Supplementary Table 1 Atomic coordinates and equivalent displacement parameters for the subcell of $Co_6Al_{11-x}Si_{6+x}$, (space group *Pnma*, a = 21.3536(9) Å, b = 4.0420(3) Å, c = 7.2723(3) Å)

Atom	Site	x	y	z	B_{eq}*
Co1	4*c*	0.54226(3)	¼	0.49382(9)	0.58(1)
Co2	4*c*	0.22763(3)	¼	0.87550(9)	0.61(1)
Co3	4*c*	0.91187(3)	¼	0.46546(9)	0.55(1)
Al1	4*c*	0.03447(8)	¼	0.4378(2)	0.95(3)
Al2	4*c*	0.11743(8)	¼	0.7250(2)	0.91(3)
Al3	4*c*	0.27541(9)	¼	0.5637(2)	1.02(3)
Al4	4*c*	0.79738(7)	¼	0.3137(2)	0.77(3)
Al5	4*c*	0.43629(8)	¼	0.6941(2)	0.90(3)
Si1	4*c*	0.64451(6)	¼	0.4283(2)	0.86(3)
Si2	4*c*	0.45372(6)	¼	0.3242(2)	0.61(2)
Al6**	4*c*	0.1665(2)	¼	0.3940(6)	2.15(9)
Si3**	8*d*	0.32961(9)	0.4322(6)	0.8611(3)	1.16(4)

*$B_{eq} = 1/3(B_{11}(a^*)^2a^2 + \ldots + B_{23}b^*c^*bc\cos\alpha)$
**Occupancy 0.50

Supplementary Table 2 Atomic coordinates and equivalent displacement parameters for $Co_6Al_{11-x}Si_{6+x}$ (space group $Cmc2_1$, a = 8.0839(6) Å, b = 14.5445(6) Å, c = 21.3536(9) Å)

Atom	Site	x	y	z	B_{eq}/B_{iso}
Co1a	4*a*	0	0.11818(4)	0.54320(4)	0.44(1)
Co1b	4*a*	½	0.12601(5)	0.54130(3)	0.61(1)
Co1c	8*b*	0.25068(6)	0.37188(3)	−0.04220(2)	0.536(9)
Co2a	4*a*	0	0.30975(5)	0.22564(3)	0.56(1)
Co2b	4*a*	½	0.31620(4)	0.22948(3)	0.58(1)
Co2c	8*b*	0.24445(6)	0.56261(3)	0.27231(2)	0.480(9)
Co3a	4*a*	0	0.10700(5)	0.91111(3)	0.51(1)
Co3b	4*a*	½	0.10836(5)	0.91275(3)	0.52(1)
Co3c	8*b*	0.24274(6)	0.35786(3)	−0.41175(2)	0.455(9)
Al1a	4*a*	0	0.0901(1)	0.03744(8)	0.98(4)
Al1b	4*a*	½	0.0965(1)	0.03168(8)	1.05(4)
Al1c	8*b*	0.2530(1)	0.34438(7)	0.46557(5)	0.69(2)
Al2a	4*a*	0	0.2310(1)	0.12011(8)	0.69(3)
Al2b	4*a*	½	0.2448(1)	0.11492(8)	0.61(3)
Al2c	8*b*	0.2469(1)	0.48707(7)	0.38253(5)	0.74(2)
Al3a	4*a*	0	0.1529(1)	0.26891(7)	0.46(3)
Al3b	4*a*	½	0.1604(1)	0.28149(7)	0.47(3)
Al3c	8*b*	0.2456(1)	0.40704(7)	0.22435(6)	0.88(2)
Al4a	4*a*	0	0.0296(1)	0.79798(8)	0.81(3)
Al4b	4*a*	½	0.0325(1)	0.79676(8)	0.80(3)
Al4c	8*b*	0.2411(1)	0.28261(7)	−0.29739(5)	0.55(2)
Al5a	4*a*	0	0.2230(1)	0.43839(8)	0.56(3)
Al5b	4*a*	½	0.2219(1)	0.43382(8)	0.69(3)
Al5c	8*b*	0.2415(2)	0.47172(7)	0.06359(6)	0.97(2)
Si1a	4*a*	0	0.08561(9)	0.64497(7)	0.66(3)
Si1b	4*a*	½	0.0940(1)	0.64266(7)	0.78(3)
Si1c	8*b*	0.2566(1)	0.33853(7)	−0.14521(5)	0.80(2)
Si2a	4*a*	0	0.03663(9)	0.45336(7)	0.53(3)
Si2b	4*a*	½	0.03691(9)	0.45489(7)	0.58(3)
Si2c	8*b*	0.2478(1)	0.28731(6)	0.04658(5)	0.60(2)
Al6b	4*a*	½	0.0710(1)	0.16608(8)	0.85(2)**
Al6c*	8*b*	0.2756(4)	0.3235(2)	0.3326(2)	2.68(6)**
Si3a*	4*a*	0.0667(3)	0.2997(2)	0.3293(1)	1.53(4)**
Si3b*	8*b*	0.6050(3)	0.3096(2)	0.3294(1)	1.78(4)**
Si3c	8*b*	0.3423(1)	0.55612(6)	0.17003(5)	0.15(1)**

*Occupancy 0.50

**Refined with isotropic displacement parameters

Supplementary Table 3 Atomic coordinates and equivalent displacement parameters for $Co_6Al_{11-x}Si_{6+x}$, described in the space group $P2_1/c$ ($mP92$, a = 8.0839(3) Å, b = 21.3536(9) Å, c = 8.3200(3) Å, β = 119.07(1)°)

Atom	Site	x	y	z	B_{eq}
Co1a	4*e*	0.11939(9)	0.54291(2)	0.48884(4)	0.34(2)
Co1b	4*e*	0.6252(1)	0.54156(2)	0.49908(5)	0.48(2)
Co2a	4*e*	0.31363(8)	0.22636(2)	0.87130(4)	0.40(2)
Co2b	4*e*	0.80980(8)	0.22893(2)	0.87974(5)	0.38(2)
Co3a	4*e*	0.11219(9)	0.91134(2)	0.46455(5)	0.34(2)
Co3b	4*e*	0.60259(9)	0.91238(2)	0.46635(5)	0.36(2)
Al1a	4*e*	0.0903(2)	0.03647(4)	0.4336(1)	0.82(4)
Al1b	4*e*	0.5989(2)	0.03256(4)	0.4417(1)	0.73(4)
Al2a	4*e*	0.2341(2)	0.11922(4)	0.7153(1)	0.66(3)
Al2b	4*e*	0.7403(2)	0.11586(4)	0.7349(1)	0.60(3)
Al3a	4*e*	0.1556(2)	0.27093(4)	0.5582(1)	0.59(3)
Al3b	4*e*	0.6553(2)	0.27999(4)	0.5692(1)	0.60(3)
Al4a	4*e*	0.0379(2)	0.79780(4)	0.3118(1)	0.59(3)
Al4b	4*e*	0.5265(2)	0.79689(4)	0.3156(1)	0.60(3)
Al5a	4*e*	0.2270(2)	0.43789(4)	0.6949(1)	0.70(4)
Al5b	4*e*	0.7157(2)	0.43464(4)	0.6935(1)	0.74(4)
Si1a	4*e*	0.0808(2)	0.64537(3)	0.4227(1)	0.60(3)
Si1b	4*e*	0.5962(2)	0.64371(4)	0.4338(1)	0.76(3)
Si2a	4*e*	0.0387(2)	0.45328(3)	0.32398(9)	0.49(3)
Si2b	4*e*	0.5363(2)	0.45430(3)	0.32431(9)	0.54(4)
Al6a	4*e*	0.5850(2)	0.16641(5)	0.3946(1)	1.99(3)
Si3a	4*e*	0.2222(1)	0.32954(5)	0.8560(1)	1.16(3)
Si3b	4*e*	0.9078(1)	0.32982(5)	0.8661(1)	0.97(3)