Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Quantum Spin Liquid: Design of a Quantum Spin Liquid next to a Superconducting State based on a Dimer-type ET Mott Insulator

Takaaki Hiramatsu,*[1] Yukihiro Yoshida,[1] Gunzi Saito,*[1] Akihiro Otsuka,[2] Hideki Yamochi,[2] Mitsuhiko Maesato,[3] Yasuhiro Shimizu,[4] Hiroshi Ito,[5] and Hideo Kishida[5]

[1]Department of Agriculture, Meijo University, 1-501 Shiogamaguchi, Tempaku-ku, Nagoya 468-8502, Japan
[2]Research Center for Low Temperature and Materials Sciences, Yoshida-Honmachi, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan
[3]Division of Chemistry, Graduate School of Science, Kyoto University, Oiwake-cho, Kitashirakawa, Sakyo-ku, Kyoto 606-8502, Japan
[4]Department of Physics, Graduate School of Science, Furo-cho, Chikusa-ku, Nagoya University, Nagoya 464-8602, Japan
[5]Department of Applied Physics, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

Corresponding authors Takaaki Hiramatsu; *E-mail:* htakaaki@meijo-u.ac.jp
Gunzi Saito; *E-mail:* gsaito@meijo-u.ac.jp

Table S1. Crystallographic data of κ-$(ET)_2I_3$ (**a**)

Chemical formula	$C_{20}H_{16}I_3S_{16}$				
Formula weight	1149.99				
Crystal system	Monoclinic				
Space group	$P2_1/c$				
Temperature (K)	298	250	200	150	100
Crystal dimension (mm^3)	0.50 × 0.28 × 0.13				
a (Å)	16.423(1)	16.379(1)	16.336(1)	16.294(1)	16.260(1)
b (Å)	8.4972(6)	8.4773(6)	8.4593(5)	8.4439(5)	8.4332(5)
c (Å)	12.8688(9)	12.8360(9)	12.8130(8)	12.7917(8)	12.7668(8)
β (°)	108.493(1)	108.624(1)	108.773(1)	108.930(1)	109.091(1)
V ($Å^3$)	1703.1(2)	1689.0(2)	1676.5(2)	1664.8(2)	1654.3(2)
Z	2	2	2	2	2
D_{calc} (g cm^{-3})	2.243	2.261	2.278	2.294	2.309
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	3.751	3.782	3.811	3.837	3.862
No. of reflections measured	8974	8887	8833	8784	8709
No. of independent reflections	3681	3646	3623	3592	3579
R_{int}	0.0157	0.0216	0.0202	0.0126	0.0129
No. of parameters	179	179	179	179	178
Final R values ($I > 2\sigma(I)$)	0.0317	0.0272	0.0221	0.0155	0.0152
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.0722	0.0622	0.0533	0.0388	0.0380
Final R values (all data)	0.0368	0.0305	0.0241	0.0165	0.0158
Final $wR(F^2)$ values (all data)	0.0777	0.0659	0.0552	0.0396	0.0383
Goodness of fit on F^2	1.054	1.061	1.083	1.080	1.147
Largest diff. peak (e $Å^{-3}$)	2.030	1.811	1.377	0.960	0.384
Largest diff. hole (e $Å^{-3}$)	–1.423	–1.144	–0.920	–0.655	–0.760
CCDC number	1016194	1016193	1016192	1016191	1016190

Table S2. Crystallographic data of κ-$(ET)_2Ag(CN)_2 \cdot H_2O$ (**b**)

Chemical formula	$C_{22}H_{18}AgN_2OS_{16}$				
Formula weight	947.21				
Crystal system	Monoclinic				
Space group	$P2_1$				
Temperature (K)	300	250	200	150	100
Crystal dimension (mm^3)	0.34 × 0.33 × 0.11				
a (Å)	16.058(1)	16.044(1)	16.028(1)	16.016(1)	15.998(1)
b (Å)	8.6445(6)	8.6241(6)	8.5985(6)	8.5758(6)	8.5499(6)
c (Å)	12.5766(9)	12.5341(8)	12.5042(8)	12.4828(9)	12.4594(9)
β (°)	109.254(1)	109.452(1)	109.620(1)	109.747(1)	109.846(1)
V ($Å^3$)	1648.1(2)	1635.3(2)	1623.3(2)	1613.7(2)	1603.0(2)
Z	2	2	2	2	2
D_{calc} (g cm^{-3})	1.909	1.924	1.938	1.949	1.962
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	1.651	1.664	1.677	1.687	1.698
No. of reflections measured	18253	18029	17764	17730	17529
No. of independent reflections	7128	7067	7009	6969	6923
R_{int}	0.0138	0.0098	0.0104	0.0099	0.0098
No. of parameters	379	379	379	379	379
Final R values ($I > 2\sigma(I)$)	0.0203	0.0175	0.0173	0.0180	0.0197
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.0520	0.0453	0.0445	0.0457	0.0478
Final R values (all data)	0.0215	0.0183	0.0177	0.0182	0.0198
Final $wR(F^2)$ values (all data)	0.0528	0.0457	0.0447	0.0459	0.0479
Goodness of fit on F^2	1.034	1.074	1.105	1.118	1.130
Largest diff. peak (e $Å^{-3}$)	0.409	0.501	0.585	0.766	1.175
Largest diff. hole (e $Å^{-3}$)	–0.539	–0.644	–0.832	–1.060	–1.291
CCDC number	1016294	1016293	1016292	1016291	1016290

Table S3. Crystallographic data of κ-$(ET)_2Cu(CN)[N(CN)_2]$ (**c**)

Chemical formula	$C_{23}H_{16}CuN_4S_{16}$				
Formula weight	924.90				
Crystal system	Monoclinic				
Space group	$P2_1$				
Temperature (K)	298	250	200	150	100
Crystal dimension (mm^3)	0.26 × 0.05 × 0.05				
a (Å)	15.975(3)	15.964(2)	15.947(2)	15.938(2)	15.930(2)
b (Å)	8.620(2)	8.608(1)	8.593(1)	8.582(1)	8.5690(9)
c (Å)	12.885(2)	12.841(2)	12.798(2)	12.762(2)	12.726(1)
β (°)	110.882(2)	111.006(2)	111.133(2)	111.243(1)	111.355(1)
V ($Å^3$)	1657.8(5)	1647.2(4)	1635.8(4)	1626.9(3)	1617.9(3)
Z	2	2	2	2	2
D_{calc} (g cm^{-3})	1.853	1.865	1.878	1.888	1.899
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	1.694	1.705	1.717	1.727	1.736
No. of reflections measured	8547	8823	8797	8787	8788
No. of independent reflections	4332	4536	4510	4524	4521
R_{int}	0.0478	0.0430	0.0397	0.0360	0.0324
No. of parameters	397	397	397	397	397
Final R values ($I > 2\sigma(I)$)	0.0429	0.0415	0.0386	0.0369	0.0356
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.0746	0.0749	0.0649	0.0659	0.0641
Final R values (all data)	0.1008	0.0967	0.0837	0.0723	0.0618
Final $wR(F^2)$ values (all data)	0.0927	0.0962	0.0808	0.0804	0.0740
Goodness of fit on F^2	0.973	1.025	1.015	1.013	1.061
Largest diff. peak (e $Å^{-3}$)	0.420	0.581	0.650	0.570	0.696
Largest diff. hole (e $Å^{-3}$)	–0.492	–0.574	–0.687	–0.622	–0.798
CCDC number	1016174	1016173	1016172	1016171	1016170

Table S4. Crystallographic data of κ-$(ET)_2Cu[N(CN)_2]Br$ (**d**)

Chemical formula	$C_{22}H_{16}BrCuN_3S_{16}$				
Formula weight	978.90				
Crystal system	Orthorhombic				
Space group	*Pnma*				
Temperature (K)	298	250	200	150	100
Crystal dimension (mm^3)	0.36 × 0.25 × 0.03				
a (Å)	12.965(1)	12.934(1)	12.912(1)	12.899(1)	12.897(1)
b (Å)	30.028(3)	29.937(3)	29.845(3)	29.754(3)	29.651(3)
c (Å)	8.5444(9)	8.5325(8)	8.5178(8)	8.5025(8)	8.4826(8)
V ($Å^3$)	3326.5(6)	3303.9(6)	3282.3(5)	3263.3(5)	3243.7(5)
Z	4	4	4	4	4
D_{calc} (g cm^{-3})	1.954	1.968	1.981	1.992	2.004
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	2.888	2.907	2.926	2.943	2.961
No. of reflections measured	17153	17013	16907	16763	16668
No. of independent reflections	3833	3816	3787	3761	3744
R_{int}	0.0269	0.0258	0.0247	0.0233	0.0224
No. of parameters	224	224	214	214	214
Final *R* values ($I > 2\sigma(I)$)	0.0463	0.0446	0.0430	0.0427	0.0418
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.1108	0.1061	0.1045	0.1021	0.0975
Final *R* values (all data)	0.0548	0.0510	0.0477	0.0458	0.0437
Final $wR(F^2)$ values (all data)	0.1148	0.1089	0.1066	0.1034	0.0984
Goodness of fit on F^2	1.077	1.117	1.140	1.174	1.179
Largest diff. peak (e $Å^{-3}$)	0.916	1.051	1.227	1.517	1.629
Largest diff. hole (e $Å^{-3}$)	–0.719	–0.665	–0.655	–0.630	–0.573
CCDC number	1016184	1016183	1016182	1016181	1016180

Table S5. Crystallographic data of κ-$(ET)_2Cu[N(CN)_2]Cl$ (**e**)

Chemical formula	$C_{22}H_{16}ClCuN_3S_{16}$				
Formula weight	934.33				
Crystal system	Orthorhombic				
Space group	*Pnma*				
Temperature (K)	298	250	200	150	100
Crystal dimension (mm^3)	0.32 × 0.19 × 0.16				
a (Å)	12.9550(8)	12.9262(8)	12.9062(7)	12.8917(7)	12.8852(7)
b (Å)	29.937(2)	29.848(2)	29.760(2)	29.669(2)	29.576(2)
c (Å)	8.4811(5)	8.4661(5)	8.4509(5)	8.4335(5)	8.4161(5)
V ($Å^3$)	3289.2(3)	3266.4(3)	3245.8(3)	3225.6(3)	3207.3(3)
Z	4	4	4	4	4
D_{calc} (g cm^{-3})	1.887	1.900	1.912	1.924	1.935
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	1.787	1.799	1.811	1.822	1.832
No. of reflections measured	17329	17201	17091	16932	16834
No. of independent reflections	3778	3752	3734	3709	3690
R_{int}	0.0136	0.0122	0.0120	0.0108	0.0103
No. of parameters	214	224	224	205	205
Final *R* values ($I > 2\sigma(I)$)	0.0458	0.0436	0.0420	0.0422	0.0394
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.1380	0.1297	0.1258	0.1280	0.1178
Final *R* values (all data)	0.0523	0.0481	0.0452	0.0444	0.0411
Final $wR(F^2)$ values (all data)	0.1461	0.1359	0.1303	0.1307	0.1196
Goodness of fit on F^2	1.067	1.042	1.049	1.071	1.059
Largest diff. peak (e $Å^{-3}$)	2.265	2.500	2.765	3.096	3.446
Largest diff. hole (e $Å^{-3}$)	–0.721	–0.723	–0.733	–0.721	–0.748
CCDC number	1016189	1016188	1016187	1016186	1016185

Table S6. Crystallographic data of κ-$(ET)_2Cu(NCS)_2$ (**f**)

Chemical formula	$C_{22}H_{16}CuN_2S_{18}$				
Formula weight	949.18				
Crystal system	Monoclinic				
Space group	$P2_1$				
Temperature (K)	298	250	200	150	100
Crystal dimension (mm^3)	0.45 × 0.44 × 0.03				
a (Å)	16.240(1)	16.243(1)	16.267(1)	16.313(1)	16.343(1)
b (Å)	8.4425(6)	8.4301(5)	8.4210(5)	8.4112(6)	8.3968(5)
c (Å)	13.1293(9)	13.0764(8)	13.0072(8)	12.9105(9)	12.8302(8)
β (°)	110.276(1)	110.455(1)	110.715(1)	111.028(1)	111.229(1)
V ($Å^3$)	1688.5(2)	1677.6(2)	1666.6(2)	1653.5(2)	1641.2(2)
Z	2	2	2	2	2
D_{calc} (g cm^{-3})	1.867	1.879	1.891	1.906	1.920
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	1.784	1.795	1.807	1.821	1.835
No. of reflections measured	9261	9190	9136	9032	8968
No. of independent reflections	6215	6161	6104	6051	6004
R_{int}	0.0381	0.0166	0.0162	0.0271	0.0142
No. of parameters	426	426	426	388	388
Final R values ($I > 2\sigma(I)$)	0.0423	0.0338	0.0289	0.0302	0.0223
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.1062	0.0798	0.0697	0.0787	0.0640
Final R values (all data)	0.0506	0.0398	0.0329	0.0329	0.0239
Final $wR(F^2)$ values (all data)	0.1105	0.0890	0.0787	0.0872	0.0645
Goodness of fit on F^2	1.061	1.119	1.136	1.117	1.099
Largest diff. peak (e $Å^{-3}$)	0.620	0.840	0.862	1.010	0.820
Largest diff. hole (e $Å^{-3}$)	–0.574	–0.570	–0.451	–0.516	–0.356
CCDC number	1016179	1016178	1016177	1016176	1016175

Table S7. Crystallographic data of κ-$(ET)_2Cu_2(CN)_3$ (**g**)

Chemical formula	$C_{23}H_{16}Cu_2N_3S_{16}$				
Formula weight	974.43				
Crystal system	Monoclinic				
Space group	$P2_1/c$				
Temperature (K)	300	250	200	150	100
Crystal dimension (mm^3)	0.49 × 0.39 × 0.02				
a (Å)	16.080(2)	16.082(2)	16.068(2)	16.060(2)	16.075(2)
b (Å)	8.584(1)	8.591(1)	8.585(1)	8.580(1)	8.580(1)
c (Å)	13.395(2)	13.353(2)	13.307(2)	13.280(2)	13.286(2)
β (°)	113.349(1)	113.794(1)	114.200(1)	114.516(1)	114.772(1)
V ($Å^3$)	1697.4(4)	1688.0(4)	1674.4(3)	1664.9(3)	1663.8(4)
Z	2	2	2	2	2
D_{calc} (g cm^{-3})	1.907	1.917	1.933	1.944	1.945
Diffractometer	Bruker APEXII CCD area detector				
Radiation type	MoKα				
Absorption correction	Numerical				
μ (mm^{-1})	2.263	2.275	2.294	2.307	2.309
No. of reflections measured	17423	17617	17329	17490	17415
No. of independent reflections	3685	3654	3639	3612	3608
R_{int}	0.0285	0.0193	0.0287	0.0196	0.0206
No. of parameters	218	218	199	199	199
Final R values ($I > 2\sigma(I)$)	0.0275	0.0277	0.0376	0.0358	0.0298
Final $wR(F^2)$ values ($I > 2\sigma(I)$)	0.0702	0.0737	0.0989	0.0969	0.0839
Final R values (all data)	0.0304	0.0298	0.0383	0.0363	0.0302
Final $wR(F^2)$ values (all data)	0.0724	0.0748	0.0992	0.0971	0.0840
Goodness of fit on F^2	1.109	1.193	1.223	1.307	1.310
Largest diff. peak (e $Å^{-3}$)	0.471	0.365	0.608	0.580	0.487
Largest diff. hole (e $Å^{-3}$)	–0.390	–0.387	–0.423	–0.402	–0.420
CCDC number	1016299	1016298	1016297	1016296	1016295

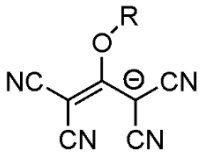

Fig. S1. Molecular structure of 2-alkoxy-1,1,3,3-tetracyanoallyl (for alkoxy = methoxy, ethoxy, butoxy, etc).