Table S2. Main crystallographic and physical properties of the $A_2BB'O_6$ double ordered perovskites: theoretical tolerance factor (f) as obtained from the definition given in equation (1) and Shannon's table; observed tolerance factor ($f$obs) as calculated from the crystallographic structures reported in the first referenc2 of each compound; reported crystallographic space group (H = hexagonal, and O = orthorhombic in the case of an unknown space group); lattice parameters (a, b, and c); tetragonal distortion (t) as defined in equation (2); representative valence states of the B and B_ atoms determined on the basis of NMR, XAS, optical conductivity, bond valence sum rule applied to neutron diffraction data and/or Mössbauer experiments (which can be found in the listed references); and magnetic ordering temperature. When no reported valence state exists in the literature, a 3+/5+ state has been assumed for the f calculation. The structural data always correspond to the first reference given for each compound. The magnetic ordering temperature (T_C/T_N) corresponds to the ferromagnetic/ferrimagnetic to paramagnetic transition. M_S and H_C refer to saturation magnetization and coercive magnetic field, respectively. The magnetic ordering type is specified (PM = paramagnetic for the whole temperature range, AFM = antiferromagnetic, C = canted ferromagnetism) if different from ferromagnetic or ferrimagnetic.

A_2	B	Cr			Mn			Fe			Co
	B′	Mo	Re	W	Mo	Re	W	Mo	Re	W	Re
Ca_2 (Ref.)		[111,112]	[113,114]	[111,115]	/	[113,116]	[117]	[111,118,119-121]	[56,122-124]	[125]	[113,116]
f/f_{obs}		0.9627/	0.9699/0.9837	0.9382/	0.9556/	0.9270/	0.9161/0.9774	0.9422/0.9531	0.9627/0.9522	0.9533/0.9979	0.9463/0.9743
Space group		O	$P2_1/n$	$P2_1/n$	/	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$	Pmn2 (O)	$P2_1/n$
Lattice (Å)											
a		5.49	5.3886	5.39	/	5.44651	5.4457	5.3875	5.390/7.623	5.4242	5.40266
b/t ($\times10^3$)		7.70/8.3	5.4604/5.1	1 5.45/4.9	/	5.639 97/9.5	5.6529/10.4	5.5027/7.5	5.5165/6.4	5.5108/4.4	5.573 47/5.9
c		5.36	7.6598	7.66	/	7.77657	7.7828	7.6767	7.6719	7.7051	7.68607
Valence (B, B′)		/	3+/5+	2+/6+	/	2+/6+	2+/6+	2.5+/5.5+	2.5+/5.5+	/	2+/6+
T_C/T_N (K)		148	360	160	/	110	16-AFM	365	540	/	130-AFM
M_S (μ_B/f.u.)(@ 5 K)		0.05	0.8	1.34)	/	0.90	/	3.51	2.24	/	0.60
H_C (kOe)		/	31.0	6.0		40.0	/	/	8.0	/	7.0
Sr_2 (Ref.)		[111, 126-128]	[113,129]	[111,115, 130]	[128,131]	[132]	[133]	[9,13,118,120,134]	[56,113,122,135]	[136,137]	[113,116]
f/f_{obs}		0.9978/0.9996	0.9947/0.9996	0.9838/	0.9905/1.0000	0.9608/0.9337	0.9495/0.9927	0.9766/0.9984	0.9850/0.9984	0.9608/0.9927	0.9808/0.9968
Space group		*I4/m*	*I4/m*	*Fm3m*	*Fm3m*	*P21/n*	*P42/n*	*I4/m*	*I4/m*	*P21/n*	*I4/m*
Lattice (Å)											
a		5.5335	5.5206	7.832	8.0056	5.651	7.9992	5.5705	5.561	5.643	5.5659
b/t ($\times10^3$)		/0.05	/0.6	//0	/0	5.6378/2.4	/0.8	/5.9	/4.6	5.5856/8.6	/7.1
c		7.8251	7.8023	/	/	7.9731	8.0058	7.9253	7.9008	7.9128	7.9508
Valence (B, B′)		3+/5+	2.5+/5.5+	2.5+/5.5+	3+/5+	2+/6+	2+/6+	2.5+/5.5+	2.5+/5.5+	2+/6+	2+/6+
T_C/T_N (K)		420	620	458	12-AFM	120-C	13-AFM	420	400	40-AFM	65-AFM
M_S (μ_B/f.u.)(@ 5 K)		0.50	0.86	1.11	/	/	/	3.0 (@ 4.2 K)	2.60	/	/
H_C (kOe)		/	17.0	0.45	/	/	/	/	2.0	/	/

Ba_2 (Ref.)	[138]	[116]	[139]	/	[140-142]	[143]	[111,118-121,144]	[56,116, 122,141,145]	[136]	[140,141]
f/f_{obs}	1.0576/	1.0655/	1.0549/	1.0497/	1.018/1.000	1.006/1.001	1.026/1.000	1.044/1.000	1.018/1.006	1.0395/
Space group	$P6_3/mmc$	H	$P\bar{6}2c$	/	$Fm3m$	$Fm\bar{3}m$	$Fm3m$	$Fm3m$	$I4/m$	$Fm3m$
Lattice (Å)										
a	5.694	4.94	5.70	/	8.1865	8.1844	8.0121	8.0518	5.7446	8.078
b/t ($\times 10^3$)	/	/	/	/	/0	/0	/0	/0	/1.7	/0
c	13.985	13.8	13.99	/	/	/	/	/	8.1099	/
Valence (B, B′)	/	/	/	/	2+/6+	2+/6+	2.5+/5.5+	2.5+/5.5+	2+/6+	2+/6+
T_C/T_N (K)	PM	PM	145-PM	/	110	45-AFM	367	303	40-AFM	40-AFM
M_S (μ_B/f.u.)	/	/	/	/	/	/	3.85	2.40	/	/
H_C (kOe)				/	/	/	/	1.80	/	/

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