

Supporting Information

Inelastic Neutron Scattering Study of Magnetic Exchange Pathways in MnS

Judith K. Clark,^a Vincent Yannello,^b Arjana M. Samarakoon,^{c,#} Cyrus Ross,^a Madeleine C. Uible,^a V. Ovidiu Garlea,^{c,*} Michael Shatruk^{a,d,*}

^a Department of Chemistry and Biochemistry, Florida State University, Tallahassee, FL 32306, United States

^b Department of Chemistry, Biochemistry and Physics, University of Tampa, Tampa, FL 33606, United States

^c Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, United States

^d National High Field Magnetic Laboratory, Tallahassee, FL 32310, United States

* Corresponding authors: mhatruk@fsu.edu, garleao@ornl.gov

Present address: Materials Science Division, Argonne National Laboratory, Lemont, IL 60439, USA

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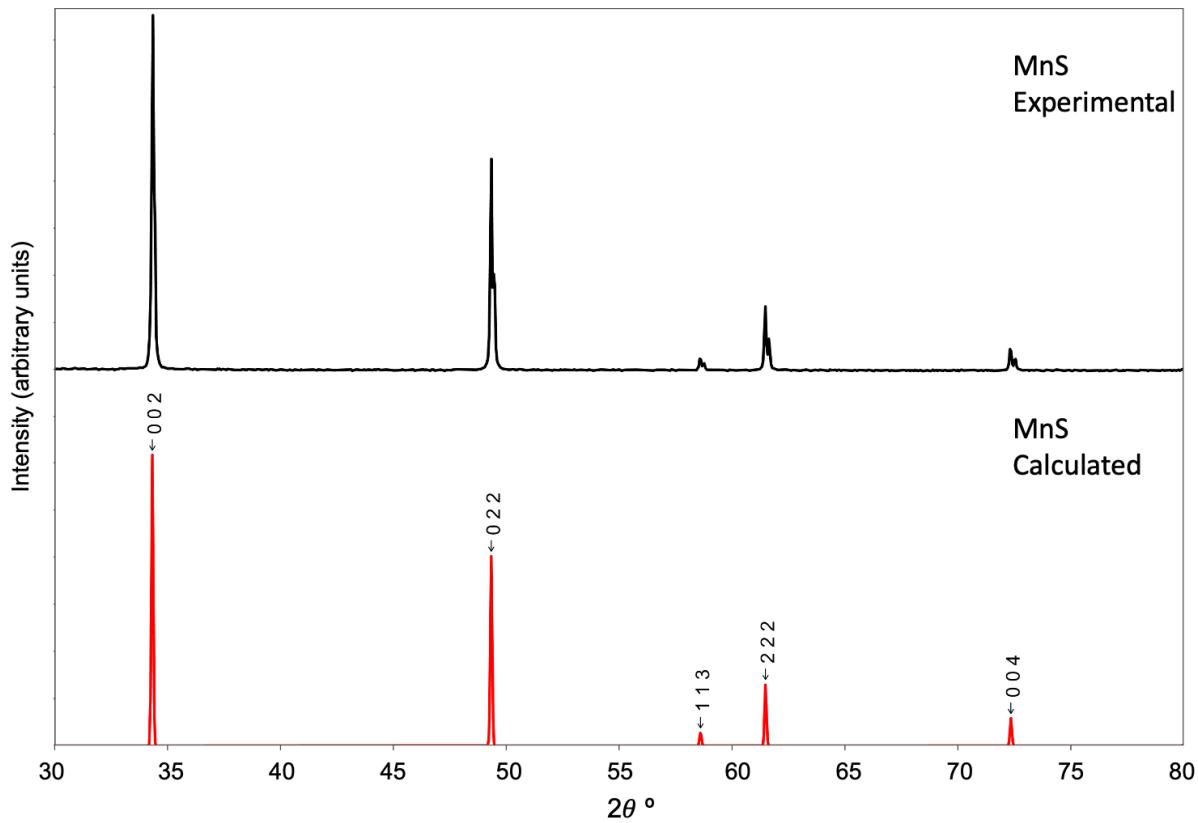


Figure S1. Experimental vs. calculated powder diffraction patterns of MnS confirming the sample purity. The refined value of the cubic lattice parameter, $a = 5.2274(5)$ Å, matches the literature value, $a = 5.221$ Å, reported in Corliss, L.; Elliott, N.; Hastings, J. *Phys. Rev.* **1956**, *104*, 924-928.

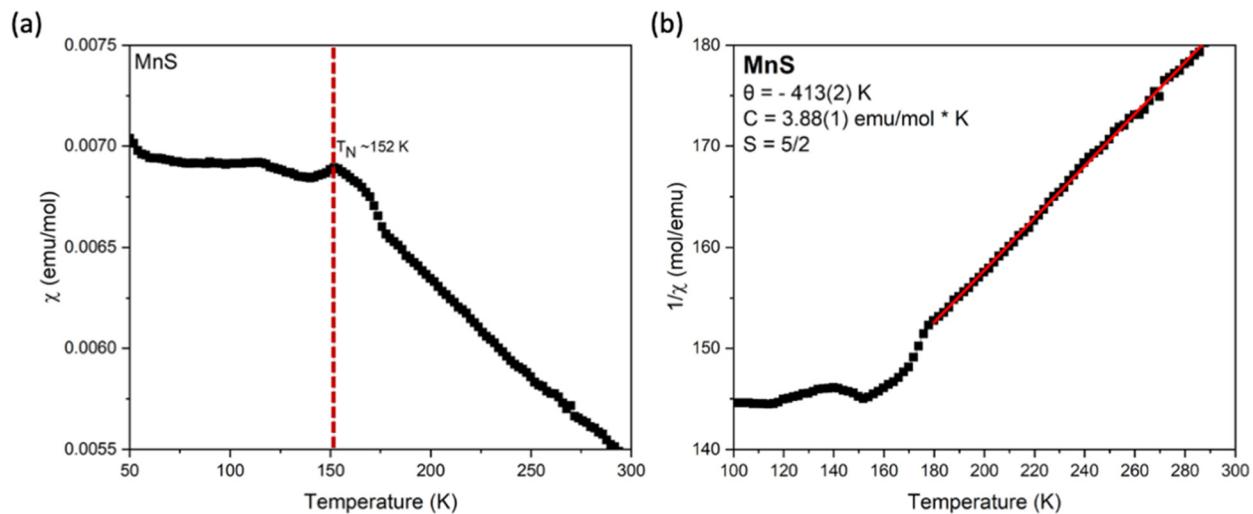


Figure S2. Temperature dependence of magnetic susceptibility (a) and inverse magnetic susceptibility (b) of MnS measured under an applied field of 100 Oe. The dashed vertical red line in panel (a) indicates the AFM ordering temperature while the solid red line in panel (b) shows the Curie-Weiss fitting.

Table S1. Calculated total energies of various spin configurations of MnS as a function of U .

Configuration of Mn spins	Energy per atom (eV) as a function of U (eV)								
	0	1	2	3	4	5	6	7	8
FM	-14.034	-13.698	-13.414	-13.160	-12.928	-12.716	-12.523	-12.193	-12.193
AFM1	-14.071	-13.738	-13.448	-13.184	-12.945	-12.728	-12.532	-12.355	-12.197
AFM2	-14.179	-13.828	-13.514	-13.233	-12.981	-12.753	-12.548	-12.364	-12.201
AFM3	-14.117	-13.781	-13.480	-13.208	-12.963	-12.741	-12.541	-12.361	-12.200
AFM4	-14.151	-13.806	-13.498	-13.222	-12.973	-12.748	-12.545	-12.363	-12.201
AFM5	-14.104	-13.766	-13.469	-13.201	-12.958	-12.738	-12.539	-12.200	-12.200

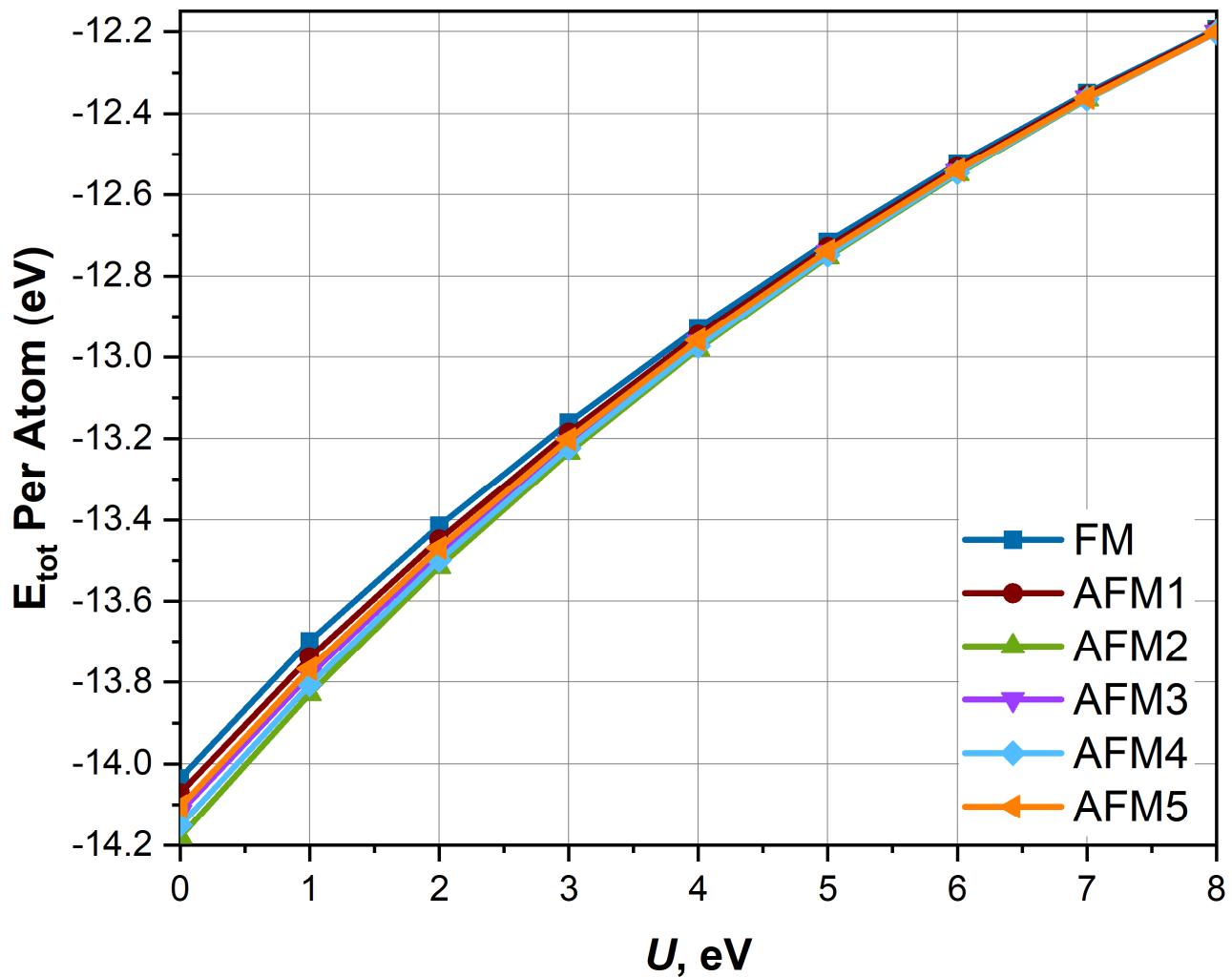


Figure S3. Calculated total energies of various spin configurations of MnS as a function of U .

Table S2. Values of J_1 , J_2 , and J_3 and their ratios calculated for MnS based on the total energies of five spin configurations AFM1-AFM5 or of one FM and four AFM spin configurations (Figure 6) as functions of the U parameter.

J_n values and their ratios	U (eV)								
	0	1	2	3	4	5	6	7	8
Spin configurations AFM1-AFM5 (12345)									
J_1 (meV)	-0.338	-0.344	-0.279	-0.230	-0.189	-0.157	-0.131	-0.109	-0.092
J_2 (meV)	2.104	1.628	1.182	0.838	0.579	0.384	0.236	0.124	0.039
J_3 (meV)	-0.340	-0.359	-0.271	-0.204	-0.155	-0.117	-0.089	-0.067	-0.050
$ J_2/J_1 $	6.227	4.731	4.244	3.650	3.059	2.445	1.805	1.136	0.426
J_1/J_3	0.995	0.958	1.028	1.124	1.224	1.338	1.472	1.636	1.846
Spin configurations FM, AFM2-AFM5 (F2345)									
J_1 (meV)	0.660	0.688	0.528	0.381	0.275	0.197	0.140	0.096	0.062
J_2 (meV)	2.104	1.628	1.182	0.838	0.579	0.384	0.236	0.124	0.039
J_3 (meV)	0.159	0.157	0.132	0.101	0.078	0.060	0.046	0.036	0.027
J_2/J_1	3.188	2.367	2.238	2.200	2.106	1.944	1.690	1.293	0.625
J_1/J_3	4.144	4.387	3.991	3.774	3.547	3.297	3.011	2.679	2.280
Spin configurations FM, AFM1, AFM3-5 (F1345)									
J_1 (meV)	0.410	0.431	0.328	0.229	0.159	0.109	0.072	0.045	0.024
J_2 (meV)	2.110	1.635	1.187	0.841	0.581	0.385	0.236	0.124	0.039
J_3 (meV)	0.034	0.028	0.031	0.025	0.019	0.016	0.013	0.010	0.008
J_2/J_1	5.142	3.794	3.620	3.670	3.643	3.525	3.274	2.776	1.635
J_1/J_3	11.935	15.657	10.426	9.314	8.197	7.019	5.759	4.416	2.962
Spin configurations FM, AFM1-3, AFM5 (F1235)									
J_1 (meV)	0.410	0.432	0.329	0.230	0.160	0.110	0.072	0.045	0.024
J_2 (meV)	1.621	1.125	0.787	0.538	0.351	0.209	0.102	0.022	-0.037
J_3 (meV)	0.034	0.027	0.031	0.025	0.019	0.016	0.013	0.010	0.008
J_2/J_1	3.953	2.603	2.392	2.340	2.192	1.906	1.411	0.500	-1.544
J_1/J_3	11.974	15.844	10.487	9.358	8.242	7.056	5.793	4.443	2.982

Table S3. Calculated total energies of various spin configurations (Figure 6) of MnO as a function of the U parameter.

Configuration of Mn spins	Energy per atom (eV) as a function of U (eV)								
	0	1	2	3	4	5	6	7	8
FM	-16.395	-16.101	-15.859	-15.640	-15.441	-15.260	-15.097	-14.953	-14.828
AFM1	-16.474	-16.189	-15.934	-15.704	-15.496	-15.208	-15.14	-14.991	-14.862
AFM2	-16.546	-16.243	-15.973	-15.731	-15.514	-15.319	-15.146	-14.993	-14.860
AFM3	-16.515	-16.222	-15.959	-15.723	-15.509	-15.318	-15.148	-14.997	-14.865
AFM4	-16.523	-16.226	-15.960	-15.722	-15.507	-15.214	-15.142	-14.990	-14.858
AFM5	-16.468	-16.177	-15.922	-15.691	-15.483	-15.295	-15.127	-14.978	-14.848

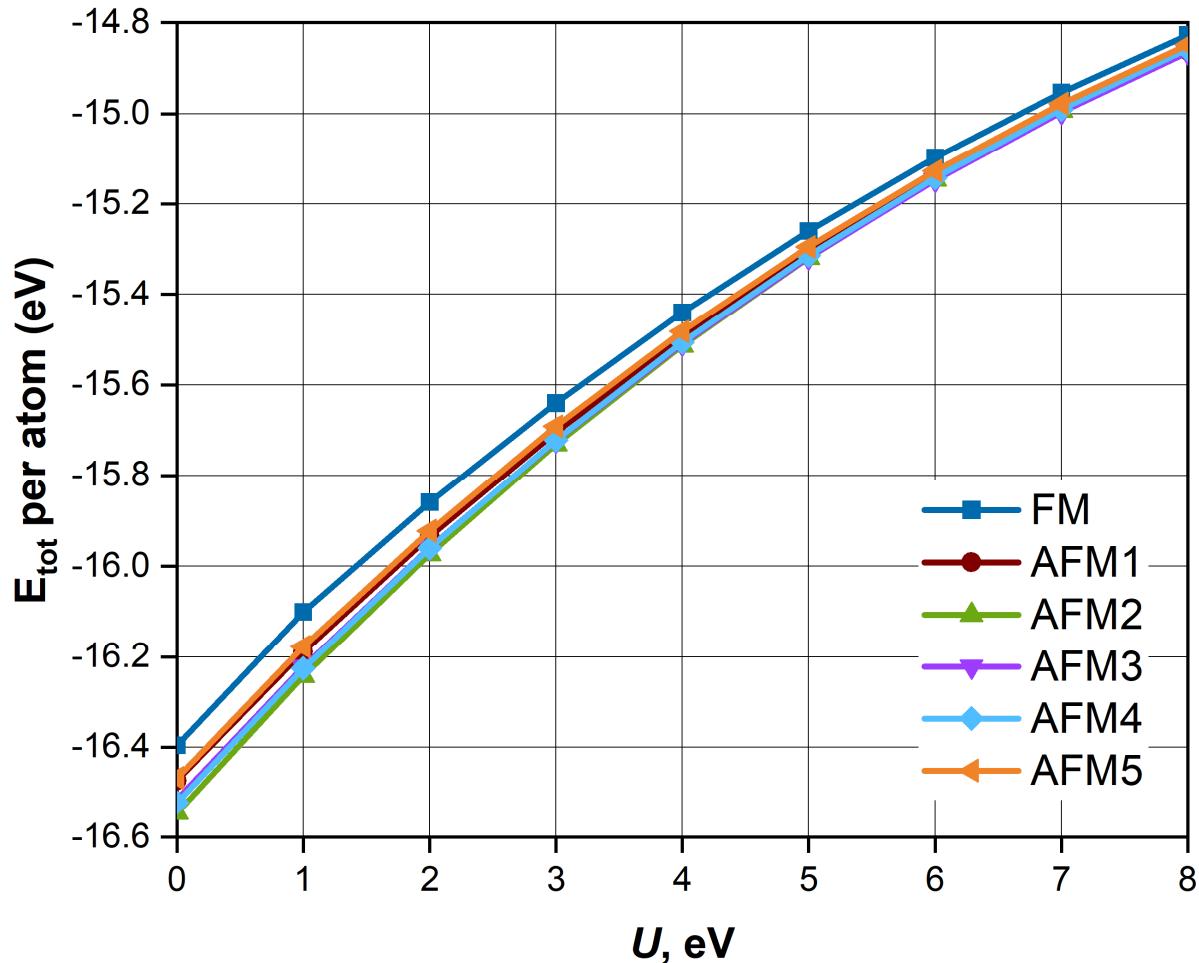


Figure S4. Calculated total energies of various spin configurations of MnO as a function of U .