Supplementary Information

Origin of Charge Density Wave in Topological Semimetals $SrAl_4$ and $EuAl_4$

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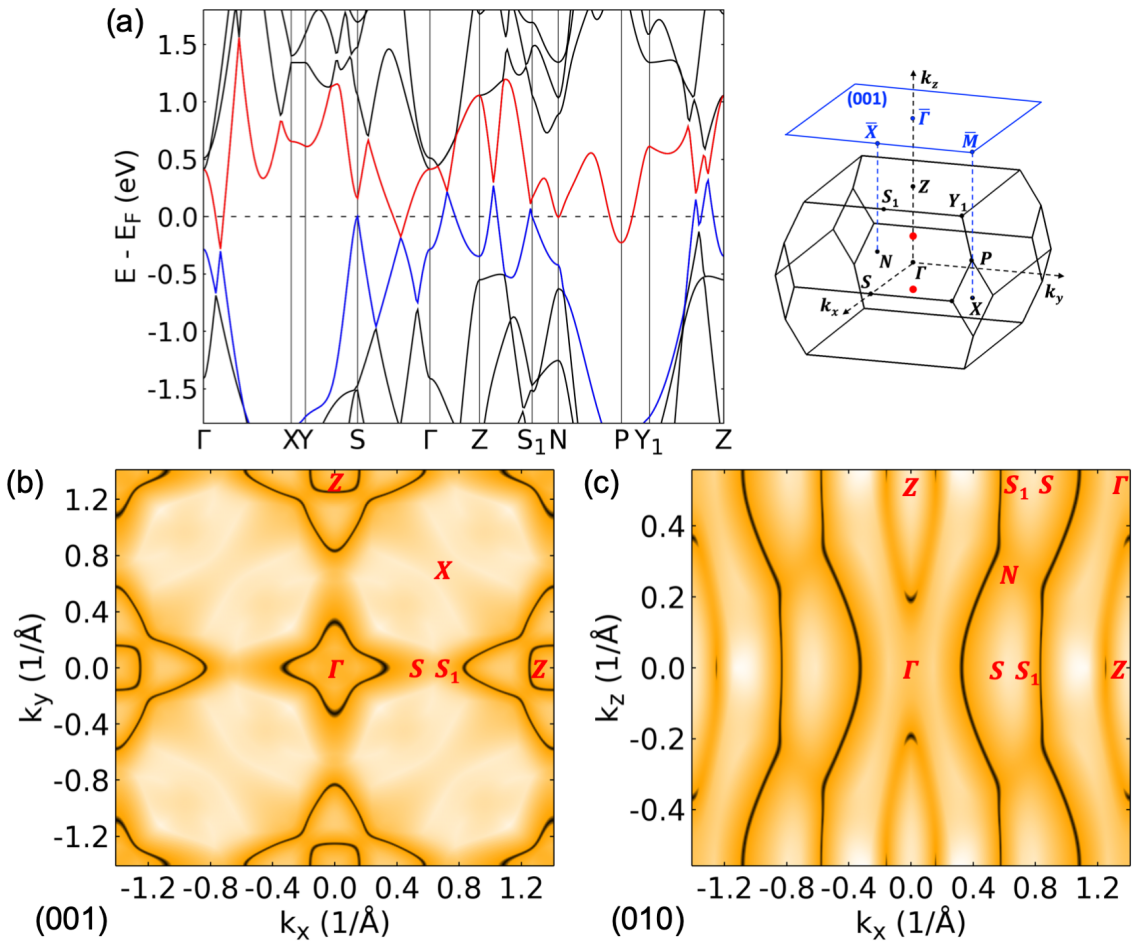
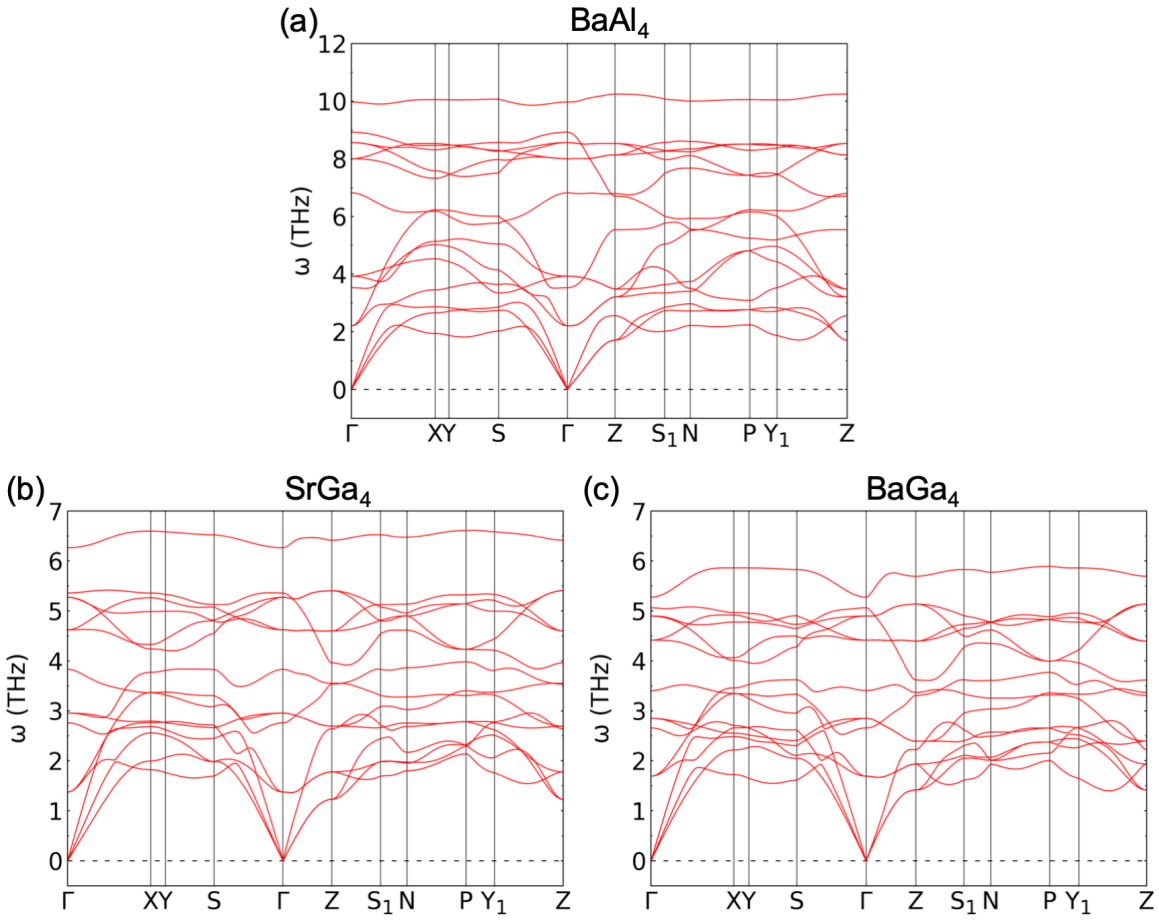
		a (Å)	*c* (Å)	*z*	4*e*-4*e* (Å)	4*d*-4*e* (Å)
$SrAl_4$	Expt[1]	4.461	11.209	0.3841	2.598	2.690
	PBE+SOC	4.456	11.250	0.3838	2.616	2.688
	δ (%)	–0.11	+0.37	–0.08	+0.69	–0.07
$BaAl_4$	Expt[2]	4.566	11.250	0.3800	2.700	2.711
	PBE+SOC	4.562	11.326	0.3812	2.691	2.722
	δ (%)	–0.09	+0.68	+0.32	–0.33	+0.41
$EuAl_4$	Expt[3]	4.402	11.163	0.380	2.679	2.636
	PBE+SOC	4.380	11.186	0.3853	2.565	2.662
	δ (%)	–0.50	+0.21	+1.39	–4.26	+0.99
$SrGa_4$	Expt[4]	4.4474	10.7300	0.38299	2.511	2.642
	PBE+SOC	4.502	10.807	0.3822	2.546	2.666
	δ (%)	+1.22	+0.72	–0.21	+1.39	+0.91
$BaGa_4$	Expt[5]	4.5661	10.7780	0.3799	2.589	2.678
	PBE+SOC	4.621	10.835	0.3788	2.627	2.699
	δ (%)	+1.20	+0.53	–0.29	+1.47	+0.78

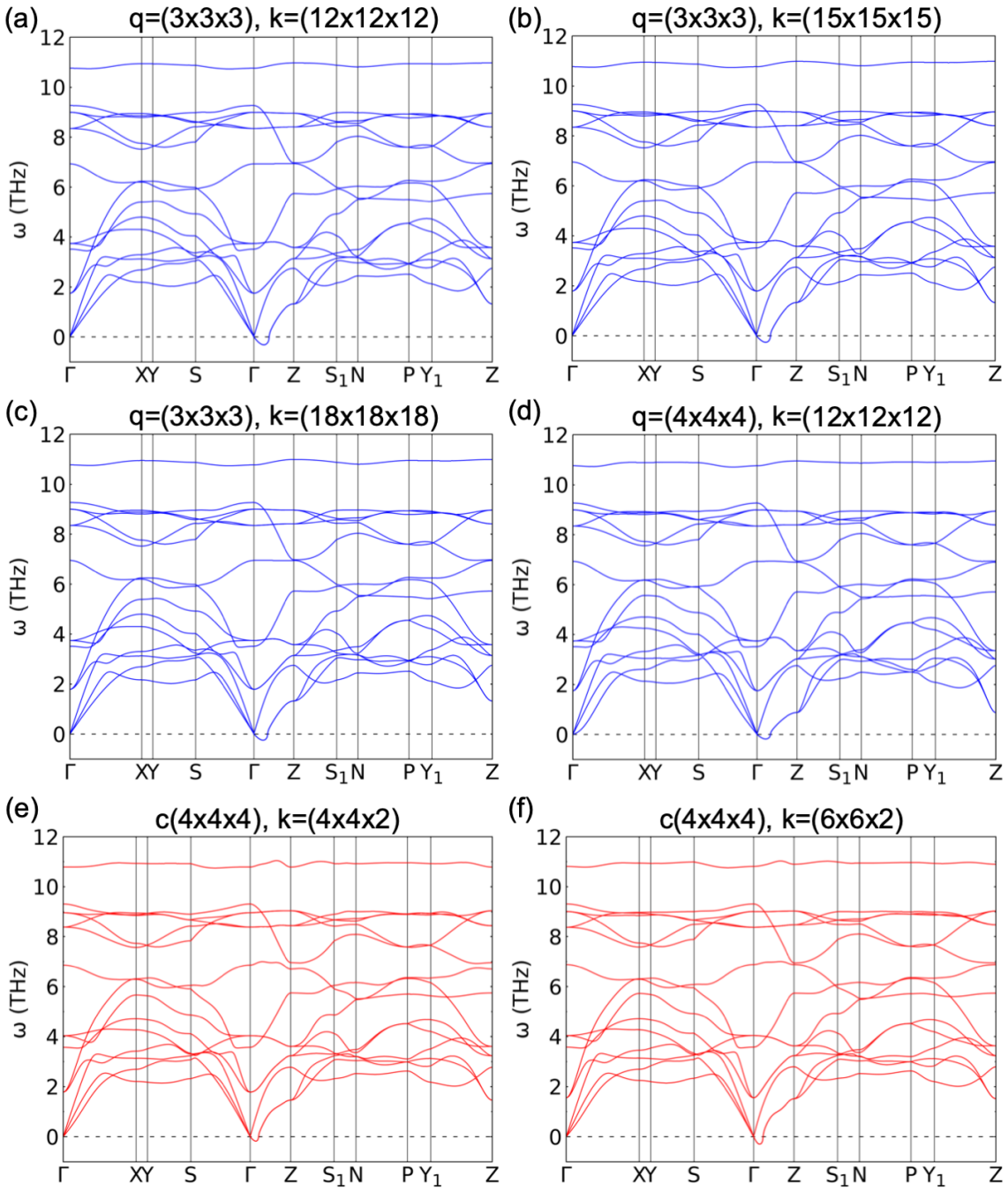
Supplementary Table 1. Fully relaxed lattice parameters and the distances between different sites (4*e*-4*e* and 4*d*-4*e*) for all the five compounds in comparison to the experimental data. The relaxations are performed in PBE exchange-correlation functional with spin-orbit coupling (SOC).



Supplementary Figure 1. Bulk band structure of $SrAl_4$ without SOC and the nodal lines with zero gap on more planes in addition to that in Fig.1(e). (a) Calculated $SrAl_4$ band structure without SOC. The top valence and conduction bands are in blue and red, respectively, with bulk Brillouin zone shown on the right. (b) Nodal lines on the (001) plane shown by the zero gap. (c) Nodal lines on the (010) plane shown by the zero gap.



Supplementary Figure 2. Phonon band dispersion of (a) $BaAl_4$, (b) $SrGa_4$ and (c) $BaGa_4$.



Supplementary Figure 3. The convergence of calculated phonon band dispersions of $SrAl_4$. (a-d) The phonon calculated in QE using DFPT with respect to increasing *k*-mesh and *q*-mesh as labeled. (e-f) The phonon calculated in VASP using the finite displacement method with increasing *k*-mesh as labeled for a large supercell of c(4x4x4) with 640 atoms. All the calculated phonon bands show the imaginary transverse acoustic mode at the small *q*-vector along the Γ-Z direction, as discussed in the main text.

Supplementary References

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