Supporting Information

Coarse-Grained Lattice Dynamics Calculations Combined with Independent Stiffness Approximation: A Comparative Study on Polymorphic Molecular Crystals

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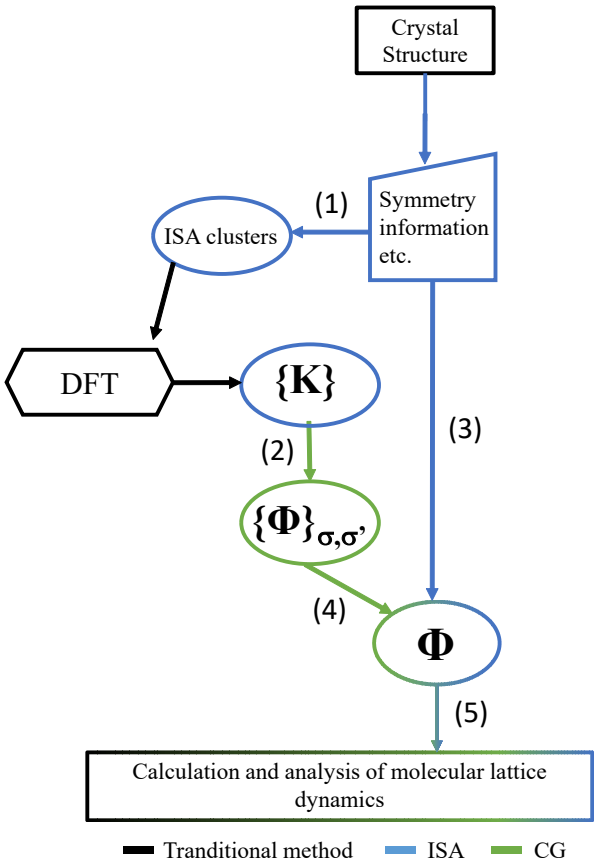

Fig. S1. Computation flow in the CG-ISA scheme. (1) Crystallographic information (unit cell and symmetry) was retrieved from the CIF file and then used to construct ISA clusters. (2) After iterative geometry optimization, the ISA clusters were submitted to force calculation (normal-mode analysis). (3) The output was processed to extract the blocks {Φ}σ,σ' of the coarse-grained stiffness matrix. (4) A set of {Φ}σ,σ' was assembled to reconstruct Φ, which was Fourier-transformed into a dynamical matrix and then diagonalized for every k-point. (5) The frequency ω(k) was plotted along the defined k-path within the first Brillouin zone (FBZ). The DFT calculations (step 2), were performed using Gaussian16w software package. For the other process (steps 1, 3, 4, and 5), we used a modeling software, a matrix calculator, and a numerical calculation package with graph-drawing functions, all of which are in-house programs developed by us (not published).

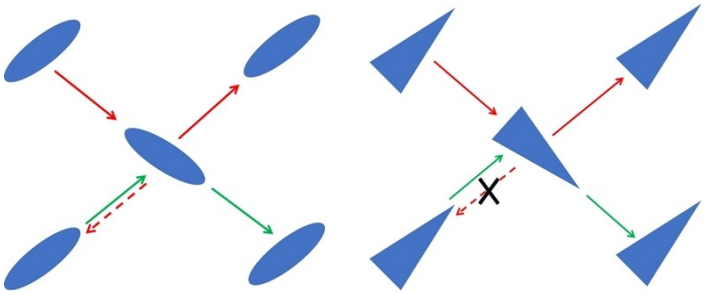

Fig. S2. Schematics for explaining the geometrical relations of molecules in a crystal with different symmetries. When the molecule possesses inversion symmetry, the interactions between the red solid line and the red dashed line are equivalent. Conversely, if the molecule lacks inversion symmetry, they are inequivalent.

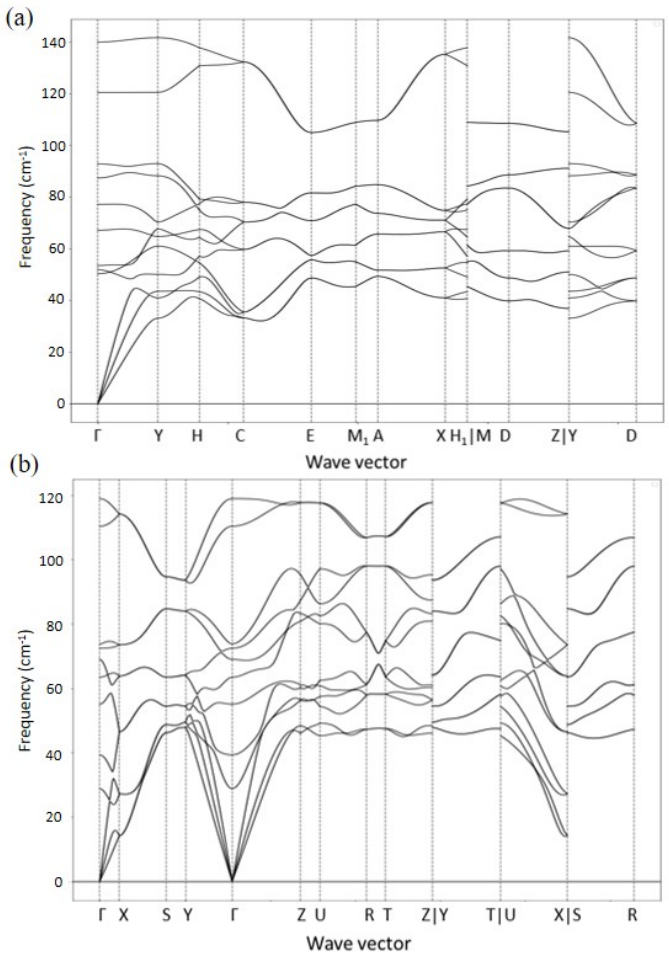

Fig. S3. Phonon band diagrams for the low-frequency region: (a) γ phase and (b) α phase. Given that the CG space dimension was 6 (α = 0) for the Z = 2 system, each diagram contains 12 ($= Z \times (6 + \alpha)$) branches.

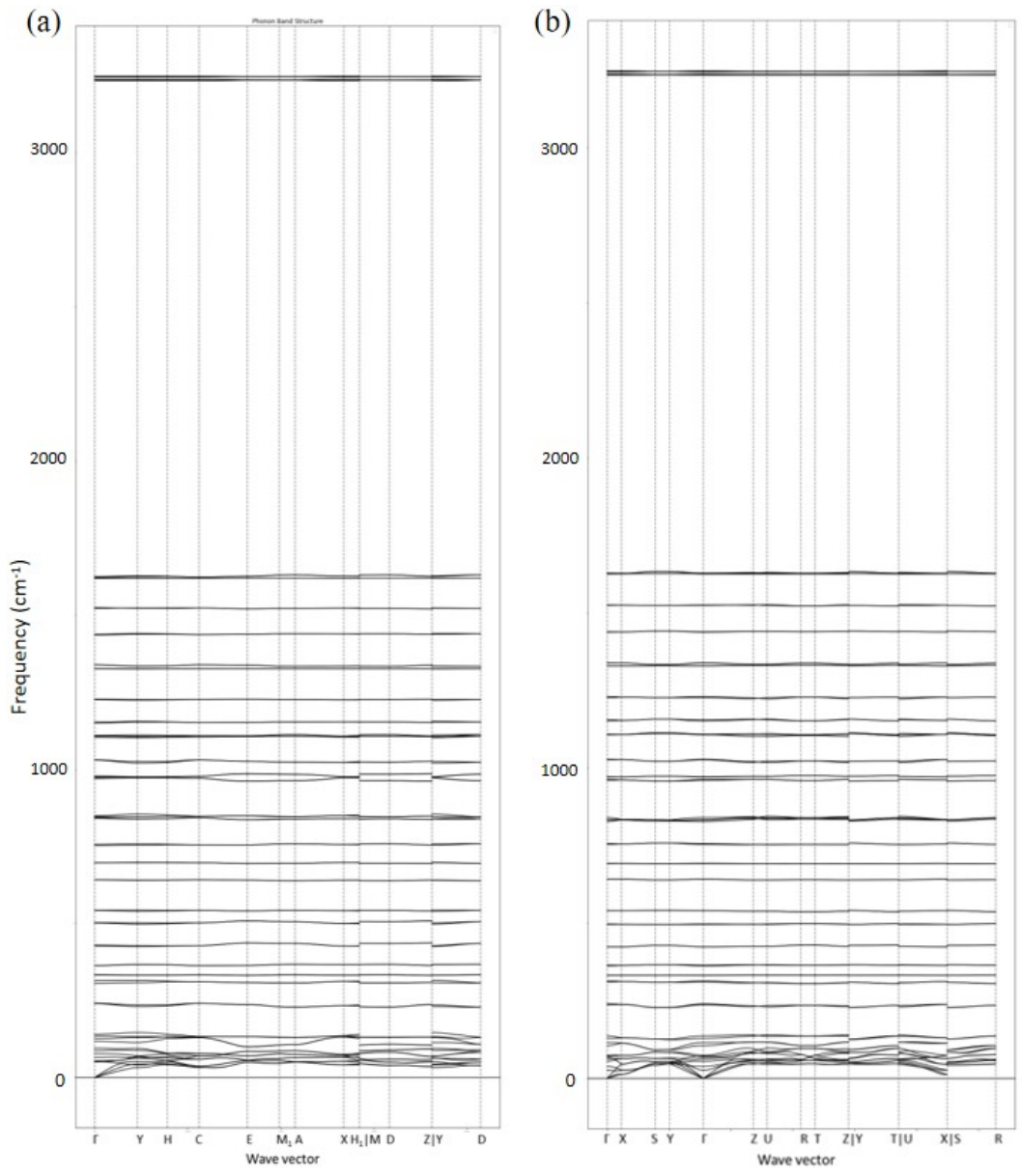

Fig. S4. Phonon band diagrams without approximation of CG: (a) γ phase and (b) α phase. Given that the CG space dimension was 36 (α = 3N – 6 = 30) for the Z = 2 system, each diagram contains 72 branches.