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

Supporting information for

Emergent high superconductivity in layered TaS_3 crystal

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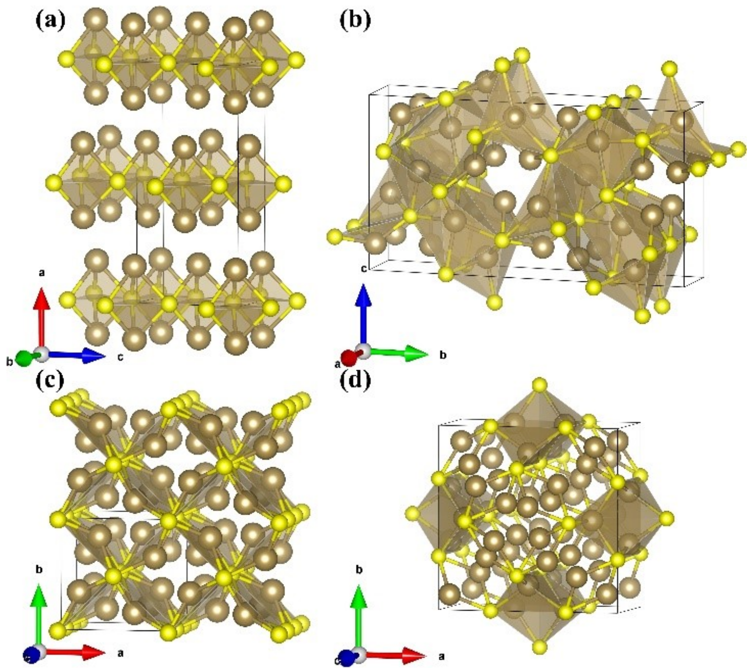

FIG. S1 The crystal structures of Ta-rich Ta-S compounds. (a) *Fmm*2 Ta_2S at 0 GPa. (b) *Cmc*2_1 Ta_2S at 60 GPa. (c) *I*4/*mcm* Ta_2S at 180 GPa. (d) $I\bar{4}2m$ Ta_3S at 40 GPa.

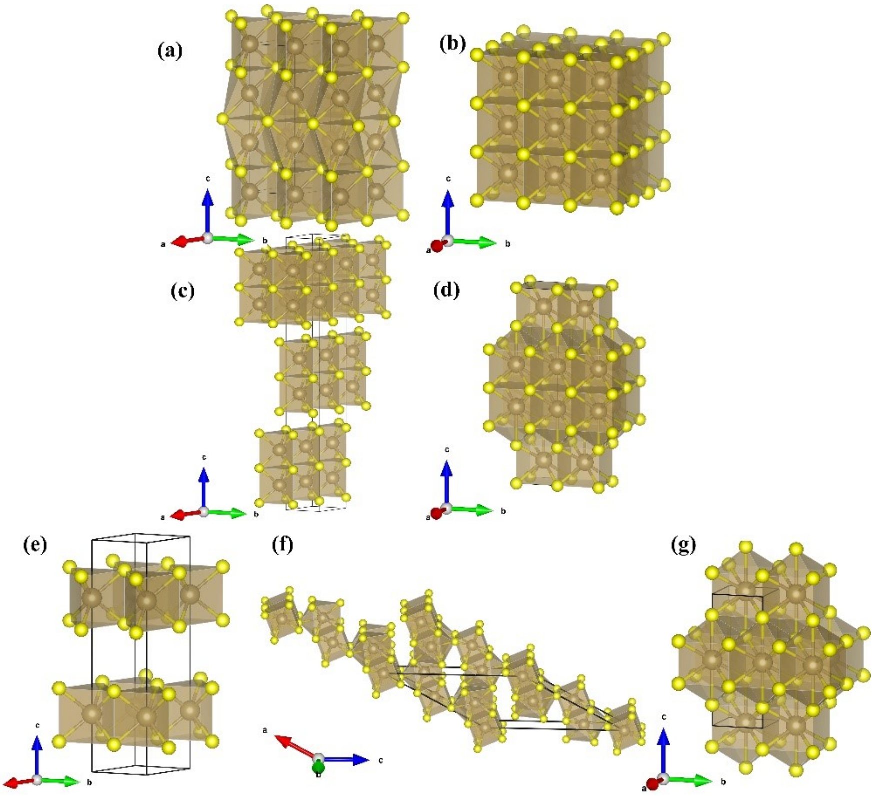

FIG. S2 The crystal structures of S-rich Ta-S compounds. (a) $P\bar{6}m2$ TaS at 60 GPa. (b) $Pm\bar{3}m$ TaS at 80 GPa. (c) $R3m$ Ta_2S_3 at 60 GPa. (d) $I4/mmm$ Ta_2S_3 at 80 GPa. (e) $P6_3/mmc$ TaS_2 at 0 GPa. (f) $C2/m$ TaS_2 at 50 GPa. (g) $I4/mmm$ TaS_2 at 60 GPa.

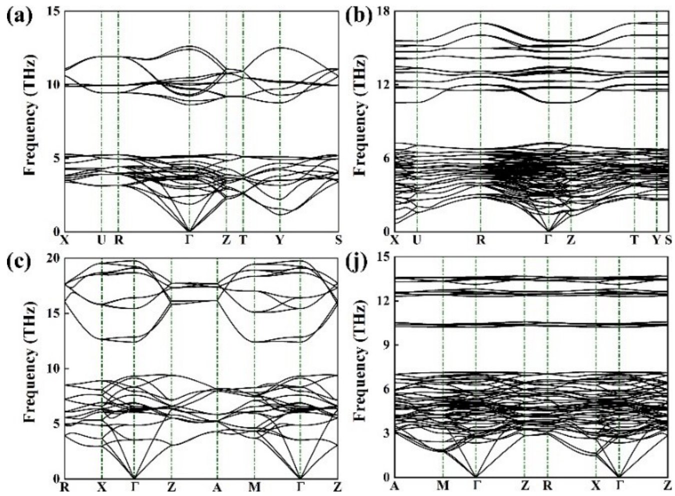

FIG. S3 Phonon dispersions of stable Ta-rich Ta-S compounds. (a) *Fmm*2 Ta_2S at 0 GPa. (b) *Cmc*2_1 Ta_2S at 60 GPa. (c) *I*4/*mcm* Ta_2S at 180 GPa. (d) $I\bar{4}2m$ Ta_3S at 60 GPa. The absence of imaginary frequencies in these structures indicates they are dynamically stable.

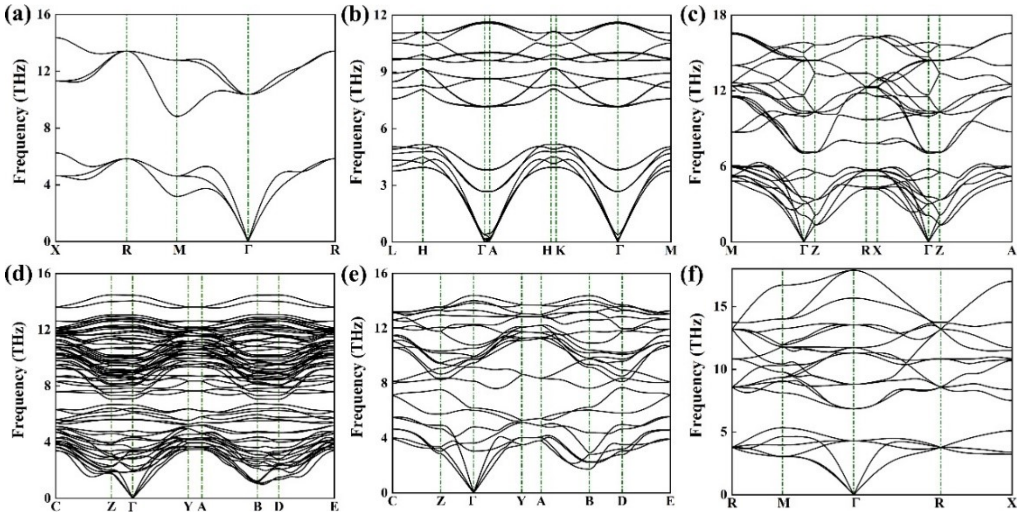

FIG. S4 Phonon dispersions of stable S-rich Ta-S compounds. (a) $Pm\bar{3}m$ TaS at 80 GPa. (b) *R*3*m* Ta_2S_3 at 60 GPa. (c) *I*4/*mmm* Ta_2S_3 at 80 GPa. (d) *P*2_1/*m*-exp TaS_3 at 20 GPa. (e) *P*2_1/*m* TaS_3 at 40 GPa. (f) $Pm\bar{3}m$ TaS_3 at 80 GPa. The absence of imaginary frequencies in these structures indicates they are dynamically stable.

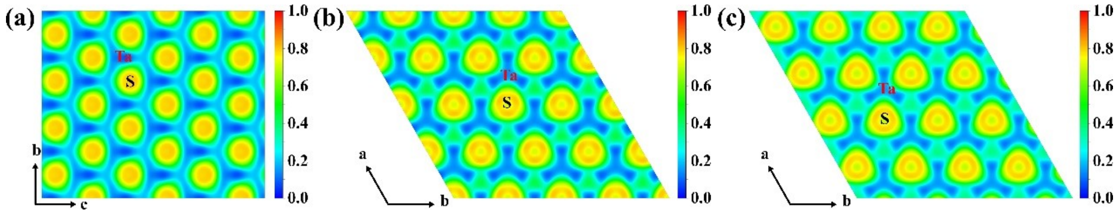

FIG. S5 Valence electron localization function (ELF) for (a) *Fmm*2 Ta_2S phase at 0 GPa, (b) *R*3*m* Ta_2S_3 at 0 GPa, and (c) *P*6$_3$/*mmc* TaS_2 at 20 GPa.

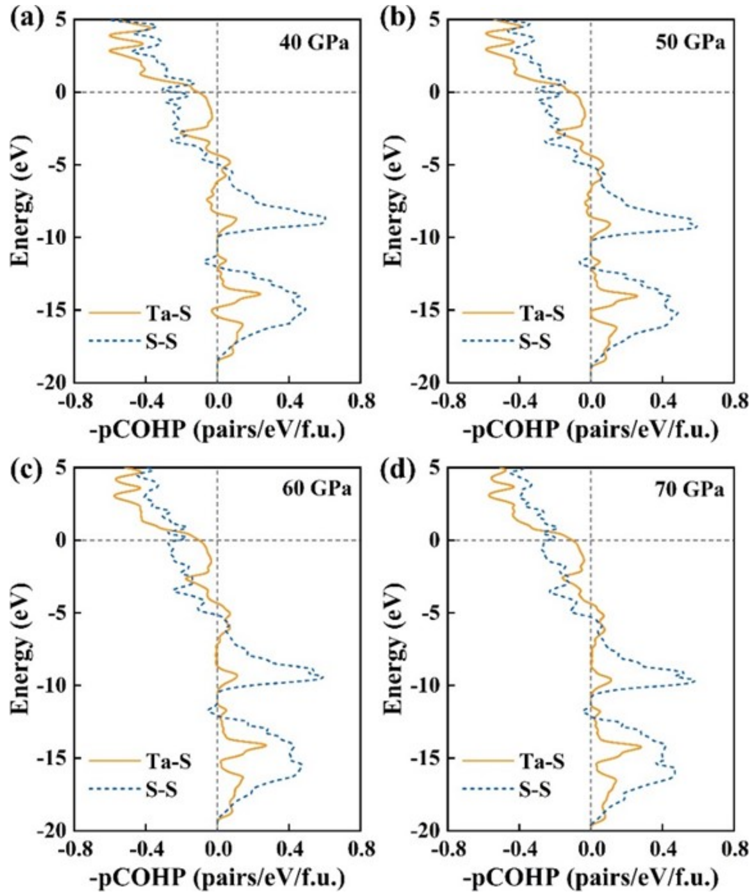

FIG. S6 Projected crystal orbital Hamiltonian population (-pCOHP) of *P*2$_1$/*m* TaS_3 at (a) 40 GPa, (b) 50 GPa, (c) 60 GPa and (d) 70 GPa. The positive and negative values of -pCOHP signify bonding and antibonding states, respectively. The Fermi level is set to zero.

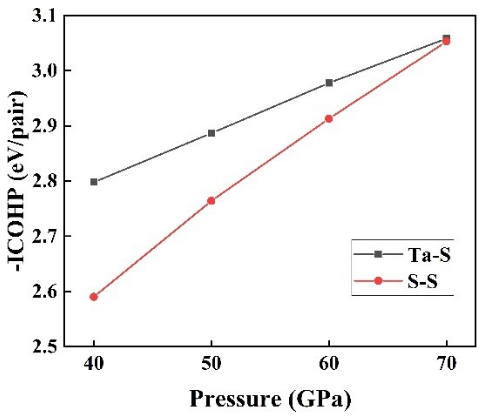

FIG. S7 The integrated COHP (ICOHP) for Ta-S and S-S bonds of $P2_1/m$ TaS_3 at the pressure range of 40 to 70 GPa.

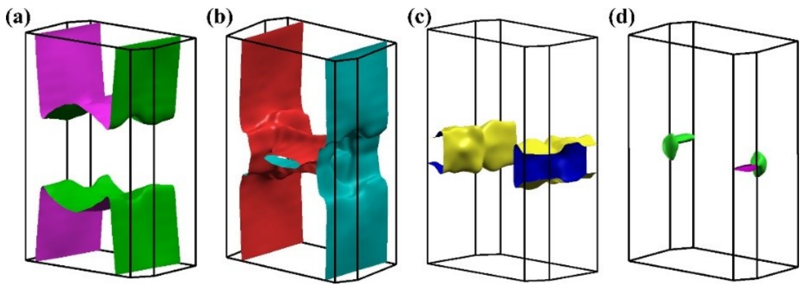

FIG. S8 The Fermi surface associated to each band crossing the Fermi level of $P2_1/m$ TaS_3 at 50 GPa.

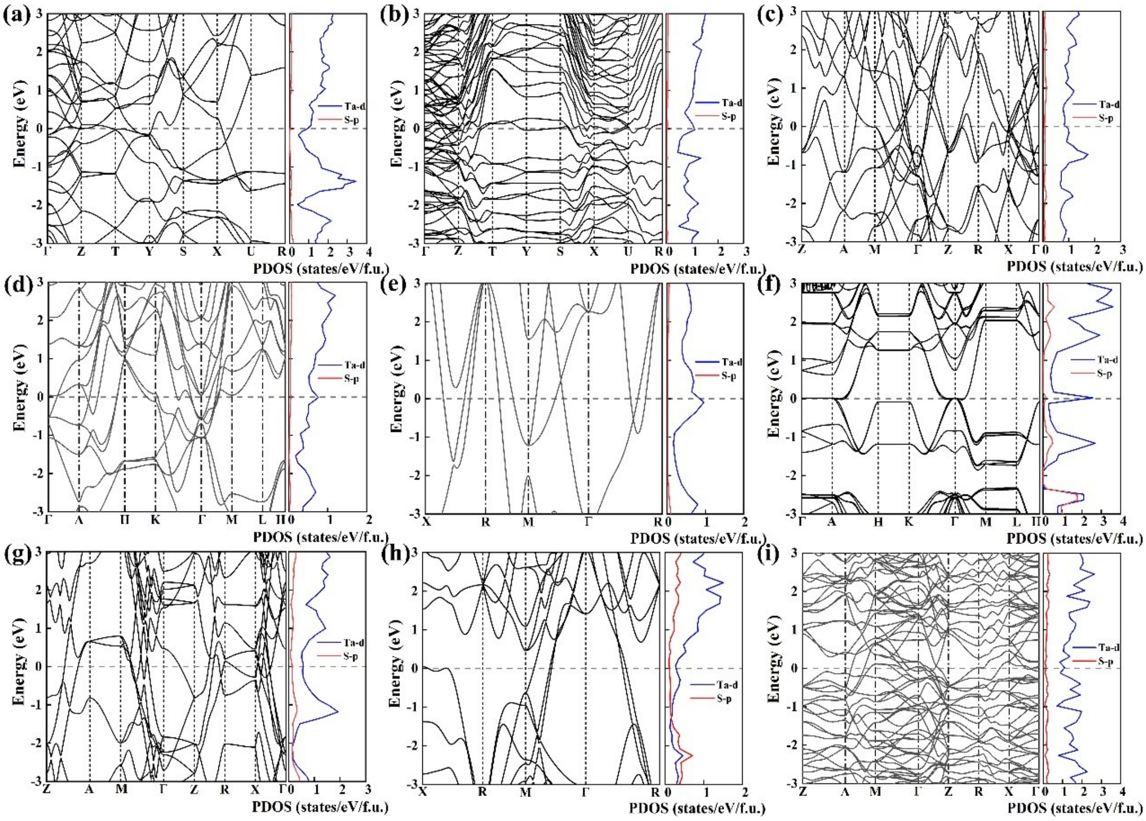

FIG. S9 The electronic band structures and projected density of states (PDOS) have been calculated at the PBE level for (a) *Fmm*2 Ta_2S at 0 GPa. (b) *Cmc*2$_1$ Ta_2S at 60 GPa. (c) *I*4/*mcm* Ta_2S at 180 GPa. (d) $P\bar{6}m2$ TaS at 60 GPa. (e) $Pm\bar{3}m$ TaS at 80 GPa. (f) *R*3*m* Ta_2S_3 at 60 GPa. (g) *I*4/*mmm* Ta_2S_3 at 80 GPa. (h) $Pm\bar{3}n$ TaS_3 at 80 GPa. (i) $I\bar{4}2m$ Ta_3S at 60 GPa.

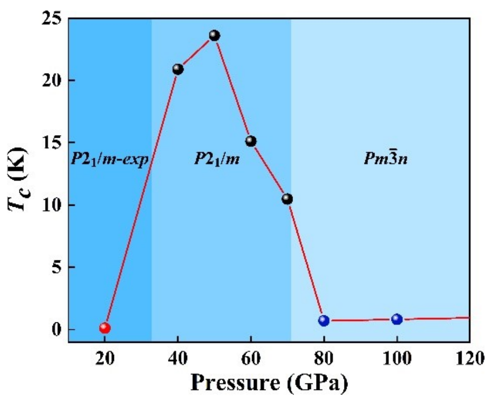

FIG. S10 The calculated superconducting T_c evolution diagram with pressure for TaS_3 using a typical choice of $\mu^* = 0.1$.

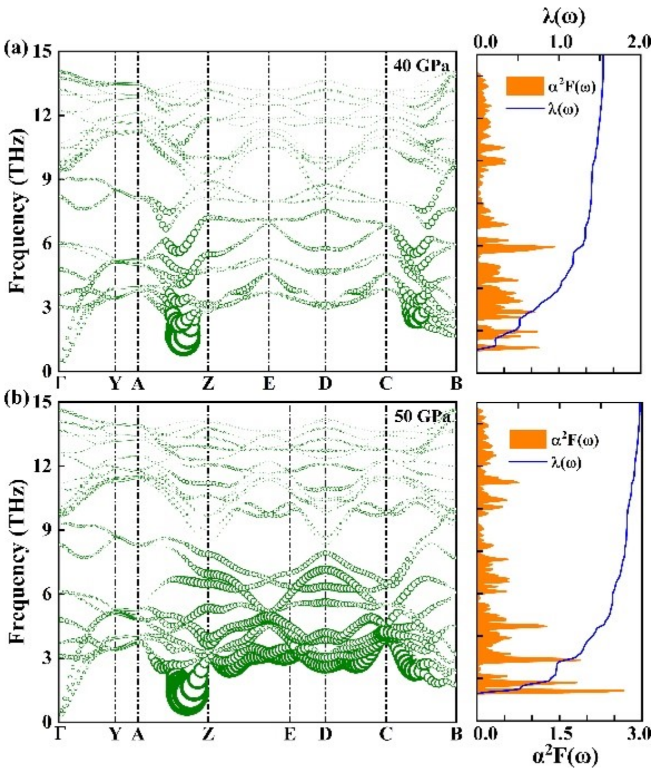

FIG. S11 The phonon dispersions, the Eliashberg spectral function $\alpha^2F(\omega)$ (orange area) and frequency-dependent electron-phonon coupling parameters $\lambda(\omega)$ (blue line) for $P2_1/m$ TaS_3 at 40 (a) and 50 GPa (b). The radius of each circle in phonon spectra is proportional to the partial electron-phonon coupling strength of each phonon mode.

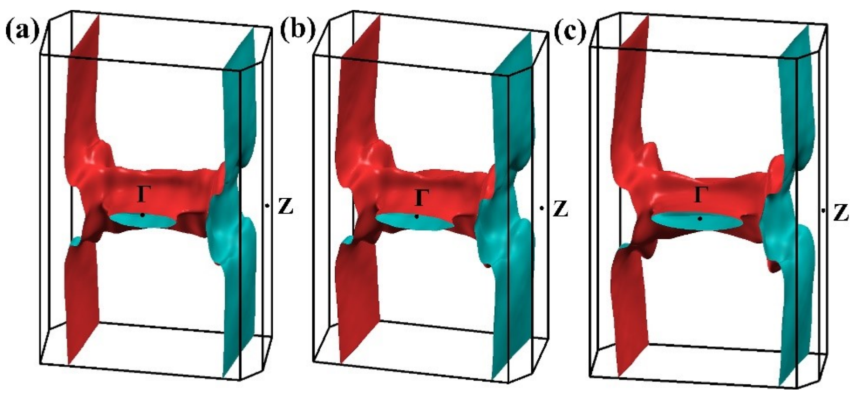

FIG. S12 The nested Fermi surface of $P2_1/m$ TaS_3 at (a) 50 GPa, (b) 60 GPa and (c) 70 GPa.

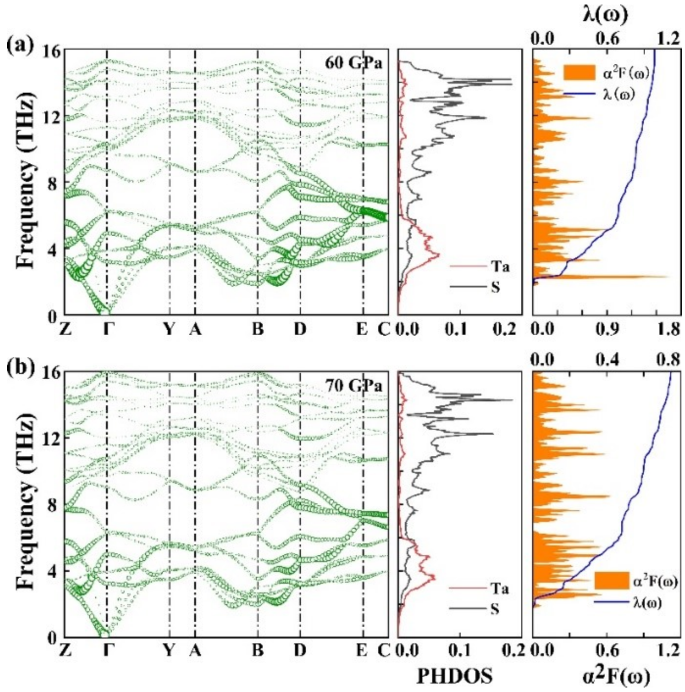

FIG. S13 The phonon dispersions, phonon density of states (PHDOS), the Eliashberg spectral function $\alpha^2F(\omega)$ (orange area) and frequency-dependent electron-phonon coupling parameters $\lambda(\omega)$ (blue line) for $P2_1/m$ TaS_3 at 60 GPa (a) and 70 GPa (b). The radius of each circle in phonon spectra is proportional to the partial electron-phonon coupling strength of each phonon mode.

Table S1 Structural information of $P2_1/m$ and $Pm\bar{3}n$ phases of TaS_3 at 60 and 100 GPa, respectively.

Phases	Pressure (GPa)	Lattice parameters (Å, °)	Wyckoff position (fractional)			
			Atoms	x	y	z
$P2_1/m$	60	a = 7.5771	S(2e)	0.1368	0.7500	0.0993
		b = 3.0328	S(2e)	0.5639	0.7500	0.7356
		c = 4.5143	S(2e)	0.8685	0.2500	0.4190
		α = γ = 90.00 β = 81.58	Ta(2e)	0.3188	0.2500	0.7926
$Pm\bar{3}n$	100	a = b = c = 4.4177	S(6d)	0.2500	0.5000	0.0000
		α =β=γ = 90.00	Ta(2a)	0.0000	0.0000	0.0000

Table S2. Superconducting properties of the metallic Ta-S phases. The μ* value for the T_c calculation is taken as 0.1.

Phase	Pressure (GPa)	λ	$\omega_{\log}$ (K)	T_c (K)
$I\bar{4}2m$ Ta_3S	20	0.34	404.72	0.6
$Fmm2$ Ta_2S	50	0.28	475.26	0.1
$I4/mcm$ Ta_2S	180	0.47	449.66	4.1
$P\bar{6}m2$ TaS	20	0.67	214.05	6.8
$Pm\bar{3}m$ TaS	80	0.70	228.50	7.9
$I4/mmm$ Ta_2S_3	80	0.40	319.53	1.2
$P2_1/m$ TaS_3	20	0.30	221.93	0.1
$P2_1/m$ TaS_3	40	1.54	154.99	20.9
$Pm\bar{3}n$ TaS_3	80	0.36	338.52	0.7