Supplementary Information For

Cobalt and zinc halide complexes of 4-chloro and 4-methylaniline: syntheses, structures and magnetic behavior.

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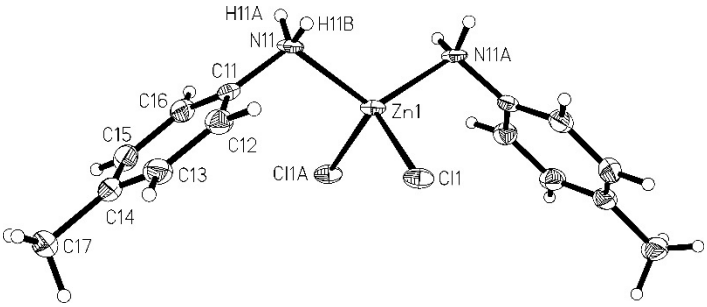

Figure S1: A plot of the molecular unit of compound **2** showing 50% probability thermal ellipsoids. Hydrogen atoms are shown as spheres of arbitrary size. The asymmetric unit, copper coordination sphere and hydrogen atoms whose positions were refined are labeled.

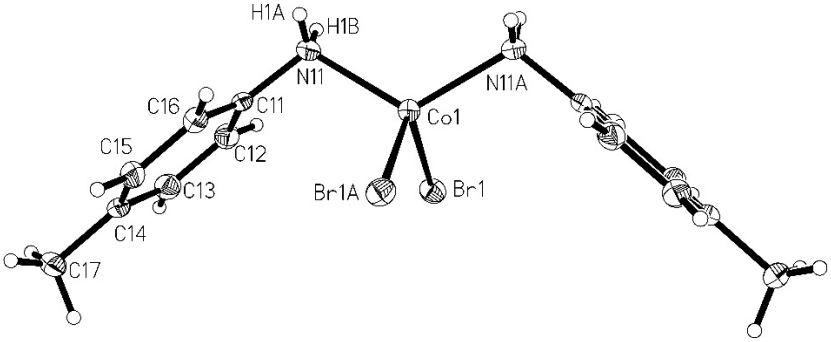

Figure S2: A plot of the molecular unit of compound **3** showing 50% probability thermal ellipsoids. Hydrogen atoms are shown as spheres of arbitrary size. The asymmetric unit, copper coordination sphere and hydrogen atoms whose positions were refined are labeled.

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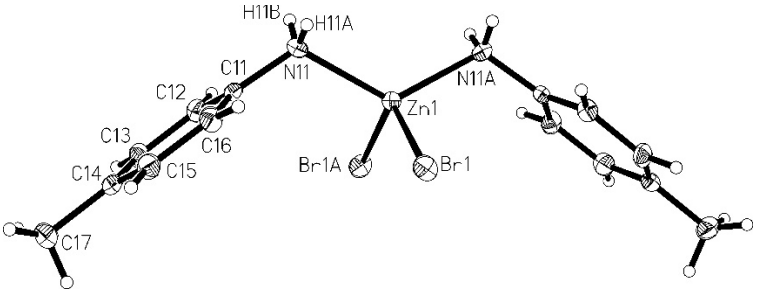

Figure S3: A plot of the molecular unit of compound **4** showing 50% probability thermal ellipsoids. Hydrogen atoms are shown as spheres of arbitrary size. The asymmetric unit, copper coordination sphere and hydrogen atoms whose positions were refined are labeled.

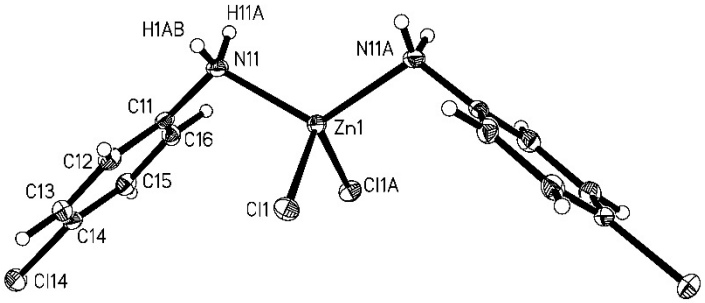

Figure S4: A plot of the molecular unit of compound **6** showing 50% probability thermal ellipsoids. Hydrogen atoms are shown as spheres of arbitrary size. The asymmetric unit, copper coordination sphere and hydrogen atoms whose positions were refined are labeled.

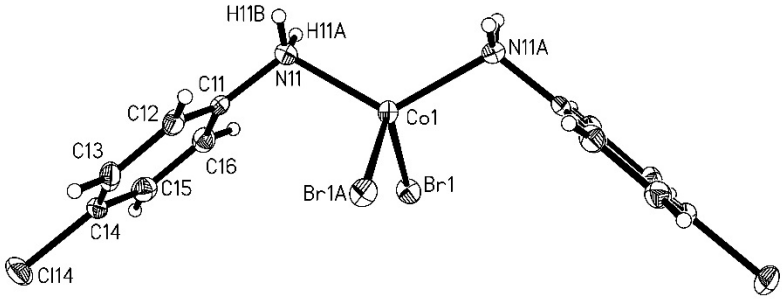

Figure S5: A plot of the molecular unit of compound **7** showing 50% probability thermal ellipsoids. Hydrogen atoms are shown as spheres of arbitrary size. The asymmetric unit, copper coordination sphere and hydrogen atoms whose positions were refined are labeled.

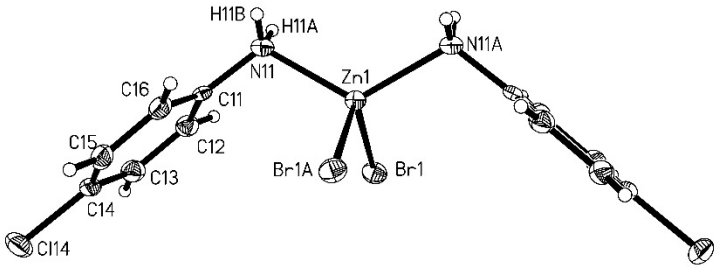

Figure S6: A plot of the molecular unit of compound **8** showing 50% probability thermal ellipsoids. Hydrogen atoms are shown as spheres of arbitrary size. The asymmetric unit, copper coordination sphere and hydrogen atoms whose positions were refined are labeled.

SI-1: SIMULATION AND MODELLING

The numerical solution of magnetic susceptibility of the S=3/2 rectangular lattice was obtained by quantum Monte Carlo simulations with the "looper" application of the ALPS libraries. [1] For all simulations, the spins were arranged on a 64x64 square lattice (ignoring 3D interaction). Along the edges two antiferromagnetic interactions *J*, *J'* were present in the single-*J* form Hamiltonian, and the ratio *J'/J* was defined as α ($0 \leq \alpha \leq 1$, step size 0.05). The scaled temperature window for the simulation was set to [0.05*T/J* to 300 *T/J*]. The simulated data for each α were fitted to the ratio of polynomial expressions with given *C*, *J* to extract the coefficients $A_{n,\alpha}$ and $B_{n,\alpha}$ according to Equation (1)

$$\chi(\alpha, T) = \frac{C}{T}\frac{1 + \sum_{n=1}^{5} A_{n,\alpha}(J/T)^n}{1 + \sum_{n=1}^{5} B_{n,\alpha}(J/T)^n} \qquad Eq.(1)$$

The empirical formulas are found to be valid for the temperature range $T/J > 0.4$ and the values of $A_{n,\alpha}$ and $B_{n,\alpha}$ are listed in Table S1. The square lattice model corresponds to the α=1 case.

Table S1. Fitted values of $A_{n,\alpha}$ and $B_{n,\alpha}$ for building the expression of susceptibility [Equation (1)] of the S=3/2 rectangle model.

α	$A_{1,\alpha}$	$A_{2,\alpha}$	$A_{3,\alpha}$	$A_{4,\alpha}$	$A_{5,\alpha}$	$B_{1,\alpha}$	$B_{2,\alpha}$	$B_{3,\alpha}$	$B_{4,\alpha}$	$B_{5,\alpha}$
0	0.396205	0.963443	0.093099	-0.00936	0.000112	2.897135	5.67027	6.98343	2.55916	-0.14614
0.05	0.769989	2.213246	0.901033	0.422884	-0.00062	3.394935	7.993201	12.28081	12.0844	5.434277
0.10	-0.65041	0.499495	-0.35749	0.192128	-0.00034	2.100521	2.458959	1.688069	-3.85436	2.256264
0.15	-0.817	0.666312	-0.34065	0.274691	-0.00054	2.058582	2.123834	1.480604	-3.00184	3.139848
0.20	-1.03865	0.839758	-0.36229	0.390542	-0.00082	1.962022	1.595264	1.156154	-2.57609	4.457846
0.25	-1.26431	1.040523	-0.39691	0.511196	-0.00121	1.861439	1.041646	0.923753	-2.11181	5.871482

0.30	-1.42706	1.210714	-0.40996	0.636565	-0.00157	1.823712	0.624263	0.887352	-1.55887	7.447175
0.35	-1.63518	1.457654	-0.46745	0.748126	-0.00233	1.740843	0.105638	0.877573	-0.86552	8.806197
0.40	-1.77652	1.657993	-0.50187	0.900245	-0.0031	1.724577	-0.25986	1.05198	-0.22754	10.81874
0.45	-1.91938	1.864754	-0.54213	1.042632	-0.00362	1.706946	-0.64516	1.257447	0.422289	12.83349
0.50	-2.01474	2.029443	-0.56131	1.273862	-0.00405	1.736645	-0.90925	1.569569	0.916277	16.24473
0.55	-2.11508	2.216227	-0.60568	1.496705	-0.00476	1.761713	-1.18457	1.957346	1.41143	19.6484
0.60	-2.25168	2.454528	-0.68157	1.706434	-0.00556	1.750198	-1.5506	2.278049	2.036454	22.95271
0.65	-2.34065	2.632797	-0.71374	1.93295	-0.00643	1.786567	-1.79948	2.629892	2.838001	26.7375
0.70	-2.46305	2.894968	-0.84127	2.240106	-0.00816	1.789328	-2.11252	3.130903	3.186335	31.66576
0.75	-2.50336	3.000677	-0.83271	2.637955	-0.00797	1.874206	-2.23455	3.559377	3.497411	38.89372
0.80	-2.5847	3.172927	-0.86155	2.877478	-0.00879	1.917893	-2.45657	3.842443	4.601108	43.5635
0.85	-2.66801	3.405841	-1.00162	3.37745	-0.011	1.960369	-2.65083	4.522825	4.491997	52.43699
0.90	-2.79326	3.682372	-1.13719	3.606621	-0.01315	1.95997	-2.98127	4.897639	5.502365	57.03889
0.95	-2.81764	3.778448	-1.12938	4.159222	-0.01308	2.060566	-3.01454	5.297742	5.955627	68.20129
1.00	-2.93113	4.10106	-1.39978	4.769242	-0.01599	2.072726	-3.28065	6.168849	5.084592	79.81426

SI-2 Magnetic data for 5 and 7

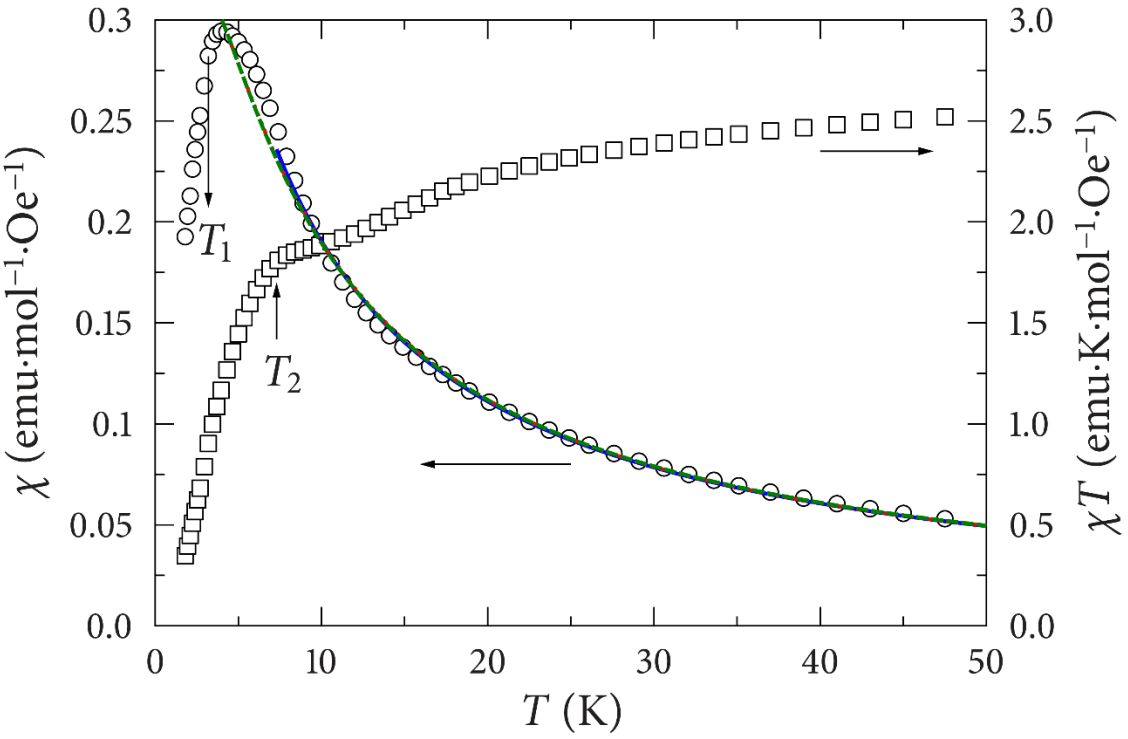

Figure S7. Temperature dependence of χ (○) and χT (□) for **5**. The solid line, dotted line, dashed line are fits using the Curie-Weiss model, 1D chain model and 2D square lattice model, respectively.

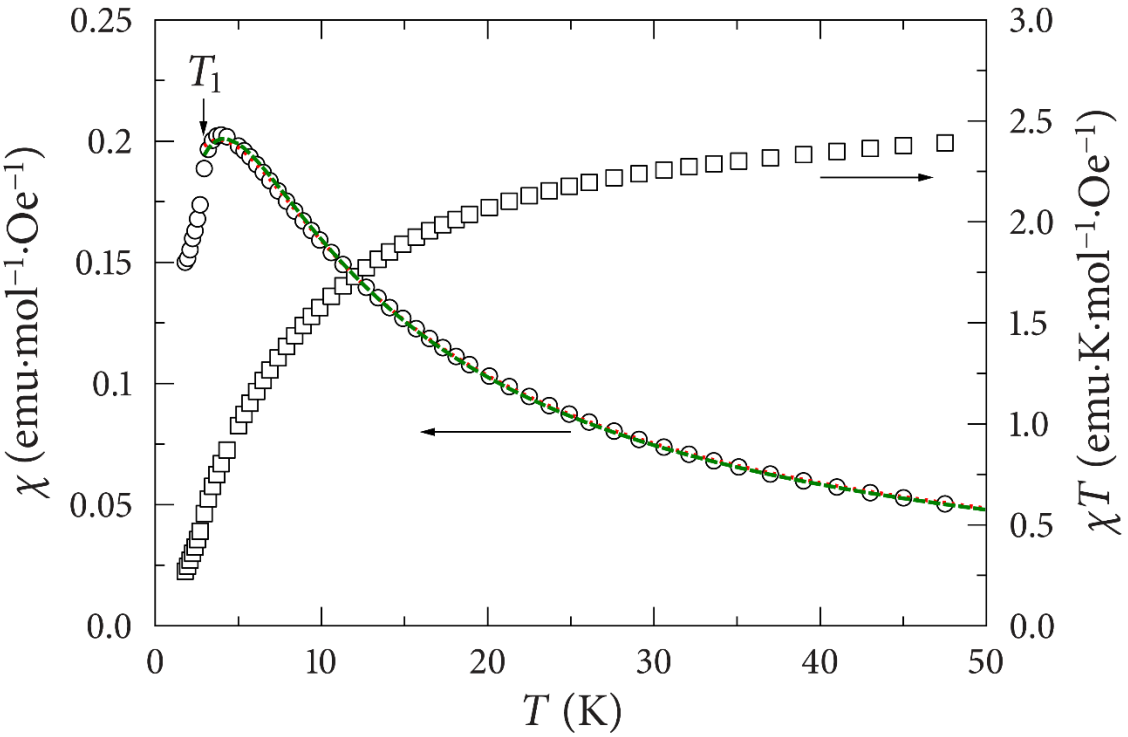

Figure S8. Temperature dependence of χ (○) and χT (□) for **7**. The dotted lines and dashed lines are fits using the 1D chain model and 2D rectangle lattice model, respectively.

1. B. Bauer, L.D. Carr, H.G. Evertz, A. Feiguin, J. Freire, S. Fuchs, L. Gamper, J. Gukelberger, E. Gull, S. Guertler, A. Hehn, R. Igarashi, S.V. Isakov, D. Koop, P.N. Ma, P. Mates, H. Matsuo, O. Parcollet, G. Pawłowski, J.D. Picon, L. Pollet, E. Santos, V.W. Scarola, U. Schollwöck, C. Silva, B. Surer, S. Todo, S. Trebst, M. Troyer, M.L. Wall, P. Werner and S. Wessel (ALPS collaboration) *J. Stat. Mech.* 2011, P05001.