Supplementary Material: Stable compositions, Structures and Electronic Properties in K–Ga Systems under Pressure *

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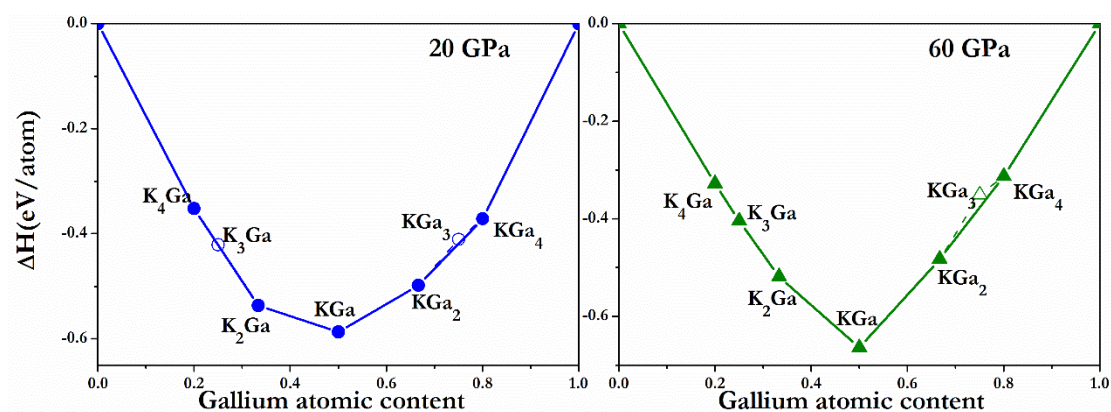**Figure S1**: Convex hull diagrams of the K−Ga system at 20 and 60 GPa.

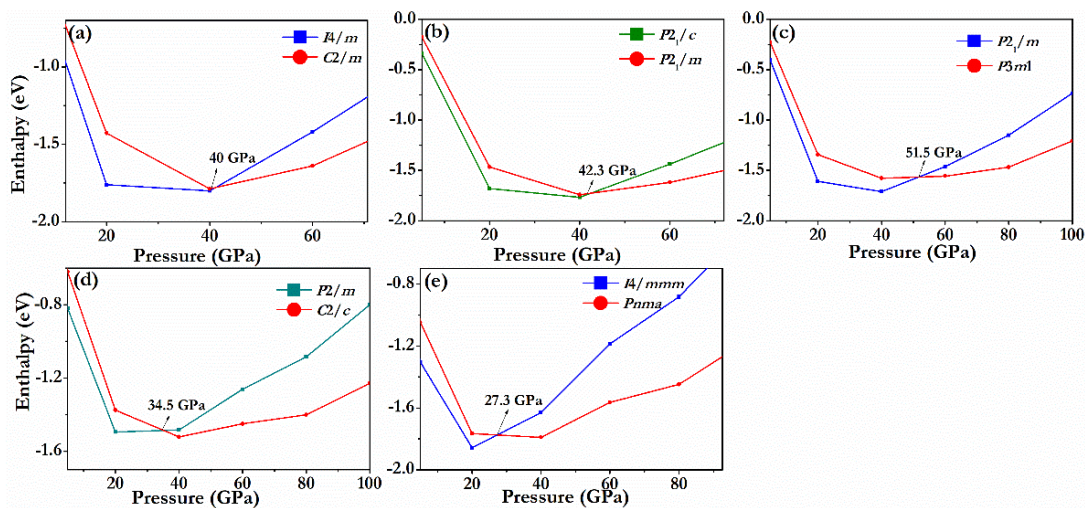**Figure S2**: Enthalpy curves for K_4Ga, K_3Ga, K_2Ga, KGa_2 and KGa_4.

Figure S3: Phonon dispersion curves for K−Ga compounds at different pressures.
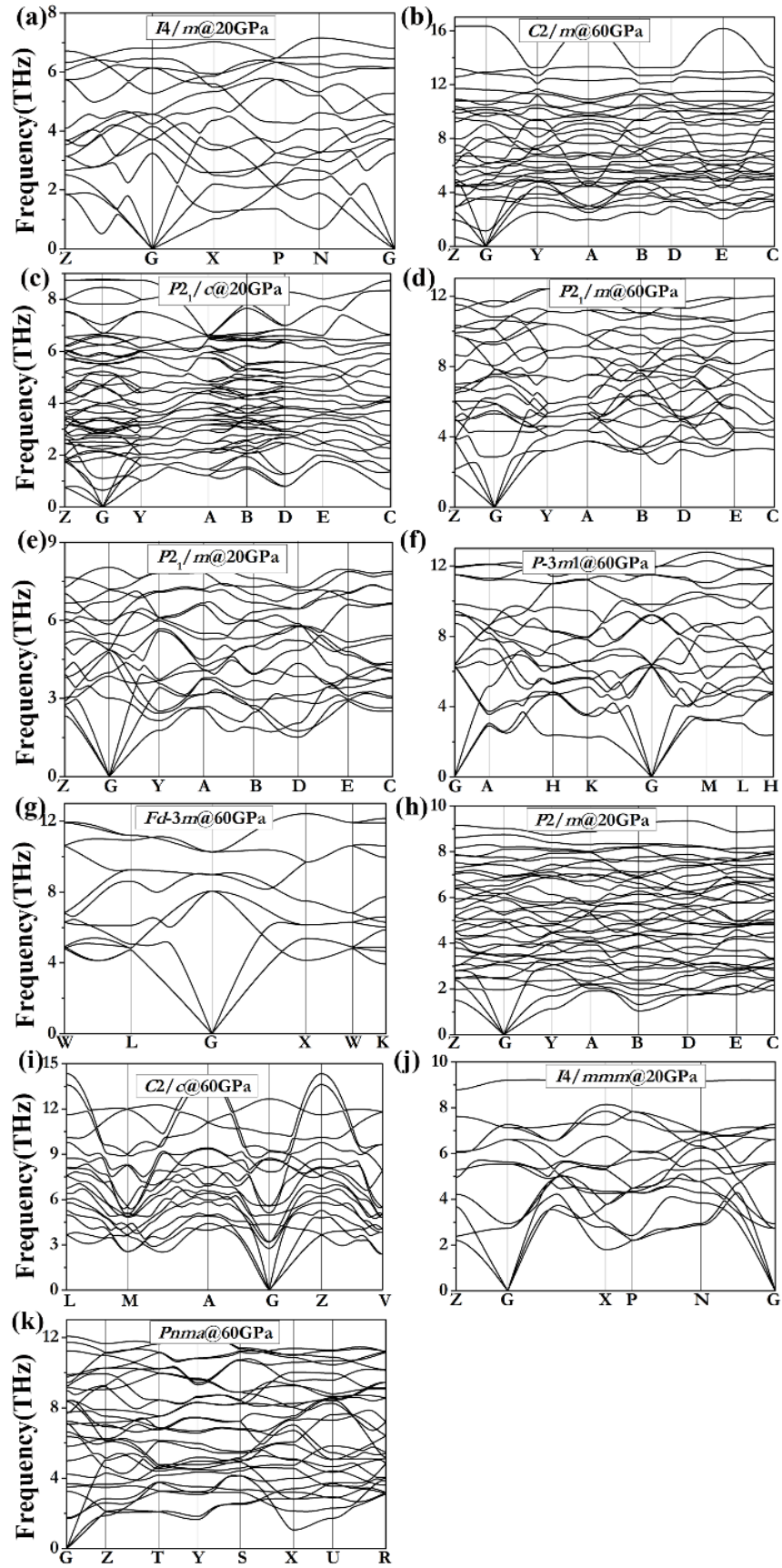
Figure S4: Calculated partial density of states (PDOS) of *Fd*-3*m*-KGa.
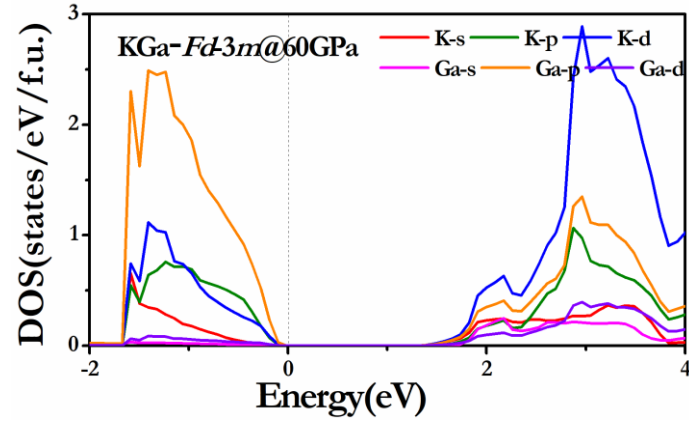

Figure S5: Calculated band structures of K-rich compounds: (a)*I*4/*m*-K$_4$Ga, (b) *C*2/*m*-K$_4$Ga, (c) *P*2$_1$/*c*-K$_3$Ga, (d) *P*2$_1$/*m*-K$_3$Ga, (e) *P*2$_1$/*m*-K$_2$Ga, (f) *P*-3*m*1-K$_2$Ga.
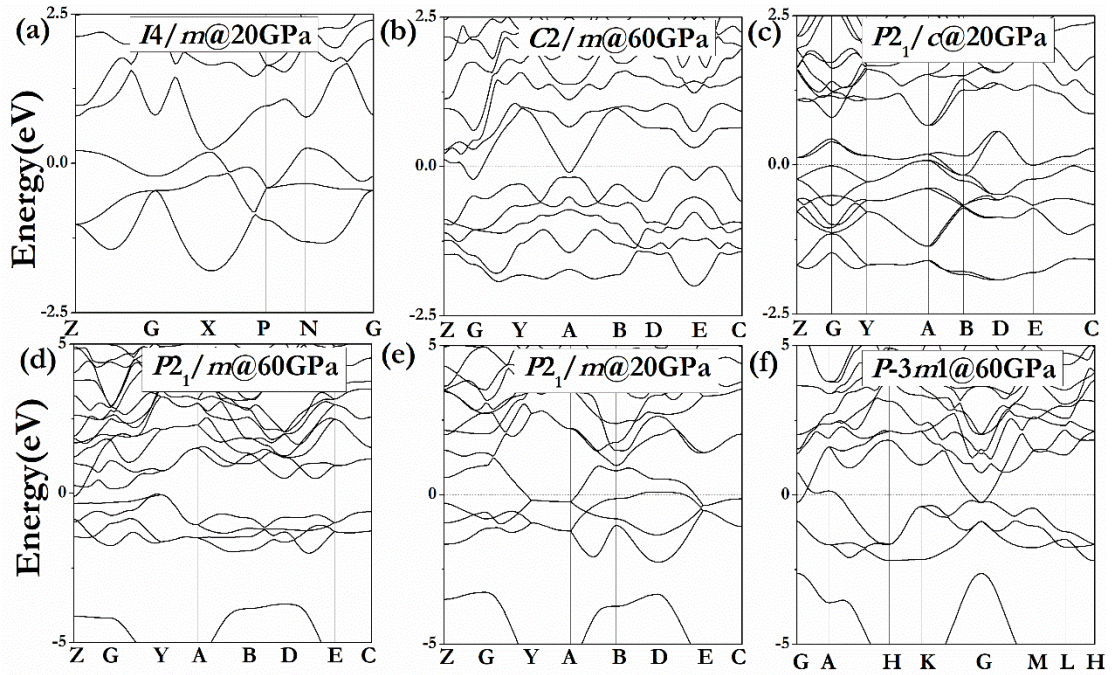
Figure S6: Calculated band structures of Ga-rich compounds: (a) *P2/m*-KGa$_2$, (b) *C2/c*-KGa$_2$, (c) *I4/mmm*-KGa$_4$, (d) *Pnma*-KGa$_4$.
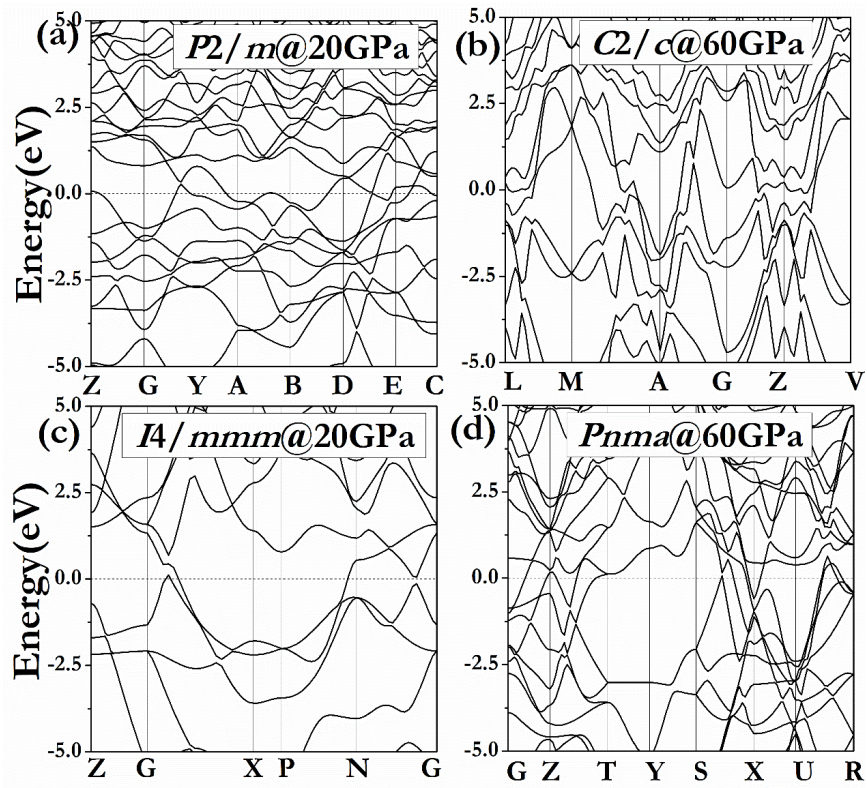

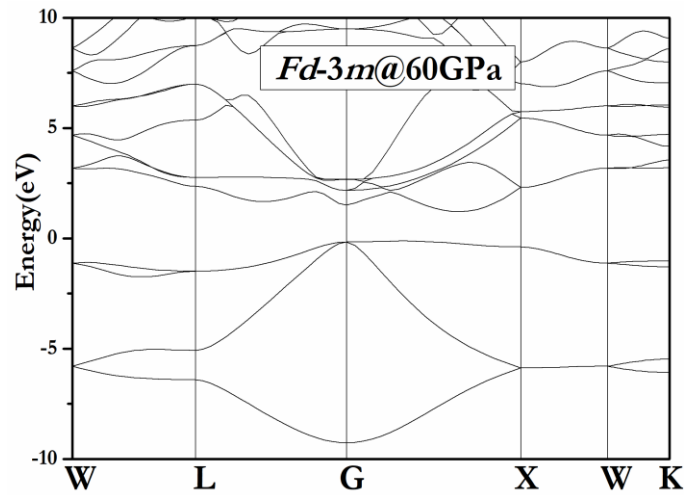**Figure S7**: Calculated band structures of *F*d-3*m*-KGa.

Figure S8: Electron localization function (ELF) of (a) $I4/m$-K_4Ga, P-$3m1$-K_2Ga and $C2/c$-KGa_2.
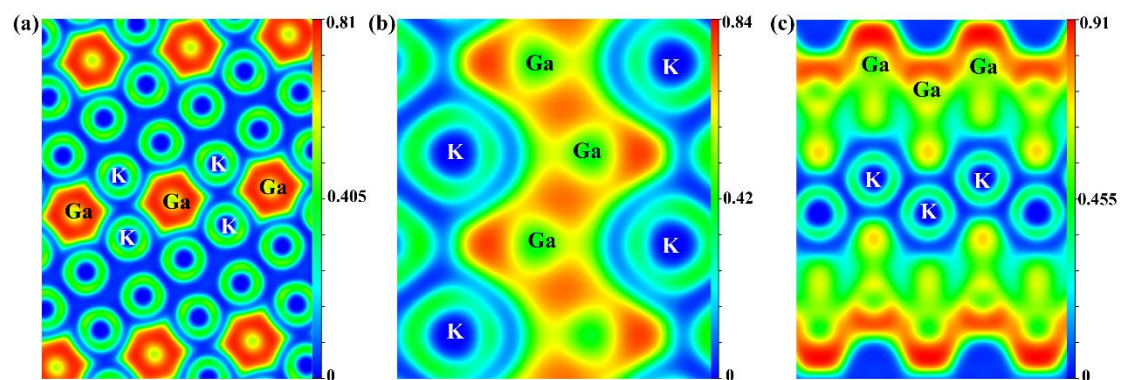

Figure S9: Electron localization function (ELF) of (a) $P2_1/c$-K_3Ga, (b) $P2_1/m$-K_3Ga and (c) Fd-$3m$-KGa.
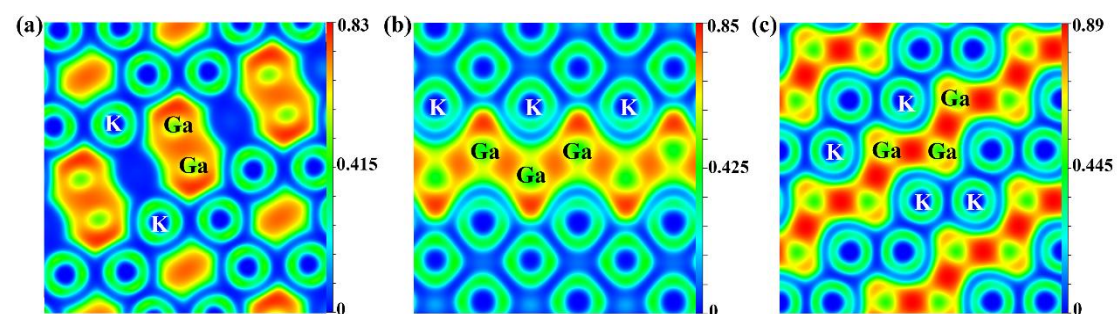
Table S1 Structural parameters of K−Ga system at selected pressures.

Space group pressure	Lattice parameters (Å, °)	Atomic coordinates (fractional)	Sites
K$_4$Ga (*I*4/*m*) 20 GPa	a=b=6.79730 α=β=γ=90	K1 0.41477 0.79496 2.00000 Ga1 0.00000 1.00000 1.00000	8h 2a
K$_4$Ga (*C*2/*m*) 60 GPa	a=9.44790 b=4.92500 c=9.10120 α=γ=90 β=133.5045	K1 0.30343 -1.00000 0.58496 K2 0.18812 -1.50000 0.12889 K3 0.00000 -1.24759 0.50000 K9 0.68093 -2.00000 0.85062 Ga1 0.44695 -1.50000 0.82018	4i 4i 4h 4i 4i
K$_3$Ga (*P*2$_1$/*c*) 20 GPa	a=5.57750 b=7.89120 c=9.87150 α=γ=90	K1 0.77864 0.25137 0.25267 K2 0.27052 0.49840 0.01945 K3 -0.13494 0.62798 -0.62484 Ga1 0.59607 0.89056 0.10810	4e 4e 4e 4e
K$_3$Ga (*P*2$_1$/*m*) 60 GPa	a=5.56840 b=4.04190 c=5.55680 α=γ=90 β=75.8530	K1 0.72871 -0.25000 -0.16915 K3 0.22588 -0.25000 -0.11132 K7 0.93540 -0.25000 0.34819 Ga1 1.55663 -0.75000 -0.39782	2e 2e 2e 2e
K$_2$Ga (*P*2$_1$/*m*) 20 GPa	a=5.64490 b=4.52120 c=5.32180 α=γ= 90 β=113.2820	K1 0.00039 0.25000 -0.30071 K5 0.68817 0.25000 0.03549 Ga1 0.38946 -0.75000 -0.59164	2e 2e 2e
K$_2$Ga (*P*-3*m*1) 100 GPa	a= b=3.98870 c=5.55120 α=β=90 γ=120	K1 2.00000 1.00000 -1.25672 K2 1.33333 0.66667 -1.41099 Ga1 1.66667 1.33333 -1.06214	2c 2d 2d
KGa (*Fd*-3*m*) 100 GPa	a=b=c=5.84330 α=β=γ=90	K1 -0.50000 0.50000 1.00000 Ga1 0.00000 0.00000 1.50000	8a 8b
KGa$_2$ (*P*2/*m*) 20 GPa	a=7.24590 b=5.48560 c=5.58290 α=γ=90 β=103.5388	K1 1.00000 1.00000 0.00000 K2 0.41054 1.00000 0.20420 K3 0.50000 0.50000 0.50000 Ga1 0.82679 0.77979 0.40204 Ga2 0.31444 0.50000 -0.05413 Ga5 1.09071 0.50000 0.23007	1a 2m 1h 4o 2n 2n
KGa$_2$ (*C*2/*c*) 60 GPa	a=2.51830 b=15.08250 c=5.07080 α=γ =90 β=121.1710	K1 2.50000 0.20451 1.25000 Ga1 2.00000 0.38684 1.25000 Ga2 2.00000 0.03435 1.25000	4e 4e 4e
KGa$_4$ (*I*4/*mmm*) 20 GPa	a= b=4.00100 c=10.69830 α=β=γ= 90	K1 2.00000 0.00000 -1.00000 Ga1 1.00000 0.00000 -0.38953 Ga9 0.50000 0.00000 -0.75000	2a 4e 4d
KGa$_4$ (*Pmma*) 60 GPa	a=4.28670 b=5.33500 c=5.90950 α=β=γ=90	K1 0.25000 0.50000 -1.12868 Ga1 0.25000 0.71158 -0.61093 Ga5 0.75000 1.00000 -0.09203 Ga9 0.25000 1.00000 -0.30676	2f 4k 2e 2e

Table S2 Bader analysis for K−Ga system at selected pressures.

Structure	Atom	Number	Charge value(e)	δ(e)
K_4Ga (*I4/m*)	Ga	1	4.8237	-1.8237
20GPa	K1	4	6.544075	+0.455925
K_4Ga (*C2/m*)	Ga	2	4.46525	-1.46525
	K1	2	6.6403	+0.3597
	K2	2	6.6013	+0.3987
60GPa	K3	2	6.6686	+0.3314
	K4	2	6.6245	+0.3755
K_3Ga (*P2₁/c*)	Ga	4	4.4502	-1.4502
	K1	4	6.5085	+0.4915
20GPa	K2	4	6.5430	+0.457
	K3	4	6.4983	+0.5017
K_3Ga (*P2₁/m*)	Ga	2	4.04005	-1.04005
	K1	2	6.6414	+0.3586
60GPa	K2	2	6.6727	+0.3273
	K3	2	6.6459	+0.3541
K_2Ga (*P2₁/m*)	Ga	2	4.05585	-1.05585
20GPa	K1	2	6.4789	+0.5211
	K2	2	6.4652	+0.5348
K_2Ga (*P-3m1*)	Ga	2	3.74885	-0.74885
100GPa	K1	2	6.63615	+0.36385
	K2	2	6.6150	+0.385
KGa (*Fd-3m*)	Ga	2	3.4521	-0.4521
100GPa	K	2	6.5479	+0.4521
	Ga1	2	3.3315	-0.3315
	Ga2	2	3.3105	-0.3105
KGa_2 (*P2/m*)	Ga3	2	3.20155	-0.20155
	Ga4	2	3.3361	-0.3361
20GPa	K1	1	6.4064	+0.5936
	K2	2	6.3880	+0.612
	K3	1	6.5483	+0.4517
KGa_2 (*C2/c*)	Ga1	2	3.3617	-0.3617
60GPa	Ga2	2	3.0718	-0.0718
	K1	2	6.5665	+0.4335
KGa_4 (*I4/mmm*)	Ga1	2	3.21405	-0.21405
20GPa	Ga2	2	3.08715	-0.08715
	K1	1	6.3975	+0.6025
	Ga1	2	3.1370	-0.1370
KGa_4 (*Pmma*)	Ga2	2	3.1592	-0.1592
	Ga3	2	3.2238	-0.2238
60GPa	Ga4	2	2.9950	+0.005
	K1	2	6.4850	+0.515