

Supplemental Material

“Superlattice Fermi surface nesting and the electron-phonon coupling in CaC₆”

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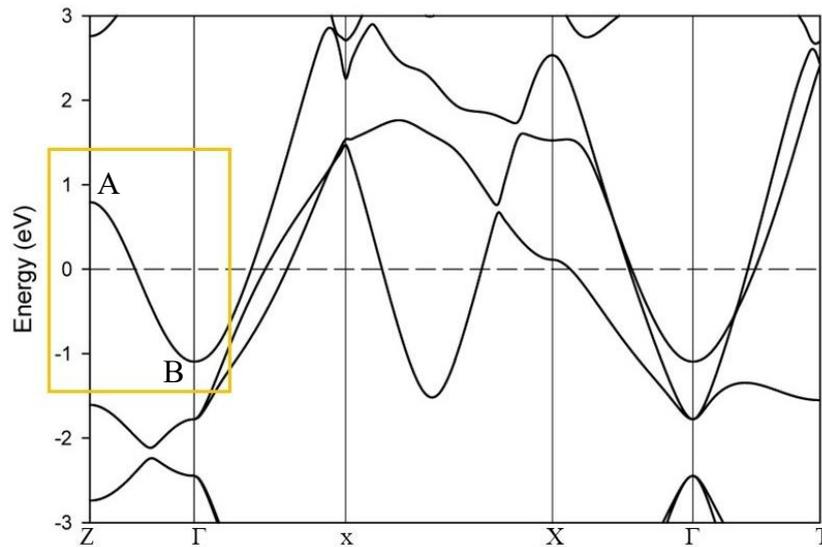


Figure S1: Calculated EBS for CaC₆ using DFT with LDA functional and $\Delta k=0.005 \text{ \AA}^{-1}$ in Space Group R-3m with rhombohedral cell dimensions ($a=b=c=5.0305 \text{ \AA}$) after original calculations by Calandra and Mauri [1]. The region along Z- Γ (yellow rectangle) shows the reciprocal space context for the cosine shaped band illustrated in detail in Figure 2a. Antibonding (A) and bonding (B) nodes are identified.

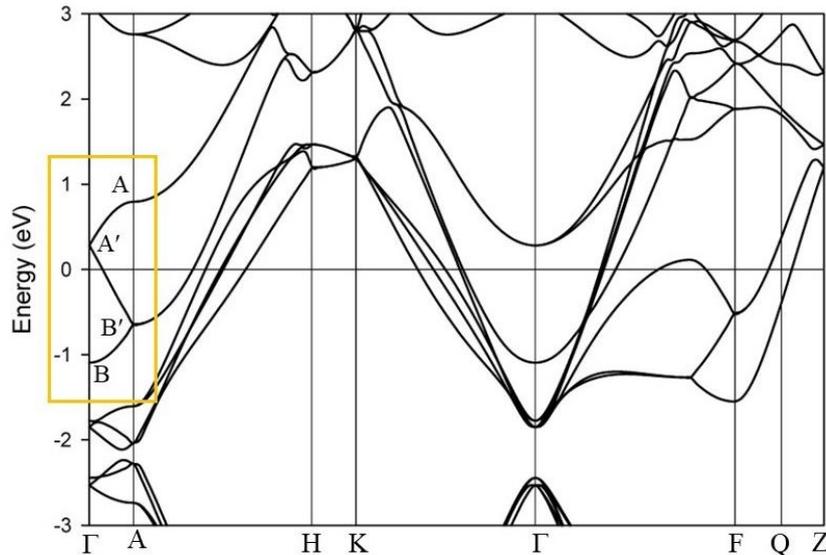


Figure S2: Calculated EBS for CaC₆ using DFT with LDA functional and $\Delta k=0.005 \text{ \AA}^{-1}$ in Space Group R-3m with hexagonal cell dimensions ($a=b=4.3054 \text{ \AA}$; $c=13.1205 \text{ \AA}$). The region along Γ -A (yellow

rectangle) shows the cosine shaped band illustrated in detail in Figure 2b. Antibonding (A, A') and bonding (B, B') nodes are identified.

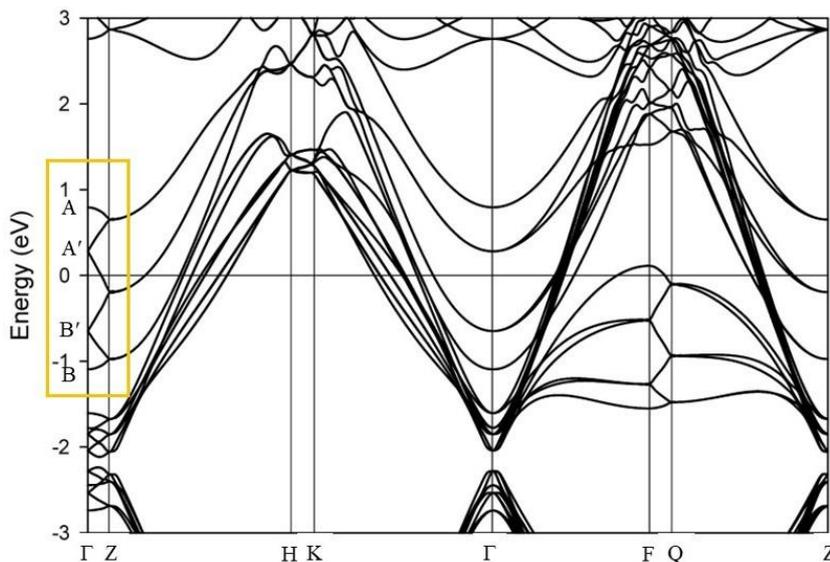
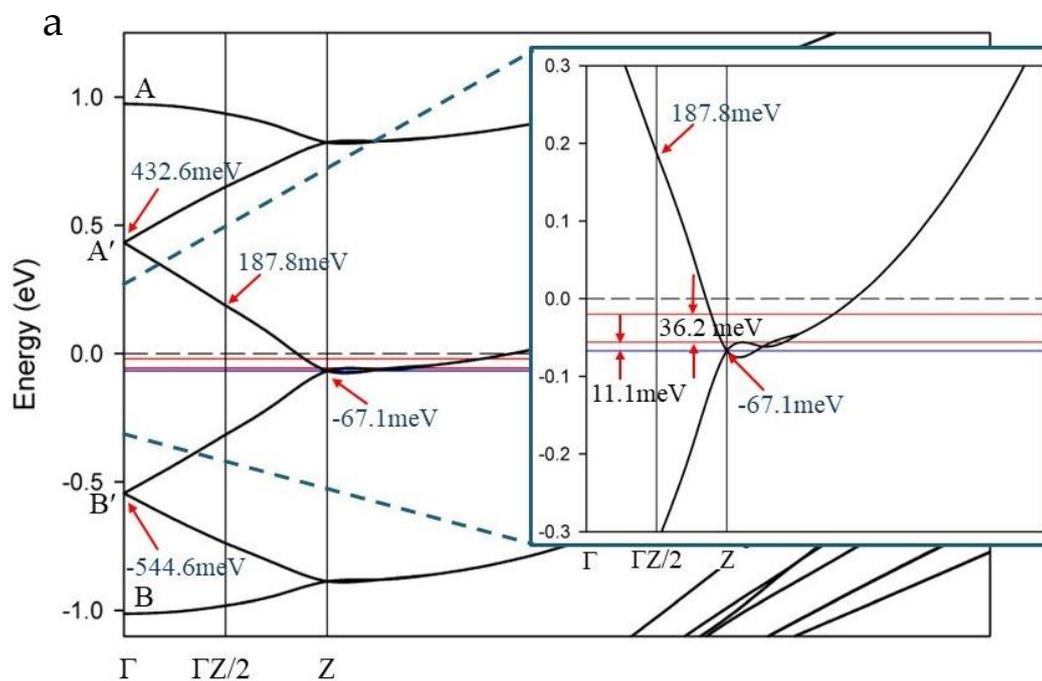


Figure S3: Calculated EBS for CaC₆ using DFT with LDA functional and $\Delta k=0.005 \text{ \AA}^{-1}$ in Space Group R-3m with hexagonal cell dimensions ($a=b= 4.3053 \text{ \AA}$; $c=26.2415 \text{ \AA}$). The region along Γ -Z (yellow rectangle) shows the cosine shaped band illustrated in detail in Figure 3a. Antibonding (A, A') and bonding (B, B') nodes along Γ are identified.



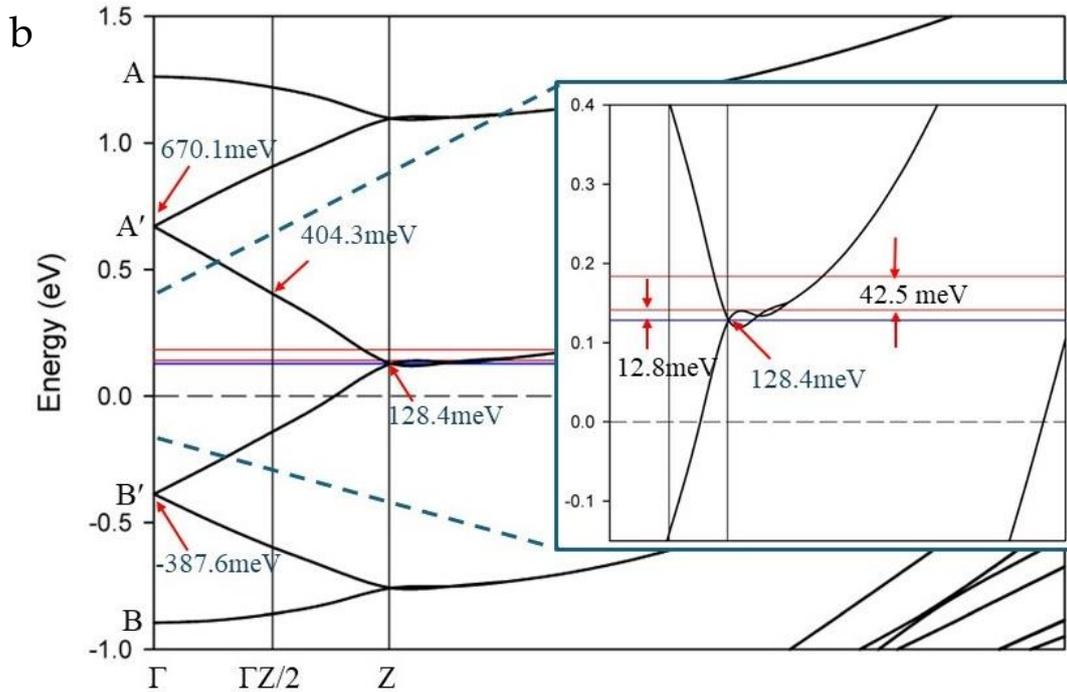


Figure S4: Electronic band structures of CaC_6 (Space Group R-3m) calculated for a $2c$ hexagonal lattice: (a) at 4 GPa and (b) at 12 GPa. In both cases, the folded cosine-shaped interlayer band along the c^* direction crosses the Fermi level (dotted horizontal line). In (a) the energy, $E_{c^*/2}$, is -67.1 meV and is at the intersection on Z where the two branches of the cosine curve from A' and B' meet. The net energy difference, ΔE , is 47.3 meV. In (b), the interlayer band along c^* is predominantly above the Fermi level and with an energy difference, ΔE , of 55.3 meV. These energy differences reflect the marginal asymmetries of the cosine band for a $2c$ superlattice at 4 GPa and 12 GPa, respectively.

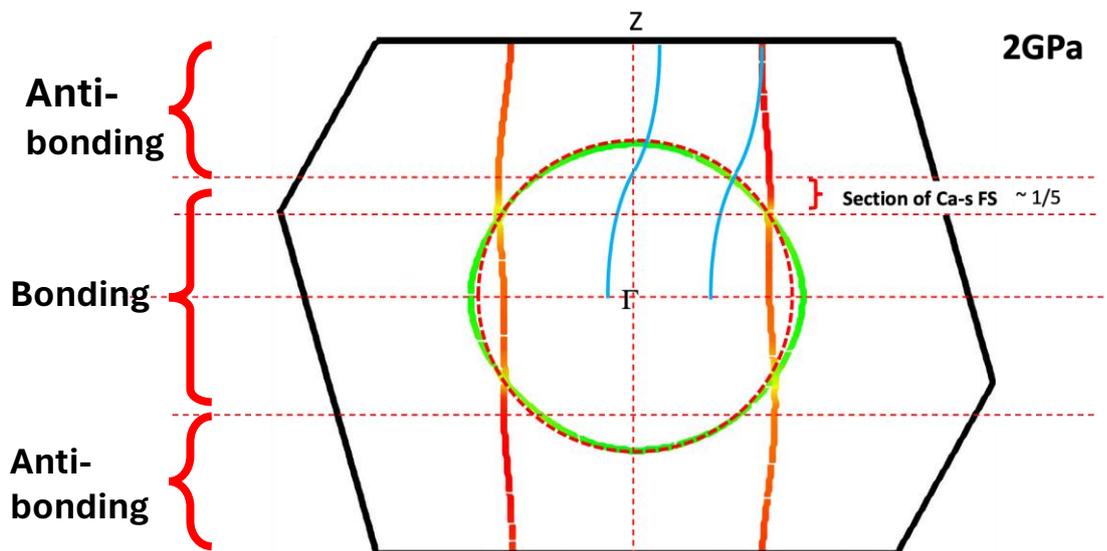


Figure S5: Schematic of the CaC_6 Fermi Surface (FS) viewed on the cross section containing the Γ -Z-T reciprocal directions for the rhombohedral unit cell (see Figure 6). The relative position of the cosine band and the FS is schematically displayed in blue. When the cosine function runs along Γ Z, the FS has pure Ca-4s orbital character. As the cosine approaches the more tubular red FS, it acquires more C- p_z orbital character.

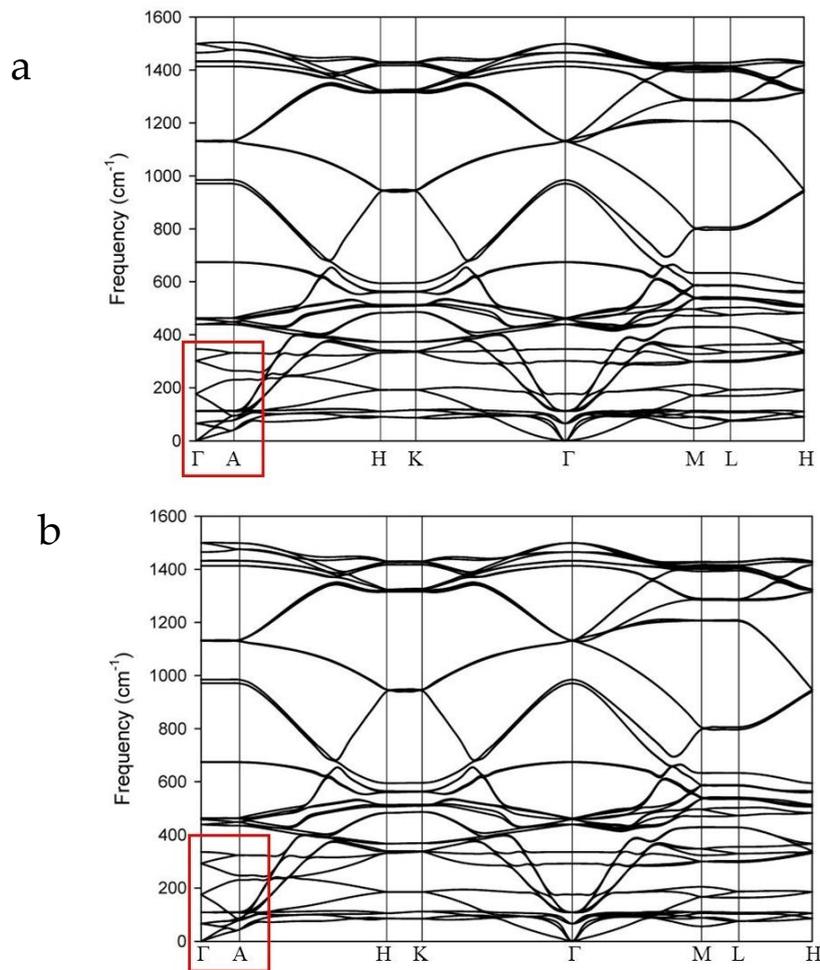


Figure S6: Calculated phonon dispersions (PDs) using DFT with LDA functional, $\Delta k=0.015 \text{ \AA}^{-1}$ and a hexagonal cell for (a) $^{40}\text{CaC}_6$ and (b) $^{44}\text{CaC}_6$. The red rectangular regions along Γ -A are shown in greater detail in Figure 9.

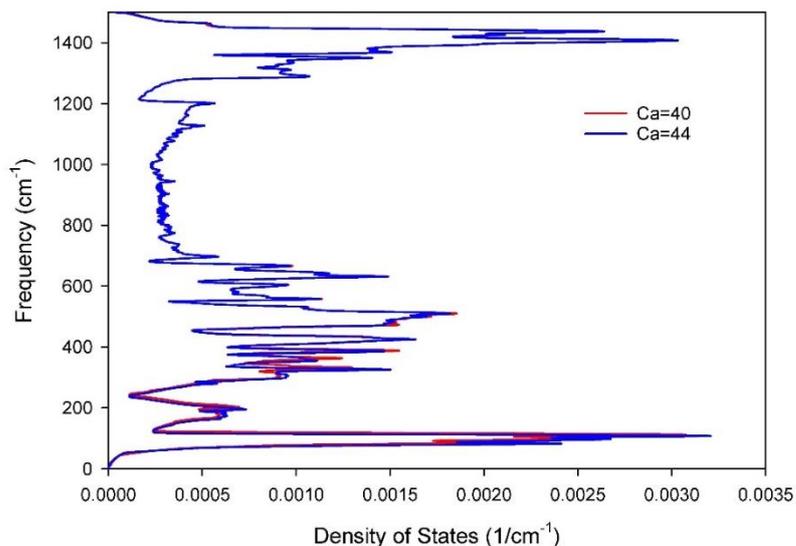


Figure S7: Calculated phonon density of states (PDOS) using DFT with LDA functional, $\Delta k=0.015 \text{ \AA}^{-1}$ and an hexagonal cell for (a) $^{40}\text{CaC}_6$ (red) and (b) $^{44}\text{CaC}_6$ (blue).

Reference:

- 1 Calandra, M., F. Mauri, Theoretical Explanation of Superconductivity in CaC_6 , *Physical Review Letters* 95 (2005) 237002.