Supplementary data - DFT Calculations

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1 Convergence

1.1 Cutoff Energy

All calculations were performed with the experimental unit cell, with full optimisation of the atomic positions within the symmetry constraints of the space-group. The PBE functional was used with a 7,6,5 Monkhurst-Pack grid. In the tables below ϵ_{ionic} and ϵ_{elec} are the ionic and electronic contributions respectively to the permittivity at zero frequency. Calculations of the ionic and electronic contributions to the permittivity and the frequencies were performed with the Density Functional Perturbation Theory (DFPT) module of the packages considered. No sum rules were used in the calculation of the phonon frequencies as calculated by the DFT packages. However, the values reported in the tables and the mean squared shifts in frequency were calculated using the PDielec package, with the translational phonon modes projected out.

In the tables the mean squared frequency shifts are calculated for all the optical phonon modes at the gamma point. The mean squared frequency shifts given in brackets are calculated ignoring the lowest non-zero phonon frequency.

1.1.1 Vasp

Table 1: Vasp cutoff energy behaviour

Cutoff Energy (eV)	Total Energy (eV)	Pressure (GPa)	$\epsilon_{\mathbf{ionic}}$	$\epsilon_{\mathbf{elec}}$	Mean Squared Frequency Shift (cm^{-2})
500	-67.471	2.393	2.854	2.437	2.43 (2.50)
550	-67.485	2.445	2.834	2.437	2.01 (2.07)
600	-67.510	2.307	2.835	2.436	1.29 (1.31)
650	-67.530	2.259	2.836	2.435	0.73 (0.73)
700	-67.546	2.319	2.847	2.434	0.26 (0.26)
900	-67.560	2.676	2.855	2.434	

Table 2: Abinit cutoff energy behaviour for FHI pseudo-potentials

Cutoff Energy (eV)	Total Energy (eV)	Pressure (GPa)	ϵ_{ionic}	ϵ_{elec}	Mean Squared Frequency Shift (cm^{-2})
1088.5	-4066.334	1.766	3.814	2.389	25.22 (5.29)
1360.6	-4066.402	1.584	3.181	2.389	14.44 (5.32)
1632.7	-4066.482	1.710	3.205	2.389	16.68 (2.47)
1904.8	-4066.510	1.864	3.206	2.389	5.89 (2.12)
2176.9	-4066.516	1.915	3.271	2.389	

Table 3: Abinit cutoff energy behaviour for ONCVPSP pseudo-potentials

Cutoff Energy (eV)	Total Energy (eV)	Pressure (GPa)	ϵ_{ionic}	ϵ_{elec}	Mean Squared Frequency Shift (cm^{-2})
1088.5	-4066.334	1.766	3.814	2.389	25.22 (5.29)
1360.6	-4066.402	1.584	3.181	2.389	14.44 (5.32)
1632.7	-4066.482	1.710	3.205	2.389	16.68 (2.47)
1904.8	-4066.510	1.864	3.206	2.389	5.89 (2.12)
2176.9	-4066.516	1.915	3.271	2.389	

1.1.2 Abinit

1.1.3 Castep

1.1.4 Quantum Espresso

Table 4: Castep cutoff energy behaviour

Cutoff Energy (eV)	Total Energy (eV)	Pressure (GPa)	ϵ_{ionic}	ϵ_{elec}	Mean Squared Frequency Shift (cm^{-2})
1000	-6564.008	-0.526	2.834	2.470	5.12
1300	-6564.171	1.494	2.813	2.469	3.06
1400	-6564.178	1.456	2.862	2.469	5.61
1500	-6564.186	1.470	2.792	2.469	0.00

Table 5: Quantum Espresso energy cutoff behaviour

Cutoff Energy (eV)	Total Energy (eV)	Pressure (GPa)	ϵ_{ionic}	ϵ_{elec}	Mean Squared Frequency Shift (cm^{-2})
680.3	-7510.348	2.782	2.837	2.454	0.95
816.3	-7510.423	2.424	2.854	2.454	0.46
952.4	-7510.524	2.513	2.827	2.453	0.05
1088.5	-7510.569	2.766	2.811	2.453	0.00