Strongly hybridized phonons and spontaneous electric polarizations in low-dimensional graphitic multilayers

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The rich interlayer couplings in graphitic multilayers and versatile controlling knobs to tune their physical properties have led to significant advancements in condensed matter physics in recent years. In this talk, I will show the spectroscopic discovery of uncharted phonon modes in one commensurate and three incommensurate DWNT crystals [1], three of which concurrently exhibit strongly reconstructed electronic band structures. Our density functional theory (DFT) calculations for the experimentally observed commensurate DWNT (7,7) @ (12,12) reveal that this new phonon mode originates from a (nearly) degenerate coupling between two transverse acoustic modes (ZA modes) of constituent inner and outer nanotubes having approximately trigonal and pentagonal rotational symmetry along the nanotube circumferences. Such coupling strongly hybridizes the two phonon modes in different layers and leads to the formation of a unique lattice motion featuring evenly distributed vibrational amplitudes over inner and outer nanotubes, distinct from any known phonon modes in 1D systems.

I will also share our direct optical nanoscopy imaging of spontaneous electric polarizations and polarization switching in tetralayer graphene [2]. We visualize opposite out-of-plane electric polarizations and its switching in adjacent polar stacking orders of tetralayer graphene that lack inversion and mirror symmetries by their own but are mutually transformable by the two symmetries with the nanometer scale resolution. This observation, in conjunction with our DFT calculations, further consolidates our assignment of the polar stacking orders in tetralayer graphene and rationalizes their formation and distribution we observed in other multilayer graphene samples. We also demonstrate a reversable polarization switching between two polar stacking orders in tetralayer graphene by an atomic force microscopy (AFM) tip manipulation.

References

[1] S. Sun et al., Strongly hybridized phonons in one-dimensional van der Waals crystals. Physical Review Letters (2025) (in press) arXiv:2408.08596
[2] Z. Zhou, X. Peng et al., Manuscript to be submitted (2025)

Figures

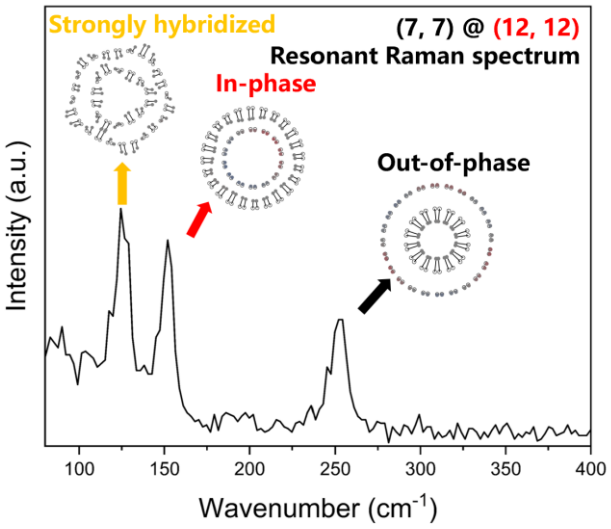

Figure 1: Strongly hybridized phonons observed in an commensurate double-walled carbon nanotubes (DWNTs) with chirality (7,7)@(12,12).