

Supplemental Information for Ferromagnetic half-metallicity in YBaCo₂O₆ and spin-states driven metal-insulator transition

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Table S1 Relative total energies ΔE (meV/f.u.) for different spin-states of YBaCo₂O₆ calculated by LSDA+ U with different U values. The total magnetic moment per formula unit (μ_B /f.u.) is listed in the round brackets. Fixed-spin-moment (FSM) results in LSDA+ U are also included.

	IS-Co ³⁺ /LS-Co ⁴⁺	IS-Co ³⁺ /IS-Co ⁴⁺	HS-Co ³⁺ /LS-Co ⁴⁺	HS-Co ³⁺ /IS-Co ⁴⁺
$U = 6$ eV	0 (3.00 μ_B)	→HS/LS	114 (5.00 μ_B)	596 (5.94 μ_B) 770 (FSM, 7.00 μ_B)
$U = 5$ eV	0 (3.00 μ_B)	→HS/LS	190 (4.91 μ_B)	→IS/LS 970 (FSM, 7.00 μ_B)
$U = 4$ eV	0 (3.00 μ_B)	→IS/LS	→IS/LS 301 (FSM, 5.00 μ_B)	→IS/LS 1138 (FSM, 7.00 μ_B)

Table S2 Relative energies ΔE (meV/f.u.) of the IS-LS and HS-LS states of YBaCo₂O₆ with different J_H .

volume expansion	spin-states	$J_H = 1$ eV	$J_H = 0.8$ eV
+0%	IS-Co ³⁺ /LS-Co ⁴⁺	0	0
	HS-Co ³⁺ /LS-Co ⁴⁺	114	76
+4%	IS-Co ³⁺ /LS-Co ⁴⁺	47	85
	HS-Co ³⁺ /LS-Co ⁴⁺	0	0

Table S3 Relative energies ΔE (meV/f.u.), local spin and orbital moments (μ_B /Co) of the IS-LS and HS-LS states of YBaCo₂O₆ calculated by LSDA+ U and LSDA+ U +SOC.

volume expansion	spin-states	LSDA+ U		LSDA+ U +SOC		
		ΔE	M_{spin}	ΔE	M_{spin}	M_{orb}
+0%	IS-Co ³⁺ /LS-Co ⁴⁺	0	1.69/1.69	0	1.67/1.67	0.29/0.30
	HS-Co ³⁺ /LS-Co ⁴⁺	114	2.80/1.70	114	2.72/1.67	0.16/0.36
+4%	IS-Co ³⁺ /LS-Co ⁴⁺	47	1.78/1.78	48	1.75/1.75	0.33/0.33
	HS-Co ³⁺ /LS-Co ⁴⁺	0	2.90/1.51	0	2.86/1.49	0.18/0.28

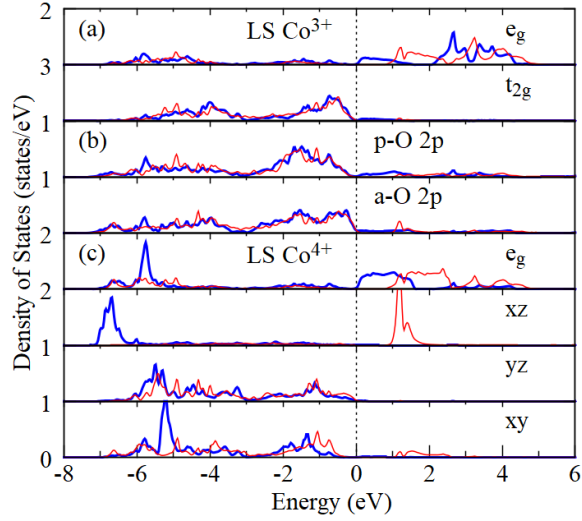


Fig. S1 DOS of YBaCo_2O_6 in the LS- Co^{3+} /LS- Co^{4+} state by LSDA+ U . It is a paramagnetic insulator.

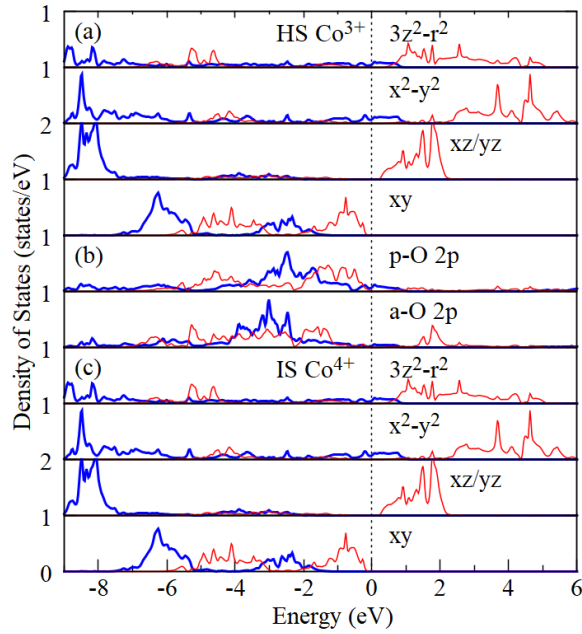


Fig. S2 DOS of YBaCo_2O_6 in the HS- Co^{3+} /IS- Co^{4+} state by LSDA+ U FSM calculation with $M_{\text{tot}} = 7.00 \mu_B/\text{f.u.}$. It is a FM half-metal.