Ab initio, artificial neural network predictions and experimental synthesis of mischmetal alloying in Sm-Co permanent magnets

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Supplementary Information

TABLES

Table S1: Total magnetization per atom and per formula unit for SmCo5 by using an 11 and a 16 e- configuration as valence, along with a U parameter of 2.22 eV for the 3d orbitals of Co and 4.7 eV for the 4f orbitals of Sm. Results are presented for simulations performed with and without considering the impact of SOC.

	11 valence e^-		16 valence e^-	
m/atom (μB/atom)	**DFT+U**	**DFT+U (SOC)**	**DFT+U**	**DFT+U (SOC)**
1a (1×RE)	-0.316	-0.301	-5.668	-5.630
2c average (2×Co)	1.748	1.736	1.783	1.780
3g average (3×Co)	1.792	1.783	1.816	1.816
Total (μB/f.u.)	8.557	8.520	3.345	3.376

Table S2: Orbital moment per atom and per formula unit for $SmCo_5$ by using an 11 and a 16-valence e^- configuration as valence, along with a U parameter of 2.22 eV for the 3d orbitals of Co and 4.7 eV for the 4f orbitals of Sm.

m_L/atom (μB/atom)	11 valence e^- (SRM)	16 valence e^-
1a (1×RE)	0.087	2.115
2c average (2×Co)	0.437	0.315
3g average (3×Co)	0.352	0.424
Total (μB/f.u.)	2.017	4.017

Table S3: Total magnetization per atom and per formula unit for $SmCo_5$ by using a 16 e^- configuration as valence, with two specified sets of values for the U parameter for the 3d orbitals of Co and the 4f orbitals of Sm. The impact of SOC was considered.

m/atom (μB/atom)	$U_{Sm} = U_{Co}$ = 0 eV	U_{Sm} = 6 eV, U_{Co} = 0 eV
1a (1×RE)	-0.162	4.796
2c average (2×Co)	1.450	1.508
3g average (3×Co)	1.467	1.526
Total (μB/f.u.)	7.138	12.390

Table S4: Orbital moment per atom and per formula unit for $SmCo_5$ by using a 16 e^- configuration as valence, with two specified sets of values for the U parameter for the 3d orbitals of Co and the 4f orbitals of Sm. The impact of SOC was considered.

m_L/atom (μB/atom)	$U_{Sm} = U_{Co}$ = 0.0 eV	U_{Sm} = 6.0 eV, U_{Co} = 0.0 eV
1a (1×RE)	-0.846	-1.423
2c average (2×Co)	0.162	0.143
3g average (3×Co)	0.151	0.127
Total (μB/f.u.)	-0.070	-0.756

Table S5: Correlation matrix for the lattice constant, *a*, *c/a* ratio, total magnetization per formula unit and relative total energy of the 34 2×2×2 supercell systems comprising the 2×2×2 dataset.

	a	*c/a*	m/f.u.	Relative E per atom
a	1.000	-0.976	0.178	-0.231
c/a	-0.976	1.000	-0.201	0.245
m/f.u.	0.178	-0.201	1.000	0.315
Relative E per atom	-0.231	0.245	0.315	1.000

Table S6: Correlation matrix for the lattice constant, *a*, *c/a* ratio, total magnetization per formula unit and relative total energy of the 400 2×2×4 supercell systems comprising the 2×2×4 dataset.

	a	*c/a*	m/f.u.	Relative E per atom
a	1.000	-0.817	0.104	0.333
c/a	-0.817	1.000	-0.192	-0.579
m/f.u.	0.104	-0.192	1.000	0.284
Relative E per atom	0.333	-0.579	0.284	1.000

Table S7: Values of enthalpy of formation per formula unit with respect to the $SmCo_5$, $LaCo_5$ and $CeCo_5$ binary compounds, total magnetization per formula unit, lattice constant, a, and c/a ratio for the 2×2×2 dataset configurations that were found unstable with respect to the binary compounds.

ΔH_{binary}/f.u. (meV)	m/f.u. (μB/f.u.)	a (Å)	c/a
38.7	7.946	5.255	0.743
44.8	8.133	5.038	0.774

Table S8: Values of enthalpy of formation per formula unit with respect to the $SmCo_5$, $LaCo_5$ and $CeCo_5$ binary compounds, total magnetization per formula unit, lattice constant, a, and c/a ratio for the 2×2×4 dataset configurations that were found unstable with respect to the binary compounds.

ΔH_{binary}/f.u. (meV)	m/f.u. (μB/f.u.)	a (Å)	c/a
13.1	7.465	5.067	0.772
7.2	7.459	5.050	0.778
1.8	7.459	5.036	0.776
8.6	7.455	5.021	0.782
2.3	7.461	5.036	0.787
39.1	7.464	5.055	0.773
17.9	7.471	5.067	0.776
0.5	7.460	5.070	0.775
10.5	7.460	5.065	0.772
15.9	7.455	5.060	0.773
31.2	7.468	5.063	0.773
2.4	7.450	5.000	0.812

FIGURES

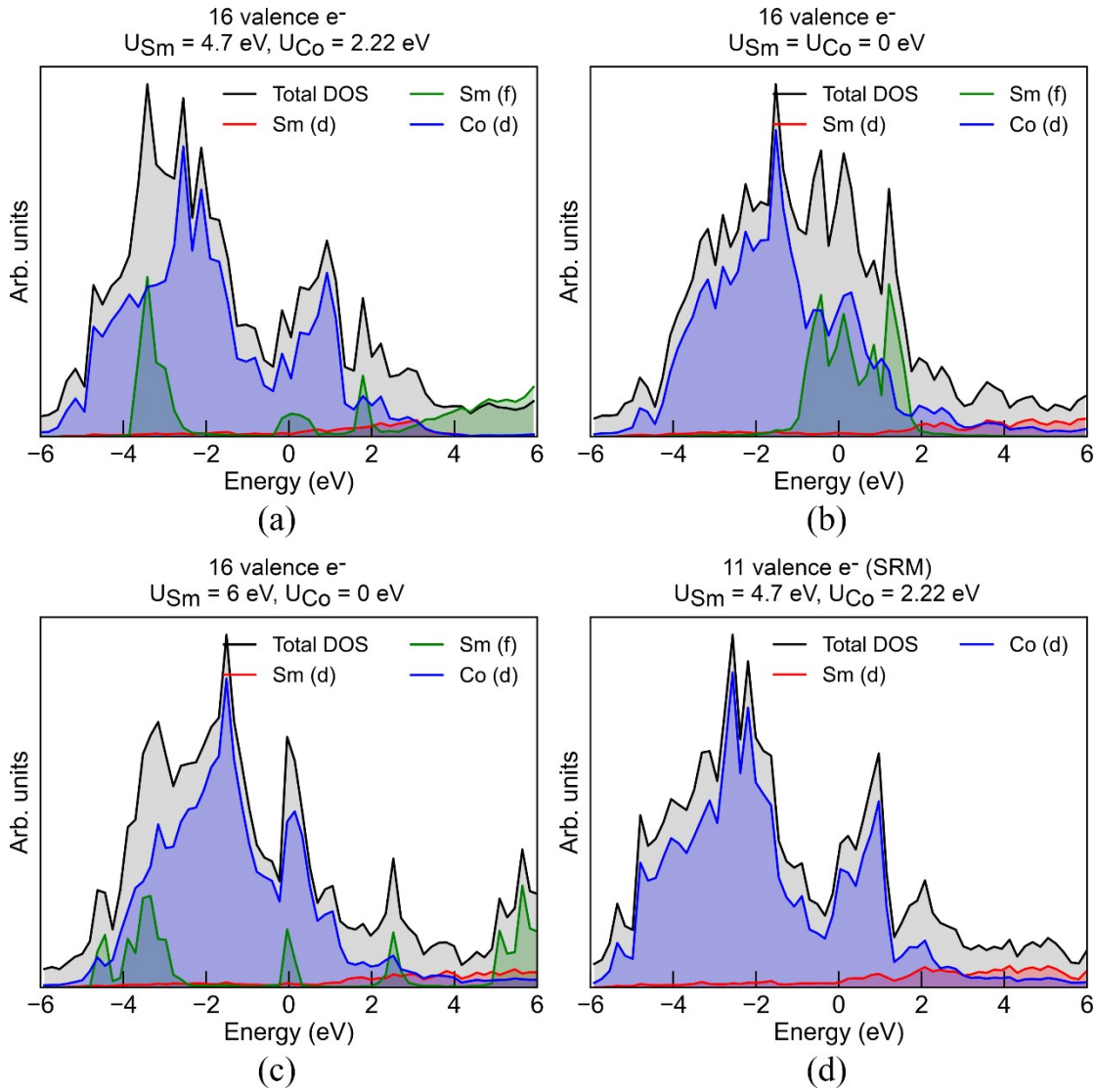

Figure S1: Electronic total and projected DOS of $SmCo_5$, as calculated by using a 16 e^- configuration as valence with three different sets of the U parameter for Sm and Co (a-c) and by using the SRM (d) with U_{Sm} = 4.7 eV, U_{Co} = 2.22 eV.

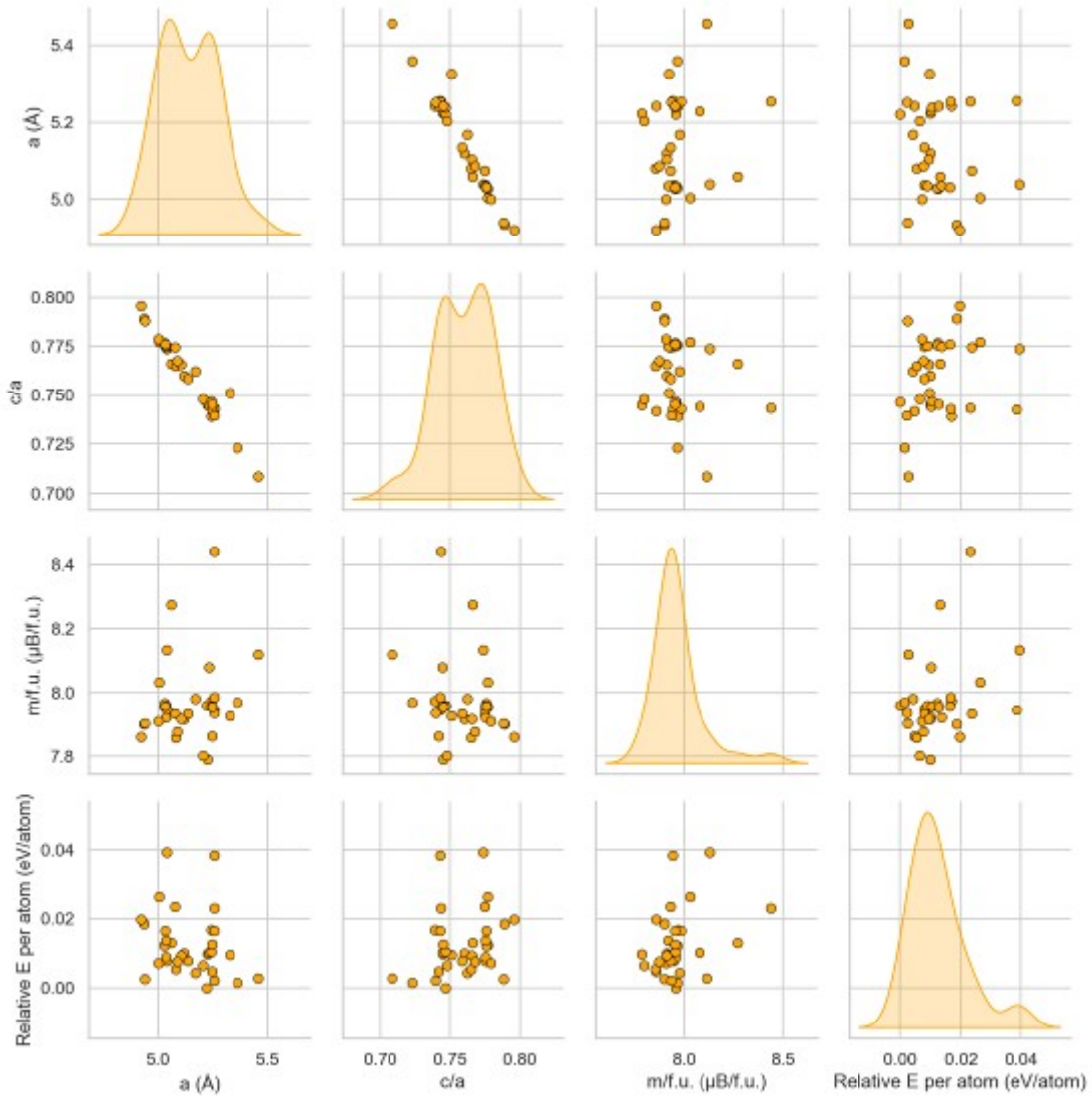

Figure S2: Scatter matrix plot with a kernel density estimation fit in the diagonal plot elements for the lattice constant, *a*, *c/a* ratio, total magnetization per formula unit and relative total energy of the 34 2×2×2 supercell systems comprising the 2×2×2 dataset.

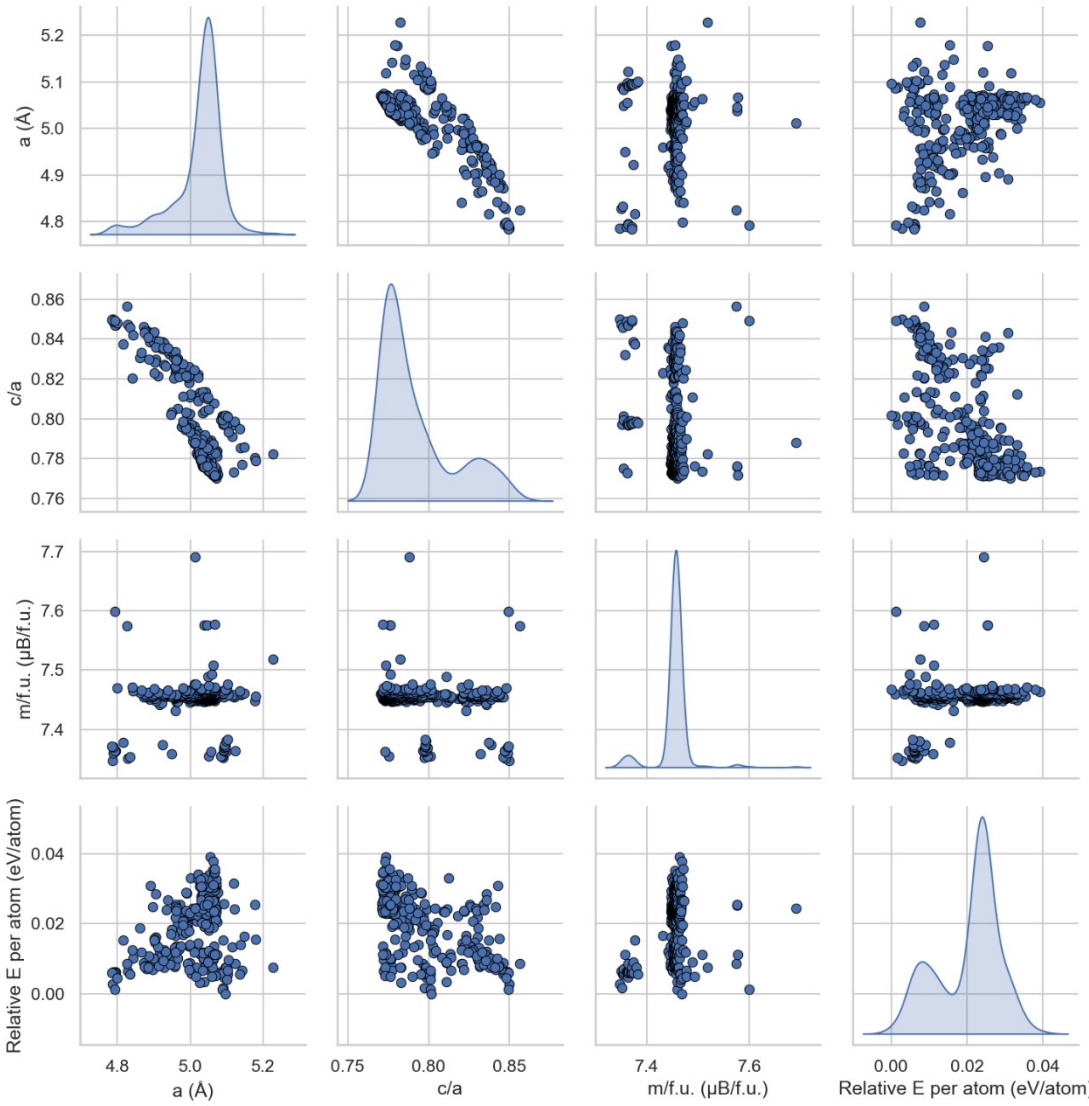

Figure S3: Scatter matrix plot with a kernel density estimation fit in the diagonal plot elements for the lattice constant, *a*, *c/a* ratio, total magnetization per formula unit and relative total energy of the 400 2×2×4 supercell systems comprising the 2×2×2 dataset.

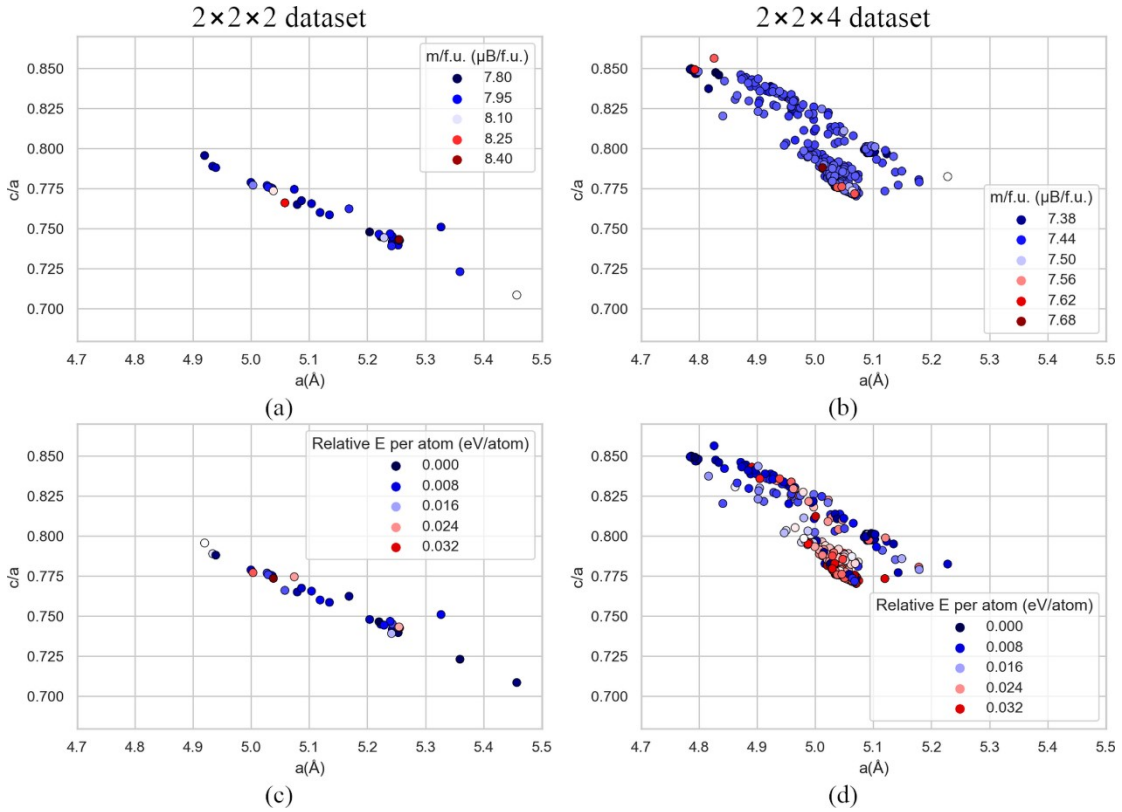

Figure S4: Scatter plot for *c/a* ratio against lattice constant, *a*, with color code for the respective values of total magnetization per formula unit (a, b) and relative energy per atom (c, d) of each configuration for the 35 48 atom configurations of the 2×2×2 dataset (a, c) and the 400 96 atom con-figurations of the 2×2×4 dataset (b, d).

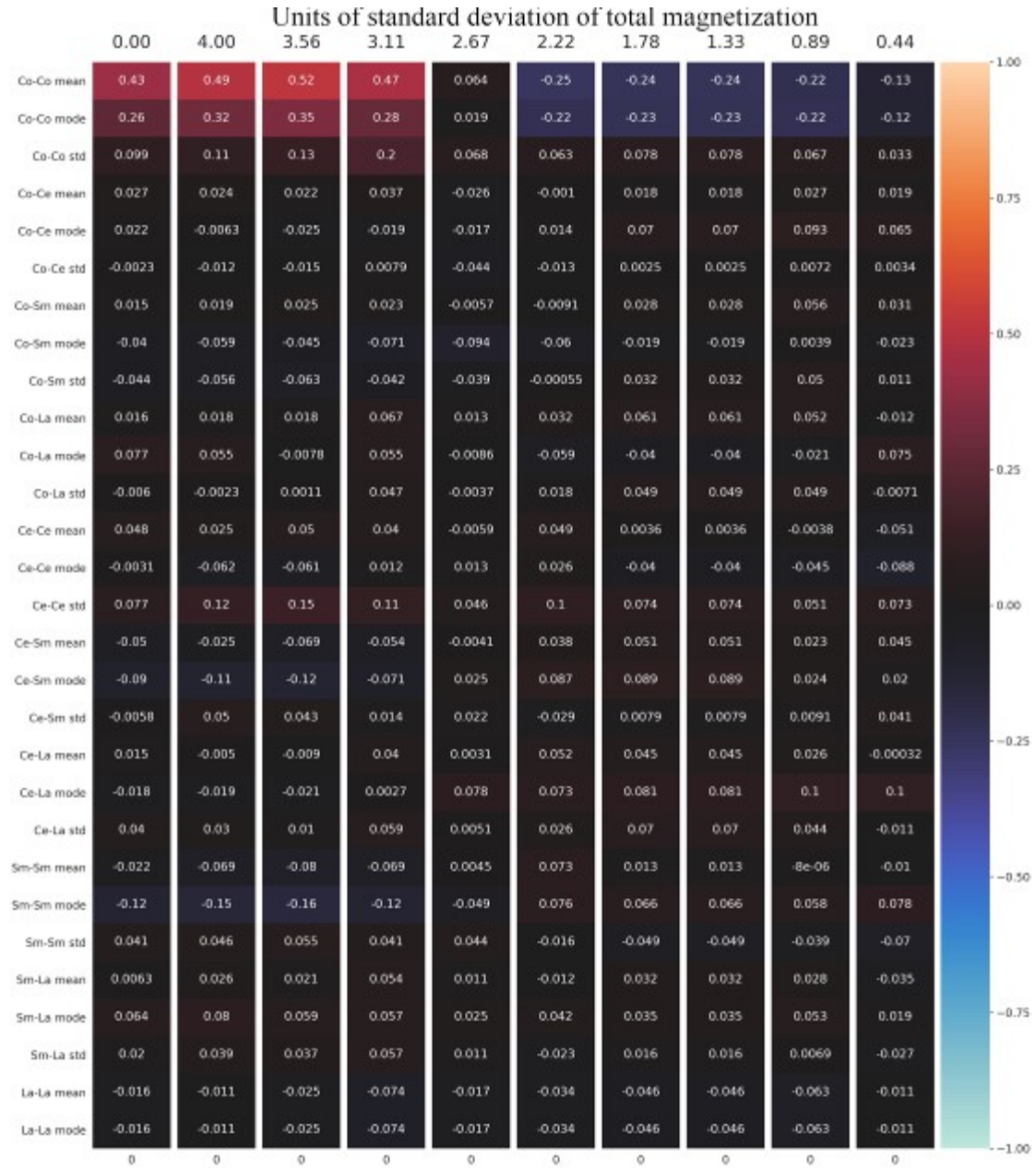

Figure S5: Correlation heatmap of the mean, the mode and the standard deviation of the interatomic distances between each atom type, filtered iteratively in each column by units of standard deviation of the total magnetization of all the cases of the 2×2×4 dataset.

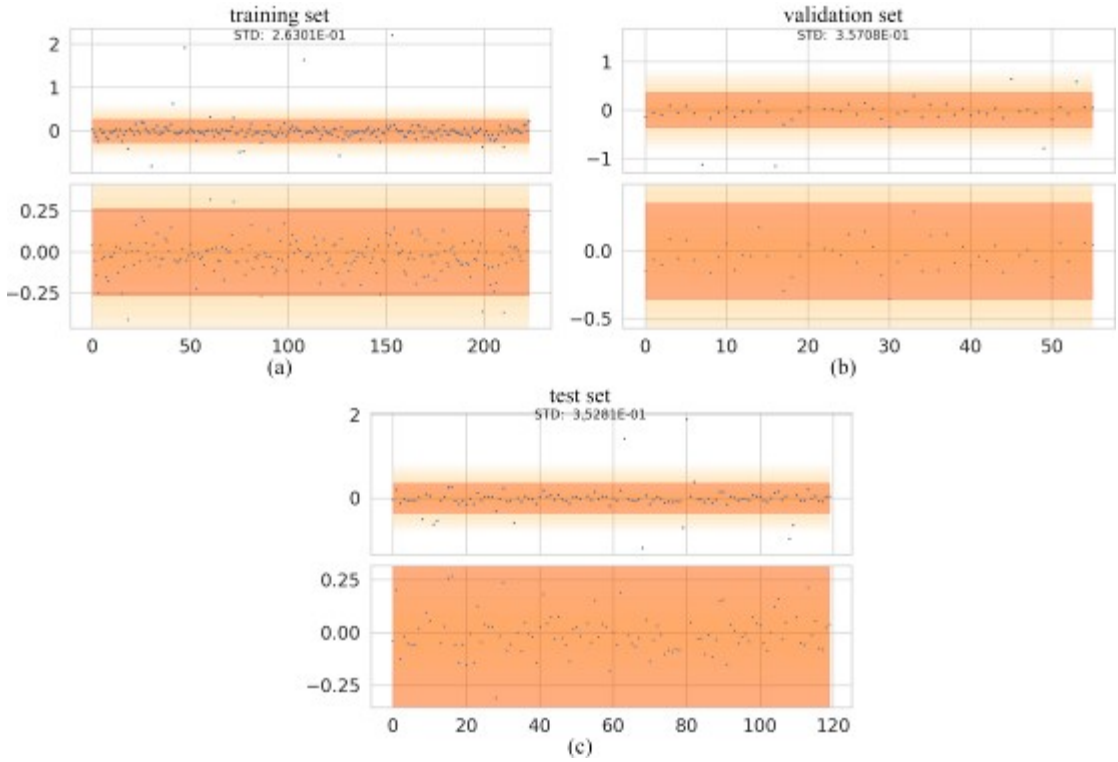

Figure S6: Residuals plot for the ANN regression model for the prediction of the total magnetization in the cases of the 2×2×4 dataset.

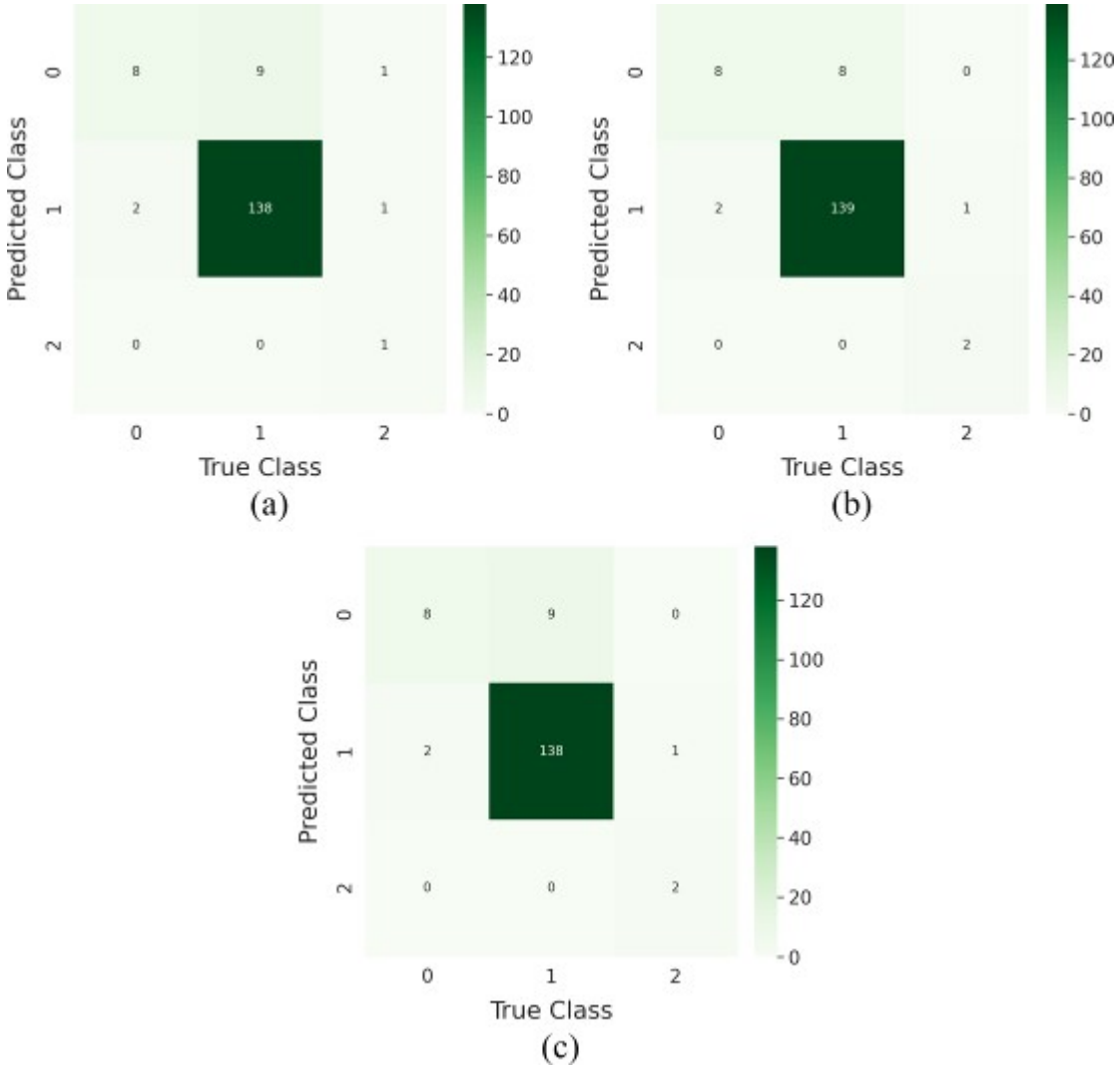

Figure S7: Confusion matrices for each one of the 3-fold cross validation steps (a-c) for the evaluation of the ANN classification model. Diagonal terms present the number of correct predictions and the rest of the terms are misclassifications. Classes 0, 1 and 2 represent the three magnetization clusters of the 2×2×4 dataset in ascending order of the magnetization value.