1. Gate-dependences of 2D-mode linewidth, 2D-mode frequency, and of G-mode linewidth.

**Fig. S1:** (a) Gate dependence of 2D-mode linewidth. The linewidth is largely constant at low doping, and starts to increase when $2|E_F|$ becomes larger than 0.7 eV. (b) Center frequency of 2D-Raman shift. It reaches a maximum at a $2|E_F|$ value that coincides with the point at which 2D-mode linewidth starts to change. (c) G-mode linewidth shows a
maximum at low doping then the G-phonon can decay into electron-hole pairs. At higher
doping, the G-mode linewidth stays at a constant value.

2. Theoretical prediction on G-mode intensity across a large doping range.

Fig. S2: Theoretical prediction on G-mode intensity with $2|E_F|$ varying from 0-3V.

G-mode Raman intensity decays slowly even when the resonant channels are completely
blocked, indicating a significant non-resonant Raman contribution from higher-energy
states.
3. Detailed theoretical evaluation of the 2D-mode resonance factor.

In this section, we show that the resonance factor $R_k$ for the dominant 2D-mode Raman scattering processes in graphene has the same phase independent of the intermediate state energies. As discussed in the main manuscript, the resonance factor for 2D-mode Raman scattering processes is given by

$$R_k = \frac{1}{(E_{ex} - E_k - i\gamma)(E_{ex} - h\Omega_D - E_{k'} - i\gamma)(E_{ex} - 2h\Omega_D - E_{k''} - i\gamma)}$$  \hspace{1cm} (SEq. 1)

First, since $E_{k'}$ and $E_{k''}$ are the energies of the second and third intermediate states where one and two charge carriers have been scattered by zone-boundary phonons, respectively, the intermediate state energies satisfy the relation

$$E_k + E_{k'} = 2E_{k''}.$$  \hspace{1cm} (SEq. 2)

as illustrated in Fig. S1.

![Figure S3. Schematic diagram showing the 2D Raman scattering processes. $E_k$, $E_{k'}$, and $E_{k''}$ are the energies of the first, second, and third intermediate states, respectively.](image)
state (after photo-excitation) has an electron-hole pair having Bloch wavevector \( \mathbf{k} \) near the Dirac point \( \mathbf{K} \). Next, either the electron or the hole in that pair is scattered into a state with Bloch wavevector \( \mathbf{k}' \) near the other Dirac point \( \mathbf{K}' \) \((\mathbf{k}' - \mathbf{k} = \mathbf{q})\). If we denote the angle that \( \mathbf{k} - \mathbf{K} \) and \( \mathbf{k}' - \mathbf{K}' \) form with the \(+k_x\) direction by \( \theta_{\mathbf{k} - \mathbf{K}} \) and \( \theta_{\mathbf{k}' - \mathbf{K}'} \), respectively, the squared electron-phonon coupling strength between these two electronic states through the highest-energy zone-boundary phonon is proportional to \( \sin^2 \left( \frac{\theta_{\mathbf{k} - \mathbf{K}} - \theta_{\mathbf{k}' - \mathbf{K}'}}{2} \right) \). Therefore, the dominant scattering occurs when \( \theta_{\mathbf{k} - \mathbf{K}} - \theta_{\mathbf{k}' - \mathbf{K}'} = \pi \), i.e., when \( \mathbf{k}' - \mathbf{K}' \) is antiparallel to \( \mathbf{k} - \mathbf{K} \). This constraint, because of the linear energy dispersion relation in graphene, is equivalent to the condition

\[
E_k + E_{k'} = 2E_0 .
\] (SEq. 3)

where \( E_0 \) is a constant that is uniquely determined by the phonon wavevector \( \mathbf{q} \).

Eliminating \( E_{k'} \) and \( E_{k'} \) in SEq. 1 using SEq. 2 and SEq. 3, we obtain

\[
R_k = \frac{1}{(E_{ex} - E_k - i\gamma)(E_{ex} - \hbar\Omega_D - E_0 - i\gamma)(E_{ex} - 2\hbar\Omega_D - 2E_0 + E_k - i\gamma)} .
\] (SEq. 4)

The dominant contribution to the integrated Raman intensity comes from processes where the second term in the denominator is of minimal magnitude, i.e.

\[
E_0 = E_{ex} - \hbar\Omega_D .
\] (SEq. 5)

Supplementary Eq. 5 is in fact a condition on the phonon wavevector \( \mathbf{q} \) because \( E_0 \) is determined by \( \mathbf{q} \). Now substituting SEq. 5 into SEq. 4, we obtain the final result of this section

\[
R_k \propto \frac{1}{(E_{ex} - E_k)^2 + \gamma^2} .
\] (SEq. 6)
Supplementary Eq. 6 demonstrates that the quantum mechanical amplitude of the dominant 2D-mode Raman scattering process has always the same phase regardless of the first intermediate state energy $E_k$ (see Fig. 3f of the main manuscript). Therefore, if part of the intermediate pathways is blocked through an electrical gating, the 2D-mode Raman intensity decreases as discussed in the main manuscript. Finally, although the discussion in this section is based on some simplifying assumptions, the findings are in good agreement with those obtained from full consideration of the matrix elements. The theory curves in Fig. 3 of the main manuscript are obtained by correctly considering all the matrix elements.