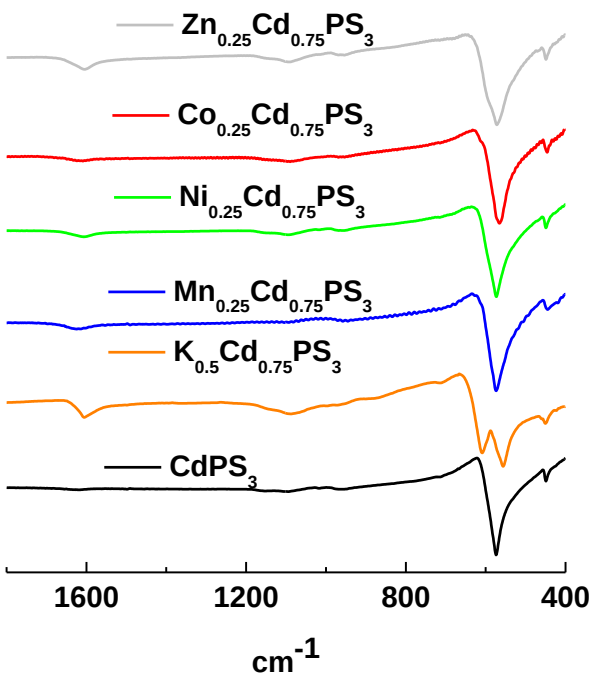

Fig. S1. FTIR

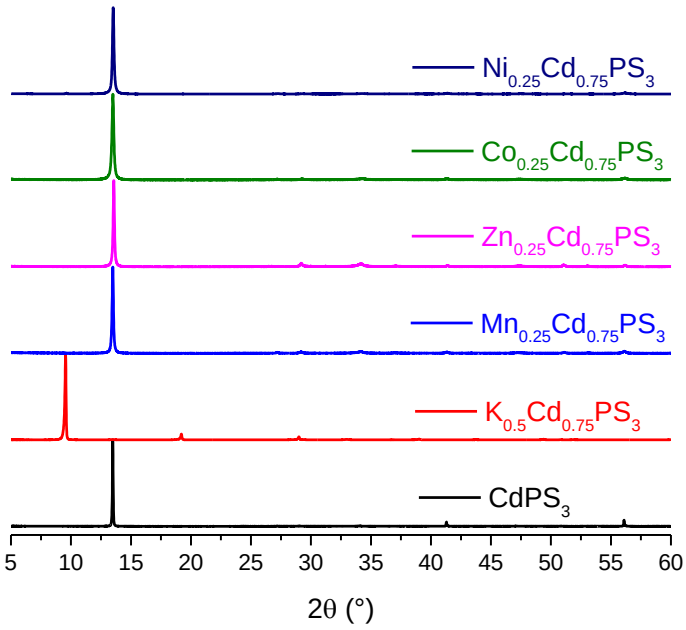

Fig. S2. Powder X-ray diffractograms of the lamellar phases.

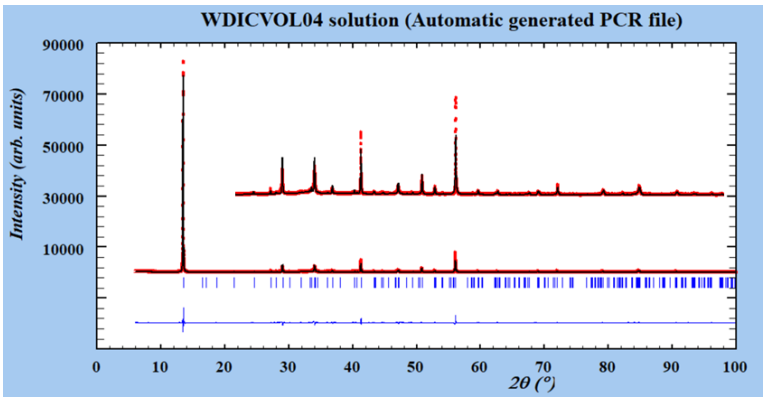

Fig. S2a: Refined powder X-ray diffractogram of $CdPS_3$

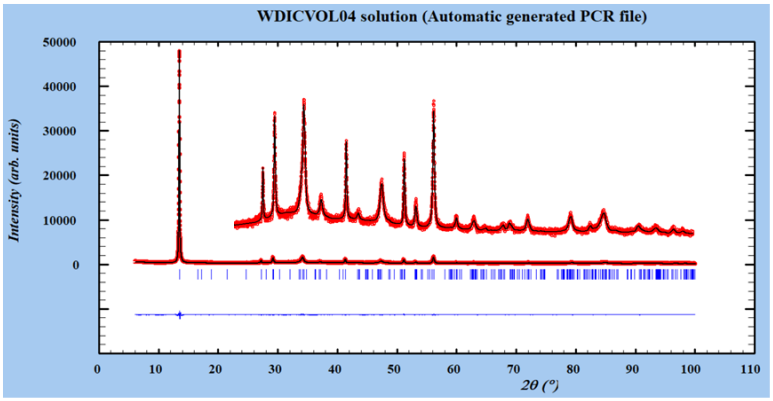

Fig. S2b: Refined powder X-ray diffractogram of $Mn_{0.25}Cd_{0.75}PS_3$

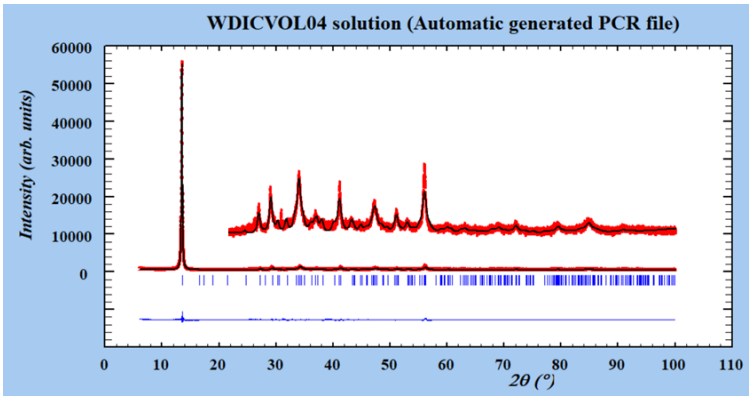

Fig. S2c: Refined powder X-ray diffractogram of $Co_{0.25}Cd_{0.75}PS_3$. Unkown small impurity line at 30.2.

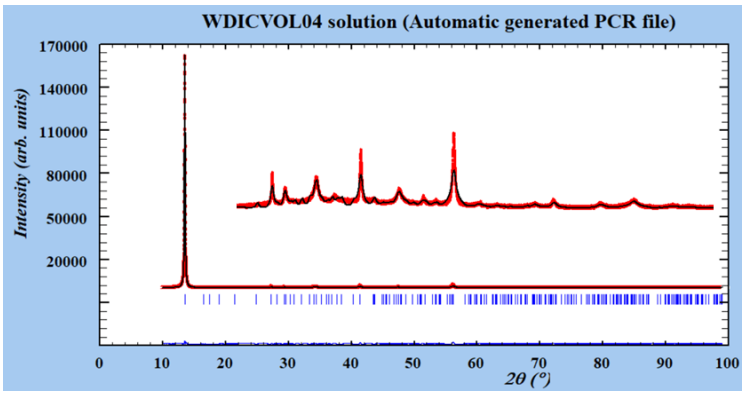

Fig. S2d: Refined powder X-ray diffractogram of $Ni_{0.25}Cd_{0.75}PS_3$

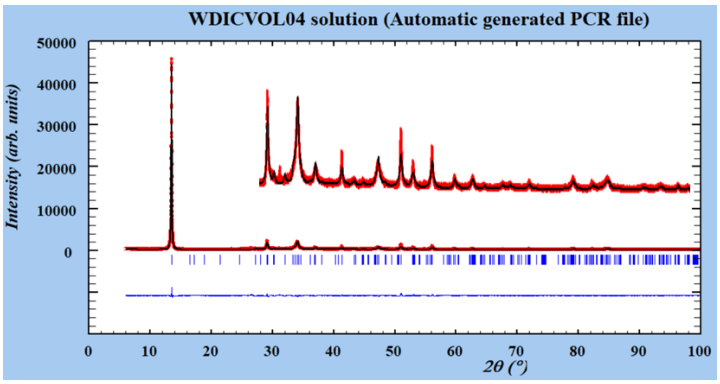

Fig. S2e: Refined powder X-ray diffractogram of $Zn_{0.25}Cd_{0.75}PS_3$. Unkown small impurity line at 30.2.

	$CdPS_3$	$Mn_{0.25}Cd_{0.75}PS_3$	$Co_{0.25}Cd_{0.75}PS_3$	$Ni_{0.25}Cd_{0.75}PS_3$	$Zn_{0.25}Cd_{0.75}PS_3$
R_{exp}	5.87	5.10	3.70	3.94	5.59
Chi^2	7.41	1.97	3.51	9.20	4.55
Rp	11.0	5.17	5.02	6.21	8.06
Rwp	16.0	7.17	6.93	11.9	11.9

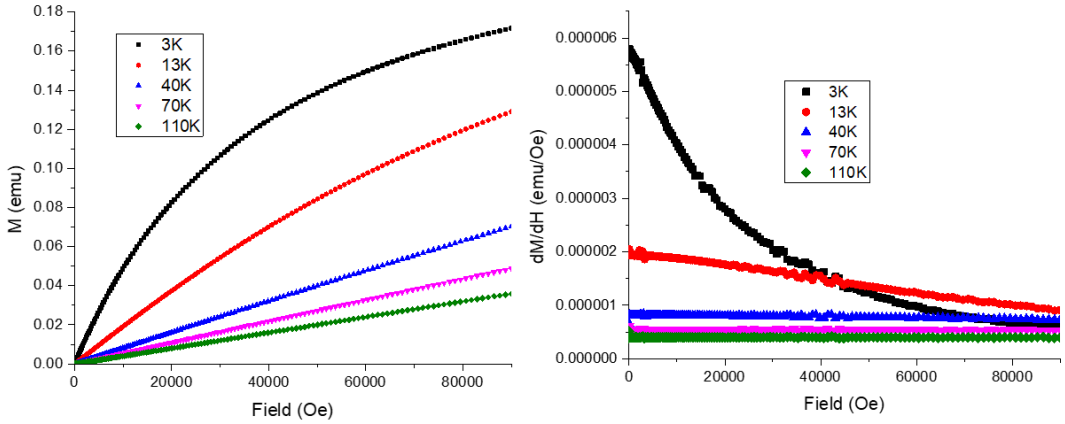

Fig. 3a. M(H) and first derivate curves for $Mn_{0.25}Cd_{0.75}PS_3$.

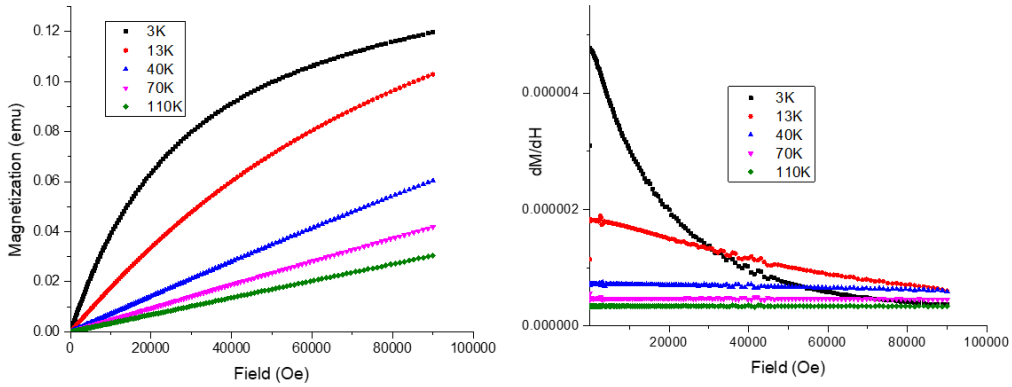

Fig S3b. M(H) and first derivate curves for $Co_{0.25}Cd_{0.75}PS_3$.

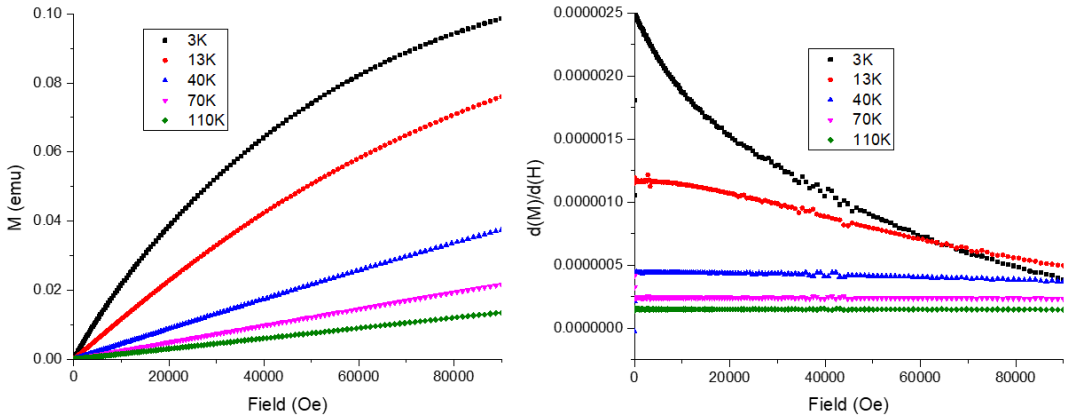

Fig. S3c. M(H) and first derivate curves for $Ni_{0.25}Cd_{0.75}PS_3$.

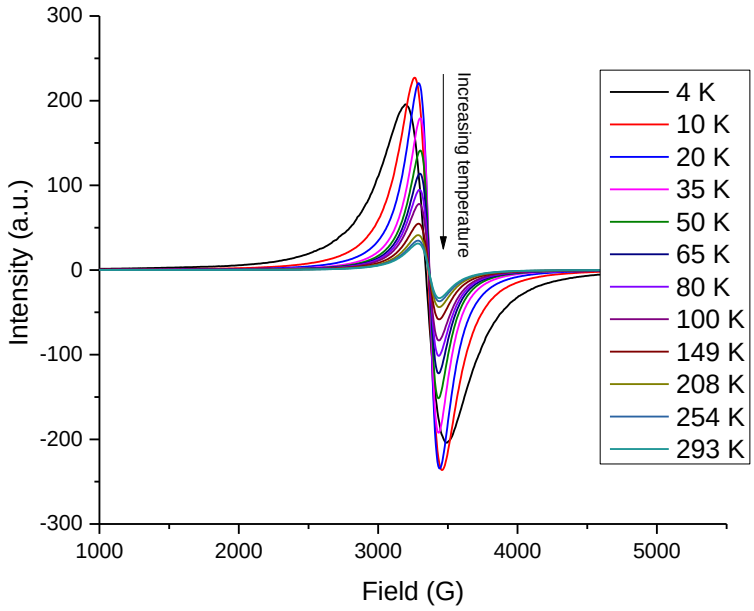

Fig. S4. EPR spectra of the $Mn_{0.25}Cd_{0.75}PS_3$. phase.

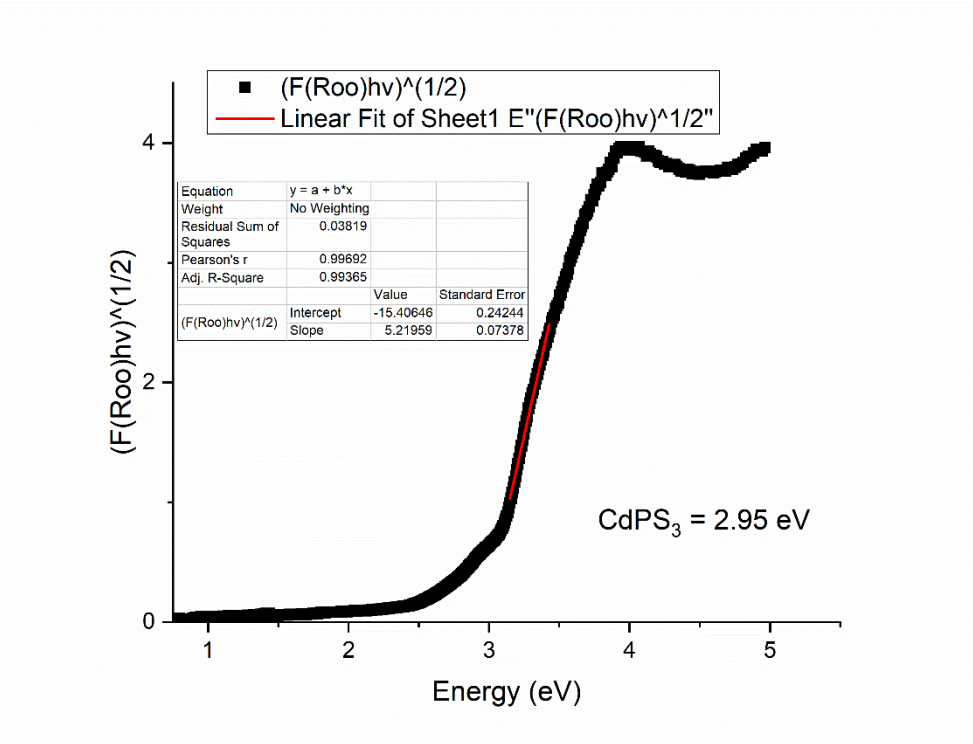

Fig. S5a Tauc plots for $CdPS_3$

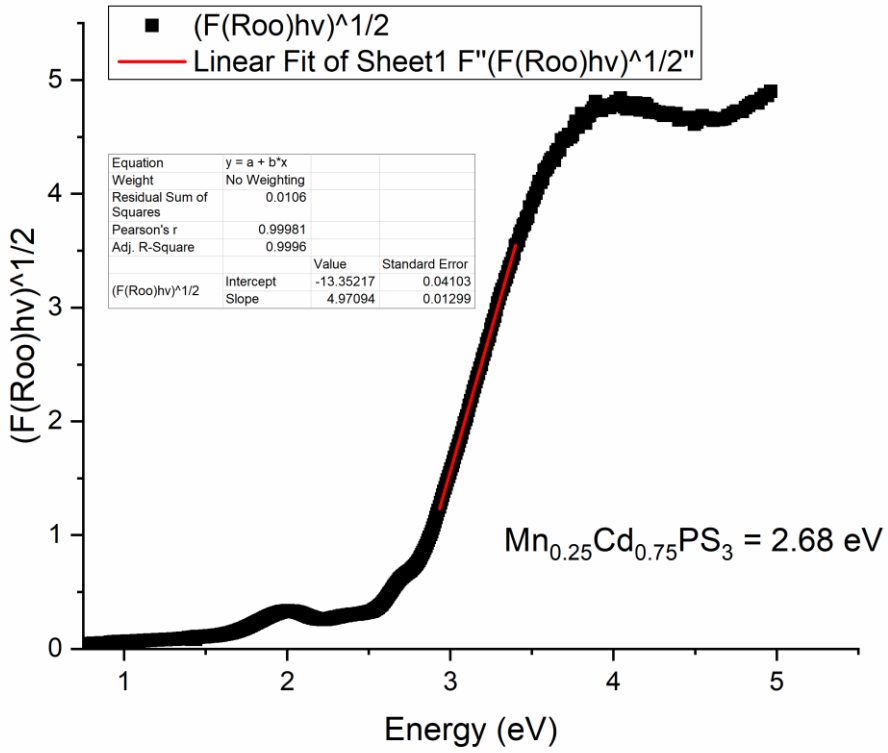

Fig. S5b Tauc plots for $Mn_{0.25}Cd_{0.75}PS_3$

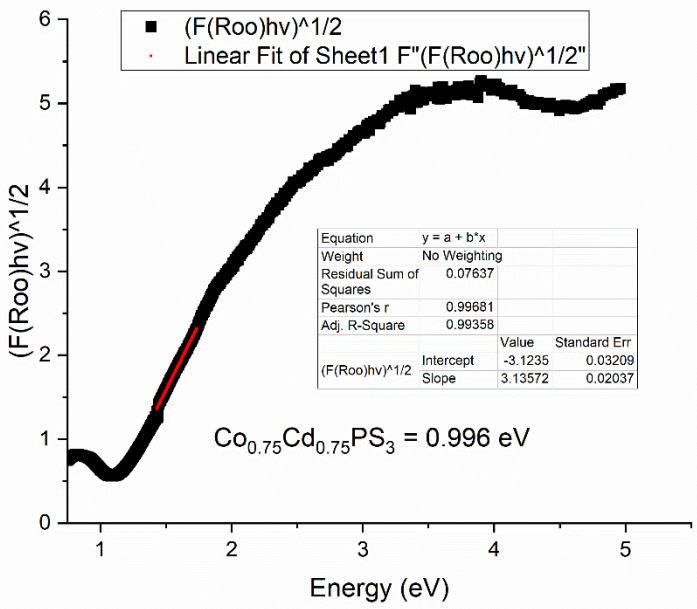

Fig. S5c Tauc plots for $Co_{0.25}Cd_{0.75}PS_3$

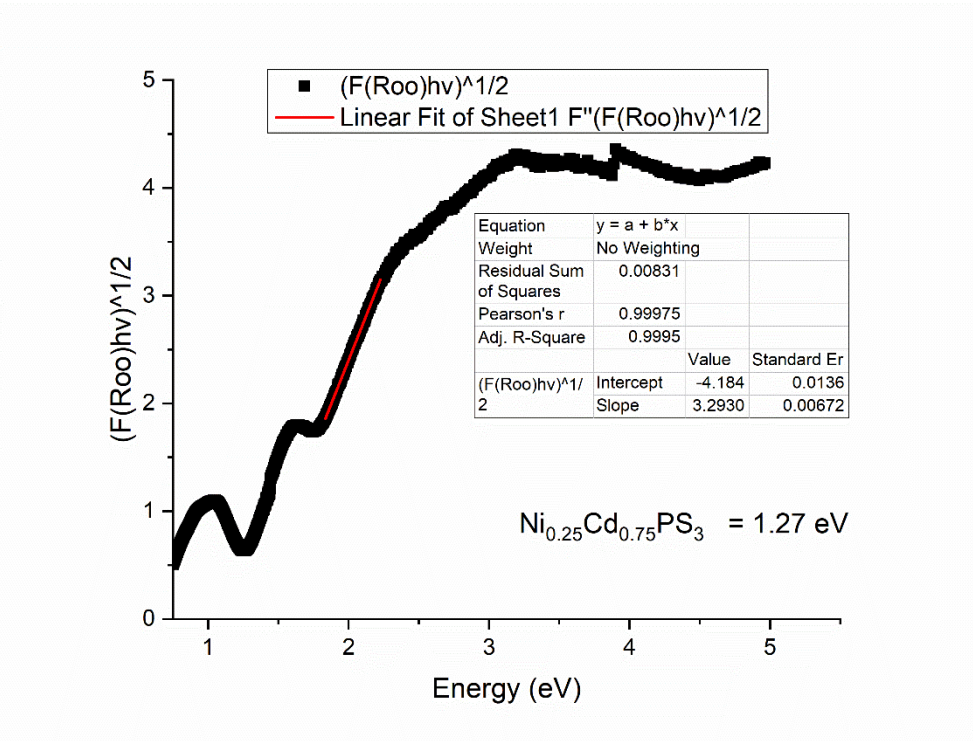

Fig. S5d Tauc plots for $Ni_{0.25}Cd_{0.75}PS_3$

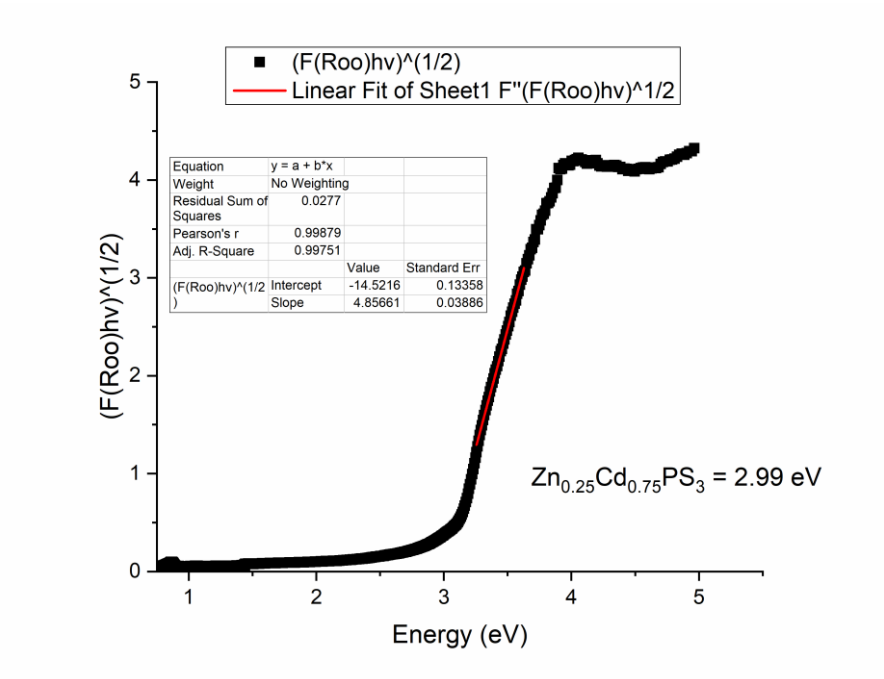

Fig. S5e Tauc plots for $Zn_{0.25}Cd_{0.75}PS_3$

Table 1: Ionic radius [1].

Element (six coordination sphere)	Ionic radii (pm)
Cd^{II}	109
K^{I}	138
Mn^{II}	97
Co^{II}	89
Ni^{II}	83
Zn^{II}	88

Equations for magnetic fittings:

$$\chi = \frac{C(x)}{T-\Theta(x)} \quad (1)$$

where the Curie constant, expressed in (emu K mol^{-1}) is given by

$$C(x) = \frac{xN_A g^2 \mu_B^2 S(S+1)}{3k_B} \quad (2)$$

Table S2. The Rushbrook and Wood b_i coefficients calculated for honeycomb lattices with S = 5/2, 3/2 and 1, and x = 0.25.

S	b_1	b_2	b_3	b_4	b_5	b_6
5/2	-17.5	4.192	-8.720	-7.706	-30.723	-86.899
3/2	-1.875	2.111	-1.496	-0.375	1.835	0.008
1	-1	0.83	-0.511	-0.035	0.264	-0.201

EPR Fittings

The spectrum was simulated by a sum of three components: an isotropic broad line and two axial sharper lines. S denote the spin value of each paramagnetic specie. The perpendicular and parallel values of the g-tensor are denoted by $g_\perp$ and $g_{/\!/}$, respectively. lwpp[G L] stands for the [Lorentzian Gaussian] line width for isotropic magnetic-field domain broadening (peak-to-peak, in Gauss). HStrain denote the Gaussian residual line width (full width at half height, in MHz), describing broadening due to unresolved hyperfine couplings. weight is a software parameter used to scale a multiple component spectrum. A linear baseline correction was used.

References.

1 G. L. Miessler, P. J. Fischer and D. a Tarr, *Inorganic Chemistry*, Fifth Edit., 2013.