Substitution Patterns in Intermetallics Understood through Chemical Pressure Analysis: Atom/dumbbell and Ru/Co ordering in derivatives of YCo_5

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SUPPORTING INFORMATION

S1. Crystallographic Tables for $Y_2Ru_{4.85}Co_{12.15}$

Table S1. Crystal Data for $Y_2Ru_{4.85}Co_{12.15}$

Chemical Formula	$Y_2Ru_{4.85}Co_{12.15}$
EDS composition	$Y_{1.83(5)}Ru_{4.42(9)}Co_{12.7(3)}$
Space group	*R-3m* (No. 166)
Unit cell *a* [Å]	8.5011(12)
c [Å]	12.3705(18)
Cell volume	774.23(19)
Z	3
Pearson Symbol	*hR*57
Cryst. Dimensions [mm^3]	0.09×0.06×0.03
Crystal color	Silver
Crystal habit	block
Data collection temp.	RT
Radiation source, λ [Å]	Mo K*α*, 0.71073
Absorption coefficient [mm^{-1}]	36.801
Absorption correction	analytical
Min/max transmission	0.140/0.339
θ_{min}, θ_{max} [°]	3.22, 28.92
Number of reflections	3980
Unique refl. [I>3σ(I), all]	383, 435
Refinement method	F^2

Rint . [I>3σ(I), all]	0.0634, 0.0651
Number of parameters	28
R[I>3σ(I)], Rw[I>3σ(I)]	0.0221, 0.0456
R(all), Rw(all)	0.0271, 0.0482
S[I>3σ(I)], S(all)	0.95, 0.94
$\Delta\rho_{max}$, $\Delta\rho_{min}$($e^-/Å^3$)	1.51, -0.92

Table S2. Refined atomic coordinates of $Y_2Ru_{4.85}Co_{12.15}$.

Site	Wyckoff position	*x*	*y*	*z*	U_{equiv}	Occupancy
Y1	6c	0.333333	0.66667	0.01100(7)	0.0061(3)	1
Co1	6c	0.00000	0.00000	0.09856(7)	0.0058(3)	0.331(13)
Ru1	6c	0.00000	0.00000	0.09856(7)	0.0058(3)	0.669(13)
Co2	18h	0.66893(9)	0.83447(5)	0.18150(5)	0.0073(3)	0.645(9)
Ru2	18h	0.66893(9)	0.83447(5)	0.18150(5)	0.0073(3)	0.355(9)
Co3	9d	0.666667	0.833333	0.833333	0.0071(4)	0.953(9)
Ru3	9d	0.666667	0.833333	0.833333	0.0071(4)	0.047(9)
Co4	18f	0.70802(9)	0.000000	0.000000	0.0069(3)	0.793(9)
Ru4	18f	0.70802(9)	0.000000	0.000000	0.0069(3)	0.207(9)

Table S3. $Y_2Ru_{4.85}Co_{12.15}$ anisotropic atomic displacement parameters.

Site	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Y1	0.0057(4)	0.0057(4)	0.0070(5)	0.00283(18)	0	0
Co/Ru1	0.0063(4)	0.0063(4)	0.0049(5)	0.00315(18)	0	0
Co/Ru2	0.0074(4)	0.0058(3)	0.0092(4)	0.00368(18)	0.0009(4)	0.00045(19)
Co/Ru3	0.0096(6)	0.0064(5)	0.0065(6)	0.0048(3)	-0.0023(6)	-0.0012(3)
Co/Ru4	0.0086(3)	0.0056(4)	0.0054(4)	0.00279(19)	-0.00020(13)	-0.0004(3)

Table S4. Selected interatomic distances for $Y_2Ru_{4.85}Co_{12.15}$.

Site	*Neighbor*	*Multiplicity*	*Distance (Å)*
Ru1/Co1	Ru1/Co1		2.4385(13)
	Co4/Ru4	×3	2.5947(4)
	Co3/Ru3	×3	2.6445(6)
	Co2/Ru2	×6	2.7654(6)
Co3/Ru3	Co4/Ru4	×2	2.4526(5)
	Co3/Ru3		2.4978(11)
	Co3/Ru3		2.4979(6)
	Co2/Ru2	×2	2.5798(7)
	Co2/Ru2	×2	2.5815(7)
Co2/Ru2	Co4/Ru4	×2	2.4256(3)
	Co2/Ru2	×2	2.4822(6)

Y1	Co3/Ru3	×3	3.0232(9)
	Co2/Ru2	×6	3.0279(9)
	Ru1/Co1		3.0403(13)
	Co3/Ru3	×3	3.1620(9)
	Co3/Ru3	×3	3.2485(9)
	Co4/Ru4	×3	3.2944(6)

S2. Powder X-ray diffraction pattern for sample used in single crystal diffraction

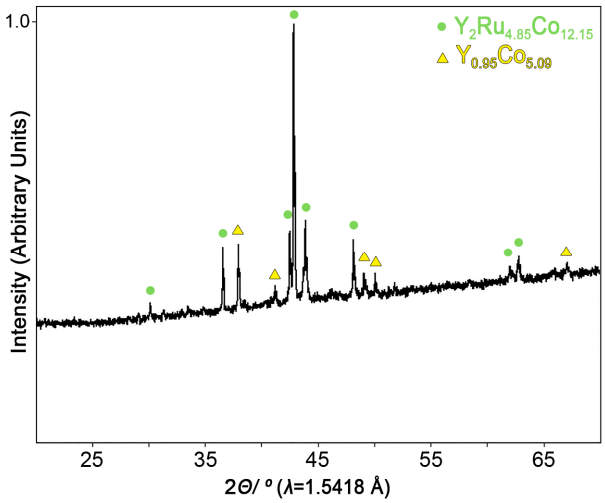

Figure S1. The powder X-ray diffraction pattern of $Y_2Ru_{4.85}Co_{12.15}$ is shown with highlighted peaks corresponding to this phase (green) and to the $CaCu_5$-type YCo_5 species (yellow).

S3. Energy Dispersive X-Ray Spectroscopy

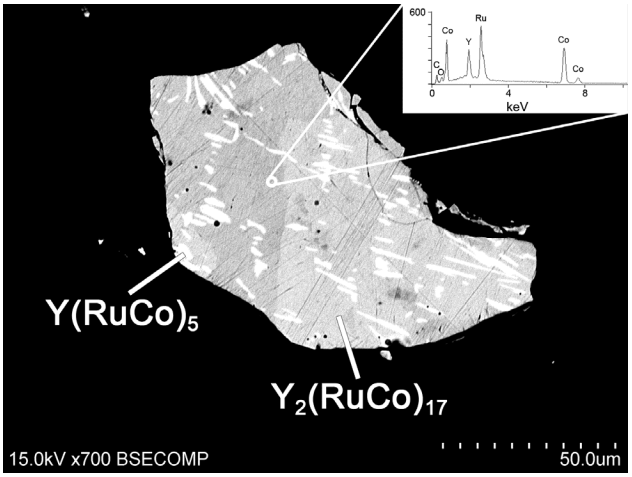

Figure S2. Back-scattered electron images of a sample of $Y_2(Ru/Co)_{17}$ used for single-crystal X-ray diffraction. A Ru-substituted $CaCu_5$-type $Y(Ru/Co)_5$ phase was detected as the minor species.

S4. Computational Details

Parameters used in the GGA-DFT calculations for all compounds are shown in Tables S5-S6, while the optimized cell parameters obtained are given in Table S7. Tables S8-S19 give the optimized atomic positions for each phase.

Table S5. Computational parameters and total energies of CP calculations with Abinit.

Structure	Energy cutoff	k-point vectors[a]	k-point shift	FFT grid	Total energy
YCo_5 (LDA)	100.00 Ha	8 0 0 0 8 0 0 0 10	0.0 0.0 0.5	125×125×100	-179.506468 Ha
YCo_5 (LDA) Spin-polarized	100.00 Ha	8 0 0 0 8 0 0 0 10	0.0 0.0 0.5	128×128×100	-179.661254 Ha
Y_2Co_{17} (LDA)	100.00 Ha	-3 3 3 3 -3 3 3 3 -3	0.5 0.5 0.5	160×160×160	-556.513847 Ha
Y_2Co_{17} (LDA) Spin-polarized	100.00 Ha	-3 3 3 3 -3 3 3 3 -3	0.5 0.5 0.5	160×160×160	-557.072519 Ha
Y_2Co_{17} (LDA) Idealized[b]	100.00 Ha	-3 3 3 3 -3 3 3 3 -3	0.5 0.5 0.5	160×160×160	-556.442723 Ha
$Y_2Ru_2Co_{15}$ (LDA)[b]	100.00 Ha	-4 4 4 4 -4 4 4 4 -4	0.5 0.5 0.5	160×160×160	-532.864740 Ha

[a]Three vectors that define a real-space super-lattice whose reciprocal lattice defines the k-point grid
[b]Hypothetical structure

Table S6. Computational parameters and total energies of Bader calculations with VASP.

Structure	Energy cutoff	k-point grid	FFT grid	Fine FFT grid	Total energy
YCo_5 (PAW-GGA) Non spin-polarized	12.31 Ha	7×7×9	30×30×24	48×48×36	-41.561728 eV
YCo_5 (PAW-GGA) Spin-polarized	12.31 Ha	7×7×9	30×30×24	48×48×40	-42.068336 eV
Y_2Co_{17} (PAW-GGA)	12.31 Ha	5×5×5	40×40×40	60×60×60	-130.914900 eV
Y_2Co_{17} (PAW-GGA) Spin-polarized	12.31 Ha	5×5×5	40×40×40	60×60×60	-133.694382 eV
Y_2Co_{17} (PAW-GGA) Idealized[a]	12.31 Ha	6×6×4	48×48×70	80×80×112	-129.181691 eV
$Y_2Ru_2Co_{15}$ (PAW-GGA)[b]	12.31 Ha	9×9×9	40×40×40	60×60×60	-135.666281 eV

[a]Hypothetical structure in which all symmetry in the $CaCu_5$-type YCo_5 compound is maintained except for the replacement of every third Y atom with a Co_2 dumbbell according to the substitution pattern in the Th_2Zn_{17}-type. Atomic positions were not allowed to relax, but the unit cell was.
[b]Modification of Th_2Zn_{17}-type Y_2Co_{17} where Ru has replaced Co on the dumbbell sites.

Table S7. Cell parameters for all DFT-optimized compounds (converted to conventional cell for convenience)

Structure	*a* (Å)	*b* (Å)	*c* (Å)	*α* (°)	*β* (°)	*γ* (°)
YCo_5 (LDA) Non spin-polarized	4.75278	4.75278	3.79758	90	90	120
YCo_5 (PAW-GGA) Non spin-polarized	4.86388	4.86388	3.86236	90	90	120
YCo_5 (LDA) Spin-polarized	4.93600	4.93600	3.88118	90	90	120
YCo_5 (PAW-GGA) Spin-polarized	4.91667	4.91667	3.94409	90	90	120
Y_2Co_{17} (LDA)	8.03297	8.03297	11.61623	90	90	120
Y_2Co_{17} (PAW-GGA)	8.20431	8.20431	11.87243	90	90	120
Y_2Co_{17} (LDA) Spin-polarized	8.11072	8.11072	11.80517	90	90	120

Y_2Co_{17} (PAW-GGA) Spin-polarized	8.30197	8.30197	12.07732	90	90	120
Y_2Co_{17} (LDA) Idealized [a]	8.02486	8.02486	11.67236	90	90	120
Y_2Co_{17} (PAW-GGA) Idealized [a]	8.19899	8.19899	11.92585	90	90	120
$Y_2Ru_2Co_{15}$ (LDA) [b]	8.08973	8.08973	11.82911	90	90	120
$Y_2Ru_2Co_{15}$ (PAW-GGA) [b]	8.25211	8.25211	12.08492	90	90	120

[a]Hypothetical structure in which all symmetry in the $CaCu_5$-type YCo_5 compound is maintained except for the replacement of every third Y atom with a Co_2 dumbbell according to the substitution pattern in the Th_2Zn_{17}-type. Atomic positions were not allowed to relax, but the unit cell was.
[b]Modification of Th_2Zn_{17}-type Y_2Co_{17} where Ru has replaced Co on the dumbbell sites.

Table S8. Fractional atomic coordinates for the LDA-DFT optimized $CaCu_5$-type compound YCo_5 with no spin polarization

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.00000
Co	0.33333	0.16667	0.00000
Co	0.66667	0.83333	0.00000
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000

Table S9. Fractional atomic coordinates for the GGA-DFT optimized $CaCu_5$-type compound YCo_5 with no spin polarization

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.00000
Co	0.33333	0.16667	0.00000
Co	0.66667	0.83333	0.00000
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000

Table S10. Fractional atomic coordinates for the LDA-DFT optimized $CaCu_5$-type compound YCo_5 with spin polarization

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.00000
Co	0.33333	0.16667	0.00000
Co	0.66667	0.83333	0.00000
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000

Co	0.50000	0.50000	0.50000

Table S11. Fractional atomic coordinates for the GGA-DFT optimized $CaCu_5$-type compound YCo_5 with spin polarization

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.00000
Co	0.33333	0.16667	0.00000
Co	0.66667	0.83333	0.00000
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000

Table S12. Fractional atomic coordinates for the LDA-DFT optimized Th_2Zn_{17}-type compound Y_2Co_{17}

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34318
Y	0.00000	0.00000	0.65682
Y	0.66667	0.33333	0.67651
Y	0.66667	0.33333	0.99016
Y	0.33333	0.66667	0.00984
Y	0.33333	0.66667	0.32349
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.16667	0.33333	0.83333
Co	0.66667	0.83333	0.83333
Co	0.16667	0.83333	0.83333
Co	0.83333	0.66667	0.16667
Co	0.33333	0.16667	0.16667
Co	0.83333	0.16667	0.16667
Co	0.28820	0.00000	0.00000
Co	0.71179	0.00000	0.00000
Co	0.00000	0.28820	0.00000
Co	0.00000	0.71179	0.00000
Co	0.71179	0.71179	0.00000
Co	0.28820	0.28820	0.00000
Co	0.95487	0.33333	0.33333
Co	0.37846	0.33333	0.33333
Co	0.66667	0.62154	0.33333
Co	0.66667	0.04513	0.33333
Co	0.37846	0.04513	0.33333
Co	0.95487	0.62154	0.33333
Co	0.62154	0.66667	0.66667
Co	0.04513	0.66667	0.66667

Co	0.33333	0.95487	0.66667
Co	0.33333	0.37846	0.66667
Co	0.04513	0.37846	0.66667
Co	0.62154	0.95487	0.66667
Co	0.50237	0.49763	0.15225
Co	0.49763	0.50237	0.84775
Co	0.50237	0.00474	0.15225
Co	0.49763	0.99525	0.84775
Co	0.99525	0.49763	0.15225
Co	0.00474	0.50237	0.84775
Co	0.16904	0.83096	0.48558
Co	0.16430	0.83570	0.18109
Co	0.16904	0.33808	0.48558
Co	0.16430	0.32859	0.18109
Co	0.66192	0.83096	0.48558
Co	0.67141	0.835700	0.18109
Co	0.83570	0.16430	0.81891
Co	0.83096	0.16904	0.51442
Co	0.83570	0.67141	0.81891
Co	0.83096	0.66192	0.51442
Co	0.32859	0.16430	0.81891
Co	0.33808	0.16904	0.51442
Co	0.00000	0.00000	0.09853
Co	0.00000	0.00000	0.90147
Co	0.66667	0.33333	0.43186
Co	0.66667	0.33333	0.23480
Co	0.33333	0.66667	0.76520
Co	0.33333	0.66667	0.56814

Table S13. Fractional atomic coordinates for the GGA-DFT optimized Th_2Zn_{17}-type compound Y_2Co_{17}

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34245
Y	0.00000	0.00000	0.65755
Y	0.66667	0.33333	0.67578
Y	0.66667	0.33333	0.99089
Y	0.33333	0.66667	0.00911
Y	0.33333	0.66667	0.32422
Co	0.83333	0.66667	0.16667
Co	0.16667	0.33333	0.83333
Co	0.33333	0.16667	0.16667
Co	0.66667	0.83333	0.83333
Co	0.83333	0.16667	0.16667

Co	0.16667	0.83333	0.83333
Co	0.50000	0.0000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.95514	0.3333	0.33333
Co	0.04486	0.66667	0.66667
Co	0.66667	0.62180	0.33333
Co	0.33333	0.37820	0.66667
Co	0.37820	0.04486	0.33333
Co	0.62180	0.95514	0.66667
Co	0.33333	0.95514	0.66667
Co	0.66667	0.04486	0.33333
Co	0.62180	0.66667	0.66667
Co	0.37820	0.33333	0.33333
Co	0.04486	0.37820	0.66667
Co	0.95514	0.62180	0.33333
Co	0.71153	0.00000	0.00000
Co	0.00000	0.71153	0.0000
Co	0.28847	0.28847	0.00000
Co	0.00000	0.28847	0.00000
Co	0.28847	0.00000	0.00000
Co	0.71153	0.71153	0.00000
Co	0.16872	0.83128	0.48623
Co	0.83130	0.16872	0.51377
Co	0.16872	0.33744	0.48523
Co	0.83128	0.66256	0.51377
Co	0.66256	0.83128	0.48623
Co	0.33744	0.16872	0.51377
Co	0.83539	0.16461	0.81956
Co	0.49795	0.50205	0.84710
Co	0.83539	0.67077	0.81956
Co	0.49795	0.99589	0.84740
Co	0.32923	0.16461	0.81956
Co	0.00411	0.50205	0.84710
Co	0.50205	0.49795	0.15290
Co	0.16461	0.83539	0.18044
Co	0.50205	0.00511	0.15290
Co	0.16461	0.32923	0.18043
Co	0.99589	0.49795	0.15290
Co	0.67077	0.83539	0.18044
Co	0.00000	0.00000	0.09875
Co	0.00000	0.00000	0.90125
Co	0.66667	0.33333	0.43209

Co	0.66667	0.33333	0.23458
Co	0.33333	0.66667	0.76542
Co	0.33333	0.66667	0.56791

Table S14. Fractional atomic coordinates for the LDA-DFT optimized Th_2Zn_{17}-type compound Y_2Co_{17} with spin polarization

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34456
Y	0.00000	0.00000	0.65544
Y	0.66667	0.33333	0.67789
Y	0.66667	0.33333	0.98878
Y	0.33333	0.66667	0.01122
Y	0.33333	0.66667	0.32211
Co	0.16667	0.33333	0.83333
Co	0.66667	0.83333	0.83333
Co	0.16667	0.83333	0.83333
Co	0.83333	0.66667	0.16667
Co	0.33333	0.16667	0.16667
Co	0.83333	0.16667	0.16667
Co	0.28819	0.00000	0.00000
Co	0.71181	0.00000	0.00000
Co	0.00000	0.28819	0.00000
Co	0.00000	0.71181	0.00000
Co	0.71181	0.71181	0.00000
Co	0.28819	0.28819	0.00000
Co	0.95486	0.33333	0.33333
Co	0.37847	0.33333	0.33333
Co	0.66667	0.62153	0.33333
Co	0.66667	0.04514	0.33333
Co	0.37847	0.04514	0.33333
Co	0.95486	0.62153	0.33333
Co	0.62153	0.66667	0.66667
Co	0.04514	0.66667	0.66667
Co	0.33333	0.95486	0.66667
Co	0.33333	0.37847	0.66667
Co	0.04514	0.37847	0.66667
Co	0.62123	0.95486	0.66667
Co	0.50172	0.49878	0.15210
Co	0.49828	0.50172	0.84790
Co	0.50172	0.00345	0.15210
Co	0.49828	0.99656	0.84790
Co	0.99656	0.49828	0.15210
Co	0.00345	0.50172	0.84790

Co	0.16390	0.83161	0.48543
Co	0.16494	0.83506	0.18124
Co	0.16839	0.33678	0.48543
Co	0.16494	0.32989	0.18124
Co	0.66322	0.83161	0.48543
Co	0.67011	0.83506	0.18124
Co	0.83506	0.16494	0.81876
Co	0.83161	0.16839	0.51457
Co	0.83506	0.67011	0.81876
Co	0.83161	0.66322	0.51457
Co	0.32989	0.16494	0.81876
Co	0.33678	0.16839	0.51457
Co	0.00000	0.00000	0.09681
Co	0.00000	0.00000	0.90319
Co	0.66667	0.33333	0.43014
Co	0.66667	0.33333	0.23653
Co	0.33333	0.66667	0.76348
Co	0.33333	0.66667	0.56986

Table S15. Fractional atomic coordinates for the GGA-DFT optimized Th_2Zn_{17}-type compound Y_2Co_{17} with spin polarization

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34436
Y	0.00000	0.00000	0.65564
Y	0.66667	0.33333	0.67769
Y	0.66667	0.33333	0.98897
Y	0.33333	0.66667	0.01103
Y	0.33333	0.66667	0.32231
Co	0.83333	0.66667	0.16667
Co	0.16667	0.33333	0.83333
Co	0.33333	0.16667	0.16667
Co	0.66667	0.83333	0.83333
Co	0.83333	0.16667	0.16667
Co	0.16667	0.83333	0.83333
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.95487	0.33333	0.33333
Co	0.04513	0.66667	0.66667
Co	0.66667	0.62153	0.33333
Co	0.33333	0.37847	0.66667
Co	0.37847	0.04513	0.33333
Co	0.62153	0.95487	0.66667

Co	0.33333	0.95487	0.66667
Co	0.66667	0.04513	0.33333
Co	0.62153	0.66667	0.66667
Co	0.37847	0.33333	0.33333
Co	0.04513	0.37847	0.66667
Co	0.95487	0.62153	0.33333
Co	0.71180	0.00000	0.00000
Co	0.00000	0.71180	0.00000
Co	0.28820	0.28820	0.00000
Co	0.00000	0.28820	0.00000
Co	0.28820	0.00000	0.00000
Co	0.71180	0.71180	0.00000
Co	0.16820	0.83180	0.48611
Co	0.83180	0.16820	0.51389
Co	0.16820	0.33640	0.48611
Co	0.83180	0.66361	0.51389
Co	0.66361	0.83180	0.48611
Co	0.33640	0.16820	0.51389
Co	0.83486	0.16514	0.81944
Co	0.49847	0.50153	0.84722
Co	0.83486	0.66973	0.81944
Co	0.49847	0.50153	0.84722
Co	0.83486	0.66973	0.81944
Co	0.49847	0.99694	0.84722
Co	0.33027	0.16514	0.81944
Co	0.00306	0.50153	0.84722
Co	0.50153	0.49847	0.15278
Co	0.16514	0.83486	0.18056
Co	0.50153	0.00306	0.15278
Co	0.16514	0.33027	0.18056
Co	0.99694	0.49847	0.15278
Co	0.66973	0.83486	0.18056
Co	0.00000	0.00000	0.09668
Co	0.00000	0.00000	0.90333
Co	0.66667	0.33333	0.43001
Co	0.66667	0.33333	0.23666
Co	0.33333	0.66667	0.76334
Co	0.33333	0.66667	0.56999

Table S16. Fractional atomic coordinates for the idealized Y_2Co_{17} model with LDA-DFT-optimized cell parameters

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34264
Y	0.00000	0.00000	0.65736
Y	0.66667	0.33333	0.67597
Y	0.66667	0.33333	0.99069
Y	0.33333	0.66667	0.00931
Y	0.33333	0.66667	0.32403
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.16667	0.33333	0.83333
Co	0.66667	0.83333	0.83333
Co	0.16667	0.83333	0.83333
Co	0.83333	0.66667	0.16667
Co	0.33333	0.16667	0.16667
Co	0.83333	0.16667	0.16667
Co	0.33333	0.00000	0.00000
Co	0.66667	0.00000	0.00000
Co	0.00000	0.33333	0.00000
Co	0.00000	0.66667	0.00000
Co	0.66667	0.66667	0.00000
Co	0.33333	0.33333	0.00000
Co	0.00000	0.33333	0.33333
Co	0.33333	0.33333	0.33333
Co	0.66667	0.66667	0.33333
Co	0.66667	0.00000	0.33333
Co	0.33333	0.00000	0.33333
Co	0.00000	0.66667	0.33333
Co	0.66667	0.66667	0.66667
Co	0.00000	0.66667	0.66667
Co	0.33333	0.00000	0.66667
Co	0.33333	0.33333	0.66667
Co	0.00000	0.33333	0.66667
Co	0.66667	0.00000	0.66667
Co	0.50000	0.50000	0.16667
Co	0.50000	0.50000	0.83333
Co	0.50000	0.00000	0.16667
Co	0.50000	0.00000	0.83333
Co	0.00000	0.50000	0.16667
Co	0.00000	0.50000	0.83333
Co	0.16667	0.83333	0.50000

Co	0.16667	0.83333	0.16667
Co	0.16667	0.33333	0.50000
Co	0.16667	0.33333	0.16667
Co	0.66667	0.83333	0.50000
Co	0.66667	0.83333	0.16667
Co	0.83333	0.16667	0.83333
Co	0.83333	0.16667	0.50000
Co	0.83333	0.66667	0.83333
Co	0.83333	0.66667	0.50000
Co	0.33333	0.16667	0.833333
Co	0.33333	0.16667	0.50000
Co	0.00000	0.00000	0.09876
Co	0.00000	0.00000	0.90124
Co	0.66667	0.33333	0.43209
Co	0.66667	0.33333	0.23457
Co	0.33333	0.66667	0.76543
Co	0.33333	0.66667	0.56791

Table S17. Fractional atomic coordinates for idealized Y_2Co_{17} model with GGA-DFT-optimized cell parameters

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34264
Y	0.00000	0.00000	0.65736
Y	0.66667	0.33333	0.67597
Y	0.66667	0.33333	0.99069
Y	0.33333	0.66667	0.00931
Y	0.33333	0.66667	0.32403
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.16667	0.33333	0.83333
Co	0.66667	0.83333	0.83333
Co	0.16667	0.83333	0.83333
Co	0.83333	0.66667	0.16667
Co	0.33333	0.16667	0.16667
Co	0.83333	0.16667	0.16667
Co	0.33333	0.00000	0.00000
Co	0.66667	0.00000	0.00000
Co	0.00000	0.33333	0.00000
Co	0.00000	0.66667	0.00000
Co	0.66667	0.66667	0.00000
Co	0.33333	0.33333	0.00000
Co	0.00000	0.33333	0.33333

Co	0.33333	0.33333	0.33333
Co	0.66667	0.66667	0.33333
Co	0.66667	0.00000	0.33333
Co	0.33333	0.00000	0.33333
Co	0.00000	0.66667	0.33333
Co	0.66667	0.66667	0.66667
Co	0.00000	0.66667	0.66667
Co	0.33333	0.00000	0.66667
Co	0.33333	0.33333	0.66667
Co	0.00000	0.33333	0.66667
Co	0.66667	0.00000	0.66667
Co	0.50000	0.50000	0.16667
Co	0.50000	0.50000	0.83333
Co	0.50000	0.00000	0.16667
Co	0.50000	0.00000	0.83333
Co	0.00000	0.50000	0.16667
Co	0.00000	0.50000	0.83333
Co	0.16667	0.83333	0.50000
Co	0.16667	0.83333	0.16667
Co	0.16667	0.33333	0.50000
Co	0.16667	0.33333	0.16667
Co	0.66667	0.83333	0.50000
Co	0.66667	0.83333	0.16667
Co	0.83333	0.16667	0.83333
Co	0.83333	0.16667	0.50000
Co	0.83333	0.66667	0.83333
Co	0.83333	0.66667	0.50000
Co	0.33333	0.16667	0.833333
Co	0.33333	0.16667	0.50000
Co	0.00000	0.00000	0.09876
Co	0.00000	0.00000	0.90124
Co	0.66667	0.33333	0.43209
Co	0.66667	0.33333	0.23457
Co	0.33333	0.66667	0.76543
Co	0.33333	0.66667	0.56791

Table S18. Fractional atomic coordinates for the LDA-DFT optimized compound $Y_2Ru_2Co_{15}$ in the Th_2Zn_{17}-type

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34662
Y	0.00000	0.00000	0.65338
Y	0.66667	0.33333	0.67995
Y	0.66667	0.33333	0.98672

Y	0.33333	0.66667	0.01328
Y	0.33333	0.66667	0.32005
Ru	0.00000	0.00000	0.09808
Ru	0.00000	0.00000	0.90192
Ru	0.66667	0.33333	0.43141
Ru	0.66667	0.33333	0.23525
Ru	0.33333	0.66667	0.76475
Ru	0.33333	0.66667	0.56859
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.16667	0.33333	0.83333
Co	0.66667	0.83333	0.83333
Co	0.16667	0.83333	0.83333
Co	0.83333	0.66667	0.16667
Co	0.33333	0.16667	0.16667
Co	0.83333	0.16667	0.16667
Co	0.29681	0.00000	0.00000
Co	0.70318	0.00000	0.00000
Co	0.00000	0.29681	0.00000
Co	0.00000	0.70318	0.00000
Co	0.70318	0.70318	0.00000
Co	0.29681	0.29681	0.00000
Co	0.96348	0.33333	0.33333
Co	0.36985	0.33333	0.33333
Co	0.66667	0.63015	0.33333
Co	0.66667	0.03652	0.33333
Co	0.36985	0.03652	0.33333
Co	0.96348	0.63015	0.33333
Co	0.63015	0.66667	0.66667
Co	0.03652	0.66667	0.66667
Co	0.33333	0.96345	0.66667
Co	0.33333	0.36985	0.66667
Co	0.03652	0.36985	0.66667
Co	0.63015	0.96348	0.66667
Co	0.50162	0.49838	0.15458
Co	0.49838	0.50162	0.84542
Co	0.50162	0.00325	0.15458
Co	0.49838	0.99675	0.84542
Co	0.99675	0.49838	0.15458
Co	0.00325	0.50162	0.84542
Co	0.16829	0.83171	0.48791
Co	0.16504	0.83496	0.17876

Co	0.16829	0.33658	0.48791
Co	0.16504	0.33009	0.17876
Co	0.66342	0.83171	0.48791
Co	0.66991	0.83496	0.17876
Co	0.83496	0.16504	0.82124
Co	0.83171	0.16829	0.51209
Co	0.83496	0.66991	0.82124
Co	0.83171	0.66342	0.51209
Co	0.33009	0.16504	0.82124
Co	0.33658	0.16829	0.51209

Table S19. Fractional atomic coordinates for the GGA-DFT optimized compound $Y_2Ru_2Co_{15}$ in the Th_2Zn_{17}-type

Element	*x*	*y*	*z*
Y	0.00000	0.00000	0.34647
Y	0.00000	0.00000	0.65353
Y	0.66667	0.33333	0.67980
Y	0.66667	0.33333	0.98687
Y	0.33333	0.66667	0.01314
Y	0.33333	0.66667	0.32020
Ru	0.00000	0.00000	0.09817
Ru	0.00000	0.00000	0.90183
Ru	0.66667	0.33333	0.43151
Ru	0.66667	0.33333	0.23516
Ru	0.33333	0.66667	0.76484
Ru	0.33333	0.66667	0.56849
Co	0.83333	0.66667	0.16667
Co	0.16667	0.33333	0.83333
Co	0.33333	0.16667	0.16667
Co	0.66667	0.83333	0.83333
Co	0.83333	0.16667	0.16667
Co	0.16667	0.83333	0.83333
Co	0.50000	0.00000	0.50000
Co	0.00000	0.50000	0.50000
Co	0.50000	0.50000	0.50000
Co	0.96302	0.33333	0.33333
Co	0.03698	0.66667	0.66667
Co	0.66667	0.62969	0.33333
Co	0.33333	0.37032	0.66667
Co	0.37032	0.03698	0.33333
Co	0.62969	0.96302	0.66667
Co	0.33333	0.96302	0.66667
Co	0.66667	0.03698	0.33333

Co	0.62969	0.66667	0.66667
Co	0.37032	0.33333	0.33333
Co	0.03698	0.37032	0.66667
Co	0.96302	0.62969	0.33333
Co	0.70365	0.00000	0.00000
Co	0.00000	0.70365	0.00000
Co	0.29635	0.00000	0.00000
Co	0.70365	0.70365	0.00000
Co	0.16841	0.83160	0.48820
Co	0.83160	0.16841	0.51180
Co	0.16841	0.33681	0.48820
Co	0.83160	0.66319	0.51180
Co	0.66319	0.83160	0.48820
Co	0.33681	0.16841	0.51180
Co	0.83507	0.16493	0.82153
Co	0.49826	0.50174	0.84514
Co	0.83507	0.67014	0.82153
Co	0.49826	0.99652	0.84514
Co	0.32986	0.16493	0.82153
Co	0.00348	0.50174	0.84514
Co	0.50174	0.49826	0.15487
Co	0.16493	0.83507	0.17847
Co	0.50174	0.00348	0.15487
Co	0.16493	0.32986	0.17487
Co	0.99652	0.49826	0.15487
Co	0.67014	0.83507	0.17847

S5. Magnetization on atoms in spin-polarized calculations

Table S20. Magnetization of atoms[a] in LDA-DFT optimized $CaCu_5$-type compound YCo_5 with spin polarization, calculated in ABINIT.

Site	Wyckoff Position	magnetization
Y	1a	-0.67171153
Co1	2c	2.07990169
Co2	3g	2.09615244

[a]Wigner-Seitz radii of 1.889 Å and 1.355 Å for Y and Co, respectively, were used to determine atomic volumes for integration of spin density

Table S21. Magnetization of atoms[a] in GGA-DFT optimized $CaCu_5$-type compound YCo_5 with spin polarization, calculated in VASP.

Site	Wyckoff Position	magnetization
Y	1a	-0.233
Co1	2c	1.477
Co2	3g	1.490

[a]Wigner-Seitz radii of 1.906 Å and 1.367 Å for Y and Co, respectively, were used to determine atomic volumes for integration of spin density

Table S22. Magnetization of atoms[a] in LDA-DFT optimized Th_2Zn_{17}-type compound Y_2Co_{17} with spin polarization, calculated in ABINIT.

Site	Wyckoff Position	magnetization
Y	6c	-0.69004085
Co1	6c	2.06452439
Co2	18h	2.00759545
Co3	9d	1.99636877
Co4	18f	1.96757142

[a]Wigner-Seitz radii of 1.779 Å and 1.276 Å for Y and Co, respectively, were used to determine atomic volumes for integration of spin density

Table S23. Magnetization of atoms[a] in GGA-DFT optimized Th_2Zn_{17}-type compound Y_2Co_{17} with spin polarization, calculated in VASP.

Site	Wyckoff Position	magnetization
Y	6c	-0.240
Co1	6c	1.629
Co2	18h	1.489
Co3	9d	1.531
Co4	18f	1.534

[a]Wigner-Seitz radii of 1.907 Å and 1.368 Å for Y and Co, respectively, were used to determine atomic volumes for integration of spin density

S6. CP schemes with varying ionicity (50% of Bader charge used for paper)

Radial electron density profiles were generated for all symmetry-distinct sites in each structure for charges equal to 100%, 75%, 50%, and 0% of the Bader charge on each site. Below are CP schemes that demonstrate the weak dependence of the results on the ionicity parameter.

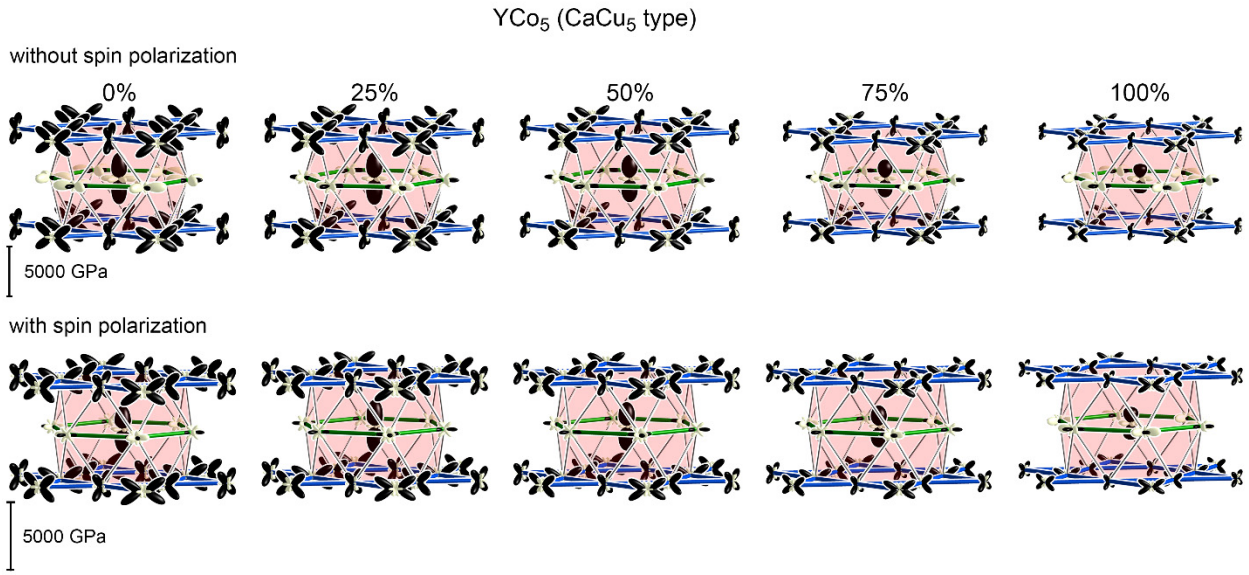

Figure S3. Chemical pressure schemes for YCo_5 in the $CaCu_5$ type, without (above) and with (below) spin-polarization included. Radial electron density files for atoms at 0, 25, 50, 75, and 100% of the calculated Bader charges were used to generate these schemes; the results for 50% ionicity are shown in the main text.

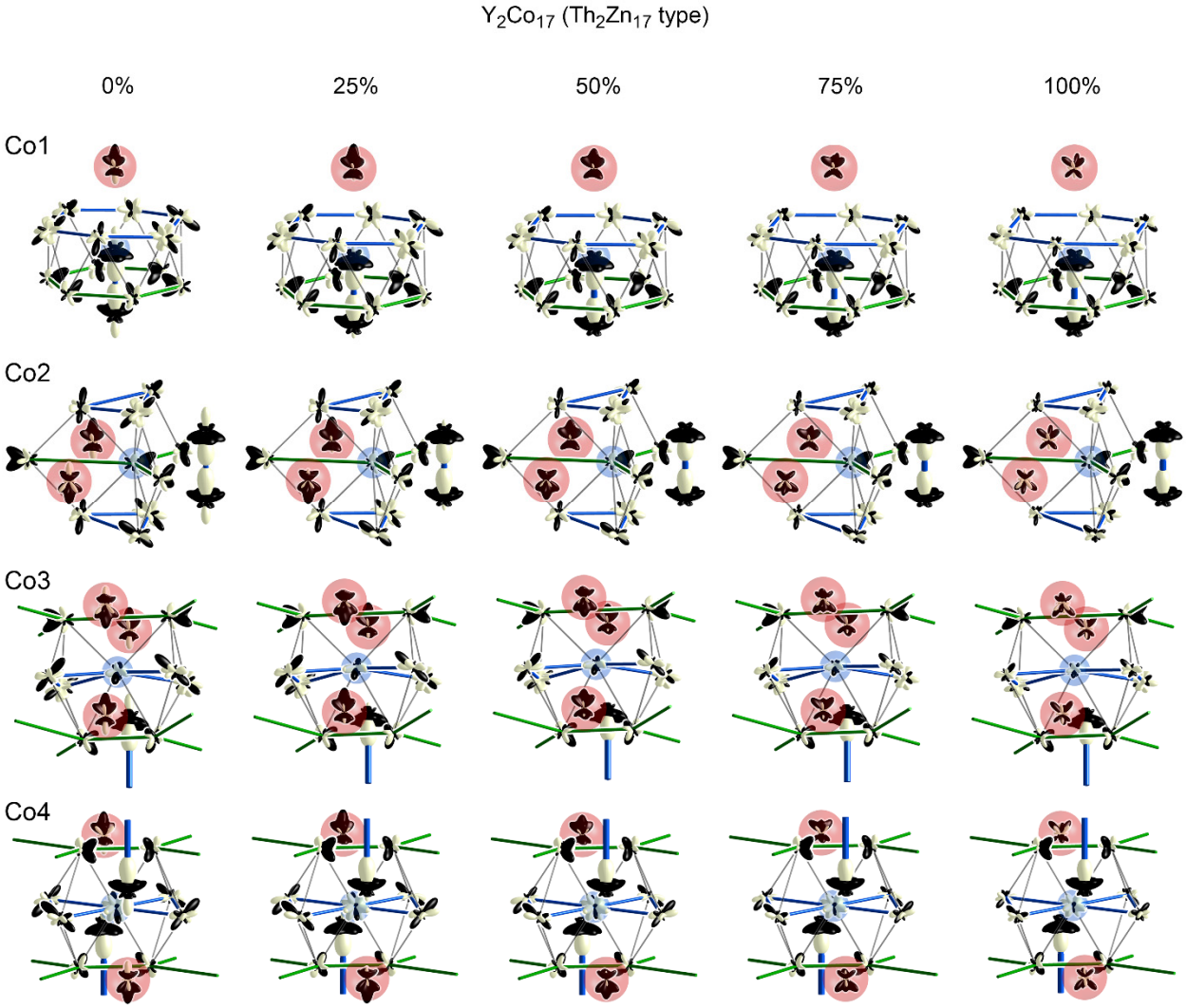

Figure S4. Chemical pressure schemes for all Co sites in Y_2Co_{17} in the Th_2Zn_{17} type without spin-polarization. The results for 50% ionicity are presented in the main text.

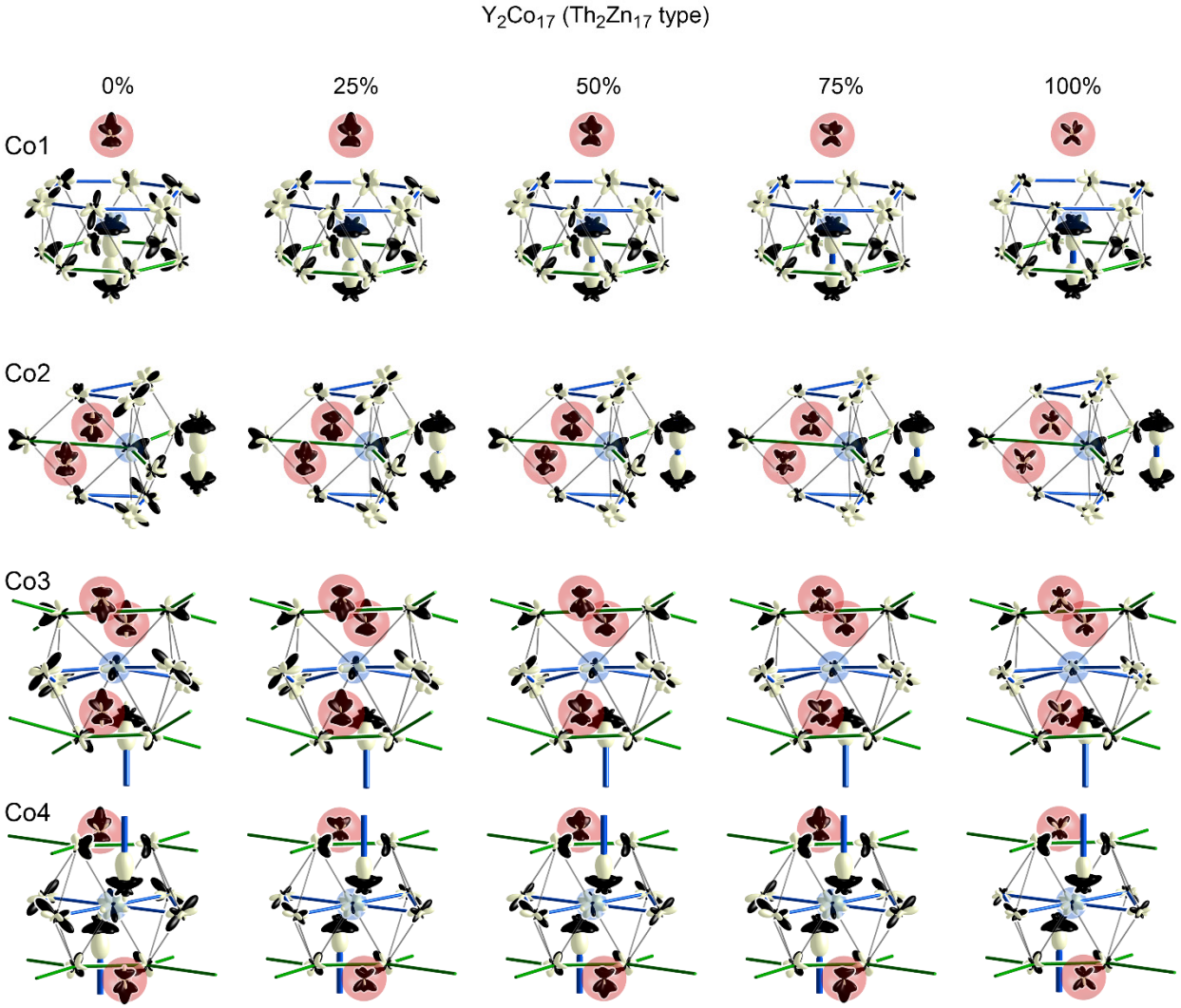

Figure S5. Chemical pressure schemes for all Co sites in Y_2Co_{17} in the Th_2Zn_{17} type with spin-polarization. The results for 50% ionicity are shown in the main text.