Spectroscopic Signatures of Electronic Excitations in Raman Scattering in Thin Films of Rhombohedral Graphite

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ABSTRACT: Rhombohedral graphite features peculiar electronic properties, including persistence of low-energy surface bands of a topological nature. Here, we study the contribution of electron–hole excitations toward inelastic light scattering in thin films of rhombohedral graphite. We show that, in contrast to the featureless electron–hole contribution toward Raman spectrum of graphitic films with Bernal stacking, the inelastic light scattering accompanied by electron–hole excitations in crystals with rhombohedral stacking produces distinct features in the Raman signal which can be used both to identify the stacking and to determine the number of layers in the film.

KEYWORDS: Rhombohedral graphite, graphene, Raman spectroscopy, electronic Raman scattering, optical absorption, stacking-dependent properties

After seven decades of intense studies,1–3 graphite still surprises us by the richness of physical phenomena and optoelectronic effects it can host. The agility of graphite is largely due to the van der Waals (vdW) nature of its interlayer bonding, coexisting with a substantial hybridization between the electronic states in the consecutive layers. On the one hand, a weak vdw bonding between honeycomb graphene layers in graphite allows for the formation of various stacking configurations: naturally appearing Bernal, rhombohedral and turbostratic graphites,4,5 or designer-twisted graphene bilayers.6 On the other hand, the interlayer hybridization of carbon $P_z$ orbitals with a characteristic energy $\sim 0.3$–0.4 eV, combined with a peculiar Dirac-like bands of electrons near the Fermi level in graphene,7–9 makes electronic properties of various graphitic structures distinctively different. As a result, over the years many extreme regimes of quantum transport and correlations were found in ultrathin graphitic films10–17 and artificially fabricated structures18–22.

Rhombohedral is a structural phase of graphite which has a specific “ABC” stacking of consecutive honeycomb layers of carbon atoms, such that every atom has a nearest neighbor from an adjacent layer either directly above or underneath it with which their $P_z$ orbitals hybridize with a coupling $\gamma \approx 0.39$ meV.23 This distinguishes it from Bernal’s “ABA” stacking, where half the carbons find the closest neighbors in the consecutive layer, whereas the other half appears to be between the empty centers of the honeycombs in the layers above and below. This difference between the crystalline structures of rhombohedral and Bernal graphite is depicted in Figure 1a, together with the intra- and interlayer hopping couplings between carbon $P_z$ orbitals, marked according to the Slonczewski–Weiss–McClure (SWMcC) tight-binding model parametrization.24,25 The difference in the lattice structures determines the difference between electron band structures3,26,27 of these two phases of graphite, illustrated in Figure 1b for their 15-layer-thick films. The most pronounced difference in these spectra is related to the existence of low-energy subbands (0’ and 0’’), confined to the top/bottom surfaces of a film of ABC graphite,26,27 which are almost flat over the momentum range, $\sim p_z = \gamma_1 /v$ around $K$ and $K’$ valleys ($v \approx 10^6$ cm/s), and the Dirac velocity of electrons in graphene. These low-energy surface bands, already established using angle-resolved photoemission spectroscopy,28–30 raise expectations for the formation of strongly correlated (magnetic17 or superconducting31) states, reigniting the interest in ABC graphitic films.32–36

Raman spectroscopy is currently one of the methods of choice used for the identification of structural properties of atomically thin films of van der Waals materials,37–39 providing information about the number of layers in the film, strain and doping. Usually, such Raman spectroscopy detects phonon
excitations in the lattice, which also stands for the ABC graphitic films6,40,41 but with a lesser clarity of interpreting the data as compared to Bernal graphite.42 At the same time, it has been demonstrated that Raman scattering enables one to detect the presence of stacking faults in thin films of rhombohedral graphite. The peculiar dispersion of electrons in thin films of rhombohedral graphite produces peaks in their Raman response at energies

\[ \alpha_n \approx 4\xi \sin \left( \frac{n + \frac{1}{2}) \pi}{2N + 1} \right), \quad 1 \leq n \leq \left\lfloor \frac{N}{2} \right\rfloor \]

where \( \lfloor x \rfloor \) is the greatest integer less than or equal to \( x \). These features, which are related to van Hove singularities in the vicinity of subband edges in the thin film spectrum, can be used for the identification of the number of layers, \( N \), in the ABC graphitic films. In addition, we find that Raman can detect the presence of stacking faults in thin films of rhombohedral graphite.

To model optical properties of graphitic films, we use a brute-force diagonalization of a hybrid “k-p” tight-binding model (HkpTB) in which the intralayer hopping of electrons between carbon atoms is taken into account in a continuous description of sublattice Bloch states using k-p theory near the

**K and K’ valleys**, combined with interlayer hopping introduced in the spirit of a tight binding model. The Hamiltonian, written on the basis \( \{ \phi_{A12}, \phi_{B12}, \phi_{A22}, ..., \phi_{BNN}, \phi_{NNN} \} \) of Bloch states \( \phi_{ij} \) and constructed of \( P_j \) orbitals on \( A \) and \( B \) sublattice of honeycomb lattice of the \( j \)th graphene layer, reads

\[
\hat{H} = \hat{H}_0 + \hat{H}_T
\]

\[
\hat{H}_0 = v_N \bigotimes \begin{pmatrix} 0 & \hat{\kappa}^\dagger \\ \hat{\kappa} & 0 \end{pmatrix}, \quad \hat{\kappa} = \xi \hat{p} + ip_j
\]

\[
\hat{H}_T = \begin{pmatrix} 0 & \hat{T}_{2,1} & \cdots & \hat{T}_{2,3} & \cdots & \hat{T}_{N-1,1} \\ \hat{T}_{1,2} & 0 & \cdots & 0 & \cdots & \hat{T}_{N-1,N} \\ \vdots & \vdots & \ddots & \vdots & \cdots & \vdots \\ \hat{T}_{N-1,2} & \cdots & \cdots & 0 & \cdots & \hat{T}_{N-1,N} \\ 0 & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & 0 & \cdots & 0 & 0 & 0 \end{pmatrix}
\]

Here, \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) is the vector of Pauli matrices, \( \hat{I}_N \) is the \( N \times N \) identity matrix, \( \xi = \pm 1 \) denotes the two inequivalent valleys \( K = \left( \frac{4\pi}{3a}, 0 \right) \) and \( K' = \left( -\frac{4\pi}{3a}, 0 \right) \) \( (a = 2.46 \text{ Å is the graphene lattice constant}) \), and \( p = (p_x, p_y) \) is the electron momentum in the valency. The \( 2 \times 2 \) matrices \( T_{\sigma+1} \) describe the

**Figure 2.** Spectral density of electronic Raman scattering \( g_\sigma(\omega) \) (for excitation with photons with \( \Omega = 2 \text{ eV} \)) and absorption coefficient \( g_\sigma(\omega) \) of 10-layer rhombohedral (ABC, left panel) and Bernal (ABA, right panel) graphite. For absorption, we assumed level broadening of 3 meV and plot the spectrum from \( \omega > 50 \text{ meV} \). The spectral density of Raman reflects the joint density of states of electrons and holes. It features distinct peaks that correspond to van Hove singularities in the subbands preceded by steps that arise from the subband edge. In the models that ignore trigonal warping, those two features would overlay but for finite \( \gamma_1 \) and \( \gamma_2 \), the subband van Hove singularities in conduction/valence lie above/below the corresponding subband edges.
electron hopping between consecutive layers. For the rhombohedral stacking

$$\hat{T}_{j,j+1} = \hat{T} \equiv \begin{pmatrix} -\nu_4\kappa' & \nu_4\kappa \\ \gamma_4 & -\nu_4\kappa \end{pmatrix}$$

$$\nu_4(\pm) = \frac{\sqrt{3}}{2\hbar}\gamma_4(\pm) \gamma_4 \approx 0.26 \text{ eV} \text{ and } \gamma_4 \approx 0.2 \text{ eV}, \text{ whereas for Bernal stacking}$$

$$\hat{T}_{j,j+1} = \begin{cases} \hat{T} & \text{for odd } j \\ \hat{T}^+ & \text{for even } j \end{cases}$$

The absorption of incident light, arriving perpendicular to the film and characterized by vector potential $A_0 = E_0/\omega$ with in-plane polarization $I = (I_x, I_y)$, by a thin graphitic film (undoped and with the thickness less than attenuation length) is described by absorption coefficient:

$$g_\omega(\omega) \approx \frac{2\alpha}{n_0} \sum_{n',m' \neq n} \int \frac{(p, m')^+ |\partial \hat{A}_z / \partial p| (p, n')^-)^2}{\omega - \epsilon_{p,n'} - \epsilon_{p,m'} + i0}$$

Here, $|p, n'\rangle$ are states (with momentum $p$) in the $n'$th subband on the conduction/valence ($s = \pm$) band side at energy $\epsilon_{p,n}$ and $\alpha = \frac{\hbar^2}{4\pi e^2 \hbar c} = \frac{1}{137}$ is the fine structure constant.

By inspection of the matrix structure of the operator $\frac{\partial \hat{A}_z}{\partial p}$ and the eigenstates of the Hamiltonian in eq 2, we find that (similarly to bilayer graphene) the dominant valence-conduction band transition is such that $0^- (n^-) \rightarrow n^+(0^+)$ and $n^- \rightarrow (n + 1)^+$ and $n^- \rightarrow (n - 1)^-$, resulting in distinct features at $\omega_n \approx \gamma_1 \sin \left(\frac{\pi + \pi n}{2N + 1}\right)$ and $\omega_n' \approx 4\gamma_1 \sin \left(\frac{n + 1 + \pi n}{2N + 1}\right)$, respectively, with $0 \leq n \leq N$ (for $N \gg 1$), marked in Figure 2. In ref 50, some IR absorption features have been observed in rhombohedral graphite flakes identified as with 4, 5, and 6 layers that were interpreted as the $0^- (1^-) \rightarrow 1^+ (0^+)$ sequence. Whereas for small $N$, formulas for $\omega_n$ and $\omega_n'$ overestimate the peak positions, computing absorption spectra numerically would reproduce the measured spectra once we take $\gamma_1 = 0.32$ eV, which is less than the values 0.38–0.4 eV typically quoted for bilayer graphene and in Slonczewski–Weiss–McClure Bernal graphite.$^{23}$

In inelastic scattering, a photon with energy $\Omega$, arriving to the sample at normal incidence, scatters to a photon with energy $\Omega' = \Omega - \omega$, leaving behind an electron–hole excitation with energy $\omega$. Specifically for graphite, the amplitude of this process is dominated by the sum of two amplitudes presented in the form of Feynman diagrams in the inset in Figure 2a, which would cancel each other (due to the opposite sign, $\Omega$ and $-\Omega'$, of the energy mismatch in the intermediate state) for nonrelativistic electron in a simple metal with a parabolic dispersion, but for Dirac electrons generate an amplitude:

$$R \approx \frac{1}{2\epsilon_\omega \Omega} (l \times I') \hat{l}_z \otimes \sigma_z$$

(4)

The latter expression means that the main contribution to Raman comes from $n^- \rightarrow n^+$ intersubband transitions and indicates that in the measurements the inelastic light scattering would not be linearly polarized in the direction perpendicular to the linear polarization of the incoming photon. Then, spectral density of Raman scattering

Figure 3. Raman signature of rhombohedral graphite. (a) Spectral density, $g_R(\omega)$, of electron excitations in Raman scattering of photons with $\Omega = 2$ eV as a function of film thickness. (b) THz absorption, $g_\omega(\omega)$, for the same films. The consecutive curves are shifted up by $8 \times 10^{-10}$ eV cm$^{-1}$ in (a) and by $2\hbar^2/4\pi e^2 \hbar c$ in (b). (c) Electronic Raman scattering on 50-layer-thick rhombohedral (blue) and Bernal (green) graphite films.

Figure 4. Rhombohedral graphitic film with a stacking fault. Spectral density of Raman, $g_R(\omega)$, and infrared absorption coefficient, $g_\omega(\omega)$, of a 20-layer thick rhombohedral film with a fault at layers 10/11.
in a film with the Fermi level at the edge between the $n = 0^\pm$ subbands is

$$g_\alpha(\omega) = \frac{1}{\pi} \int \frac{dq}{(2\pi\hbar)^2} \delta(\Omega - \epsilon_q) = \frac{\Omega^2}{(2\pi\hbar)^2} \epsilon_w(\omega)$$

$$w(\omega) = \frac{2}{\pi\hbar} \sum_{n=\pm 1} \int dp(p, m^+\pi^+p, n) |\tilde{f}|^2 \delta(\epsilon_{p,n} - \epsilon_{p,n} - \omega)$$

(5)

with an overall quantum efficiency $I = \int d\omega g_\alpha(\omega) \sim 10^{-10}$, which was proven to be in the measurable range by the earlier studies of graphene.\textsuperscript{43,47,51}

Figure 2 exemplifies the calculated Raman scattering and THz/IR absorption spectra in films of rhombohedral and Bernal graphite. Although their spectra are essentially featureless for Bernal stacking, those for ABC stacking show a series of peaks related to the excitations of electrons between van Hove singularities in (i) the nth valence band to the n-th conduction band transition for Raman and (ii) the n-th valence band to the (n ± 1)th conduction band transition for absorption. Figure 3 shows the Raman and THz absorption spectra for the films of rhombohedral graphite with various thicknesses, where the positions of Raman peaks coincide with the values described by eq 1. A Raman feature, interpreted as an intersubband excitation in graphite \textsuperscript{1} layers. Similarly, an $\epsilon_{p,n}$ can be identified by means of cross-polarization measurement) can be used to distinguish graphic films with ABC stacking from Bernal graphite, and even to determine their thickness. For films thicker than those described in Figure 3a, the attenuation of the photon field inside the film, described by absorption coefficient $g(\Omega) \approx \alpha \pi \approx 2\%$, would require to take into account the inhomogeneity of the excitation field profile and the outgoing photon field distribution in the calculation of the matrix elements of the Raman process. For the experiments where the Raman signal would be detected in the transmission geometry, this would simply lead to the damping of the overall Raman spectrum by the factor of $\exp(-0.046N)$, whereas for the detection of Raman signal in the reflected light the ABC film spectrum would additionally change, as shown in Figure 3c for a 50-layer thick film, losing the distinguishing features of the rhombohedral stacking. Additionally, we note that disorder and a finite scattering rate $\tau^{-1}$ for electrons would broaden the spectral features in Raman to a $\hbar \tau^{-1}$ line width.

Finally, in anticipation of possible stacking faults in rhombohedral graphite we computed Raman and absorption spectra of ABC films with an ABA stacking fault in the middle of it. Introduction of a Bernal stacking between the jth and (j + 1)th layers of a film is taken into account by a change of one of the hopping matrices, $T_{ij}\rightarrow T^{'}_{ij}$ in eq 2 from $T$ to $T^{'}$. The resulting spectra, illustrated in Figure 4 for $N = 20$ with a fault between the 10th and 11th layers show that the film appears in Raman as overlaying rhombohedral crystals of thicknesses $j$ and $(N - j - 1)$ layers. Similarly, an $M$-layer “ABC insert” in a thin film of Bernal graphite would produce features in the overall Raman spectrum of the film with the van Hove singularities peaks for the $M$-layer film of rhombohedral phase superimposed over the featureless background of Bernal graphite spectrum.

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**Notes**

The authors declare no competing financial interest.

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