Supporting Information S1A. Eight shortest X···O distances (Å) in XO_n^+ polyhedra for $X_2[M(H_2O)_6](SeO_4)_2$ Tutton's salts at 298 K*

X	M	X(10)···O(3)[a]	X(10)···O(3)[b]	X(10)···O(4)[b]	X(10)···O(4)[c]	X(10)···O(5)[c]	X(10)···O(6)[d]	X(10)···O(7)[a]	X(10)···O(8)[e]	δ(Å)[f]
K	Mg[g]	2.836(2)	2.986(2)	3.009(2)	3.451(2)	2.740(2)	2.813(2)	3.136(2)	3.032(2)	0.45
K	Co[g]	2.842(2)	2.975(2)	3.018(2)	3.426(2)	2.750(2)	2.817(2)	3.074(2)	2.977(2)	0.44
K	Ni[h]	2.841(1)	2.947(1)	3.034(1)	3.396(1)	2.746(1)	2.811(1)	3.097(1)	2.995(1)	0.41
K	Cu[i]	2.842(1)	2.992(1)	2.968(1)	3.496(1)	2.749(1)	2.794(1)	2.922(1)	3.023(1)	0.52
K	Zn[i]	2.841(1)	2.974(1)	3.018(1)	3.424(1)	2.753(1)	2.815(1)	3.080(1)	2.979(1)	0.44
Rb	Mg[j]	2.972(2)	3.026(2)	3.210(2)	3.363(2)	2.882(2)	2.946(2)	3.262(2)	3.185(2)	0.26
Rb	Mn[j]	2.998(2)	3.031(2)	3.213(2)	3.385(2)	2.891(2)	2.969(2)	3.241(2)	3.123(2)	0.28
Rb	Co[j]	2.970(2)	3.014(2)	3.214(2)	3.334(2)	2.889(2)	2.946(2)	3.208(2)	3.133(2)	0.25
Rb	Ni[h]	2.964(2)	3.004(2)	3.220(2)	3.316(2)	2.888(2)	2.943(2)	3.224(2)	3.139(1)	0.23
Rb	Cu[i]	2.972(2)	3.070(2)	3.118(2)	3.211(2)	2.955(2)	2.949(2)	3.227(2)	3.069(2)	0.16
Rb	Zn[h]	2.975(1)	3.016(1)	3.209(2)	3.335(2)	2.892(1)	2.946(1)	3.212(2)	3.131(1)	0.25
Tl	Mg[k]	3.019(2)	2.972(2)	3.143(2)	3.324(2)	2.796(2)	2.903(2)	3.362(2)	3.243(2)	0.27
Tl	Mn[k]	3.031(2)	3.001(2)	3.177(2)	3.345(2)	2.795(2)	2.918(2)	3.322(2)	3.165(2)	0.25
Tl	Co[k]	3.001(2)	2.980(2)	3.171(2)	3.298(2)	2.797(2)	2.910(2)	3.283(2)	3.174(2)	0.22
Tl	Ni[k]	3.018(2)	2.944(2)	3.181(2)	3.282(2)	2.789(2)	2.897(2)	3.290(2)	3.186(2)	0.22
Tl	Cu[k]	2.983(2)	3.081(2)	3.084(2)	3.168(2)	2.866(2)	2.923(2)	3.319(2)	3.038(2)	0.26
Tl	Zn[k]	3.008(2)	2.974(2)	3.172(2)	3.294(2)	2.808(2)	2.917(2)	3.297(2)	3.167(2)	0.22
Cs	Mg[l]	3.092(2)	3.152(2)	3.381(2)	3.391(2)	3.064(2)	3.107(2)	3.482(2)	3.372(2)	0.25*
Cs	Mn[l]	3.115(2)	3.159(2)	3.391(2)	3.401(2)	3.063(2)	3.125(2)	3.461(2)	3.334(2)	0.29
Cs	Co[l]	3.098(2)	3.138(2)	3.386(2)	3.368(2)	3.068(2)	3.109(2)	3.429(2)	3.330(2)	0.29
Cs	Ni[l]	3.082(2)	3.132(2)	3.388(2)	3.352(2)	3.066(2)	3.100(2)	3.429(2)	3.318(2)	0.29
Cs	Zn[l]	3.093(2)	3.146(2)	3.381(2)	3.364(2)	3.073(2)	3.106(2)	3.430(2)	3.322(2)	0.29

Symmetry codes for atom O: [a]x, y, z; [b]x - ½, ½ - y, z; [c]½ - x, ½ + y, 1 - z; [d]x - ½, ½ - y, z - 1; [e]½ + x, ½ - y, z. [f]δ(Å) is the deviation of the longest of the eight X···O distances from their average. [g]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 351-354. [h]Simmons, C.J. Unpublished results. [i]This work. [j]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2003**, *218*, 265-268. [k]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 360-364. [l]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2003**, *218*, 405-408. *For the cesium salts, δ′ is defined to be the deviation of the longest of the nine X···O distances (symmetry code: x - ½, ½ - y, z - 1) from their average.

Supporting Information S1B. Eight shortest X···O distances (Å) in XO_n^+ polyhedra for $X_2[M(H_2O)_6](SO_4)_2$ Tutton's salts at 298 K[*]

X	M	X(10)···O(3)[a]	X(10)···O(3)[b]	X(10)···O(4)[b]	X(10)···O(4)[c]	X(10)···O(5)[c]	X(10)···O(6)[d]	X(10)···O(7)[a]	X(10)···O(8)[e]	δ(Å)[f]
Na	Co[g]	2.961(3)	2.962(3)	3.046(2)	3.239(3)	2.770(2)	2.844(2)	3.191(5)	3.035(6)	0.23
K	Mg[h]	2.899(2)	2.944(2)	2.940(2)	3.296(2)	2.725(2)	2.815(2)	3.148(2)	3.003(2)	0.33
K	Fe[h]	2.916(2)	2.947(2)	2.944(2)	3.279(2)	2.733(2)	2.823(2)	3.093(2)	2.950(2)	0.32
K	Co[h]	2.901(2)	2.929(2)	2.950(2)	3.266(2)	2.730(2)	2.817(2)	3.094(2)	2.956(2)	0.31
K	Ni[h]	2.886(2)	2.909(2)	2.962(2)	3.256(2)	2.727(2)	2.811(2)	3.108(2)	2.965(2)	0.30
K	Cu[i]	2.902(1)	2.982(1)	2.882(1)	3.133(1)	2.778(1)	2.828(1)	3.107(1)	2.882(1)	0.20
K	Zn[i]	2.902(1)	2.927(1)	2.947(1)	3.268(1)	2.734(1)	2.818(1)	3.093(1)	2.948(1)	0.31
Rb	Mg[j]	3.011(2)	2.996(2)	3.131(2)	3.248(2)	2.864(2)	2.939(2)	3.266(2)	3.161(2)	0.19
Rb	Cr[k]	3.056(2)	3.016(2)	3.046(2)	3.134(2)	2.926(2)	2.967(2)	3.275(2)	3.014(2)	0.22
Rb	Mn[j]	3.044(2)	3.008(2)	3.135(2)	3.261(2)	2.876(2)	2.959(2)	3.253(2)	3.107(2)	0.18
Rb	Fe[j]	3.019(2)	3.007(2)	3.125(2)	3.231(2)	2.877(2)	2.949(2)	3.219(2)	3.116(2)	0.16
Rb	Co[j]	3.004(2)	2.993(2)	3.133(2)	3.223(2)	2.874(2)	2.940(2)	3.221(2)	3.119(2)	0.16
Rb	Ni[j]	2.989(2)	2.978(2)	3.139(2)	3.216(2)	2.869(2)	2.933(2)	3.224(2)	3.115(2)	0.17
Rb	Cu[i]	2.996(1)	3.049(1)	3.046(1)	3.104(1)	2.945(1)	2.957(1)	3.231(2)	3.034(2)	0.19
Rb	Zn[j]	3.005(2)	2.991(2)	3.127(2)	3.219(2)	2.877(2)	2.941(2)	3.216(2)	3.110(2)	0.16
Tl	Mg[l]	3.098(2)	2.966(2)	3.139(2)	3.151(2)	2.793(2)	2.902(2)	3.363(2)	3.220(2)	0.28
Tl	Mn[l]	3.150(2)	2.971(2)	3.154(2)	3.144(2)	2.790(2)	2.919(2)	3.333(2)	3.140(2)	0.26
Tl	Fe[l]	3.124(2)	2.967(2)	3.161(2)	3.140(2)	2.811(2)	2.923(2)	3.297(2)	3.149(2)	0.23
Tl	Co[l]	3.090(2)	2.958(2)	3.158(2)	3.139(2)	2.804(2)	2.907(2)	3.303(2)	3.146(2)	0.24
Tl	Ni[l]	3.072(2)	2.939(2)	3.137(2)	3.134(2)	2.804(2)	2.903(2)	3.285(2)	3.154(2)	0.23
Tl	Zn[l]	3.081(2)	2.973(2)	3.132(2)	3.136(2)	2.804(2)	2.908(2)	3.293(2)	3.144(2)	0.23
Cs	Mg[m]	3.114(2)	3.128(2)	3.316(2)	3.300(2)	3.054(2)	3.096(2)	3.481(2)	3.359(2)	0.19[*]
Cs	Cr[k]	3.118(2)	3.185(2)	3.241(2)	3.207(2)	3.133(2)	3.122(2)	3.469(2)	3.225(2)	0.30
Cs	Mn[m]	3.140(2)	3.139(2)	3.327(2)	3.311(2)	3.060(2)	3.116(2)	3.454(2)	3.320(2)	0.23
Cs	Fe[m]	3.124(2)	3.133(2)	3.322(2)	3.291(2)	3.062(2)	3.104(2)	3.433(2)	3.320(2)	0.24
Cs	Co[m]	3.113(2)	3.116(2)	3.323(2)	3.285(2)	3.057(2)	3.096(2)	3.423(2)	3.317(2)	0.22
Cs	Ni[i]	3.097(1)	3.110(1)	3.317(1)	3.271(1)	3.053(1)	3.088(1)	3.415(1)	3.309(1)	0.23
Cs	Cu[i]	3.095(1)	3.183(1)	3.238(1)	3.181(1)	3.151(1)	3.112(1)	3.420(1)	3.234(1)	0.29
Cs	Zn[i]	3.109(1)	3.128(1)	3.311(1)	3.279(1)	3.062(1)	3.097(1)	3.419(1)	3.309(1)	0.24

Symmetry codes for atom O: [a]x, y, z; [b]x - ½, ½ - y, z; [c]½ - x, ½ + y, 1 - z; [d]x - ½, ½ - y, z - 1; [e]½ + x, ½ - y, z. [f]δ(Å) is the deviation of the longest of the eight X···O distances from their average. [g]Lu, Y.-K.; Fend, Y.-L. *Chinese J. Inorg. Chem*. **2009**, *25*, 447-453. [h]Bosi, F.; Belardi, G.; Ballirano, P. *Am. Mineral*. **2009**, *94*, 74-82. [i]Simmons, C.J. Unpublished results. [j]Euler, H.; Barbier, B.; Klumpp, S.; Kirfel, A. *Z. Kristallogr*. **2000**, *215*, 473-476. [k]Dobe, A.; Noble, C.; Carver, G.; Tregenna-Piggott, P.L.W.; McIntyre, G.J.; Barra, A.-L.; Neels, A.; Janssen, S.; Juranyi, F. *J. Am. Chem. Soc*. **2004**, *126*, 16639-16652. [l]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 355-359. [m]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2003**, *218*, 409-413. [*]For the cesium salts, δ′ is defined to be the deviation of the longest of the nine X···O distances (symmetry code: x - ½, ½ - y, z - 1) from their average.

Supporting Information S2A. Values of Δ (Å) for non-Jahn-Teller selenate salts

X	M	M-O(7)	M-O(8)	T(K)	Form	Δ (Å)	Ave.	Ref..
K	Mg	2.096	2.095	298	B	0.110		a
K	Co	2.132	2.137	298	B	0.110		a
K	Ni	2.080	2.083	298	B	0.114		a
K	Ni	2.0832	2.0815	298	B	0.110		b
K	Ni	2.0822	2.0789	200	B	0.105		b
K	Ni	2.0841	2.0817	100	B	0.103		b
K	Zn	2.136	2.128	298	B	0.111		a
K	Zn	2.1354	2.1276	298	B	0.110		b
K	Zn	2.1349	2.1274	298	B	0.110	0.109(3)	b
K/Rb*	Zn	2.127	2.120	298	B	0.091		b
Rb	Mg	2.088	2.096	298	A	0.078		c
Rb	Mg	2.089	2.092	298	A	0.074		d
Rb	Mn	2.199	2.203	298	A	0.082		d
Rb	Co	2.121	2.127	298	A	0.070		d
Rb	Ni	2.0725	2.0785	298	A	0.067		b
Rb	Ni	2.0753	2.0780	200	A	0.063		b
Rb	Ni	2.0735	2.0798	100	A	0.065		b
Rb	Ni	2.075	2.076	298	A	0.071		e
Rb	Zn	2.1242	2.1225	298	A	0.070		b
Rb	Zn	2.1243	2.1232	200	A	0.062		b
Rb	Zn	2.1211	2.1264	100	A	0.061		b
Rb	Zn	2.123	2.125	298	A	0.078	0.070(7)	d
Tl	Mg	2.068	2.089	298	A	0.082		f
Tl	Mn	2.184	2.197	298	A	0.086		f
Tl	Co	2.103	2.108	298	A	0.077		f
Tl	Ni	2.067	2.068	298	A	0.074		f
Tl	Zn	2.112	2.116	298	A	0.061		f
Tl	Zn	2.092	2.115	298	A	0.090	0.078(10)	b
Cs	Mg	2.086	2.095	298	A	0.051		c
Cs	Mg	2.082	2.089	298	A	0.047		g
Cs	Mn	2.186	2.195	298	A	0.052		g
Cs	Co	2.112	2.116	298	A	0.044		g
Cs	Ni	2.067	2.074	298	A	0.038		g
Cs	Zn	2.110	2.119	298	A	0.040		g
Cs	Zn	2.111	2.121	298	A	0.040	0.045(6)	b

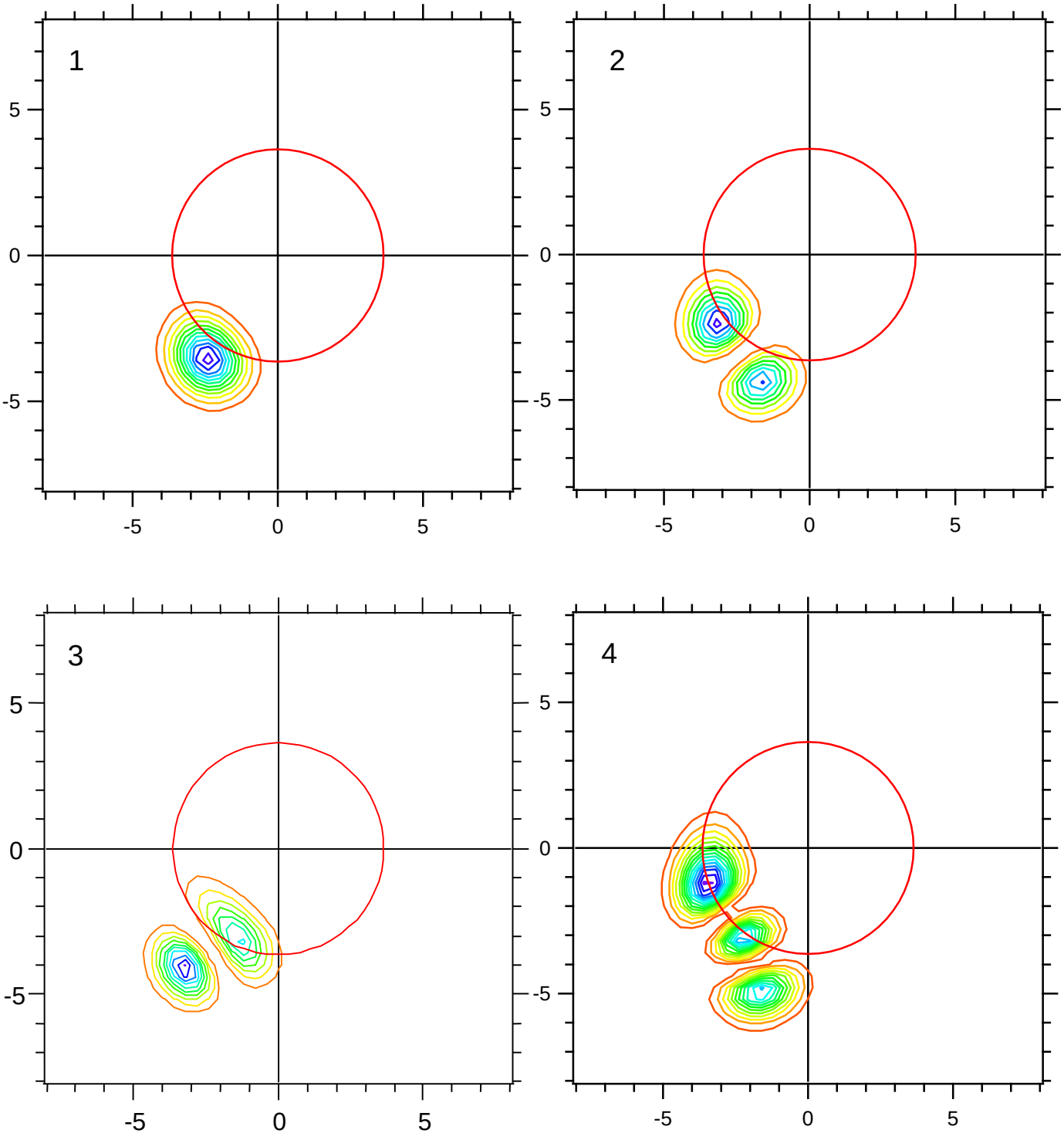
[a]Euler, H.; Barbier, B. Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 351-354. [b]Simmons, C.J. Unpublished results. [c]Micka, Z.; Prokopova, L.; Cisarova, I.; Havlicek, D. *Collect. Czech. Chem. Commun*. **1996**, *61*, 1295-1306. [d]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2003**, *218*, 265-268. [e]Fleck, M.; Kolitsch, U. *Z. Kristallogr*. **2002**, *217*, 15-16. [f]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 360-364. [g]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2003**, *218*, 405-408. *For a mixed crystal of $K_{2x}Rb_{2-2x}[Zn(H_2O)_6](SeO_4)_2$; x = 0.541.

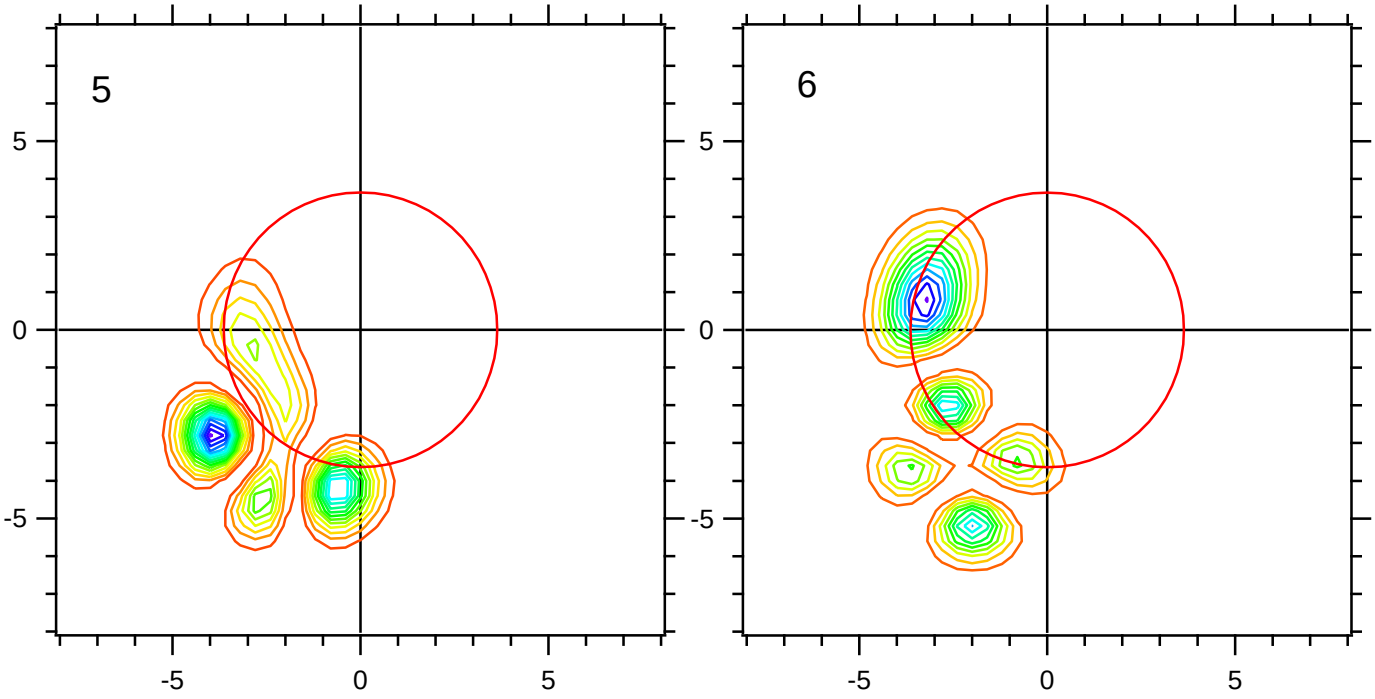
Supporting Information S2B. Values of Δ (Å) for non-Jahn-Teller sulfate salts

X	M	M-O(7)	M-O(8)	T(K)	Form	Δ(Å)	Ave.	Ref.
Na	Co	2.111	2.119	298	A	0.080		a
K	Mg	2.089	2.101	298	A	0.093		b
K	Fe	2.166	2.171	298	A	0.091		b
K	Fe	2.164	2.164	298	A	0.088		c
K	Co	2.122	2.137	298	A	0.086		b
K	Ni	2.076	2.085	298	A	0.094		b
K	Ni	2.073	2.079	298	A	0.099		d
K	Ni	2.078	2.084	298	A	0.088		e
K	Ni	2.077	2.085	200	A	0.083		e
K	Ni	2.078	2.085	95	A	0.083		e
K	Zn	2.124	2.134	298	A	0.093		b
K	Zn	2.123	2.131	298	A	0.088		c
K	Zn	2.126	2.133	298	A	0.087		e
K	Zn	2.124	2.133	260	A	0.087		e
K	Zn	2.123	2.134	120	A	0.087		e
K	Zn	2.124	2.135	95	A	0.083	0.089(4)	e
Rb	Mg	2.083	2.092	298	A	0.057		f
Rb	Mn	2.190	2.204	298	A	0.067		f
Rb	Fe	2.155	2.159	298	A	0.055		f
Rb	Co	2.110	2.122	298	A	0.051		f
Rb	Ni	2.068	2.078	298	A	0.053		f
Rb	Zn	2.112	2.125	298	A	0.053	0.056(6)	f
Tl	Mg	2.064	2.085	298	A	0.052		g
Tl	Mn	2.180	2.193	298	A	0.047		g
Tl	Fe	2.145	2.156	298	A	0.049		g
Tl	Co	2.097	2.118	298	A	0.049		g
Tl	Ni	2.064	2.069	298	A	0.043		g
Tl	Zn	2.098	2.122	298	A	0.051	0.048(3)	g
Cs	Mg	2.075	2.094	298	A	0.031		h
Cs	Mn	2.183	2.201	298	A	0.039		h
Cs	Fe	2.140	2.153	298	A	0.029		h
Cs	Co	2.105	2.120	298	A	0.028		h
Cs	Ni	2.061	2.078	298	A	0.022		h
Cs	Ni	2.064	2.077	298	A	0.023		e
Cs	Ni	2.065	2.0785	200	A	0.019		e
Cs	Ni	2.0654	2.078	100	A	0.018		e
Cs	Zn	2.099	2.126	298	A	0.027		h
Cs	Zn	2.101	2.127	298	A	0.029		e
Cs	Zn	2.1021	2.127	200	A	0.022		e
Cs	Zn	2.105	2.125	100	A	0.019	0.025(6)	e

[a]Lu, Y.-K.; Feng, Y.-L. *Chinese J. Inorg. Chem*. **2009**, *25*, 447-453. [b]Bosi, F.; Belardi, G.; Ballirano, P. *Am. Mineral*. **2009**, *94*, 74-82. [c]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 171-173. [d]Pietrashko, A.; Perekalina, Z.B.; Soboleva, L.V.; Kirpichnikova, L.F. *Crystallogr. Reports* **2000**, *45*, 479-481. [e]Simmons, C.J. Unpublished results. [f]Euler, H.; Barbier, B.; Klumpp, S.; Kirfel, A. *Z. Kristallogr*. **2000**, *215*, 473-476. [g]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2009**, *224*, 355-359. [h]Euler, H.; Barbier, B.; Meents, A.; Kirfel, A. *Z. Kristallogr*. **2003**, *218*, 409-413.

Supporting Information 3. Two-dimensional plots of the probability functions for the first six vibronic energy levels for $K_2[Cu(H_2O)_6](SeO_4)_2$. These have been calculated using the parameters from Table 5.

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