Table S1. Total and atomic magnetic moments for cubic L2$_1$ Pd$_2$CrZ (Z = Al, Ga, In, Tl, Si, Sn, P, As, Sb, Bi, Se, Te, Zn).

Compounds X$_2$YZ	Mt (μ_B/f.u.)	MCr (μ_B)	MZ (μ_B)	MPd-1 (μ_B)	MPd-2 (μ_B)
Pd$_2$CrAl	3.50	3.25	-0.02	0.13	0.13
Pd$_2$CrGa	3.53	3.30	-0.04	0.13	0.13
Pd$_2$CrIn	3.69	3.46	-0.04	0.10	0.10
Pd$_2$CrTl	3.66	3.51	-0.03	0.09	0.09
Pd$_2$CrSi	3.33	3.22	-0.04	0.07	0.07
Pd$_2$CrSn	3.55	3.48	-0.06	0.06	0.06
Pd$_2$CrP	3.22	3.27	-0.57	0.003	0.003
Pd$_2$CrAs	3.40	3.41	-0.46	0.018	0.018
Pd$_2$CrSb	3.56	3.55	-0.04	0.02	0.02
Pd$_2$CrBi	3.74	3.69	-0.04	0.04	0.04
Pd$_2$CrSe	3.56	3.58	-0.06	0.02	0.02
Pd$_2$CrTe	3.71	3.70	-0.05	0.02	0.02
Pd$_2$CrZn	3.79	3.38	-0.001	0.20	0.203.61

Table S2. Total and atomic magnetic moments, and c/a ratio for tetragonal $L1_0$ Pd_2CrZ (Z = Al, Ga, In, Tl, Si, Sn, P, As, Sb, Bi, Se, Te, Zn).

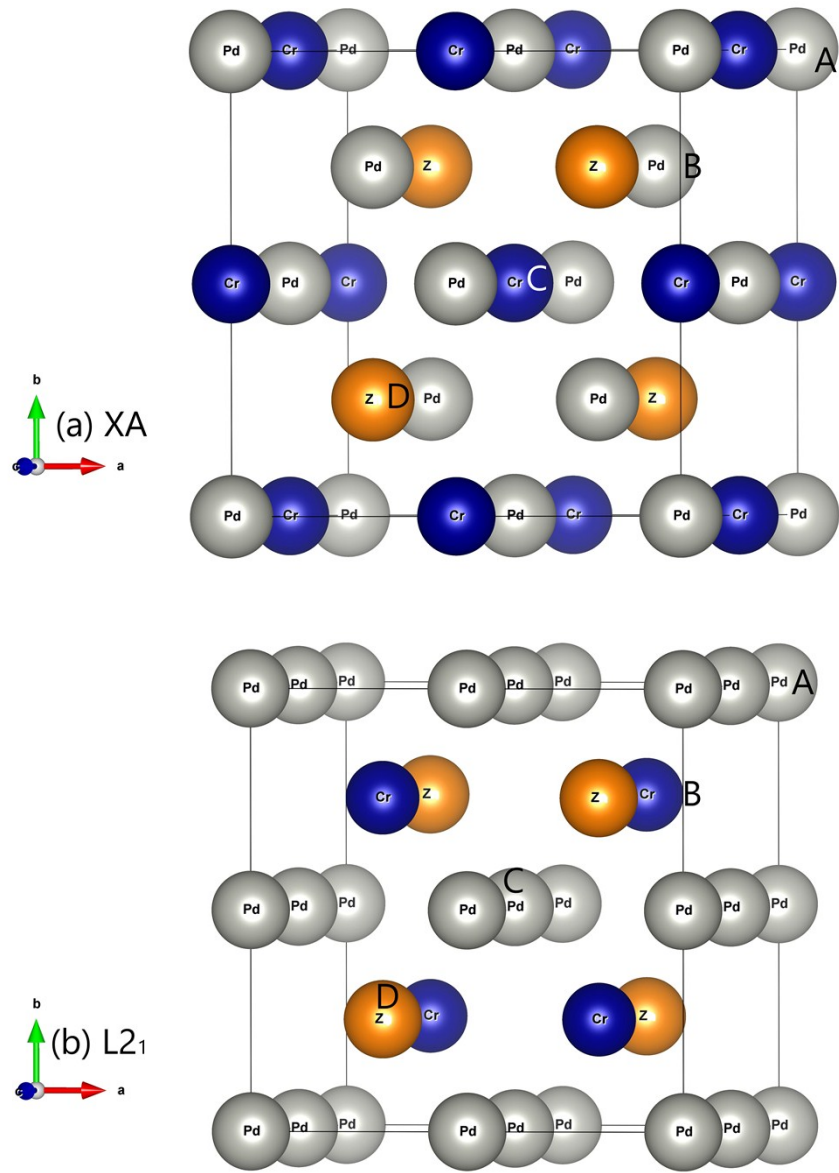| Compounds X_2YZ	Mt (μ_B/f.u.)	MCr (μ_B)	MZ (μ_B)	MPd-1 (μ_B)	MPd-2 (μ_B)	c/a
Pd_2CrAl	3.55	3.23	-0.02	0.17	0.17	1.21
Pd_2CrGa	3.52	3.27	-0.03	0.15	0.15	1.26
Pd_2CrIn	3.82	3.53	-0.01	0.15	0.15	1.22
Pd_2CrTl	3.75	3.50	-0.01	0.13	0.13	1.28
Pd_2CrSi	3.16	3.09	-0.06	0.06	0.06	1.21
Pd_2CrSn	–	–	–	–	–	–
Pd_2CrP	3.02	3.10	0.01	-0.11	-0.11	1.43
Pd_2CrAs	3.14	3.21	-0.1	0.02	0.02	1.43
Pd_2CrSb	3.41	3.46	-0.075	0.009	0.009	1.18
Pd_2CrBi	–	–	–	–	–	–
Pd_2CrSe	3.42	3.47	-0.104	0.02	0.02	1.45
Pd_2CrTe	3.67	3.62	-0.06	0.05	0.05	1.35
Pd_2CrZn	3.49	3.24	-0.009	0.12	0.12	1.22

Fig. S1. Crystal structures of XA (a) and L2$_1$ (b)-type Heusler alloys Pd$_2$CrZ.

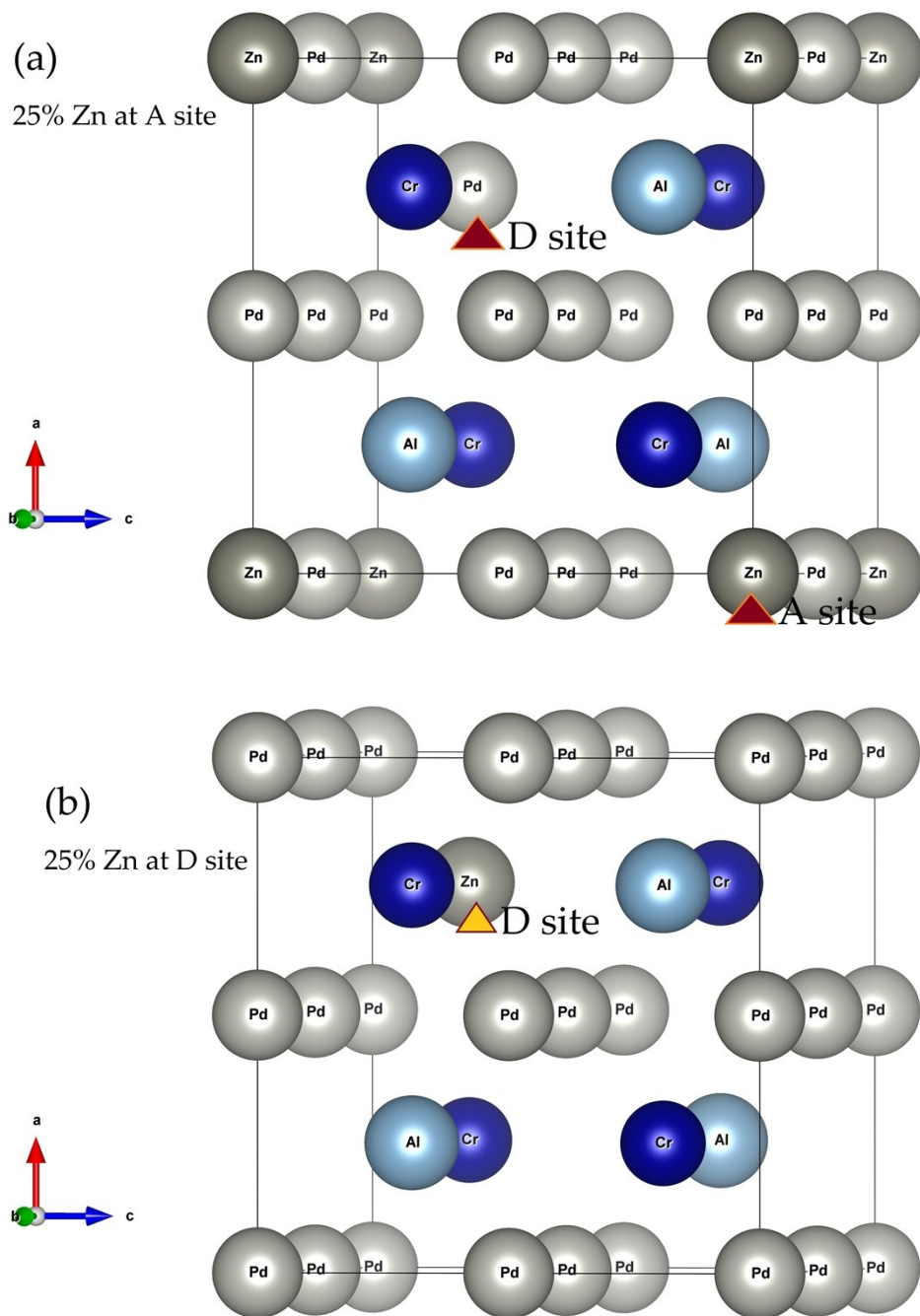Fig. S2. Crystal structures of Pd$_2$CrAl doping with 25 % Zn at Wyckoff sites (a) A or (b) D.
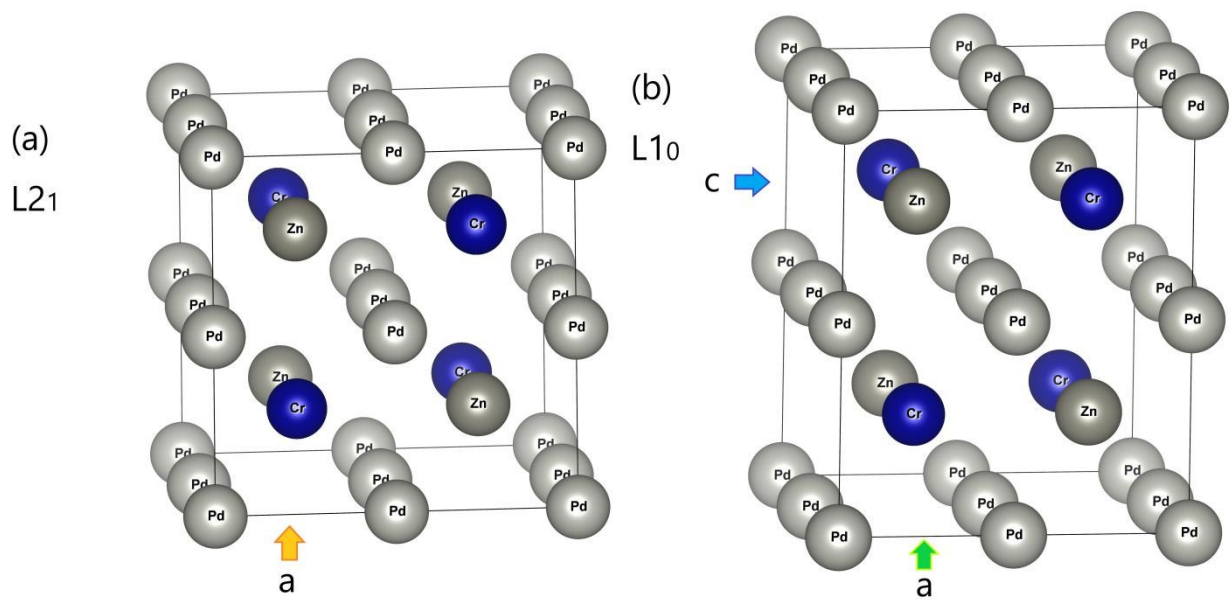
Fig. S3. Crystal structures of cubic $L2_1$-type (a) and tetragonal $L1_0$-type (b) Pd_2CrZn.
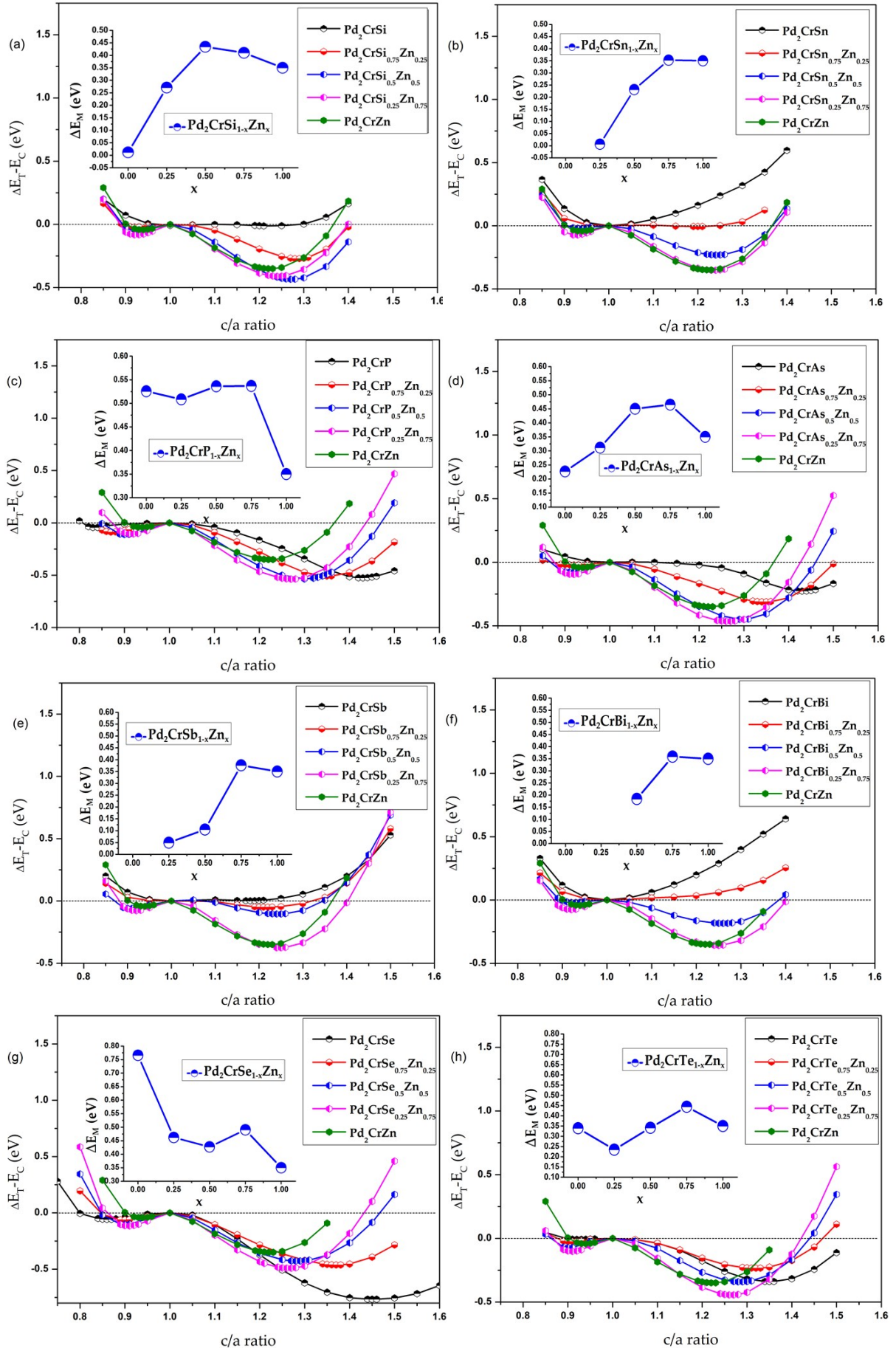
Fig. S4. The total energy differences between tetragonal and cubic states as functions of the c/a ratio of different amount of Zn doping in Pd_2CrZ (Z = Si, Sn, P, As, Sb, Bi, Se, and Te). The zero point of the total energy was set as the energy of the cubic states (c/a = 1).
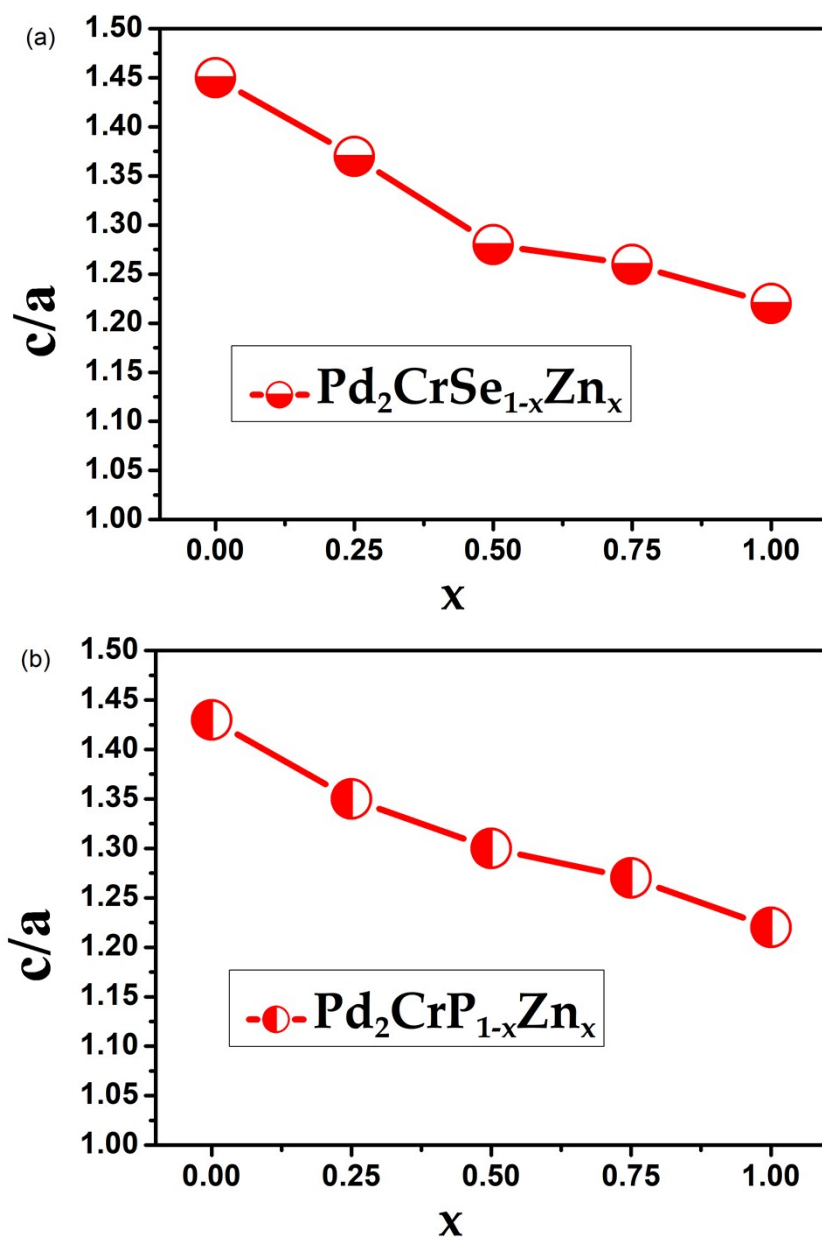
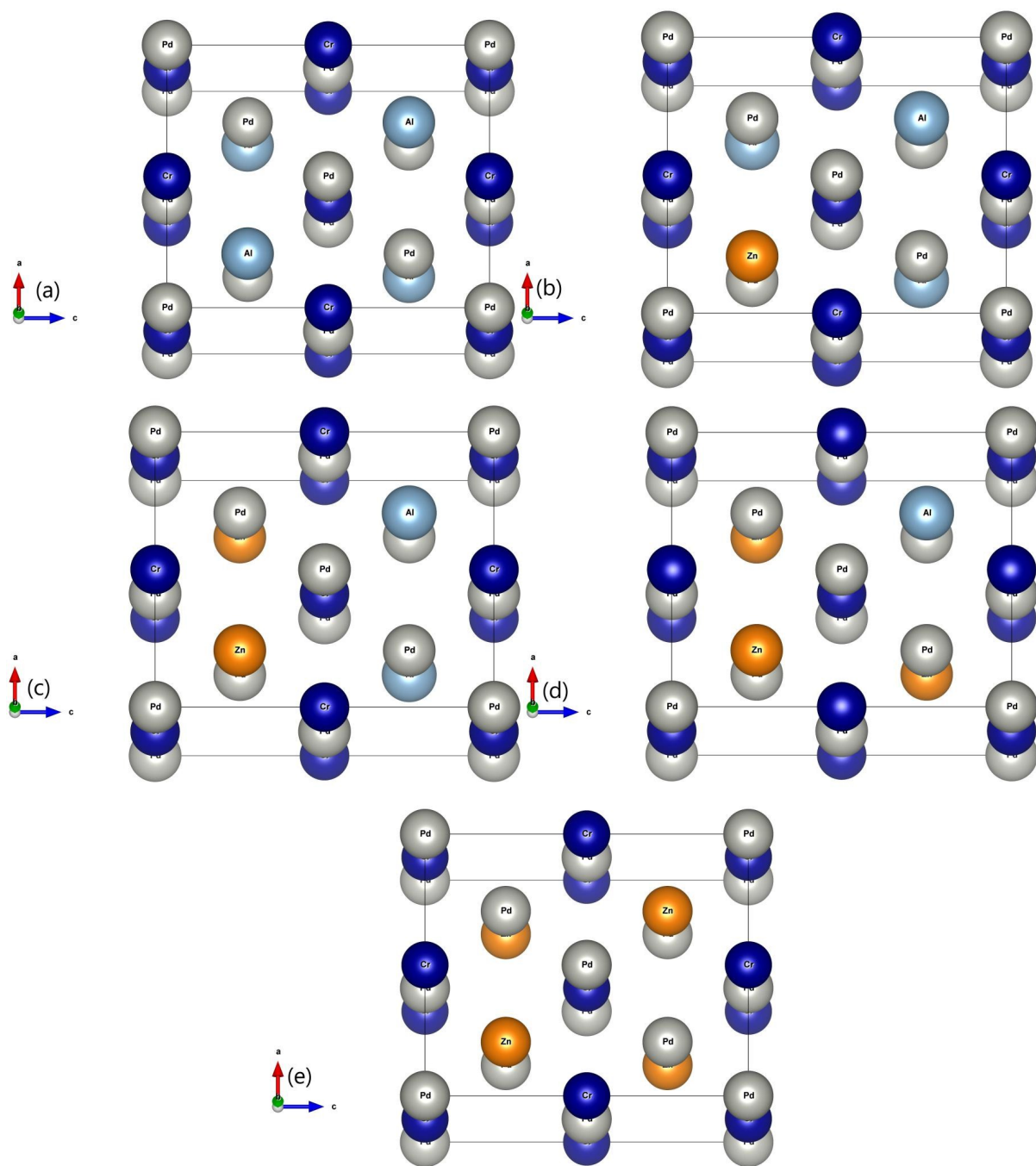Fig. S5. The c/a ratios as functions of the amount of Zn doping for Pd_2CrZ (Z = Se, P).

Fig. S6. Crystal structure of $L1_0$ type $Pd_2CrAl_xZn_{1-x}$ (x = 0, 0.25, 0.5, 0.75, 1).