Supporting Information for

Tuning of magnetic properties of the 2D CN-bridged Ni^{II}-Nb^{IV} framework by incorporation of guest cations of alkali and alkaline earth metals

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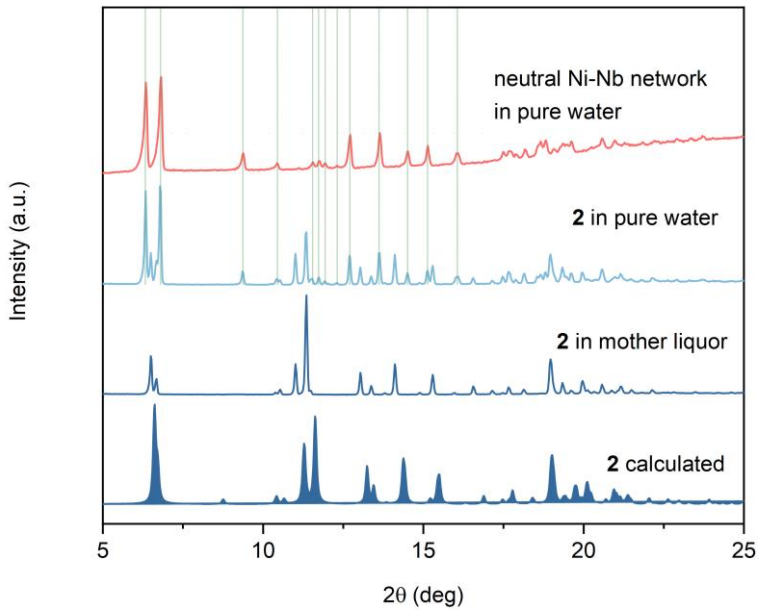

Figure S1. Powder diffraction patterns of **2**: calculated form SC-XRD model at 170 K (bottom), sample in mother liquor, the same sample immersed in pure water for a few days and neutral Ni-Nb network for comparison at room temperature (top).

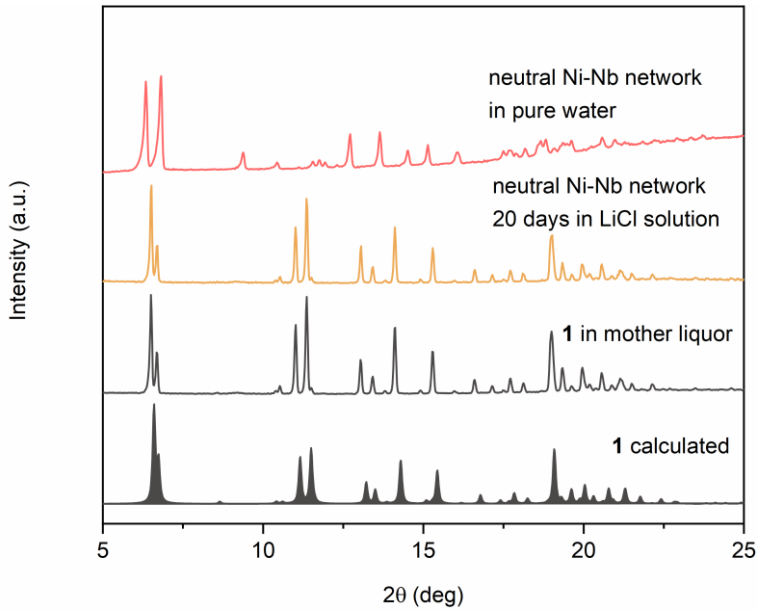

Figure S2. Powder diffraction patterns of neutral Ni-Nb compound in water (top) and the same sample immersed in LiCl water solution for 20 days in comparison with data for **1** in mother liquor at room temperature and diffractogram calculated form SC-XRD model at 100 K (bottom).

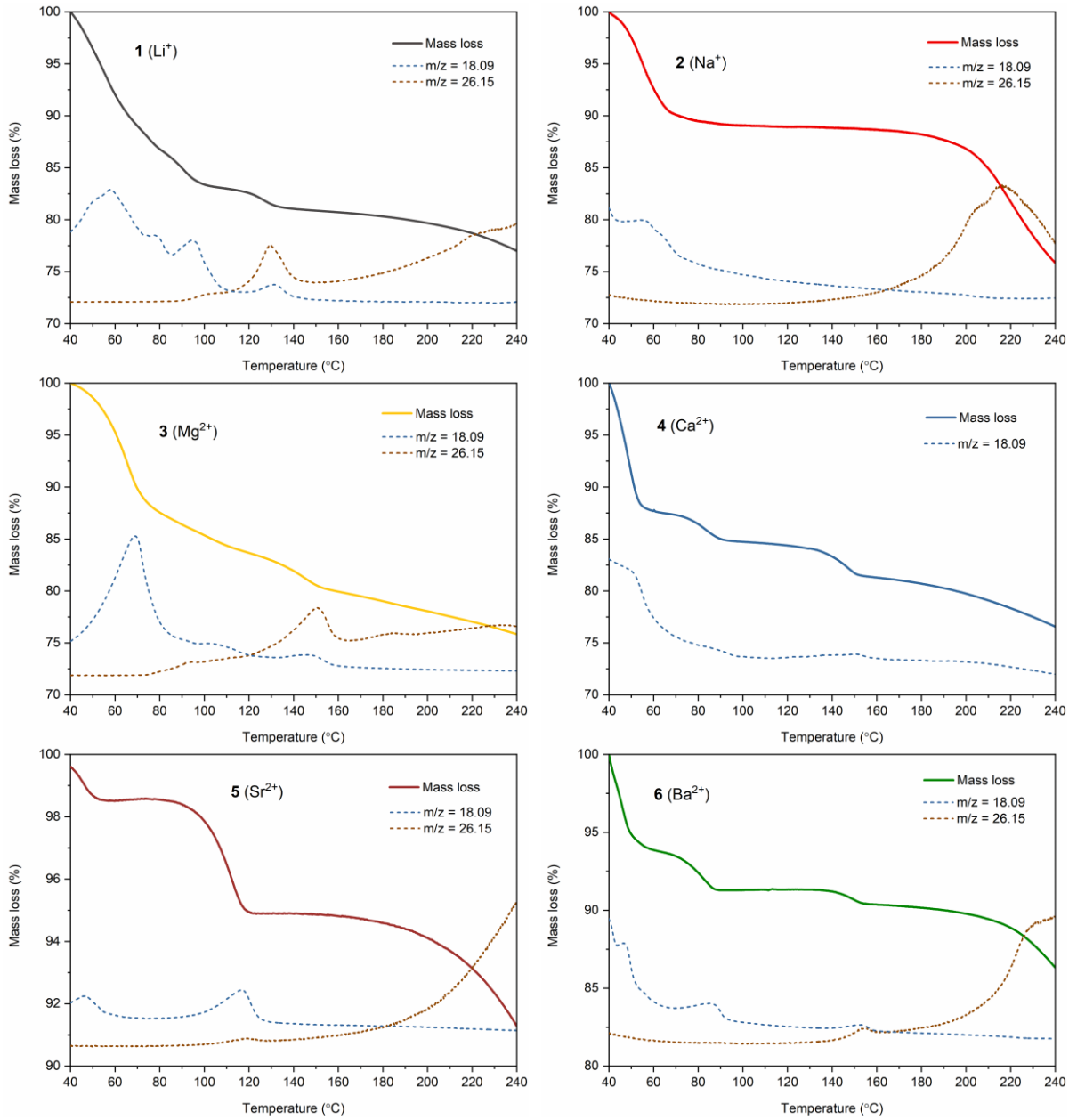

Figure S3. Termogravimetric analysis (solid lines) together with MS scan curves (dashed lines) for compounds **1-6** measured under Ar atmosphere with heating rate of 2°C/min.

Table S1. Continuous Shape Measure parameters for the octa-coordinated niobium centres in compounds **1-6**.

		SAPR-8	TDD-8	BTPR-8	JSD-8
1	Nb1	0.360	1.925	1.214	4.325
2	Nb1	0.354	1.512	1.560	3.940
3	Nb1	0.327	1.714	1.321	4.096
4	Nb1	0.308	1.977	1.331	4.382
5	Nb1	0.319	1.960	1.328	4.376
6	Nb1	0.276	1.485	1.551	3.975
	Nb2	0.555	1.251	1.214	3.683

CShM = 0 indicates an ideal geometry; SAPR-8 = square antiprism, TDD-8 = triangular dodecahedron, BTPR-8 = biaugmented trigonal prism, JSD-8 = snub disphenoid.

Table S2. Continuous Shape Measure parameters for the hexa-coordinated nickel centres in compounds **1-6**.

	1	**2**	**3**	**4**	**5**	**6**
OC-6	Ni1: 0.125 Ni2: 0.128	Ni1: 0.192 Ni2: 0.118 Ni3: 0.154	Ni1: 0.144 Ni2: 0.126	Ni1: 0.120 Ni2: 0.131	Ni1: 0.143 Ni2: 0.142	Ni1: 0.126 Ni2: 0.137 Ni3: 0.116 Ni4: 0.104 Ni5: 0.196

CShM = 0 indicates an ideal geometry; OC-6 = octahedron.

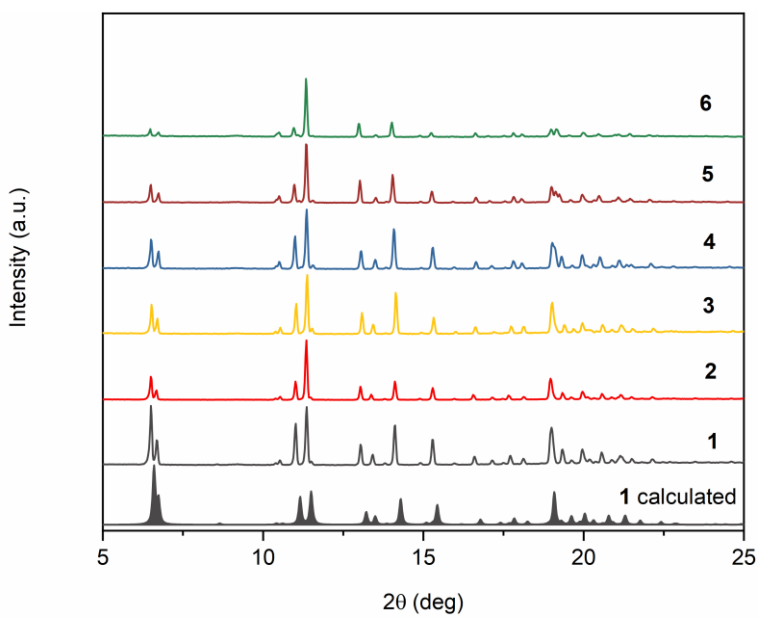

Figure S4. Powder X-Ray diffraction patterns at room temperature for crystalline samples **1-6** in mother liquor in comparison with the reference diffractogram of **1** calculated from SC-XRD.

Table S3. Selected interatomic distances and bond angles in the structures **1**-**6**.

1 (Li^+)	**2** (Na^+)	**3** (Mg^{2+})	**4** (Ca^{2+})	**5** (Sr^{2+})	**6** (Ba^{2+})	
Distance between metal centres (Å)						
Ni1-Nb1: 5.2521(5) Ni2-Nb1: 5.3750(4)	Ni1-Nb1: 5.4401(10) Ni2-Nb1: 5.2528(7) Ni3-Nb1: 5.3104(8)	Ni1-Nb1: 5.3791(6) Ni2-Nb1: 5.2459(3)	Ni1-Nb1: 5.2220(14) Ni2-Nb1: 5.3626(9)	Ni1-Nb1: 5.2187(6) Ni2-Nb1: 5.3714(4)	Ni1-Nb1: 5.2489(12) Ni2-Nb1: 5.3458(7) Ni3-Nb1: 5.3712(7)	Ni1-Nb2: 5.2067(12) Ni4-Nb2: 5.3772(7) Ni5-Nb2: 5.3799(7)
Ni-N_{CN} distance (Å)						
Ni1-N1: 2.086(4) Ni2-N2: 2.092(3)	Ni1-N2: 2.130(9) Ni2-N4: 2.097(6) Ni3-N7: 2.086(7)	Ni1-N1: 2.099(3) Ni1-N7: 2.110(3) Ni2-N2: 2.086(3)	Ni1-N2: 2.078(5) Ni2-N1: 2.096(3)	Ni1-N1: 2.074(7) Ni2-N2: 2.093(5)	Ni1-N1: 2.081(7) Ni1-N1A: 2.081(7) Ni2-N2: 2.062(7)	Ni3-N3: 2.109(7) Ni4-N4A: 2.101(7) Ni5-N5A: 2.142(7)
Ni-N_{cyclam} distance (Å)						
Ni1-N11: 2.071(4) Ni2-N21: 2.076(6) Ni2-N22: 2.073(7)	Ni1-N11: 2.078(7) Ni1-N12: 2.071(7) Ni2-N21: 2.057(8) Ni2-N22: 2.095(10) Ni3-N31: 2.082(9) Ni3-N32: 2.103(9)	Ni1-N11: 2.071(3) Ni1-N12: 2.079(3) Ni1-N13: 2.068(3) Ni1-N14: 2.076(3) Ni2-N21: 2.082(3) Ni2-N22: 2.080(3)	Ni1-N11: 2.073(4) Ni2-N21: 2.070(4) Ni2-N22: 2.065(4)	Ni1-N11: 2.074(5) Ni2-N21: 2.063(5) Ni2-N22: 2.075(5)	Ni1-N11: 2.069(8) Ni1-N12: 2.071(7) Ni1-N13: 2.089(7) Ni1-N14: 2.074(7) Ni2-N21: 2.074(8) Ni2-N22: 2.071(9)	Ni3-N31: 2.087(10) Ni3-N32: 2.080(10) Ni4-N41: 2.060(7) Ni4-N42: 2.083(7) Ni5-N51: 2.071(7) Ni5-N52: 2.067(7)
Ni-N≡C angle (°)						
Ni1-N1-C1: 151.4(4) Ni2-N2-C2: 158.7(3)	Ni1-N2-C2: 157.7(7) Ni2-N4-C4: 151.0(6) Ni3-N7-C7: 158.5(8)	Ni1-N1-C1: 159.5(3) Ni1-N7-C7: 157.4(3) Ni2-N2-C2: 151.8(2)	Ni1-N2-C2: 150.9(5) Ni2-N1-C1: 158.5(3)	Ni1-N1-C1: 150.6(6) Ni2-N2-C2: 158.3(4)	Ni1-N1-C1: 151.0(7) Ni1-N1A-C1A: 150.6(7) Ni2-N2-C2: 159.4(7)	Ni3-N3-C3: 159.1(6) Ni4-N4A-C4A: 156.0(6) Ni5-N5A-C5A: 153.0(6)

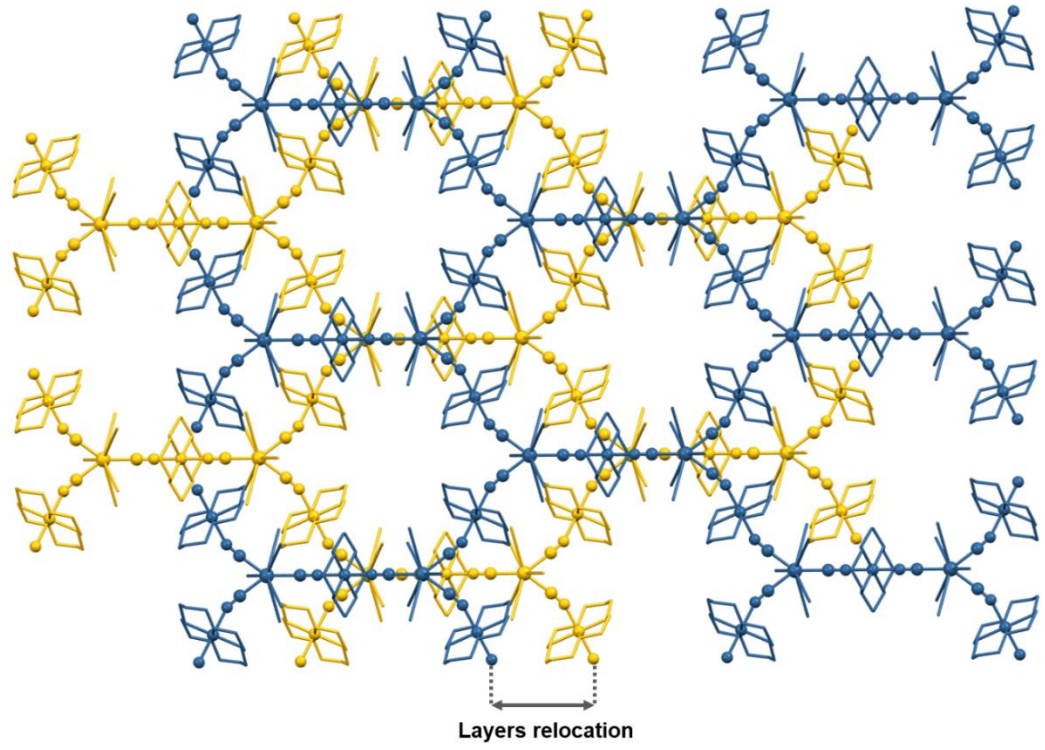

Figure S5. Structure of **1** viewed perpendicular to the layers, showing the relative relocation of two neighbouring layers (water molecules and lithium cations are omitted for clarity).

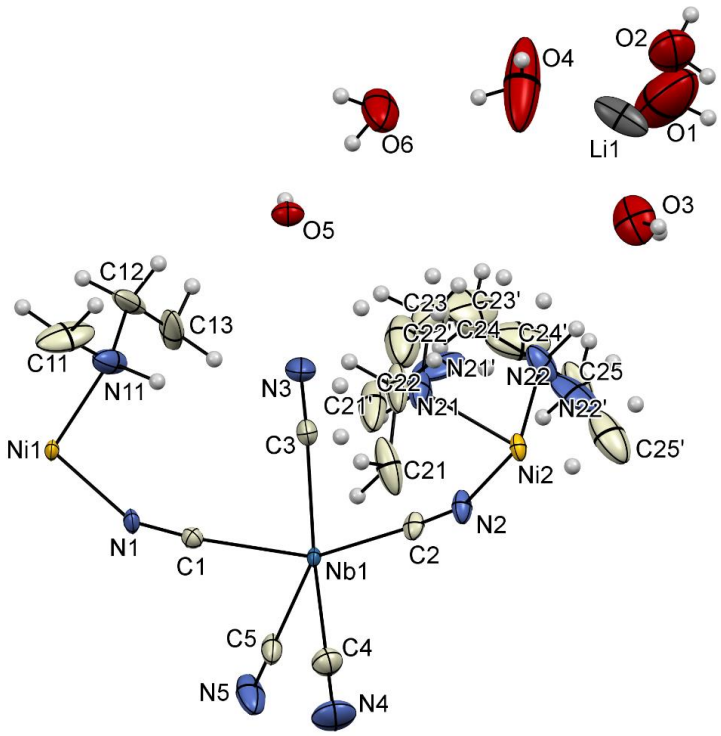

Figure S6. Asymmetric unit of **1**; thermal ellipsoids shown at 50% probability.

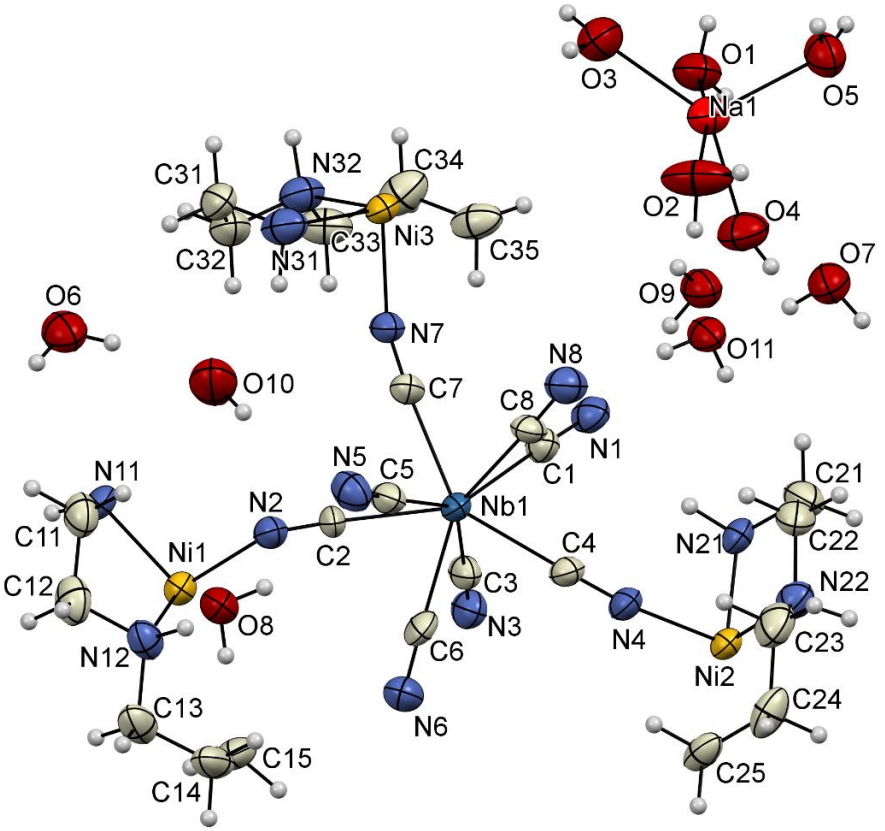

Figure S7. Asymmetric unit of **2**; thermal ellipsoids shown at 50% probability.

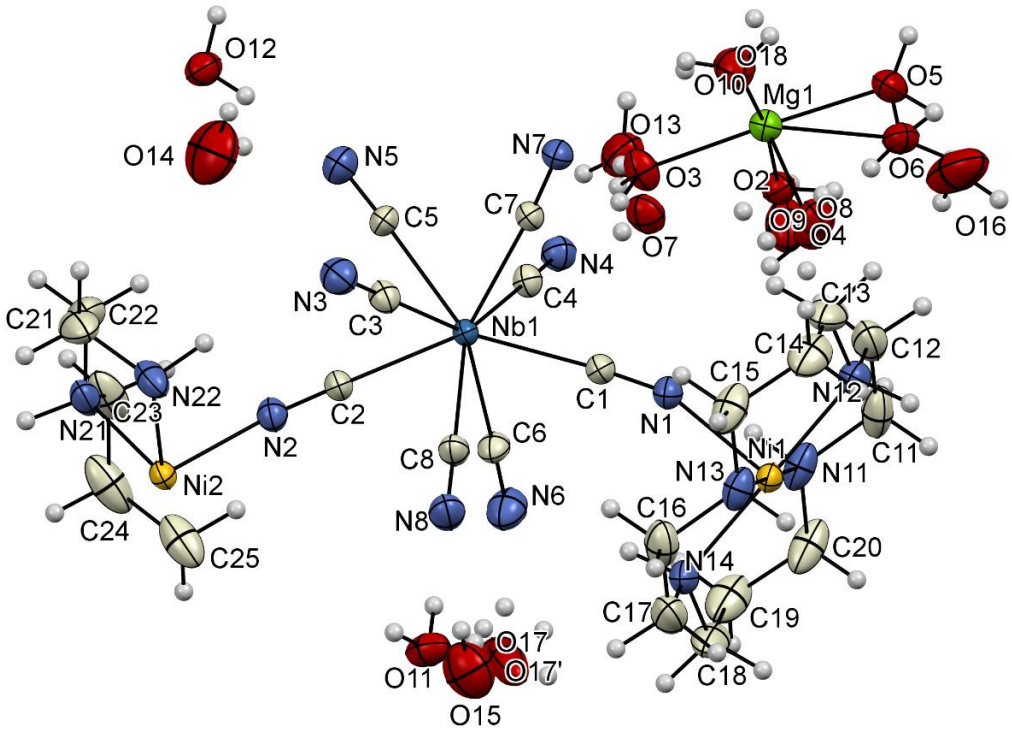

Figure S8. Asymmetric unit of **3**; thermal ellipsoids shown at 50% probability.

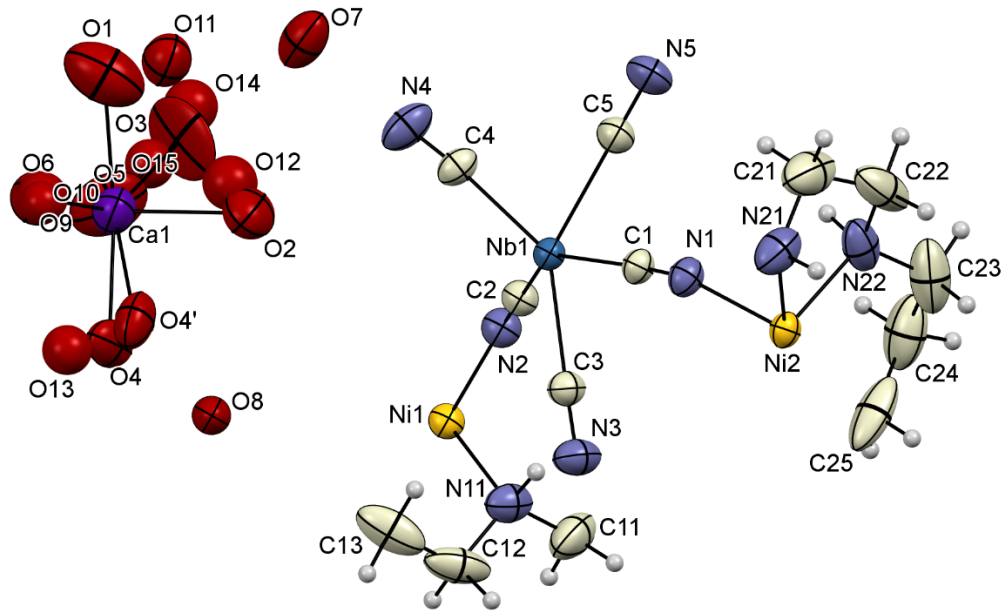

Figure S9. Asymmetric unit of **4**; thermal ellipsoids shown at 50% probability.

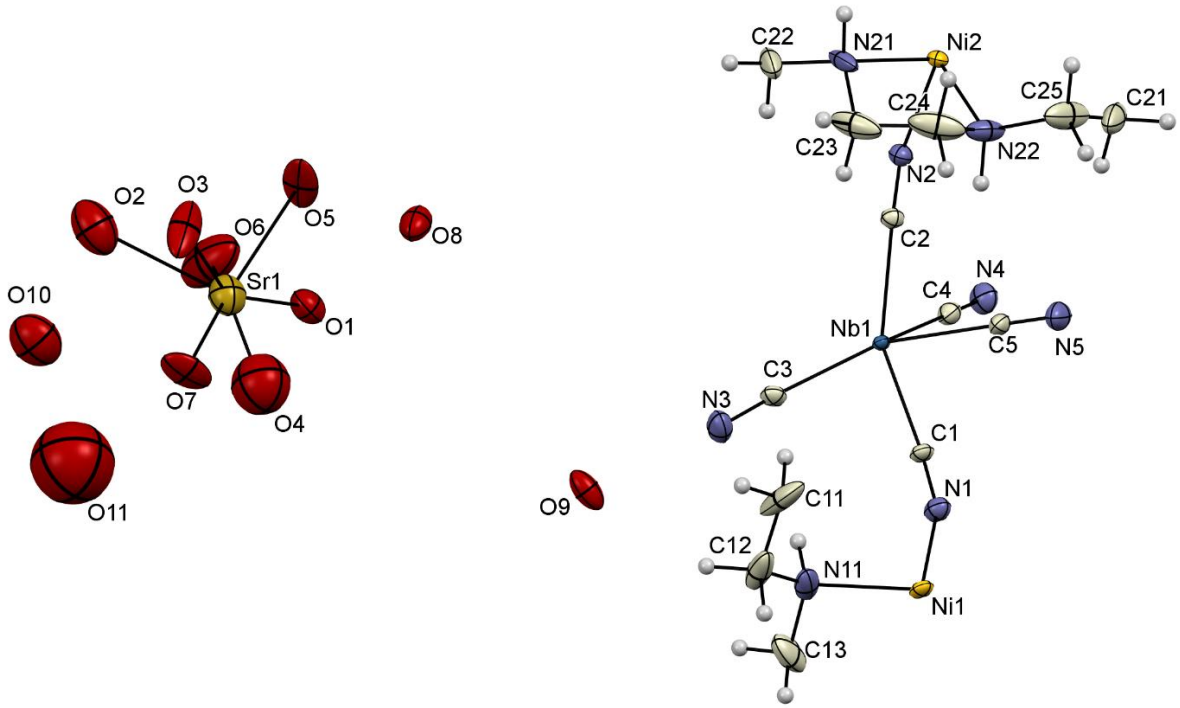

Figure S10. Asymmetric unit of **5**; thermal ellipsoids shown at 50% probability.

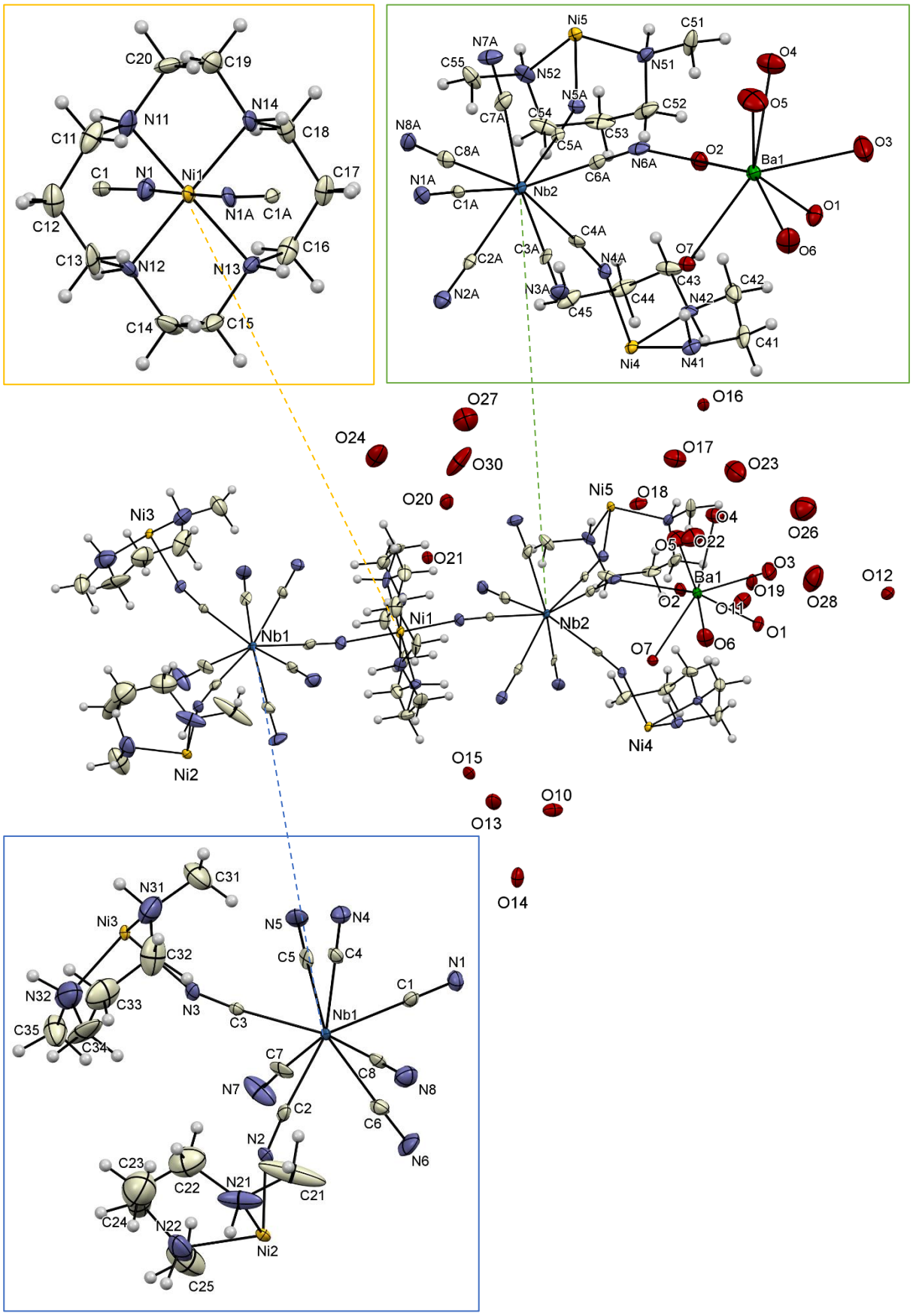

Figure S11. Asymmetric unit of **6**; thermal ellipsoids shown at 50% probability.

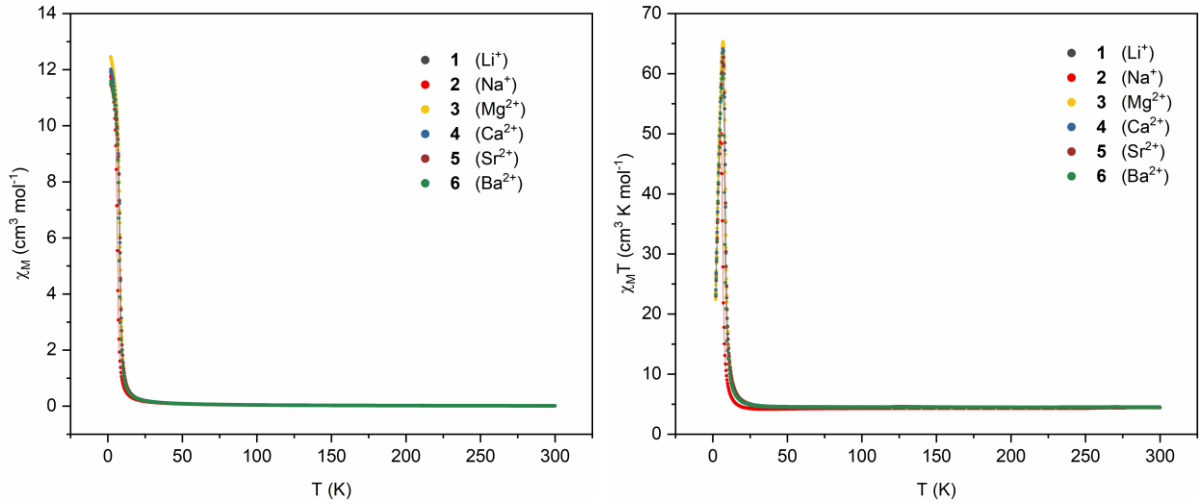

Figure S12. χ and χT vs. temperature plots in full temperature range at constant magnetic field of 1 kOe.

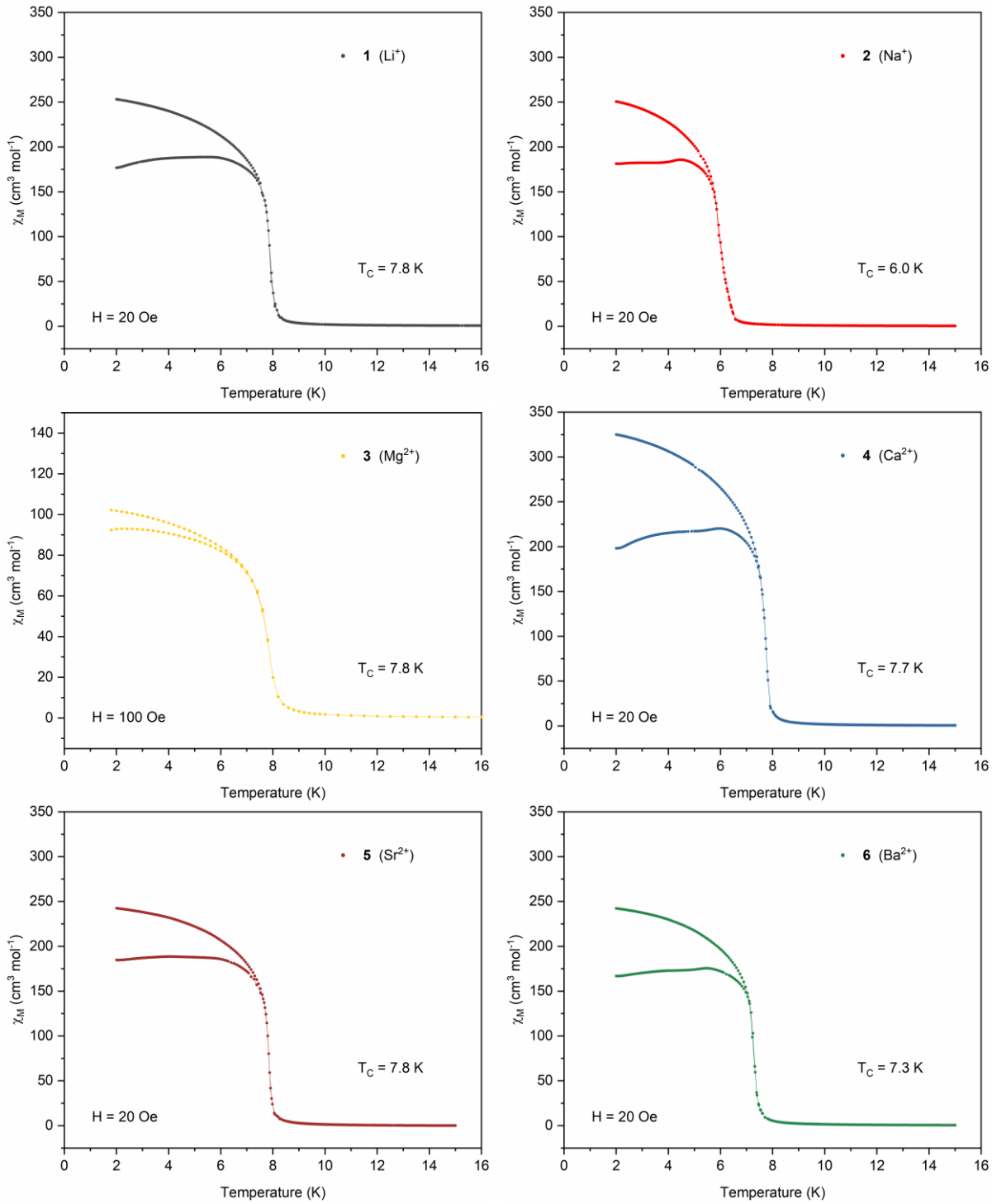

Figure S13. Magnetic susceptibility measurements on zero-field-cooled and field-cooled samples of **1**-**6**.

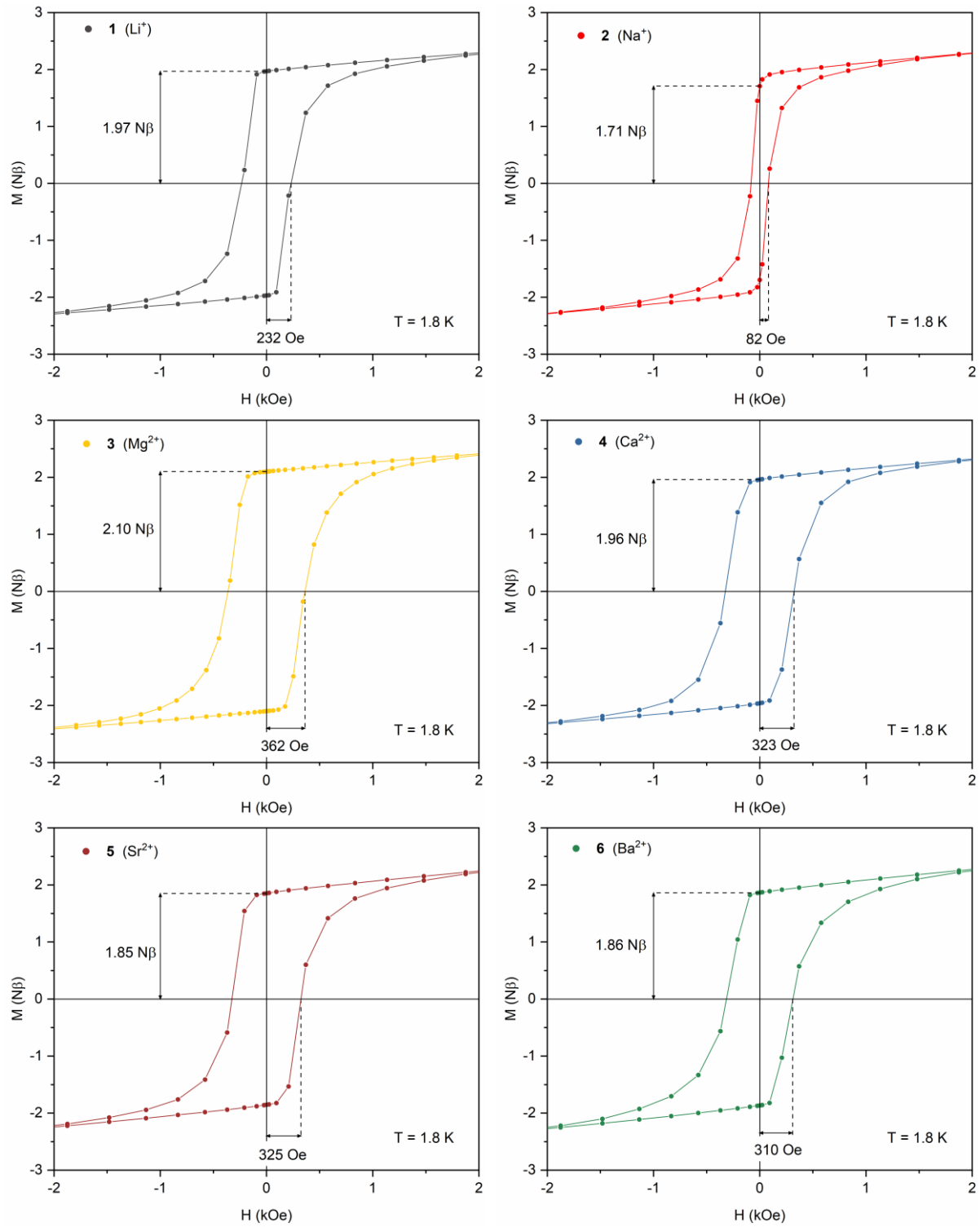

Figure S14. Magnetization vs. magnetic field measurements at 1.8 K with hysteresis parameters (remnant magnetization and coercive field).