Supplemental Material for Bond Ordering and Molecular Spin-Orbital Fluctuations in the Cluster Mott Insulator $GaTa_4Se_8$

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Supplemental material containing tables that list: (Tabel I) parameters from single domain crystallographic refinement, (Table II) crystallography refined structure for $P\bar{4}2_1m$ space group, (Table III) PDF refined structure for $P\bar{4}2_1m$ space group, (Table IV) Normalized atomic displacements at 10 K and (Table V-X) corresponding distortion modes.

TABLE I. Crystallographic refinement for single domain.

Temperature (K)	10
Crystal system	Tetragonal
Space group	$P\bar{4}2_1m$
a (Å)	10.34370(14)
b (Å)	10.34370(14)
c (Å)	20.6878(5)
Data collection diffractometer	15-ID-D, APS
Absorption correction	Multiscan
Reflections collected	13104
Independent reflections	1952 (R_{int}=0.0281)
F(000)	4760.0
Radiation	Synchrotron, λ=0.41328 Å
2θ range for data collection (deg)	3.932 to 90.234
Index ranges	$0 \leq h \leq 10$, $0 \leq k \leq 14$, $0 \leq l \leq 29$
Refinement method	Full-matrix least squares on F^2
Data, restraints, parameters	1952/0/63
Goodness of fit	1.198
R_1, $wR_2(I \geq 2\sigma)$	0.0879, 0.1018
R_1, wR_2(all)	0.0990, 0.1046
Largest diff. peak/hole	3.98/-5.61

TABLE II. Crystallography refined $GaTa_4Se_8$ structure for $P\bar{4}2_1m$ space group.

Atom	WP	x	y	z	|U|
Ta1	4e	0.35331(12)	0.14669(12)	0.17427(7)	0.0011
Ta2	4e	0.14610(12)	0.35390(12)	0.17822(7)	0.0022
Ta3	4e	0.85306(12)	0.35306(12)	0.32562(7)	0.0011
Ta4	4e	0.64616(12)	0.14616(12)	0.32212(7)	0.0023
Ta5	8f	0.85394(12)	0.15077(12)	0.42634(5)	0.0022
Ta6	8f	0.35432(14)	0.34864(13)	0.07346(6)	0.0079
Se1	4e	0.1164(4)	0.3836(4)	0.0568(2)	0.0008
Se2	4e	0.8871(4)	0.3871(4)	0.4463(2)	0.0042
Se3	4e	0.3895(4)	0.1105(4)	0.0543(4)	0.0173
Se4	4e	0.3857(4)	0.1143(4)	0.30524(18)	0.0008
Se5	4e	0.8847(4)	0.3847(4)	0.19461(18)	0.0005
Se6	4e	0.6149(4)	0.1149(4)	0.1922(2)	0.0024
Se7	4e	0.6170(4)	0.1170(4)	0.4419(2)	0.0083
Se8	4e	0.1139(4)	0.3861(4)	0.30736(18)	0.0005
Se9	8f	0.8852(4)	0.1119(4)	0.30795(13)	0.0002
Se10	8f	0.3863(4)	0.3880(4)	0.19221(13)	0.0008
Se11	8f	0.1179(4)	0.1126(4)	0.44136(14)	0.0011
Se12	8f	0.6164(4)	0.3854(4)	0.05854(17)	0.0090
Ga1	4e	0.2510(4)	0.2490(4)	0.3753(2)	0.0029
Ga2	4e	0.7512(4)	0.2512(4)	0.1250(2)	0.0043

TABLE III. PDF refined $GaTa_4Se_8$ local structure, atomic positions were constraint by $P\bar{4}2_1m$ space group.

Atom	WP	x	y	z	|U|
Ta1	4e	0.351101	0.148899	0.173364	0.002041
Ta2	4e	0.142536	0.357464	0.180630	0.002041
Ta3	4e	0.845456	0.345456	0.325379	0.002041
Ta4	4e	0.644848	0.144848	0.321580	0.002041
Ta5	8f	0.856884	0.153446	0.426199	0.002041
Ta6	8f	0.353801	0.352045	0.073713	0.002041
Se1	4e	0.114412	0.385588	0.059105	0.003523
Se2	4e	0.388703	0.111297	0.055231	0.003523
Se3	4e	0.382959	0.117041	0.305490	0.003523
Se4	4e	0.884463	0.384463	0.196939	0.003523
Se5	4e	0.611382	0.111382	0.191494	0.003523
Se6	4e	0.615178	0.115178	0.439884	0.003523
Se7	4e	0.113594	0.386406	0.309588	0.003523
Se8	4e	0.886271	0.386271	0.446034	0.003523
Se9	8f	0.884539	0.116007	0.305650	0.003523
Se10	8f	0.385064	0.388671	0.192559	0.003523
Se11	8f	0.116575	0.113089	0.442572	0.003523
Se12	8f	0.615978	0.384913	0.057510	0.003523
Ga1	4e	0.253877	0.246123	0.376226	0.001286
Ga2	4e	0.744378	0.244378	0.127202	0.001286

TABLE IV. Normalized atomic displacements at 10 K. The distortion is defined by the deviation from the cubic structure. The corresponding distortion modes are listed in Table V-X.

Atom	WP	u_x/a	u_y/b	u_z/c	$\|u\|$ (Å)
Se1	4e	-0.0033	0.0033	-0.0026	0.0718
Se2	4e	-0.0026	0.0026	-0.0001	0.0380
Se3	8f	0.0019	-0.0010	0.0011	0.0310
Se4	4e	-0.0009	-0.0009	0.0032	0.0671
Se5	4e	-0.0032	-0.0032	-0.0012	0.0529
Se6	8f	0.0018	0.0001	-0.0009	0.0261
Ta1	8f	-0.0039	0.0018	-0.0003	0.0444
Ta2	4e	-0.0005	0.0005	-0.0019	0.0395
Ta3	4e	0.0013	-0.0013	0.0016	0.0385
Ta4	8f	-0.0014	-0.0033	0.0001	0.0370
Ta5	4e	-0.0008	0.0008	-0.0020	0.0426
Ta6	4e	0.0014	-0.0014	0.0020	0.0451
Ga1	4e	-0.0012	0.0012	0.0000	0.0175
Ga2	4e	-0.0010	0.0010	0.0003	0.0158
Se7	8f	-0.0014	0.0003	-0.0011	0.0268
Se8	4e	0.0003	-0.0003	-0.0021	0.0433
Se9	4e	0.0001	-0.0001	0.0003	0.0061
Se10	8f	-0.0029	0.0024	-0.0012	0.0457
Se11	4e	0.0010	-0.0010	-0.0001	0.0155
Se12	4e	-0.0006	0.0006	-0.0022	0.0470

TABLE V. Normalized atomic displacements of Γ_1 mode.

Atom	WP	δ_x/a	δ_y/b	δ_z/c
Se1	4e	-0.0091	0.0091	-0.0046
Se2	4e	0.0091	-0.0091	-0.0046
Se3	8f	0.0091	0.0091	0.0046
Se4	4e	-0.0091	-0.0091	0.0046
Se5	4e	0.0091	0.0091	0.0046
Se6	8f	0.0091	0.0091	0.0046
Ta1	8f	0.0025	0.0025	0.0012
Ta2	4e	-0.0025	0.0025	0.0012
Ta3	4e	0.0025	-0.0025	0.0012
Ta4	8f	-0.0025	0.0025	0.0012
Ta5	4e	-0.0025	0.0025	0.0012
Ta6	4e	0.0025	-0.0025	0.0012
Ga1	4e	0.0000	0.0000	0.0000
Ga2	4e	0.0000	0.0000	0.0000
Se7	8f	-0.0030	-0.0030	-0.0015
Se8	4e	0.0030	-0.0030	0.0015
Se9	4e	-0.0030	0.0030	0.0015
Se10	8f	-0.0030	-0.0030	-0.0015
Se11	4e	-0.0030	0.0030	0.0015
Se12	4e	0.0030	-0.0030	0.0015

TABLE VI. Normalized atomic displacements of Γ_3 mode.

Atom	WP	δ_x/a	δ_y/b	δ_z/c
Se1	4e	0.0029	-0.0029	-0.0029
Se2	4e	-0.0029	0.0029	-0.0029
Se3	8f	-0.0029	-0.0029	0.0029
Se4	4e	0.0029	0.0029	0.0029
Se5	4e	-0.0029	-0.0029	0.0029
Se6	8f	-0.0029	-0.0029	0.0029
Ta1	8f	-0.0059	-0.0059	-0.0059
Ta2	4e	0.0059	-0.0059	0.0059
Ta3	4e	-0.0059	0.0059	0.0059
Ta4	8f	0.0059	-0.0059	0.0059
Ta5	4e	0.0059	-0.0059	0.0059
Ta6	4e	-0.0059	0.0059	0.0059
Ga1	4e	0.0000	0.0000	0.0000
Ga2	4e	0.0000	0.0000	0.0000
Se7	8f	0.0026	0.0026	-0.0026
Se8	4e	-0.0026	0.0026	0.0026
Se9	4e	0.0026	-0.0026	0.0026
Se10	8f	0.0026	0.0026	-0.0026
Se11	4e	0.0026	-0.0026	0.0026
Se12	4e	-0.0026	0.0026	0.0026

TABLE VII. Normalized atomic displacements of Δ_3 mode.

Atom	WP	δ_x/a	δ_y/b	δ_z/c
Se1	4e	-0.0125	0.0125	-0.0028
Se2	4e	0.0125	-0.0125	-0.0028
Se3	8f	-0.0042	-0.0042	-0.0013
Se4	4e	0.0125	0.0125	-0.0028
Se5	4e	-0.0125	-0.0125	-0.0028
Se6	8f	0.0042	0.0042	0.0013
Ta1	8f	-0.0009	-0.0009	-0.0017
Ta2	4e	0.0013	-0.0013	-0.0010
Ta3	4e	-0.0013	0.0013	-0.0010
Ta4	8f	-0.0009	0.0009	-0.0017
Ta5	4e	-0.0013	0.0013	0.0010
Ta6	4e	0.0013	-0.0013	0.0010
Ga1	4e	0.0000	0.0000	-0.0025
Ga2	4e	0.0000	0.0000	0.0025
Se7	8f	-0.0021	-0.0021	0.0008
Se8	4e	0.0083	-0.0083	0.0023
Se9	4e	-0.0083	0.0083	0.0023
Se10	8f	0.0021	0.0021	-0.0008
Se11	4e	0.0083	-0.0083	-0.0023
Se12	4e	-0.0083	0.0083	-0.0023

TABLE VIII. Normalized atomic displacements of X_3 mode.

Atom	WP	δ_x/a	δ_y/b	δ_z/c
Se1	4e	0.0027	-0.0027	-0.0053
Se2	4e	-0.0027	0.0027	-0.0053
Se3	8f	0.0027	0.0027	-0.0053
Se4	4e	0.0027	0.0027	0.0053
Se5	4e	-0.0027	-0.0027	0.0053
Se6	8f	0.0027	0.0027	-0.0053
Ta1	8f	-0.0049	-0.0049	-0.0006
Ta2	4e	-0.0049	0.0049	-0.0006
Ta3	4e	0.0049	-0.0049	-0.0006
Ta4	8f	0.0049	-0.0049	0.0006
Ta5	4e	-0.0049	0.0049	-0.0006
Ta6	4e	0.0049	-0.0049	-0.0006
Ga1	4e	0.0000	0.0000	0.0007
Ga2	4e	0.0000	0.0000	0.0007
Se7	8f	-0.0019	-0.0019	-0.0053
Se8	4e	-0.0019	0.0019	-0.0053
Se9	4e	0.0019	-0.0019	-0.0053
Se10	8f	-0.0019	-0.0019	-0.0053
Se11	4e	0.0019	-0.0019	-0.0053
Se12	4e	-0.0019	0.0019	-0.0053

TABLE IX. Normalized atomic displacements of X_5 mode.

Atom	WP	δ_x/a	δ_y/b	δ_z/c
Se1	4e	-0.0074	0.0074	-0.0051
Se2	4e	-0.0074	0.0074	0.0051
Se3	8f	0.0034	-0.0034	0.0000
Se4	4e	-0.0074	-0.0074	0.0051
Se5	4e	-0.0074	-0.0074	-0.0051
Se6	8f	0.0034	-0.0034	0.0000
Ta1	8f	-0.0077	0.0077	0.0000
Ta2	4e	0.001	-0.001	-0.0055
Ta3	4e	0.001	-0.001	0.0055
Ta4	8f	-0.0077	-0.0077	0.0000
Ta5	4e	0.001	-0.001	-0.0055
Ta6	4e	0.001	-0.001	0.0055
Ga1	4e	-0.0033	0.0033	0.0000
Ga2	4e	-0.0033	0.0033	0.0000
Se7	8f	-0.0052	0.0052	0.0000
Se8	4e	0.0006	-0.0006	-0.0033
Se9	4e	0.0006	-0.0006	0.0033
Se10	8f	-0.0052	0.0052	0.0000
Se11	4e	0.0006	-0.0006	0.0033
Se12	4e	0.0006	-0.0006	-0.0033

TABLE X. Normalized atomic displacements of W_4 mode.

Atom	WP	δ_x/a	δ_y/b	δ_z/c
Se1	4e	-0.0061	0.0061	0.0064
Se2	4e	-0.0061	0.0061	-0.0064
Se3	8f	0.0041	-0.0041	0.0000
Se4	4e	0.0061	0.0061	0.0064
Se5	4e	0.0061	0.0061	-0.0064
Se6	8f	-0.0041	0.0041	0.0000
Ta1	8f	-0.0033	0.0033	0.0000
Ta2	4e	0.0006	-0.0006	0.0015
Ta3	4e	0.0006	-0.0006	-0.0015
Ta4	8f	0.0033	0.0033	0.0000
Ta5	4e	-0.0006	0.0006	-0.0015
Ta6	4e	-0.0006	0.0006	0.0015
Ga1	4e	-0.0014	0.0014	0.0000
Ga2	4e	0.0014	-0.0014	0.0000
Se7	8f	0.0119	-0.0119	0.0000
Se8	4e	0.0000	0.0000	-0.0009
Se9	4e	0.0000	0.0000	0.0009
Se10	8f	-0.0119	0.0119	0.0000
Se11	4e	0.0000	0.0000	-0.0009
Se12	4e	0.0000	0.0000	0.0009