Support Information

Quasiparticle Effects on the Linear and Nonlinear Susceptibility of $ZnGeP_2$

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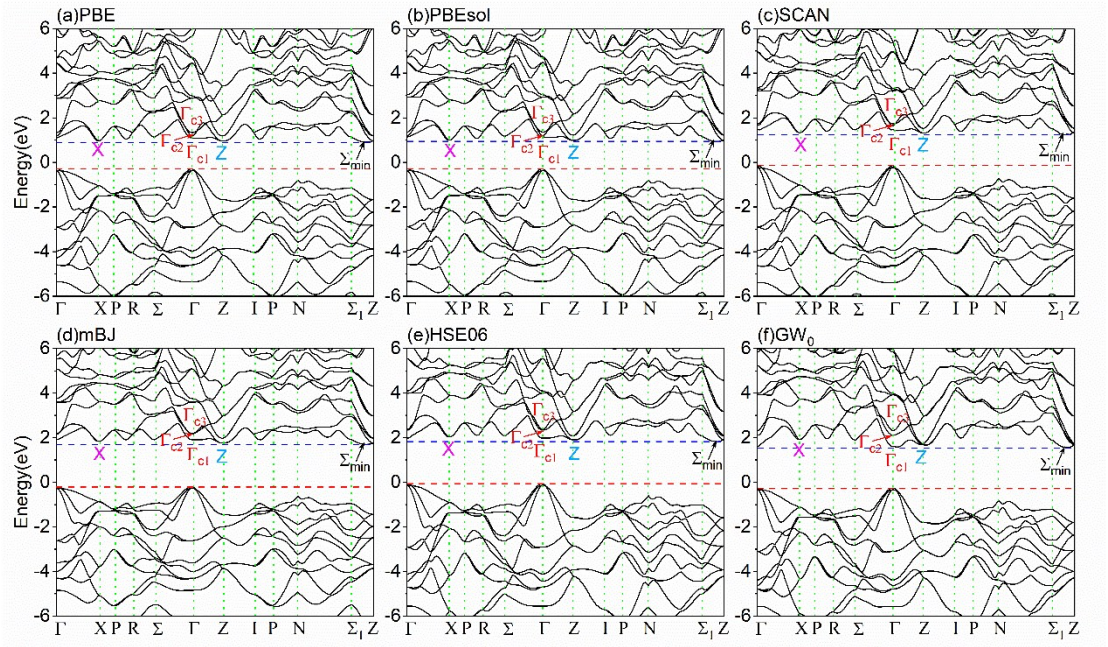

Fig. S1 Band structure in the Brillouin zone corresponding to the primitive unit cell of the body centered tetragonal phase $ZnGeP_2$ calculated with the (a) PBE, (b) PBEsol, (c) SCAN meta-GGA, (d) mBJ, (e) HSE06, (f) GW0. The zero of energy corresponds to the Fermi level.

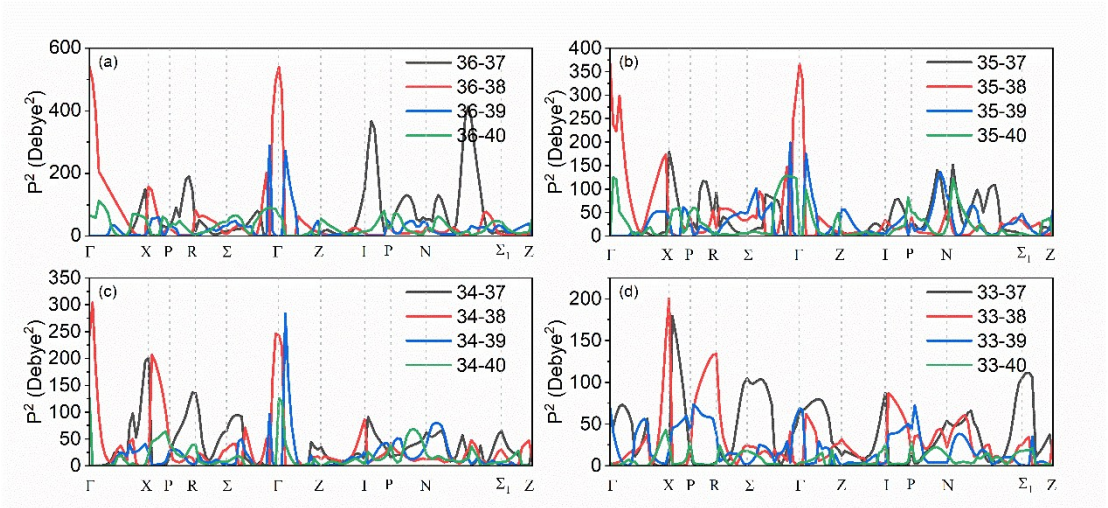

Fig. S2 Transition diploe moment between a few valence bands and conduction bands in $ZnGeP_2$. The highest occupancy valence band is 36.

Table S1 The transition dipole moment for the peaks in the imaginary part of dielectric function.

Peak	Initial band	Final band	P^2(Debye^2)
A	36	38	539.916
B	36	37	413.215
C1	35	37	178.736
C2	36	37	341.737
D1	36	39	21.047
	36	40	32.971
	35	39	34.427
	35	40	21.417
D2	36	40	28.639
	35	40	51.031
	34	40	40.602
	33	40	28.078
	36	39	38.059
	35	39	28.713
	34	39	27.693
	33	39	43.113
D3	31	37	0.549
	31	38	11.629
	32	37	176.4
	32	38	5.511
E1	36	39	0.134
	36	40	12.953
	35	39	4.532
	35	40	0.126
E2	33	41	16.115
	33	42	27.640
	34	41	26.312
	34	42	16.385

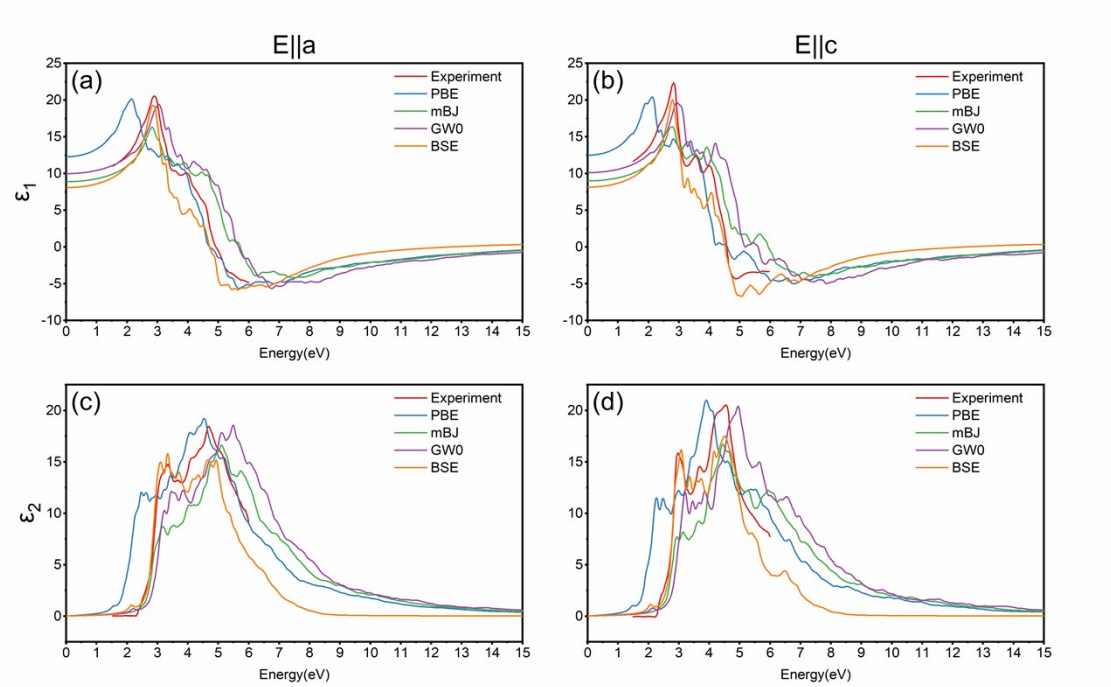

Fig. S3 Real part $\varepsilon_1(\omega)$ (top) and imaginary part $\varepsilon_2(\omega)$ (bottom) of the linear dielectric function of $ZnGeP_2$. The dielectric functions for the electric field polarization along the z-direction (E||c-axis) (right) and x-direction (E||a-axis) (left) are calculated with different methods and determined with experimentally (red line). Only direct interband optical transitions are considered in the calculation. The experimental data in the energy range 1.5-6.0 eV are taken from[1].

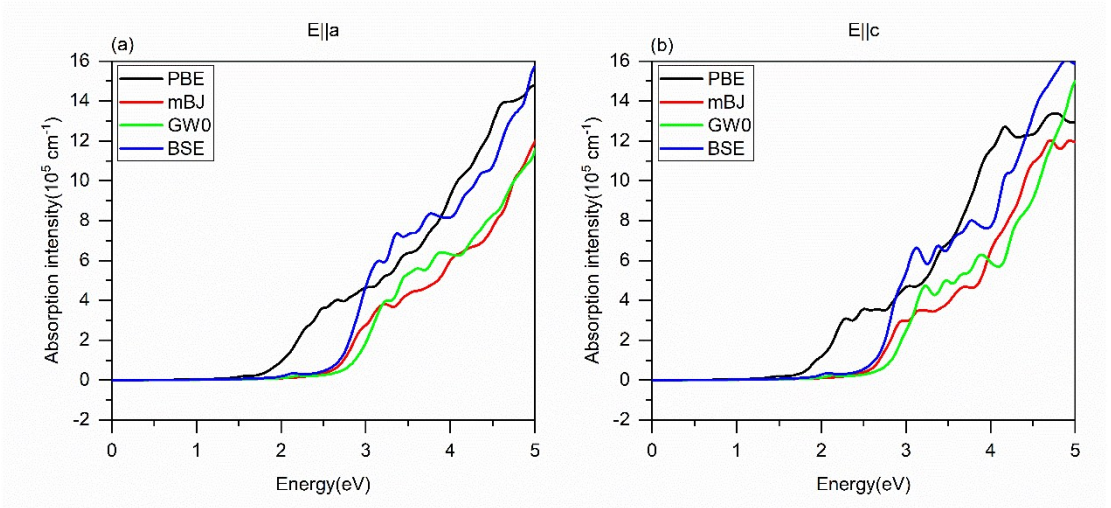

Fig. S4 The calculated $\varepsilon||a$ (ordinary) and $\varepsilon||c$ (extraordinary) optical absorption coefficients along the z-axis of $ZnGeP_2$ unit-cell as a function of phonon energy using the PBE, mBJ, GW_0 and BSE.

1. V. Blickle, K. Flock, N. Dietz and D. E. Aspnes, *Appl. Phys. Lett.*, 2002, **81**, 628-630.