Optical Phonon Anomaly in Bilayer Graphene with Ultrahigh Carrier Densities

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Introduction

Electron-phonon coupling (EPC) is an important effect in monolayer graphene (MLG). Interesting phenomena such as the renormalization of the phonon energy, the Kohn anomalies, and the breakdown of the adiabatic (Born-Oppenheimer) approximation have been reported. AB-stacked bilayer graphene (BLG) is a unique platform where both the band structure and the doping level (i.e., the Fermi level E_F) can be tuned through the applied electrical gates, allowing for the control of a delicate interplay between electrons, phonons, and photons.

Previous investigations of BLG mainly focused on situations with charge carriers near the charge neutrality Dirac point E_D (i.e., $|E_F-E_D|<0.4$ eV). Recent progress in fabricating electrolytic and ionic-liquid gates provides the possibilities of doping MLG and BLG with ultrahigh charge-carrier densities of $|n| > 10^{14}$ cm^{-2} and of tuning the Fermi level close to the van Hove singularity (VHS) point at M in the first Brillouin zone (BZ). Such a high carrier density will embark interesting technological applications including supercapacitors, transparent electrodes, and high performance organic thin film transistors. In this regard, understanding the carrier dynamics will be a crucial step for the potential electronic device applications. Distinct many-body effects and superconducting instability have been observed in doped graphene when the Fermi energy approaches the VHS point. Since superconductivity also occurs in graphene-related systems such as graphite-intercalation compounds (GICs), the study of EPC in BLG might shed new light on the underlying mechanism of superconductivity in GICs.

In this work, we show that the band structure of BLG at high doping levels plays a critical role in the EPC of the two long-wavelength high-energy symmetric (S) and antisymmetric (AS) optical modes in the high carrier density regime.

Methods

- Quantum Espresso code, with plane wave cutoff 70 Ry
- Norm-conserving pseudopotential for carbon
- Frozen-phonon method to calculate the EPC matrix elements

$$g^{\nu}_{mn}(\mathbf{k},\mathbf{q}) = \sqrt{\frac{\hbar}{2M\omega^{\nu}_{\mathbf{q}}}}\langle m\ \mathbf{k}+\mathbf{q}|\frac{\delta V_{scf}}{\delta u^{\nu}_{\mathbf{q}}}|n\ \mathbf{k}\rangle$$

- Phonon self-energy:

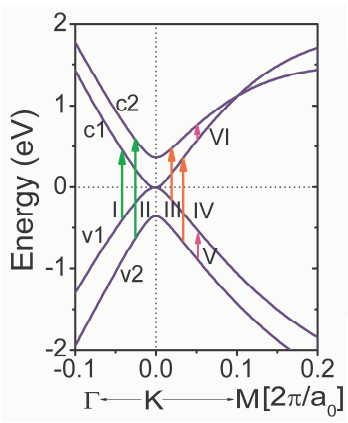
$$\Pi_{\mathbf{q}\nu}(\omega) = 2\sum_{mn}\int \frac{d\mathbf{k}}{\Omega_{BZ}}|g^{\nu}_{mn}(\mathbf{k},\mathbf{q})|^2\frac{[f(\epsilon_{n\mathbf{k}+\mathbf{q}})-f(\epsilon_{m\mathbf{k}})][\epsilon_{n\mathbf{k}+\mathbf{q}}-\epsilon_{m\mathbf{k}}]}{(\epsilon_{n\mathbf{k}+\mathbf{q}}-\epsilon_{m\mathbf{k}})^2-(\hbar\omega+i\eta)^2}$$

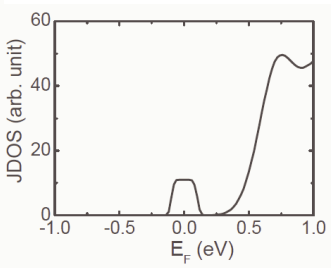
The phonon linewidth γ and frequency shift Δω are determined by

$$\gamma = -2\mathrm{Im}(\Pi_{\mathbf{q}\nu}(\omega_0)) \qquad \Delta\omega = \frac{1}{\hbar}[\mathrm{Re}(\Pi_{\mathbf{q}\nu}(\omega_0)|_{E_F} - \Pi_{\mathbf{q}\nu}(\omega_0)|_{E_F=0}]$$

Results

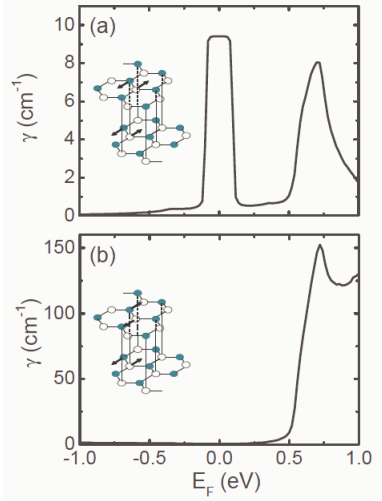
Band dispersions of BLG



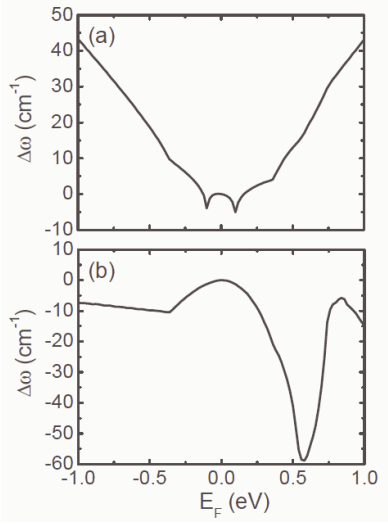


$$N(E_F,\omega_0) = \frac{4\pi}{N_k}\sum_{\mathbf{kj}'}[f(\epsilon_{\mathbf{kj}})-f(\epsilon_{\mathbf{kj}'})]\delta[\epsilon_{\mathbf{kj}}-\epsilon_{\mathbf{kj}'}+\hbar\omega_0]$$

Phonon Linewidth as a Function of Doping



Phonon Frequency Shift as a Function of Doping



Summary

- The phonon linewidths and frequency shifts for the long-wavelength high-energy optical modes in bilayer graphene exhibit a distinct dependence on the electron and hole doping due the intriguing interplay between the unique band structure and the phonon modes in this system.
- In particular, we predict that the linewidth for the antisymmetric mode could be significantly enhanced when the Fermi level is tuned to be 0.5 eV above the neutrality point.

Bibliography

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