Reactive Molten-Flux Assisted Syntheses of Single Crystals of $Cs_{19}Ln_{19}Mn_{10}Te_{48}$ (*Ln* = Pr and Gd) Crystallizing in a New Structure Type

Gopabandhu Panigrahi,[a] Subhendu Jana,[a] S. Narayanswamy,[b] Pinaki P. Bhattacharjee,[b] Manish K. Niranjan,[c] and Jai Prakash[a*]

[a]*Department of Chemistry, Indian Institute of Technology Hyderabad, Kandi, Sangareddy, Telangana 502284, India*

[b]*Department of Materials Science & Metallurgical Engineering, Indian Institute of Technology Hyderabad, Kandi, Sangareddy, Telangana 502284, India*

[c]*Department of Physics, Indian Institute of Technology Hyderabad, Kandi, Sangareddy, Telangana 502284, India*

Electronic Supplementary Information (ESI)

The following reactants were used for the attempts to synthesize $Cs_{19}Ln_{19}Mn_{10}Te_{48}$ (*Ln* = Sm and Yb): cesium chloride powder (SRL, 99.9 % purity), samarium chunks (Alfa Aesar, 99.9 % purity), ytterbium chunks (Alfa Aesar, 99.9 % purity), manganese (II) oxide powder (Alfa Aesar, 99.9% purity), and tellurium ingot (Sigma-Aldrich, 99.999 % purity). These metals were handled inside an Ar-filled dry glove box due to the air and moisture sensitivity. The thin oxidized layer on the surface of the *Ln* chunks was first removed, and then small shiny pieces were cut for the reactions. The CsCl powder was dried in an oven overnight and then stored in the glove box prior to use for the reactions. The other reactants were used as obtained.

***Synthesis of single crystals of* $Cs_{19}Sm_{19}Mn_{10}Te_{48}$.** Black plate-shaped crystals of $Cs_{19}Pr_{19}Mn_{10}Te_{48}$ were obtained by the high-temperature molten flux reaction of Sm (41.8 mg, 0.278 mmol), MnO (4.9 mg, 0.069 mmol), and Te (53.2 mg, 0.417 mmol) along with an excess of cesium chloride (100 mg, 0.594 mmol) as a reactant and molten flux. All these reactants were weighed and then transferred in a carbon-coated fused silica tube [6 mm outer diameter (OD)] inside the argon-filled glove box.

The fused silica tube containing the reaction mixture was then connected to a vacuum line and evacuated (*ca*. 10^{-4} Torr) using a diffusion pump and sealed by a flame torch. The evacuated sealed tube was placed inside a programmable muffle furnace for heat treatment. The temperature of the furnace was slowly ramped up to 1273 K (in 54 h), and the reaction mixture was soaked for 48 h at 1273 K. The reaction mixture was then allowed to cool down to 1123 K at a rate of 3 K/h and annealed for 48 h before cooling the furnace to 773 K at a rate of 4 K/h. The furnace was finally switched off at 773 K to cool down the reaction mixture to room temperature by radiative cooling.

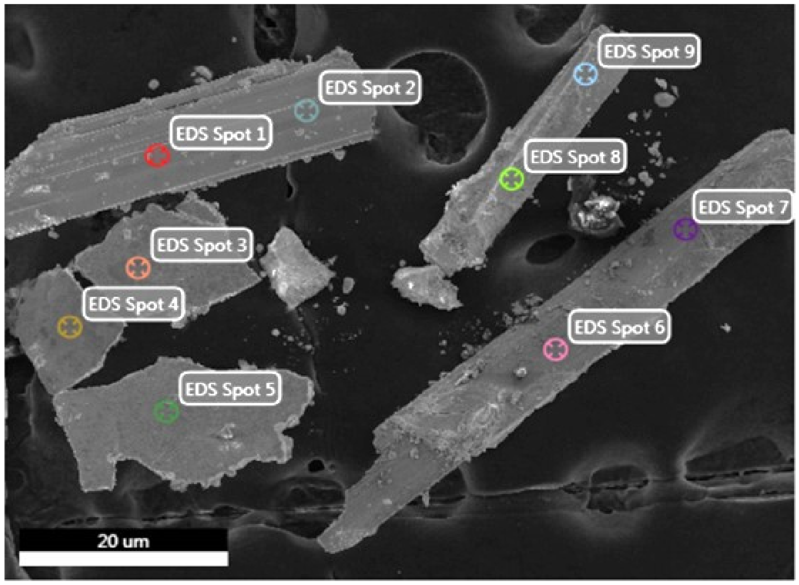

Fig SI 1. The SEM image of $Cs_{19}Sm_{19}Mn_{10}Te_{48}$ crystals.

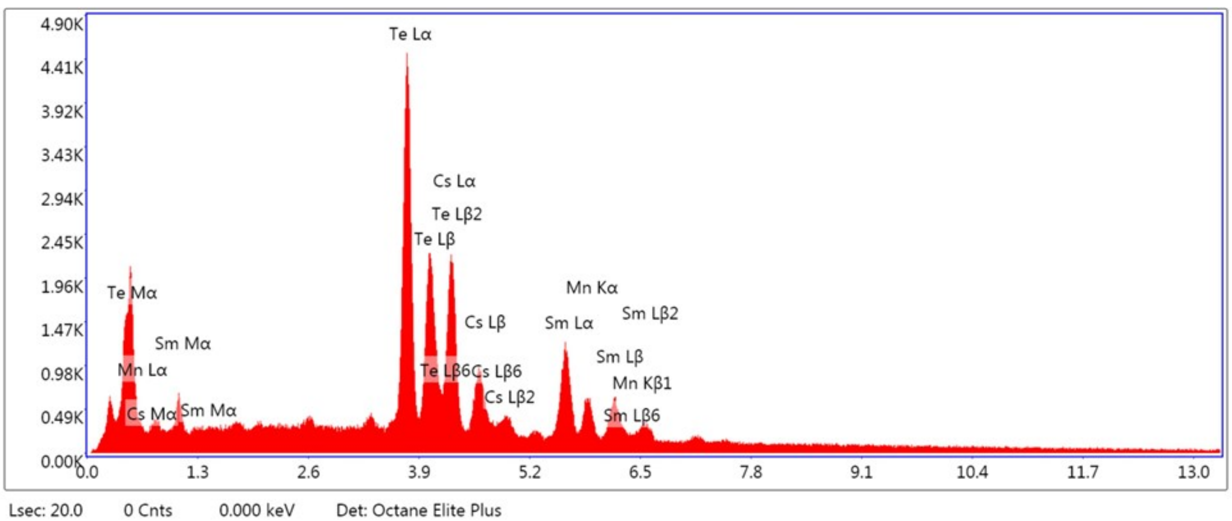

Fig SI 2. The EDS (spot 1) spectrum of $Cs_{19}Sm_{19}Mn_{10}Te_{48}$ crystal.

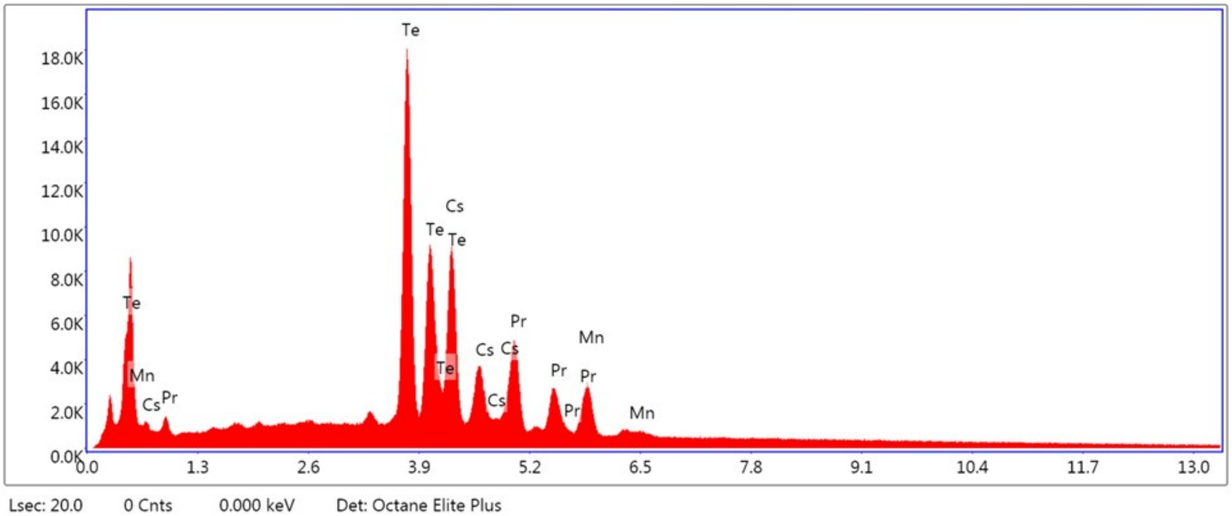

Fig SI 3. The EDS (spot 1) spectrum of $Cs_{19}Pr_{19}Mn_{10}Te_{48}$ crystal.

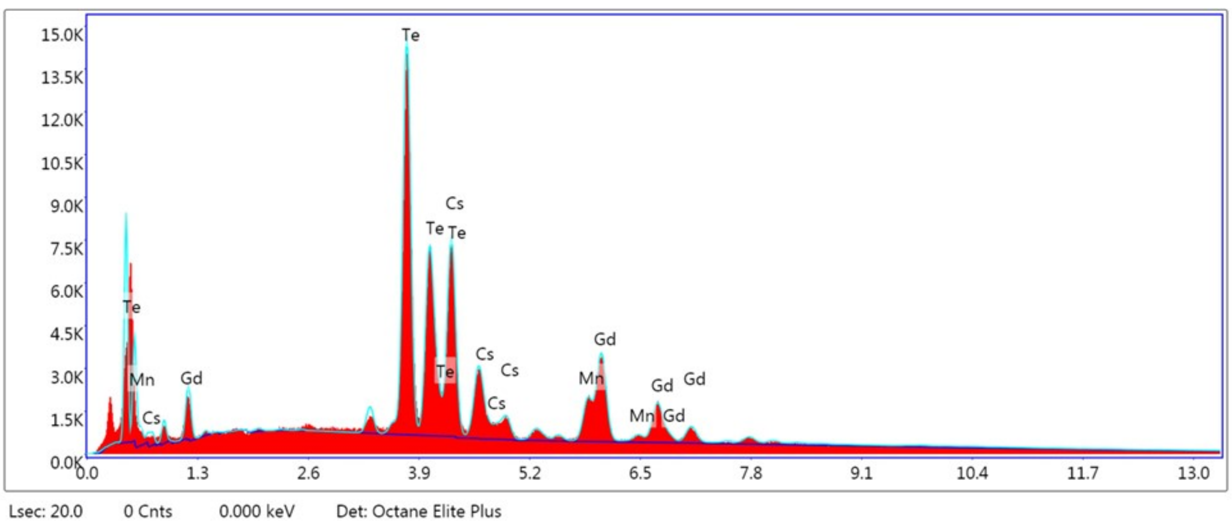

Fig SI 4. The EDS (spot 1) spectrum of $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ crystal

***Synthesis of single crystals of* $\mathbf{Cs_{19}Yb_{19}Mn_{10}Te_{48}}$.** Black plate-shaped crystals of $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ were synthesized by the high-temperature molten flux reaction of Yb (45.2 mg, 0.261 mmol), MnO (4.6 mg, 0.065 mmol), and Te (50.1 mg, 0.392 mmol) along with an excess of cesium chloride (100 mg, 0.594 mmol) that acted a molten flux as well as a source of cesium. The heating profile of this reaction is the same as employed for the Sm-compound.

Attempt to synthesize single crystals of $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ by using Mn-Metal. We also try to synthesize $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ crystals by the reaction of elements instead of MnO as a source of Mn. But black needle-shaped crystals of $CsGdMnTe_3$ were obtained (instead of the target phase) by the high-temperature molten flux reaction of Gd (43.4 mg, 0.275 mmol), Mn (3.8 mg, 0.068 mmol), and Te (52.8 mg, 0.413 mmol) along with an excess of cesium chloride (100 mg, 0.594 mmol) that acted a motel flux as well as a source of cesium. The composition of $CsGdMnTe_3$ was established by the EDS study (Fig. SI3).

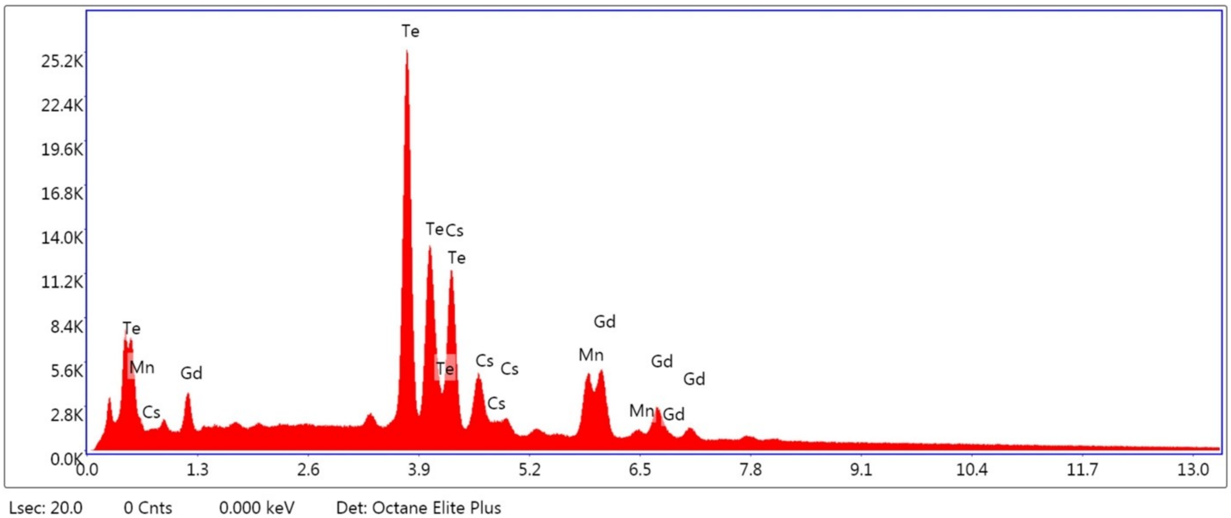

Fig. SI 5. The EDS spectrum of $CsGdMnTe_3$ Crystal.

Table SI 1: Fractional atomic coordinates and U_{iso}/U_{eq} values for $Cs_{19}Ln_{19}Mn_{10}Te_{48}$ structure.

$Cs_{19}Ln_{19}Mn_{10}Te_{48}$ (*Ln* = Pr and Gd)					
Atoms	**Wyckoff Position**	***x* (Pr/Gd)**	***y* (Pr/Gd)**	***z* (Pr/Gd)**	**U_{iso}/U_{eq} (Pr/Gd)**
Cs(1)	4*i*	0.05410(3)/0.05360(2)	0.000000	0.22564(4)/0.22609(3)	0.0307(3)/0.0293(2)
Cs(2)	4*i*	0.15450(3)/0.15433(2)	0.000000	0.70265(3)/0.70229(3)	0.0256(2)/0.0263(2)
Cs(3)	4*i*	0.21339(3)/0.21332(2)	0.000000	0.46351(3)/0.46368(3)	0.0250(2)/0.0266(2)
Cs(4)	4*i*	0.25854(3)/0.25933(2)	0.000000	0.19427(4)/0.19347(3)	0.0311(3)/0.0302(2)
Cs(5)	4*i*	0.42661(3)/0.42557(2)	0.000000	0.37513(4)/0.37503(3)	0.0346(3)/0.0317(2)
Cs(6)	4*i*	0.46860(3)/0.46846(2)	0.000000	0.11719(3)/0.11671(3)	0.0257(3)/0.0263(2)
Cs(7)	4*i*	0.63402(3)/0.63481(2)	0.000000	0.35402(4)/0.35371(3)	0.0299(3)/0.0300(2)
Cs(8)	4*i*	0.67856(2)/0.67800(2)	0.000000	0.07541(3)/0.07479(3)	0.0226(2)/0.0235(2)
Cs(9)	4*i*	0.88906(4)/0.88942(3)	0.000000	0.04304(4)/0.04262(3)	0.0411(4)/0.0389(3)
Cs(10)	2*c*	0.00000/0.00000	0.000000	0.50000/0.50000	0.0282(4)/0.0246(3)

*Ln(*1)	4*i*	0.03466(2)/0.03424(2)	0.000000	0.36724(3)/0.36778(2)	0.01697(18)/0.01553(15)
Ln(2)	4*i*	0.05751(2)/0.05802(2)	0.000000	0.76507(3)/0.76507(2)	0.01514(18)/0.01428(15)
Ln(3)	4*i*	0.11478(2)/0.11523(2)	0.000000	0.51217(3)/0.51228(2)	0.01444(17)/0.01468(15)
Ln(4)	4*i*	0.25350(2)/0.25289(2)	0.000000	0.65292(3)/0.65269(2)	0.01387(17)/0.01417(15)
Ln(5)	4*i*	0.27132(2)/0.27133(2)	0.000000	0.04814(3)/0.04740(2)	0.01485(17)/0.01586(15)
Ln(6)	4*i*	0.31738(2)/0.31714(2)	0.000000	0.41540(3)/0.41589(2)	0.01359(17)/0.01385(15)
Ln(7)	4*i*	0.36946(2)/0.36992(2)	0.000000	0.17608(3)/0.17586(2)	0.01449(17)/0.01513(15)
Ln(8)	4*i*	0.56804(2)/0.56746(2)	0.000000	0.07190(3)/0.07178(2)	0.01528(18)/0.01527(15)
Ln(9)	4*i*	0.65508(2)/0.65545(2)	0.000000	0.21250(3)/0.21214(2)	0.01567(18)/0.01613(15)
Ln(10)	2*a*	0.00000/0.00000	0.000000	0.00000/0.00000	0.0166(3)/0.0160(2)
Mn(1)	4*i*	0.12030(6)/0.11998(5)	0.000000	0.13563(8)/0.13581(6)	0.0179(5)/0.0184(5)
Mn(2)	4*i*	0.19211(6)/0.19260(5)	0.000000	0.28624(8)/0.28591(6)	0.0188(5)/0.0180(5)
Mn(3)	4*i*	0.43392(6)/0.43376(5)	0.000000	0.55487(8)/0.55479(6)	0.0192(5)/0.0190(5)
Mn(4)	4*i*	0.49745(6)/0.49715(5)	0.000000	0.29323(8)/0.29354(6)	0.0178(5)/0.0182(5)
Mn(5)	4*i*	0.81418(6)/0.81442(5)	0.000000	0.12042(8)/0.11973(6)	0.0193(5)/0.0198(5)
Te(1)	4*i*	0.02452(2)/0.02465(2)	0.000000	0.08618(3)/0.08563(3)	0.0177(2)/0.0164(2)
Te(2)	4*i*	0.02757(2)/0.02832(2)	0.000000	0.68025(3)/0.68019(2)	0.0172(2)/0.0157(2)
Te(3)	4*i*	0.08856(2)/0.08889(2)	0.000000	0.84709(3)/0.84657(3)	0.0164(2)/0.01572(19)
Te(4)	4*i*	0.09347(2)/0.09391(2)	0.000000	0.42141(3)/0.42133(3)	0.0168(2)/0.0167(2)
Te(5)	4*i*	0.11344(2)/0.11194(2)	0.000000	0.06010(3)/0.05977(3)	0.0166(2)/0.0165(2)
Te(6)	4*i*	0.13434(3)/0.13440(2)	0.000000	0.26439(3)/0.26255(3)	0.0178(2)/0.0178(2)
Te(7)	4*i*	0.13659(2)/0.13762(2)	0.000000	0.59790(3)/0.59687(2)	0.0144(2)/0.01390(19)
Te(8)	4*i*	0.17349(2)/0.17206(2)	0.000000	0.80419(3)/0.80451(3)	0.0159(2)/0.01581(19)
Te(9)	4*i*	0.17717(3)/0.17747(2)	0.000000	0.16200(3)/0.16293(3)	0.0171(2)/0.0170(2)
Te(10)	4*i*	0.20118(2)/0.20266(2)	0.000000	0.36160(3)/0.36161(2)	0.0156(2)/0.01508(19)
Te(11)	4*i*	0.20932(2)/0.20946(2)	0.000000	0.00433(3)/0.00365(2)	0.0148(2)/0.0165(2)
Te(12)	4*i*	0.22751(2)/0.22704(2)	0.000000	0.56934(3)/0.56978(2)	0.0141(2)/0.01378(19)
Te(13)	4*i*	0.28959(2)/0.28957(2)	0.000000	0.33027(3)/0.33107(3)	0.0163(2)/0.01529(19)
Te(14)	4*i*	0.33743(2)/0.33803(2)	0.000000	0.09146(3)/0.09065(3)	0.0164(2)/0.0169(2)
Te(15)	4*i*	0.34137(2)/0.34167(2)	0.000000	0.50139(3)/0.50112(3)	0.0158(2)/0.0159(2)
Te(16)	4*i*	0.40094(2)/0.40122(2)	0.000000	0.25817(3)/0.25741(2)	0.0161(2)/0.01504(19)
Te(17)	4*i*	0.43134(2)/0.43020(2)	0.000000	0.47931(3)/0.47893(3)	0.0172(2)/0.0158(2)
Te(18)	4*i*	0.45387(2)/0.45472(2)	0.000000	0.01331(3)/0.01244(3)	0.0173(2)/0.0163(2)
Te(19)	4*i*	0.48668(2)0.48533(2)	0.000000	0.21794(3)/0.21782(3)	0.0168(2)/0.0158(2)
Te(20)	4*i*	0.51012(3)/0.50951(2)	0.000000	0.41521(3)/0.41407(3)	0.0197(2)/0.0165(2)
Te(21)	4*i*	0.55495(3)/0.55516(2)	0.000000	0.31435(3)/0.31678(3)	0.0184(2)/0.0171(2)

Te(22)	4*i*	0.59433(3)/0.59371(2)	0.000000	0.16066(3)/0.16095(3)	0.0173(2)/0.0165(2)
Te(23)	4*i*	0.71754(2)/0.71851(2)	0.000000	0.26003(3)/0.25973(3)	0.0157(2)/0.0163(2)
Te(24)	4*i*	0.75648(2)/0.75636(2)	0.000000	0.10472(3)/0.10255(3)	0.0163(2)/0.0171(2)

Table SI 2: Atomic displacement parameters (Å^2) for $Cs_{19}\mathit{Ln}_{19}Mn_{10}Te_{48}$

$Cs_{19}Pr_{19}Mn_{10}Te_{48}$						
	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Cs01	0.0269(6)	0.0267(7)	0.0393(7)	0.000	0.0080(5)	0.000
Cs02	0.0284(6)	0.0279(6)	0.0213(5)	0.000	0.0075(4)	0.000
Cs03	0.0268(6)	0.0273(6)	0.0220(5)	0.000	0.0082(4)	0.000
Cs04	0.0267(6)	0.0278(7)	0.0400(7)	0.000	0.0091(5)	0.000
Cs05	0.0360(7)	0.0312(7)	0.0385(7)	0.000	0.0134(6)	0.000
Cs06	0.0272(6)	0.0296(7)	0.0211(5)	0.000	0.0065(4)	0.000
Cs07	0.0233(6)	0.0272(7)	0.0400(7)	0.000	0.0070(5)	0.000
Cs08	0.0181(5)	0.0223(6)	0.0272(6)	0.000	0.0015(4)	0.000
Cs09	0.0568(10)	0.0387(9)	0.0300(7)	0.000	0.0166(7)	0.000
Cs10	0.0321(9)	0.0219(9)	0.0316(9)	0.000	0.0084(7)	0.000
Pr01	0.0215(5)	0.0129(4)	0.0155(4)	0.000	-0.0035(3)	0.000
Pr02	0.0151(4)	0.0127(4)	0.0170(4)	0.000	-0.0020(3)	0.000
Pr03	0.0164 (4)	0.0099(4)	0.0165(4)	0.000	-0.0011(3)	0.000
Pr04	0.0144(4)	0.0120(4)	0.0146(4)	0.000	-0.0018(3)	0.000
Pr05	0.0179(4)	0.0107(4)	0.0152(4)	0.000	-0.0022(3)	0.000
Pr06	0.0140(4)	0.0106(4)	0.0158(4)	0.000	-0.0005(3)	0.000
Pr07	0.0145(4)	0.0129(4)	0.0156(4)	0.000	-0.0012(3)	0.000
Pr08	0.0152(4)	0.0143(4)	0.0157(4)	0.000	-0.0018(3)	0.000
Pr09	0.0195(4)	0.0122(4)	0.0145(4)	0.000	-0.0030(3)	0.000
Pr10	0.0160(6)	0.0152(6)	0.0178(6)	0.000	-0.0027(5)	0.000
Mn01	0.0207(13)	0.0157(13)	0.0163(12)	0.000	-0.0031(10)	0.000
Mn02	0.0216(13)	0.0176(13)	0.0166(12)	0.000	-0.0017(10)	0.000
Mn03	0.0218 (13)	0.0171 (14)	0.0179 (12)	0.000	-0.0012 (10)	0.000
Mn04	0.0210 (13)	0.0143 (13)	0.0176 (12)	0.000	-0.0012 (10)	0.000
Mn05	0.0204 (13)	0.0185 (14)	0.0182 (12)	0.000	-0.0022 (10)	0.000
Te01	0.0156 (5)	0.0166 (5)	0.0211 (5)	0.000	0.0022 (4)	0.000
Te02	0.0182 (5)	0.0159 (5)	0.0167 (5)	0.000	-0.0028 (4)	0.000
Te03	0.0167 (5)	0.0164 (5)	0.0162 (5)	0.000	0.0015 (4)	0.000

Te04	0.0180 (5)	0.0154 (5)	0.0163 (5)	0.000	-0.0024 (4)	0.000
Te05	0.0162 (5)	0.0178 (6)	0.0156 (5)	0.000	0.0007 (4)	0.000
Te06	0.0182 (5)	0.0173 (6)	0.0179 (5)	0.000	0.0011 (4)	0.000
Te07	0.0153 (5)	0.0136 (5)	0.0140 (5)	0.000	0.0004 (4)	0.000
Te08	0.0162 (5)	0.0157 (5)	0.0158 (5)	0.000	0.0013 (4)	0.000
Te09	0.0187 (5)	0.0162 (5)	0.0162 (5)	0.000	0.0001 (4)	0.000
Te10	0.0151 (5)	0.0163 (5)	0.0155 (5)	0.000	0.0016 (4)	0.000
Te11	0.0178 (5)	0.0111 (5)	0.0154 (5)	0.000	0.0011 (4)	0.000
Te12	0.0145 (5)	0.0136 (5)	0.0142 (5)	0.000	0.0010 (4)	0.000
Te13	0.0174 (5)	0.0142 (5)	0.0173 (5)	0.000	0.0024 (4)	0.000
Te14	0.0165 (5)	0.0163 (5)	0.0160 (5)	0.000	-0.0013 (4)	0.000
Te15	0.0175 (5)	0.0123 (5)	0.0180 (5)	0.000	0.0030 (4)	0.000
Te16	0.0157 (5)	0.0151 (5)	0.0176 (5)	0.000	0.0014 (4)	0.000
Te17	0.0172 (5)	0.0142 (5)	0.0203 (5)	0.000	0.0016 (4)	0.000
Te18	0.0174 (5)	0.0184 (6)	0.0155 (5)	0.000	-0.0009 (4)	0.000
Te19	0.0167 (5)	0.0174 (5)	0.0160 (5)	0.000	0.0006 (4)	0.000
Te20	0.0212 (5)	0.0177 (6)	0.0203 (5)	0.000	0.0021 (4)	0.000
Te21	0.0188 (5)	0.0176 (6)	0.0184 (5)	0.000	0.0001 (4)	0.000
Te22	0.0192 (5)	0.0160 (5)	0.0159 (5)	0.000	-0.0027 (4)	0.000
Te23	0.0172 (5)	0.0164 (5)	0.0130 (5)	0.000	-0.0013 (4)	0.000
Te24	0.0164 (5)	0.0156 (5)	0.0169 (5)	0.000	0.0010 (4)	0.000
$Cs_{19}Gd_{19}Mn_{10}Te_{48}$						
	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Cs1	0.0240 (6)	0.0267 (5)	0.0383 (6)	0.000	0.0085 (4)	0.000
Cs2	0.0264 (6)	0.0308 (5)	0.0224 (5)	0.000	0.0059 (4)	0.000
Cs3	0.0293 (6)	0.0300 (6)	0.0214 (5)	0.000	0.0063 (4)	0.000
Cs4	0.0241 (6)	0.0271 (5)	0.0401 (6)	0.000	0.0066 (4)	0.000
Cs5	0.0318 (6)	0.0330 (6)	0.0319 (6)	0.000	0.0096 (5)	0.000
Cs6	0.0274 (6)	0.0307 (5)	0.0214 (5)	0.000	0.0058 (4)	0.000
Cs7	0.0220 (6)	0.0282 (5)	0.0399 (6)	0.000	0.0038 (4)	0.000
Cs8	0.0186 (5)	0.0239 (5)	0.0278 (5)	0.000	0.0019 (4)	0.000
Cs9	0.0524(8)	0.0373(6)	0.0293(6)	0.000	0.0140(5)	0.000
Cs10	0.0244(8)	0.0246(7)	0.0248(7)	0.000	0.0027(6)	0.000
Gd1	0.0171(4)	0.0141(3)	0.0140(4)	0.000	-0.0039(3)	0.000
Gd2	0.0129(4)	0.0134(3)	0.0155(4)	0.000	-0.0023(3)	0.000
Gd3	0.0142(4)	0.0128(3)	0.0159(4)	0.000	-0.0032(3)	0.000
Gd4	0.0132(4)	0.0130(3)	0.0153(4)	0.000	-0.0029(3)	0.000

Gd5	0.0184(4)	0.0138(3)	0.0142(4)	0.000	-0.0029(3)	0.000
Gd6	0.0129(4)	0.0117(3)	0.0160(4)	0.000	-0.0024(3)	0.000
Gd7	0.0127(4)	0.0144(3)	0.0172(4)	0.000	-0.0028(3)	0.000
Gd8	0.0143(4)	0.0144(3)	0.0161(4)	0.000	-0.0021(3)	0.000
Gd9	0.0181(4)	0.0140(3)	0.0148(4)	0.000	-0.0040(3)	0.000
Gd10	0.0142(5)	0.0155(5)	0.0172(5)	0.000	-0.0024(4)	0.000
Mn1	0.0179(12)	0.0166(11)	0.0194(12)	0.000	-0.0029(9)	0.000
Mn2	0.0187(12)	0.0157(11)	0.0180(12)	0.000	-0.0041(9)	0.000
Mn3	0.0234(13)	0.0146(11)	0.0176(11)	0.000	-0.0041(9)	0.000
Mn4	0.0191(12)	0.0153(11)	0.0188(12)	0.000	-0.0032(9)	0.000
Mn5	0.0177(12)	0.0177(11)	0.0224(12)	0.000	-0.0036(9)	0.000
Te1	0.0148(5)	0.0145(5)	0.0198(5)	0.000	0.0014(4)	0.000
Te2	0.0144(5)	0.0167(5)	0.0148(5)	0.000	-0.0032(4)	0.000
Te3	0.0148(5)	0.0159(5)	0.0160(5)	0.000	0.0000(4)	0.000
Te4	0.0169(5)	0.0166(5)	0.0154(5)	0.000	-0.0032(4)	0.000
Te5	0.0144(5)	0.0175(5)	0.0172(5)	0.000	-0.0001(4)	0.000
Te6	0.0161(5)	0.0180(5)	0.0186(5)	0.000	-0.0003(4)	0.000
Te7	0.0125(5)	0.0146(4)	0.0138(4)	0.000	-0.0017(3)	0.000
Te8	0.0139(5)	0.0167(5)	0.0161(5)	0.000	-0.0008(4)	0.000
Te9	0.0159(5)	0.0160(5)	0.0183(5)	0.000	-0.0011(4)	0.000
Te10	0.0141(5)	0.0153(4)	0.0153(5)	0.000	-0.0008(3)	0.000
Te11	0.0182(5)	0.0153(5)	0.0156(5)	0.000	-0.0006(4)	0.000
Te12	0.0132(5)	0.0135(4)	0.0141(5)	0.000	-0.0008(3)	0.000
Te13	0.0155(5)	0.0135(4)	0.0168(5)	0.000	0.0011(4)	0.000
Te14	0.0168(5)	0.0160(5)	0.0170(5)	0.000	-0.0024(4)	0.000
Te15	0.0152(5)	0.0144(4)	0.0177(5)	0.000	0.0001(4)	0.000
Te16	0.0135(5)	0.0150(5)	0.0161(5)	0.000	-0.0003(4)	0.000
Te17	0.0140(5)	0.0161(5)	0.0165(5)	0.000	-0.0011(4)	0.000
Te18	0.0164(5)	0.0177(5)	0.0141(5)	0.000	-0.0015(4)	0.000
Te19	0.0139(5)	0.0170(5)	0.0161(5)	0.000	0.0006(4)	0.000
Te20	0.0162(5)	0.0177(5)	0.0150(5)	0.000	-0.0007(4)	0.000
Te21	0.0153 (5)	0.0180 (5)	0.0175 (5)	0.000	-0.0005 (4)	0.000
Te22	0.0169 (5)	0.0159 (5)	0.0158 (5)	0.000	-0.0022 (4)	0.000
Te23	0.0175 (5)	0.0162 (5)	0.0141 (5)	0.000	-0.0024 (4)	0.000
Te24	0.0160 (5)	0.0175 (5)	0.0172 (5)	0.000	-0.0007 (4)	0.000

Table SI 3: Interatomic distances (Å) for $Cs_{19}Pr_{19}Mn_{10}Te_{48}$

Cs01—Te06	3.869 (2)	Cs09—Cs09^{iii}	4.5959 (9)
Cs01—Te19^{i}	3.8840 (15)	Cs09—Cs09^{iv}	4.5959 (9)
Cs01—Te19^{ii}	3.8840 (15)	Cs10—Te20^{v}	3.8866 (11)
Cs01—Te22^{i}	3.8882 (17)	Cs10—Te20^{i}	3.8866 (11)
Cs01—Te22^{ii}	3.8882 (17)	Cs10—Te20^{vi}	3.8866 (11)
Cs01—Te21^{ii}	3.9332 (16)	Cs10—Te20^{ii}	3.8866 (11)
Cs01—Te21^{i}	3.9332 (16)	Cs10—Te17^{v}	3.9551 (11)
Cs01—Mn04^{i}	4.406 (3)	Cs10—Te17^{i}	3.9551 (11)
Cs01—Mn04^{ii}	4.406 (3)	Cs10—Te17^{vi}	3.9551 (11)
Cs01—Cs01^{iii}	4.5959 (9)	Cs10—Te17^{ii}	3.9551 (11)
Cs01—Cs01^{iv}	4.5959 (9)	Cs10—Mn03^{i}	4.456 (3)
Cs01—Cs06^{i}	5.788 (2)	Cs10—Mn03^{v}	4.456 (3)
Cs02—Te08	3.6811 (19)	Cs10—Mn03^{vi}	4.456 (3)
Cs02—Te13^{v}	3.7522 (15)	Cs10—Mn03^{ii}	4.456 (3)
Cs02—Te13^{vi}	3.7522 (15)	Pr01—Te20^{ii}	3.1550 (12)
Cs02—Te07	3.7866 (18)	Pr01—Te20^{i}	3.1550 (12)
Cs02—Te16^{vi}	3.8282 (15)	Pr01—Te21^{i}	3.1857 (12)
Cs02—Te16^{v}	3.8282 (15)	Pr01—Te21^{ii}	3.1857 (12)
Cs02—Cs02^{iv}	4.5959 (9)	Pr01—Te04	3.2125 (18)
Cs02—Cs02^{iii}	4.5959 (9)	Pr01—Te02^{xii}	3.2263 (18)
Cs02—Cs05^{vi}	5.044 (2)	Pr02—Te03	3.1649 (17)
Cs02—Cs05^{v}	5.044 (2)	Pr02—Te16^{v}	3.1698 (11)
Cs02—Cs04^{v}	5.715 (2)	Pr02—Te16^{vi}	3.1698 (11)
Cs02—Cs04^{vi}	5.715 (2)	Pr02—Te19^{vi}	3.1850 (12)
Cs03—Te10	3.6610 (18)	Pr02—Te19^{v}	3.1850 (12)
Cs03—Te15^{vi}	3.7442 (15)	Pr02—Te02	3.2368 (17)
Cs03—Te15^{v}	3.7442 (15)	Pr03—Te15^{v}	3.1455 (11)
Cs03—Te12	3.8069 (18)	Pr03—Te15^{vi}	3.1455 (11)
Cs03—Te12^{v}	3.8606 (15)	Pr03—Te07	3.1583 (16)
Cs03—Te12^{vi}	3.8606 (15)	Pr03—Te17^{vi}	3.1828 (11)
Cs03—Cs03^{iii}	4.5959(9)	Pr03—Te17^{v}	3.1828(11)
Cs03—Cs03^{iv}	4.5959(9)	Pr03—Te04	3.3256(16)
Cs03—Cs03^{v}	4.707(3)	Pr04—Te12	3.1370(16)
Cs03—Cs03^{vi}	4.707(3)	Pr04—Te13^{vi}	3.1496(11)
Cs03—Cs07^{i}	5.637(2)	Pr04—Te13^{v}	3.1496(11)
Cs03—Cs07^{ii}	5.637(2)	Pr04—Te10^{v}	3.1989(11)

Cs04—Te09	3.863(2)	Pr04—Te10^{vi}	3.1989(11)
Cs04—Te08^{v}	3.9121(15)	Pr04—Te23^{xi}	3.2945(16)
Cs04—Te08^{vi}	3.9121(15)	Pr05—Te11	3.1607(17)
Cs04—Te23^{ii}	3.9259(16)	Pr05—Te11^{xiii}	3.1614(11)
Cs04—Te23^{i}	3.9259(16)	Pr05—Te11^{xiv}	3.1614(11)
Cs04—Te24^{i}	3.9544(16)	Pr05—Te24^{ii}	3.1909(12)
Cs04—Te24^{ii}	3.9544(16)	Pr05—Te24^{i}	3.1909(12)
Cs04—Mn05^{ii}	4.517(3)	Pr05—Te14	3.3218(17)
Cs04—Mn05^{i}	4.517(3)	Pr06—Te12^{vi}	3.1919(11)
Cs04—Cs04^{iii}	4.5959(9)	Pr06—Te12^{v}	3.1919(11)
Cs04—Cs04^{iv}	4.5959(9)	Pr06—Te15	3.1923(17)
Cs05—Te17	3.738(2)	Pr06—Te07^{v}	3.2119(11)
Cs05—Te02^{v}	3.8264(17)	Pr06—Te07^{vi}	3.2119(11)
Cs05—Te02^{vi}	3.8264(17)	Pr06—Te13	3.2139(17)
Cs05—Te07^{vi}	3.9322(16)	Pr07—Te08^{vi}	3.1737(11)
Cs05—Te07^{v}	3.9322(16)	Pr07—Te08^{v}	3.1737(11)
Cs05—Te20	4.024(2)	Pr07—Te16	3.1746(17)
Cs05—Te16	4.269(2)	Pr07—Te03^{v}	3.1817(12)
Cs05—Cs05^{iv}	4.5959(9)	Pr07—Te03^{vi}	3.1817(11)
Cs05—Cs05^{iii}	4.5959(9)	Pr07—Te14	3.2649(17)
Cs06—Te19	3.6473(19)	Pr08—Te01^{viii}	3.1411(11)
Cs06—Te01^{vii}	3.7268(15)	Pr08—Te01^{vii}	3.1411(11)
Cs06—Te01^{viii}	3.7268(15)	Pr08—Te18^{x}	3.1421(17)
Cs06—Te18	3.7399(18)	Pr08—Te05^{vii}	3.1809(11)
Cs06—Te03^{vi}	3.8339(15)	Pr08—Te05^{viii}	3.1809(11)
Cs06—Te03^{v}	3.8339(15)	Pr08—Te22	3.3139(17)
Cs06—Cs06^{iv}	4.5959(9)	Pr09—Te09^{viii}	3.1635(12)
Cs06—Cs06^{iii}	4.5959(9)	Pr09—Te09^{vii}	3.1635(12)
Cs06—Cs09^{i}	4.930(2)	Pr09—Te06^{vii}	3.1709(12)
Cs06—Cs09^{ii}	4.930(2)	Pr09—Te06^{viii}	3.1709(12)
Cs07—Te21	3.828(2)	Pr09—Te23	3.2367(17)
Cs07—Te10^{vii}	3.8744 (15)	Pr09—Te22	3.2423 (18)
Cs07—Te10^{viii}	3.8744 (15)	Pr10—Te01	3.2050 (14)
Cs07—Te04^{vii}	3.9532 (17)	Pr10—Te01^{xv}	3.2050 (14)
Cs07—Te04^{viii}	3.9532 (17)	Pr10—Te18^{xiv}	3.2152 (9)
Cs07—Te06^{viii}	3.9641 (16)	Pr10—Te18^{i}	3.2152 (9)
Cs07—Te06^{vii}	3.9641 (16)	Pr10—Te18^{ii}	3.2152 (9)

Cs07—Mn02^{vii}	4.454 (3)	Pr10—Te18^{xiii}	3.2152 (9)
Cs07—Mn02^{viii}	4.454 (3)	Mn01—Te05	2.709 (3)
Cs07—Cs07^{iv}	4.5959 (9)	Mn01—Te09	2.732 (3)
Cs07—Cs07^{iii}	4.5959 (9)	Mn01—Te22^{i}	2.7873 (19)
Cs08—Te24	3.6878 (18)	Mn01—Te22^{ii}	2.7873 (19)
Cs08—Te05^{viii}	3.8075 (14)	Mn02—Te10	2.707 (3)
Cs08—Te05^{vii}	3.8075 (14)	Mn02—Te06	2.735 (3)
Cs08—Te11^{viii}	3.8202 (15)	Mn02—Te23^{ii}	2.7914 (19)
Cs08—Te11^{vii}	3.8202 (15)	Mn02—Te23^{i}	2.7914 (19)
Cs08—Te09^{vii}	3.8798 (15)	Mn03—Te17	2.713 (3)
Cs08—Te09^{viii}	3.8798 (15)	Mn03—Te20^{xi}	2.728 (3)
Cs08—Mn01^{viii}	4.300 (3)	Mn03—Te04^{vi}	2.8017 (19)
Cs08—Mn01^{vii}	4.300 (3)	Mn03—Te04^{v}	2.8017 (19)
Cs08—Cs08^{iii}	4.5959 (9)	Mn04—Te19	2.711 (3)
Cs08—Cs08^{iv}	4.5959 (9)	Mn04—Te21	2.718 (3)
Cs08—Cs09^{ix}	5.557 (2)	Mn04—Te02^{vi}	2.7870 (18)
Cs09—Te05^{x}	3.706 (2)	Mn04—Te02^{v}	2.7870 (18)
Cs09—Te14^{viii}	3.8629 (18)	Mn05—Te24	2.698 (3)
Cs09—Te14^{vii}	3.8629 (18)	Mn05—Te08^{xi}	2.722 (3)
Cs09—Te03^{xi}	3.996 (2)	Mn05—Te14^{vii}	2.7860 (19)
Cs09—Te18^{vii}	4.0230 (18)	Mn05—Te14^{viii}	2.7860 (19)
Cs09—Te18^{viii}	4.0230 (18)		

Symmetry codes: (i) *x*-1/2, *y*-1/2, *z*; (ii) *x*-1/2, *y*+1/2, *z*; (iii) *x*, *y*-1, *z*; (iv) *x*, *y*+1, *z*; (v) -*x*+1/2, -*y*+1/2, -*z*+1; (vi) -*x*+1/2, -*y*-1/2, -*z*+1; (vii) *x*+1/2, *y*+1/2, *z*; (viii) *x*+1/2, *y*-1/2, *z*; (ix) -*x*+3/2, -*y*-1/2, -*z*; (x) -*x*+1, -*y*, -*z*; (xi) -*x*+1, -*y*, -*z*+1; (xii) -*x*, -*y*, -*z*+1; (xiii) -*x*+1/2, -*y*-1/2, -*z*; (xiv) -*x*+1/2, -*y*+1/2, -*z*; (xv) -*x*, -*y*, -*z*.

Table SI 4: Interatomic distances (Å) for $Cs_{19}Gd_{19}Mn_{10}Te_{48}$

Cs1—Te6	3.8062 (14)	Cs9—Te18^{viii}	4.0184 (13)
Cs1—Te19^{i}	3.8606 (11)	Cs9—Cs9^{iii}	4.5030 (2)
Cs1—Te19^{ii}	3.8606 (11)	Cs9—Cs9^{iv}	4.5030 (2)
Cs1—Te22^{i}	3.8728 (12)	Cs10—Te20^{v}	3.8811 (8)
Cs1—Te22^{ii}	3.8728 (12)	Cs10—Te20^{i}	3.8811 (8)
Cs1—Te21^{ii}	3.9452 (11)	Cs10—Te20^{vi}	3.8811 (8)
Cs1—Te21^{i}	3.9452 (11)	Cs10—Te20^{ii}	3.8811 (8)
Cs1—Mn4^{i}	4.387 (2)	Cs10—Te17^{v}	3.9339 (8)

Cs1—Mn4ii	4.387 (2)	Cs10—Te17i	3.9339 (8)
Cs1—Cs1iii	4.5030 (2)	Cs10—Te17vi	3.9339 (8)
Cs1—Cs1iv	4.5030 (2)	Cs10—Te17ii	3.9339 (8)
Cs1—Cs6i	5.6749 (13)	Cs10—Mn3i	4.445 (2)
Cs2—Te8	3.6542 (13)	Cs10—Mn3v	4.445 (2)
Cs2—Te13v	3.7385 (11)	Cs10—Mn3vi	4.445 (2)
Cs2—Te13vi	3.7385 (11)	Cs10—Mn3ii	4.445 (2)
Cs2—Te7	3.7610 (13)	Gd1—Te20ii	3.0957 (8)
Cs2—Te16vi	3.8275 (11)	Gd1—Te20i	3.0957 (8)
Cs2—Te16v	3.8275 (11)	Gd1—Te21i	3.1251 (8)
Cs2—Cs2iv	4.5030 (2)	Gd1—Te21ii	3.1251 (8)
Cs2—Cs2iii	4.5030 (2)	Gd1—Te4	3.1650 (11)
Cs2—Cs5vi	4.8911 (13)	Gd1—Te2xii	3.1811 (11)
Cs2—Cs5v	4.8911 (13)	Gd2—Te3	3.0909 (11)
Cs2—Cs4v	5.5997 (13)	Gd2—Te16v	3.1045 (8)
Cs2—Cs4vi	5.5997 (13)	Gd2—Te16vi	3.1045 (8)
Cs3—Te10	3.6296 (13)	Gd2—Te19vi	3.1242 (8)
Cs3—Te15vi	3.7285 (11)	Gd2—Te19v	3.1242 (8)
Cs3—Te15v	3.7285 (11)	Gd2—Te2	3.1812 (11)
Cs3—Te12	3.7755 (13)	Gd3—Te7	3.0806 (11)
Cs3—Te12v	3.8617 (11)	Gd3—Te15v	3.0828 (8)
Cs3—Te12vi	3.8617 (11)	Gd3—Te15vi	3.0828 (8)
Cs3—Cs3iii	4.5030 (2)	Gd3—Te17vi	3.1214 (8)
Cs3—Cs3iv	4.5030 (2)	Gd3—Te17v	3.1214 (8)
Cs3—Cs3v	4.6055 (17)	Gd3—Te4	3.2876 (11)
Cs3—Cs3vi	4.6055 (17)	Gd4—Te12	3.0641 (11)
Cs3—Cs7i	5.5065 (13)	Gd4—Te13vi	3.0886 (8)
Cs3—Cs7ii	5.5065 (13)	Gd4—Te13v	3.0886 (8)
Cs4—Te9	3.8117 (14)	Gd4—Te10v	3.1326 (8)
Cs4—Te8v	3.8842 (11)	Gd4—Te10vi	3.1326 (8)
Cs4—Te8vi	3.8842 (11)	Gd4—Te23xi	3.2540 (11)
Cs4—Te23ii	3.9174 (12)	Gd5—Te11	3.0924 (11)
Cs4—Te23i	3.9174 (12)	Gd5—Te11xiii	3.0974 (8)
Cs4—Te24i	3.9470 (11)	Gd5—Te11xiv	3.0974 (8)
Cs4—Te24ii	3.9470 (11)	Gd5—Te24ii	3.1272 (8)
Cs4—Mn5i	4.490 (2)	Gd5—Te24i	3.1272 (8)
Cs4—Mn5ii	4.490 (2)	Gd5—Te14	3.2802 (11)

Cs4—Cs4^{iii}	4.5030 (2)	Gd6—Te12^{vi}	3.1241 (8)
Cs4—Cs4^{iv}	4.5030 (2)	Gd6—Te12^{v}	3.1241 (8)
Cs5—Te17	3.7010 (14)	Gd6—Te15	3.1250 (11)
Cs5—Te2^{vi}	3.8139 (12)	Gd6—Te7^{v}	3.1445 (8)
Cs5—Te2^{v}	3.8139 (12)	Gd6—Te7^{vi}	3.1445 (8)
Cs5—Te7^{vi}	3.9160 (12)	Gd6—Te13	3.1492 (11)
Cs5—Te7^{v}	3.9160 (12)	Gd7—Te16	3.0995 (11)
Cs5—Te20	3.9640 (14)	Gd7—Te8^{vi}	3.1086 (8)
Cs5—Te16	4.2266 (14)	Gd7—Te8^{v}	3.1086 (8)
Cs5—Cs5^{iii}	4.5030 (2)	Gd7—Te3^{v}	3.1166 (8)
Cs5—Cs5^{iv}	4.5030 (2)	Gd7—Te3^{vi}	3.1166 (8)
Cs5—Cs10^{vii}	5.7645 (10)	Gd7—Te14	3.2249 (11)
Cs6—Te19	3.6115 (13)	Gd8—Te18^{x}	3.0663 (11)
Cs6—Te1^{vii}	3.7094 (11)	Gd8—Te1^{viii}	3.0780 (8)
Cs6—Te1^{viii}	3.7094 (11)	Gd8—Te1^{vii}	3.0780 (8)
Cs6—Te18	3.7110 (13)	Gd8—Te5^{vii}	3.1144 (8)
Cs6—Te3^{vi}	3.8310 (11)	Gd8—Te5^{viii}	3.1144 (8)
Cs6—Te3^{v}	3.8310 (11)	Gd8—Te22	3.2770 (11)
Cs6—Cs6^{iv}	4.5030 (2)	Gd9—Te9^{viii}	3.1059 (8)
Cs6—Cs6^{iii}	4.5030 (2)	Gd9—Te9^{vii}	3.1059 (8)
Cs6—Cs9^{i}	4.8091 (14)	Gd9—Te6^{vii}	3.1144 (8)
Cs6—Cs9^{ii}	4.8091 (14)	Gd9—Te6^{viii}	3.1144 (8)
Cs7—Te21	3.7607 (14)	Gd9—Te23	3.1951 (11)
Cs7—Te10^{vii}	3.8451 (11)	Gd9—Te22	3.2015 (11)
Cs7—Te10^{viii}	3.8451 (11)	Gd10—Te1	3.1397 (9)
Cs7—Te4^{vii}	3.9519 (12)	Gd10—Te1^{xv}	3.1398 (9)
Cs7—Te4^{viii}	3.9519 (12)	Gd10—Te18^{xiv}	3.1431 (7)
Cs7—Te6^{viii}	3.9640 (12)	Gd10—Te18^{i}	3.1431 (7)
Cs7—Te6^{vii}	3.9640 (12)	Gd10—Te18^{ii}	3.1431 (7)
Cs7—Mn2^{vii}	4.436 (2)	Gd10—Te18^{xiii}	3.1431 (7)
Cs7—Mn2^{viii}	4.436 (2)	Mn1—Te5	2.705 (2)
Cs7—Cs7^{iv}	4.5030 (2)	Mn1—Te9	2.718 (2)
Cs7—Cs7^{iii}	4.5030 (2)	Mn1—Te22^{i}	2.7602 (15)
Cs8—Te24	3.6409 (13)	Mn1—Te22^{ii}	2.7602 (15)
Cs8—Te5^{viii}	3.7811 (11)	Mn2—Te10	2.696 (2)
Cs8—Te5^{vii}	3.7811 (11)	Mn2—Te6	2.720 (2)
Cs8—Te11^{viii}	3.8177 (11)	Mn2—Te23^{ii}	2.7670 (15)

Cs8—Te11^{vii}	3.8177 (11)	Mn2—Te23^{i}	2.7670 (15)
Cs8—Te9^{vii}	3.8796 (11)	Mn3—Te17	2.703 (2)
Cs8—Te9^{viii}	3.8796 (11)	Mn3—Te20^{xi}	2.724 (2)
Cs8—Mn1^{viii}	4.297 (2)	Mn3—Te4^{vi}	2.7753 (15)
Cs8—Mn1^{vii}	4.297 (2)	Mn3—Te4^{v}	2.7753 (15)
Cs8—Cs8^{iv}	4.5030 (2)	Mn4—Te19	2.701 (2)
Cs8—Cs8^{iii}	4.5030 (2)	Mn4—Te21	2.711 (2)
Cs8—Cs9^{ix}	5.4141 (14)	Mn4—Te2^{vi}	2.7584 (14)
Cs9—Te5^{x}	3.6602 (14)	Mn4—Te2^{v}	2.7584 (14)
Cs9—Te14^{viii}	3.8293 (12)	Mn5—Te24	2.681 (2)
Cs9—Te14^{vii}	3.8293 (12)	Mn5—Te8^{xi}	2.710 (2)
Cs9—Te3^{xi}	3.9736 (14)	Mn5—Te14^{vii}	2.7606 (15)
Cs9—Te18^{vii}	4.0184 (13)	Mn5—Te14^{viii}	2.7606 (15)

Symmetry codes: (i) *x*-1/2, *y*-1/2, *z*; (ii) *x*-1/2, *y*+1/2, *z*; (iii) *x*, *y*-1, *z*; (iv) *x*, *y*+1, *z*; (v) -*x*+1/2, -*y*+1/2, -*z*+1; (vi) -*x*+1/2, -*y*-1/2, -*z*+1; (vii) *x*+1/2, *y*+1/2, *z*; (viii) *x*+1/2, *y*-1/2, *z*; (ix) -*x*+3/2, -*y*-1/2, -*z*; (x) -*x*+1, -*y*, -*z*; (xi) -*x*+1, -*y*, -*z*+1; (xii) -*x*, -*y*, -*z*+1; (xiii) -*x*+1/2, -*y*-1/2, -*z*; (xiv) -*x*+1/2, -*y*+1/2, -*z*; (xv) -*x*, -*y*, -*z*.

The orientations of magnetic moments of Mn atoms in the $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ system as calculated by the DFT studies.

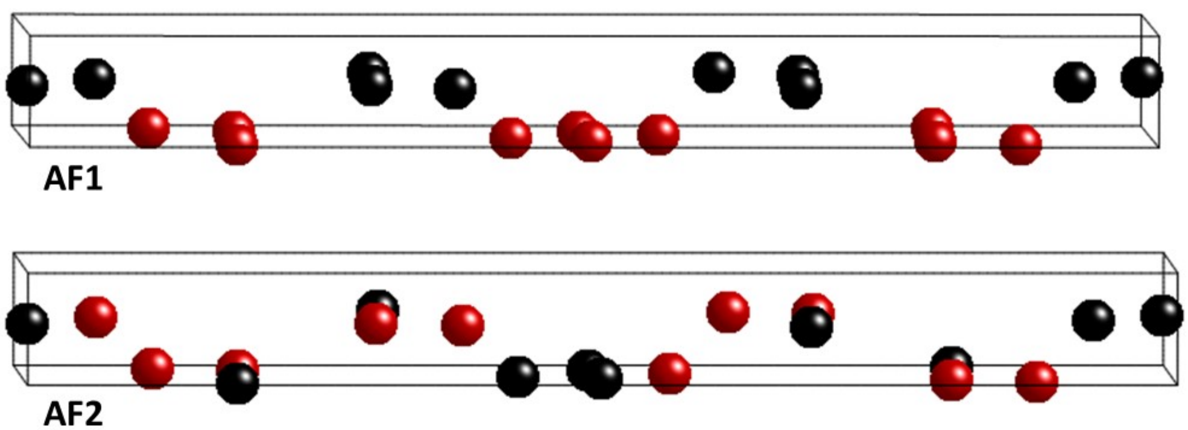

Fig. SI 6: The orientations of magnetic moments of Mn atoms in the $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ system with AF1 and AF2 antiferromagnetic configurations. The red and black balls indicate Mn atoms with spin-up and spin-down orientations, respectively. (Only Mn atoms are shown in the unit cell).

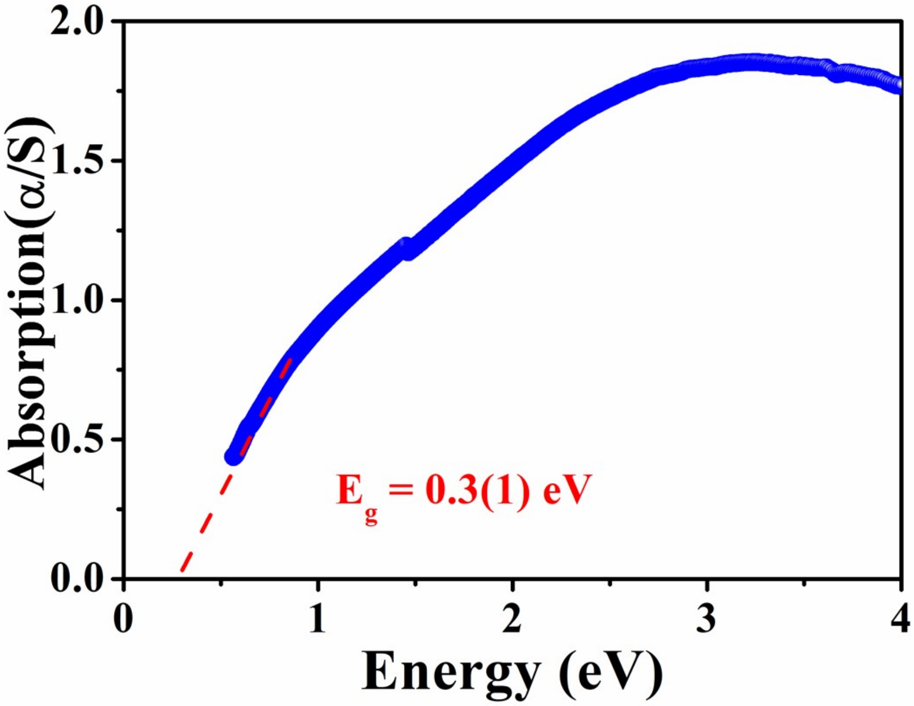

Fig. SI 7 Absorption spectrum of polycrystalline $Cs_{19}Gd_{19}Mn_{10}Te_{48}$ compound