Many-Body Effects in Suspended Graphene Probed through Magneto-Phonon Resonances

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Micro-magneto-Raman scattering spectroscopy is made use of to probe magneto-phonon resonances (MPR) in suspended mono- to pentalayer graphene. MPR correspond to avoided crossings between zone-center optical phonons (G-mode) and optically active inter-Landau level (LL) transitions and provide a tool to perform LL spectroscopy at a fixed energy ($\approx$197 meV) set by the G-mode phonon. Using a single-particle effective bilayer model, the velocity parameter associated with each MPR is readily extracted. A single velocity parameter slightly above the bulk graphite value suffices to fit all MPR for $N \geq 2$ layer systems. In contrast, in monolayer graphene, it is found that the velocity parameter increases significantly from $(1.23 \pm 0.01) \times 10^6$ m s$^{-1}$ up to $(1.45 \pm 0.02) \times 10^6$ m s$^{-1}$ as the first to third optically active inter-LL transitions couple to the G-mode phonon. This result is understood as a signature of enhanced many-body effects in unscreened graphene.

Pristine suspended monolayer graphene is a well-defined, unscreened 2D electronic system, in which a wealth of intriguing electronic, optical, and mechanical properties have been uncovered. In particular, speculative deviations from a simple one-electron picture of graphene’s band structure (i.e., the Dirac cones) emerge at low carrier densities (below a few $10^{11}$ cm$^{-2}$). Due to electron–electron interactions, the velocity parameter diverges logarithmically as the Fermi energy approaches the Dirac point, reaching values, well above the Fermi velocities of bulk graphite and supported graphene ($=1 \times 10^6$ m s$^{-1}$).

In the presence of a transverse magnetic field $B$, the electronic states of graphene merge into discrete, highly degenerate Landau levels (LL). The energy and lifetime of LL can be probed using magneto-transport measurements, scanning tunnelling spectroscopy, and magneto-optical spectroscopies. Recently, micro-magneto-Raman spectroscopy (MMRS) has been used to probe the electronic dispersion of suspended graphene layers and also to demonstrate that electronic excitations between LL may be strongly affected by many-body effects. First, as $B$ decreases, the energy of a given LL also decreases and electron–electron interactions lead to a logarithmic divergence of the corresponding velocity parameter.

Second, inter-LL excitations lead to the formation of magneto-excitons, whose binding energies depend on the index $n$ of the electron and hole LL they arise from, leading to $n$-dependent $v_F$.

Thus far, Raman signatures of many-body effects have predominantly appeared on the electronic Raman scattering response of graphene under a transverse magnetic field. Recently, many-body effects have also been unveiled by monitoring magneto-phonon resonances (MPR) between optically active inter-LL transitions and zone-center optical phonons (i.e., Raman G-mode phonons) in graphene encapsulated in hexagonal boron nitride (BN) films. As recently suggested by Sonntag et al., more prominent effects are expected in suspended graphene, where electron–electron interactions are minimized screened.

In this letter, we report the results of MMRS measurements performed on suspended mono- to pentalayer graphene. For each number of layers $N$, we resolve a set of well-defined MPR. Using a single-particle effective bilayer model, we readily extract $v_F$ associated with each MPR. While a single parameter slightly above the bulk graphite value ($\approx 1.05 \times 10^6$ m s$^{-1}$) suffices to fit all MPR for $N \geq 2$ layer systems, we find that $v_F$ increases significantly up to $(\approx 1.45 \times 10^6$ m s$^{-1}$ in monolayer graphene. This result is understood as a signature of enhanced many-body effects in unscreened graphene.

The low-energy electronic electronic bands and magneto-optical response of mono- and $N$-layer graphene have been extensively discussed using an effective bilayer model. To lowest order, this model uses only two parameters, namely, the single-particle Fermi velocity $v_F = \frac{2}{\hbar} \frac{\alpha}{\gamma_0}$ where $\gamma_0$ is the nearest neighbor hopping parameter and $a = 0.142$ nm is the C–C bond length, and the nearest neighbor interlayer coupling constant $\gamma_1$. The band structure of Bernal- or equivalently AB-stacked $N \geq 2$ layer systems exhibits $2N$ bands, including one pair of linear (monolayer-like) bands only present for odd

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$N$ and a set of $[N/2]$ effective bilayer bands (here $[\cdot ]$ denotes the integer part) with a rescaled $\tilde{\gamma}_1 = 2\gamma_1 \cos \theta$,\textsuperscript{[21–26]} which corresponds to half the energy gap between their split-off bands. The angle $\theta$ represents the quantized transverse momenta $\theta = k_c c/2$ at which 2D cuts are made in the 3D electronic dispersion of graphite, with $c/2 = 0.34$nm being the interlayer separation in bulk graphite. The band structure of mono- to trilayer graphene and corresponding values of $\tilde{\gamma}_1$ and $\theta$ are shown in Figure 1a–c.

In the presence of a transverse magnetic field, the effective mono- and bilayers give rise to independent sets of Landau fans. The energy $E_{\pm n} = E_{\pm n}(B)$ of the $n$th electron ($n \geq 0$) or hole ($n \leq 0$) LL arising from the gapless bands in an effective bilayer is given by:\textsuperscript{[19,27]}

$$E_{\pm n} = \sqrt{\frac{\tilde{\gamma}_1^2}{2} + (|n| + \frac{1}{2}) E_1^2} - \sqrt{\frac{\tilde{\gamma}_1^2}{4} + (|n| + \frac{1}{2}) E_0^2 + E_1^2}.$$  \hspace{1cm} (1)

Where $E_1 = \nu_F \sqrt{2 e B}$. At $\theta = \pi/2$, one obtains the well-known LL fan of monolayer graphene $E_{\pm n} = \sqrt{n} E_1$. For $\theta \neq \pi/2$, nearly linear scalings of $E_{\pm n}(B)$, characteristic of effective bilayers, are observed. The corresponding LL fans are shown in Figure 1d–f for mono- to trilayer graphene, respectively. As previously established,\textsuperscript{[28–30]} optically allowed transitions, such that $\delta|n| = \pm 1$, couple to the graphene G-mode phonons (with energy $\hbar \omega_G \approx 1585$ cm$^{-1}$ or equivalently $\approx 197$ meV) and give rise to series of MPR,\textsuperscript{[12,31,32]} as indicated by the vertical arrows in Figure 1d–f. In contrast, symmetric inter-LL transitions ($\delta|n| = 0$, denoted $L_n$, with $n \geq 1$) are Raman-allowed\textsuperscript{[5,13,14]} but are not expected to couple to optical phonons.\textsuperscript{[28]} In the following, the energy of the optically active inter-LL transitions associated with a given value of $\theta$ will be simply denoted

$$T_n = |\Delta \omega_{\pm(n+1)} - \Delta \omega_{\mp n}|, \quad n \geq 0$$  \hspace{1cm} (2)

and the $(n+1)$th MPR ($n \geq 0$) occurs when $T_n = \hbar \omega_G$.

Figure 2 shows MMRS data for a suspended graphene monolayer with minimal native doping (spatially averaged over our laser spot size) well below $10^{13}$ cm$^{-2}$. MPR appear as anticrossings involving significant frequency shifts and line broadening of the G-mode feature. Due to electronic broadening in graphene,\textsuperscript{[36,37]} antisymmetric MPR are not fully resolved, particularly so at low field. The third and second MPR, involving transitions $T_2$ and $T_1$, appear at $B_2 = 1.45 \pm 0.1$ T and $B_1 = 2.75 \pm 0.1$ T, respectively. Remarkably, for the first time in suspended graphene, we report the $n = 0$ MPR at $B_0 = 19.4 \pm 0.1$ T. The prominence of the $T_0$ MPR originates from the fact that the coupling strength in MPR theory grows with the LL degeneracy,\textsuperscript{[32]} which is proportional to $B$. As a result, MPR involving LL with higher $n$ are fainter because these resonances occur at lower $B$.

The resonant fields observed here are considerably lower than in substrate-supported systems\textsuperscript{[12,15,16]} as discussed in more detail later. As shown in Figure 2b, near $B_0$, the G-mode exhibits

Figure 1. Single-particle low-energy electronic structure and LL of mono-, bi-, and trilayer graphene. a–c) The electronic dispersions obtained from the effective bilayer model described in the text are shown, respectively. d–f) The corresponding dispersion of the LL arising from the gapless bands is shown for mono-, bi-, and trilayer graphene, respectively. Red lines in (d) and (f) correspond to monolayer-like LL ($\theta = \pi/2$), whereas dark yellow lines in (e) and (f) correspond to LL arising from effective bilayers obtained at quantized values of $\theta \neq \pi/2$. The vertical arrows indicate the optically allowed inter-LL transitions $T_n$ that give rise to the MPR. The calculations are performed with $\nu_F = 1.05 \times 10^6$ m s$^{-1}$ and $\gamma_1 = 400$ meV.
Figure 2. MPR in suspended monolayer graphene. a,b) Raman G-mode spectra at selected values of the transverse magnetic field B. The spectra are acquired for the same duration and incident laser power and offset for clarity. The dark and light gray lines are Lorentzian fits to the data. c) G-mode frequency $\omega_{GC}$ (red circles), extracted from Lorentzian fits of the G-mode spectra, overlaid on a false-color plot of the Raman spectra as a function of B. The red and grey lines are calculated dispersions of the $T_0$ (optically allowed) and of the $L_1$ (symmetric, Raman allowed) transitions, respectively. The insets in (c) and (d) show close-ups as a function of the magnetic $\omega_B$ MPR in suspended monolayer graphene. a,b) Raman G-mode spectra at selected values of the transverse magnetic field $B$. c) G-mode frequency $\omega_{GC}$ (red circles), extracted from Lorentzian fits of the G-mode spectra, overlaid on a false-color plot of the Raman spectra as a function of B. The red and grey lines are calculated dispersions of the $T_0$ (optically allowed) and of the $L_1$ (symmetric, Raman allowed) transitions, respectively. The insets in (c) and (d) show close-ups as a function of the magnetic field. The resonant fields $B_n$ are indicated. The insets in (c) and (d) show close-ups on the regions boxed with thin dotted lines.

a double-Lorentzian lineshape, with a broad, prominent feature that shows an avoided crossing with the $T_0$ transition and a fainter subfeature (not shown in Figure 2c,d) that is virtually independent on B. The integrated intensity of this uncoupled subfeature is about one order of magnitude smaller than that of the coupled G-mode feature (Figure 2b); its frequency at $1589.2 \pm 0.8 \text{ cm}^{-1}$ is slightly upshifted and its full width at half maximum ($6.5 \pm 1.5 \text{ cm}^{-1}$) is significantly lower as compared with the G-mode feature recorded at $B=0\text{T}$ ($\omega_{GC} = 1587.0 \pm 0.1 \text{ cm}^{-1}$, $\Gamma_G = 13.6 \pm 0.1 \text{ cm}^{-1}$). These characteristics suggest a slight doping with a Fermi level slightly above $\hbar \omega_{GC}/2$ (i.e., $\approx 100 \text{ meV}$). We therefore suggest that local inhomogeneities at the nanometer scale may result in an uncoupled G-mode subfeature due to local Pauli blocking of the $T_0$ transition. Importantly, Kim et al. reported a similar uncoupled G-mode feature in SiO$_2$-supported graphene, however with a large integrated intensity, comparable with that of the coupled G-mode feature that follows an anticrossing with the $T_0$ transition. Here, the much lower spectral weight of the uncoupled G-mode subfeature illustrates the superior spatial homogeneity of suspended graphene. We suggest that MMRS near the $n=0$ MPR can be used as a quantitative probe of residual electron and hole puddles in high-quality graphene.

In Figure 2c, one can clearly see an increase in the phonon linewidth, a signature of a variation of the phonon lifetime induced by an interaction, at $B_1 = 4.7 \pm 0.1 \text{ T}$. This resonance implies the G-mode phonon and the symmetric inter-LL excitation $L_1$. Although symmetric electronic excitations should not couple to the optical phonons, a similar MPR has been observed in graphene on graphite and on graphene encapsulated in hexagonal BN. Interestingly, both in suspended graphene and in BN-encapsulated graphene, these nominally forbidden MPR are much fainter than the allowed MPR involving $T_1$ and have similar strength. Here, as we investigate suspended graphene, substrate-induced effects cannot account for this unexpected coupling. Our results point toward an intrinsic effect, possibly originating from LL mixing induced by Coulomb interactions.

We now consider how MPR evolve for thicker graphene stacks, as the electronic structure gets increasingly complex. Figure 3 shows MPR data for suspended bilayer and trilayer graphene. Data for tetra- and pentalayer graphene are shown in Figure 4. These data were acquired using a lower spectral resolution that for $N=1, 2, 3$. As a result, we have not attempted to resolve signatures of pronounced anticrossings at high fields for $N=4$ and $N=5$. For $N=2$, three MPR are resolved at $B_1 = 7.9 \pm 0.1 \text{ T}$, $B_2 = 10.8 \pm 0.1 \text{ T}$, and $B_3 = 16.6 \pm 0.1 \text{ T}$ and are assigned to the $T_0$, $T_2$, and $T_1$ MPR of bilayer graphene ($\theta = \pi/3$), respectively. In trilayer graphene (Figure 3c,d), we are able to resolve the first three monolayer-like MPR (involving transitions $T_0$, $T_1$, and $T_2$ for $\theta = \pi/2$) and the MPR associated with $T_1$, $T_2$, and $T_1$ for the effective bilayer ($\theta = \pi/4$). At high fields $B>14 \text{ T}$, the G-mode lineshape becomes more complex.
Figure 3. MPR in suspended a,b) bilayer and c,d) trilayer graphene. a,c) G-mode frequency $\omega_G$ (red circles), extracted from a Lorentzian fit of the G-mode spectra, overlaid on a false-color plot of the Raman spectra as a function of the transverse magnetic field $B$. The red and dark yellow dashed lines are calculated dispersions of the $T_n$ transitions for monolayer-like and effective bilayer bands. The single $v_F$ extracted from all MPR is indicated in both cases in units of $v_F = 1.00 \times 10^6 \text{ m s}^{-1}$. b,d) Lorentzian full width at half maximum $\Gamma_G$ as a function of the magnetic field. The resonant fields $B_n$ are indicated with color-coded values of $\theta$. In the trilayer case (c,d), the G-mode spectra are fit to a double Lorentzian (red and orange symbols) for $13 \text{T} < B < 21 \text{T}$ and $26 \text{T} < B < 29 \text{T}$ and to a triple Lorentzian (red, yellow, and dark yellow) for $21 \text{T} < B < 26 \text{T}$ near the first monolayer-like and second bilayer-like MPR. The insets in (a,b,d) show close-ups on the regions boxed with red dotted lines in (a,d) and on the most prominent G-mode subfeature in (c).

Figure 4. MPR in suspended a,b) tetralayer and c,d) pentalayer graphene. a,c) G-mode frequency $\omega_G$ (red circles), extracted from a Lorentzian fit of the G-mode spectra as a function of the transverse magnetic field $B$. The red, dark yellow, and orange dashed lines are calculated dispersions of the $T_n$ transitions for monolayer-like and effective bilayer bands with color-coded values of $\theta$. The single $v_F$ extracted from all MPR is indicated in both cases in units of $v_F = 1.00 \times 10^6 \text{ m s}^{-1}$. b,d) Lorentzian full width at half maximum $\Gamma_G$ as a function of the magnetic field. The resonant fields $B_n$ are indicated with color-coded values of $\theta$. 

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due to the overlap between the second bilayer-like MPR (T1, \( \theta = \pi/4 \)) and first monolayer-like MPR (T0, \( \theta = \pi/2 \)). These spectra are well fit to a bilayer Lorentzian form for for \( 13 \text{T} < B < 21 \text{T} \) and \( 26 \text{T} < B < 29 \text{T} \) and to a triple Lorentzian (red, yellow, and dark yellow) for \( 21 \text{T} < B < 26 \text{T} \). The narrow, central feature (red symbols in Figure 3c) is assigned to the second bilayer-like MPR (T1, \( \theta = \pi/4 \)), whereas the broader and shifted features (orange symbols for double-Lorentzian fits and then dark yellow and yellow symbols for triple-Lorentzian fits in Figure 3c) are assigned to the first monolayer-like MPR (T0, \( \theta = \pi/2 \)).

In tetralayer graphene (Figure 4a,b), under transverse magnetic fields up to \( B = 29 \text{T} \), we observe six MPR associated with gapless effective bilayer bands at \( \theta = \pi/5 \) and \( \theta = 2\pi/5 \), respectively.\(^{[43]}\) Up to nine MPR are observed in the same window for pentalayer graphene (Figure 4c,d). These MPR are associated with the effective monolayer (\( \theta = \pi/2 \)), with two other subsets of MPR stemming from the gapless effective bilayer bands at \( \theta = \pi/3 \) and \( \theta = \pi/6 \), respectively. Let us note that as the number of electronic subbands increase, MPR involving distinct values of \( \theta \) and \( n \) may be nearly degenerate. As a result, we are not able to clearly resolve \( T_1, \theta = 2\pi/5 \) and \( T_3, \theta = \pi/5 \) near 12 T for \( N = 4 \) (Figure 4c,d). Similarly, for \( N = 5 \) (Figure 4c,d), the MPR associated with \( T_2, \theta = \pi/3 \) and \( T_4, \theta = \pi/6 \) near 11 T as well as with \( T_0, \theta = \pi/2 \) and \( T_1, \theta = \pi/6 \) near 27–28 T appear single, slightly broadened MPR. Thus, the growing complexity of the LL spectrum with increasing \( N \) prevents detailed studies of MPR for \( N > 5 \), until the limit of the LL fan diagram of bulk graphite is reached.

As shown in Figure 5a, from the data in Figure 2–4, and using the effective bilayer model (Equation (2)), we can now determine, for each MPR, the velocity parameters \( v_F, n \) evaluated at resonant fields \( B_n \). For simplicity, we have considered a constant \( \gamma_1 = 400 \text{meV} \) for all \( N \) and have attempted a global fit of all the \( B_n \) measured for a given \( N \). For \( N = 2 \) to \( N = 5 \), this procedure yields a very good agreement between Equation (2) and our experiments with a constant \( v_F \) that decreases smoothly from \( (1.08 \pm 0.01) \times 10^5 \text{m}^\text{s}^{-1} \) for \( N = 2 \) down to \( (1.05 \pm 0.01) \times 10^5 \text{m}^\text{s}^{-1} \) for \( N = 5 \). These values converge toward the bulk graphite value of \( (1.02 \pm 0.01) \times 10^5 \text{m}^\text{s}^{-1} \) and are in excellent agreement with previous electronic Raman measurements on the same samples.\(^{[14]}\)

In contrast, a single velocity parameter would grossly fail to fit the three observed MPR (involving \( T_n \) with \( n = 0, 1, 2 \)) in monolayer graphene. Instead, using Equation (2), we determine that \( v_F \) increases significantly from \( v_{F,0} = (1.23 \pm 0.01) \times 10^5 \text{m}^\text{s}^{-1} \) up to \( v_{F,2} = (1.45 \pm 0.02) \times 10^5 \text{m}^\text{s}^{-1} \) as \( B_n \) decreases from 19.4 T down to 1.40 T. Interestingly, our value of \( v_{F,1} = (1.35 \pm 0.1) \times 10^5 \text{m}^\text{s}^{-1} \) inferred from the \( n = 1 \) MPR exactly matches a recent report in neutral suspended graphene grown by chemical vapor deposition and carefully cleaned after transfer.\(^{[18]}\)

The extracted \( v_F, n \) can be viewed as effective velocity parameters that account for the combination of two effects: 1) a logarithmic divergence of the band velocity as \( B_n \), decreases (self-energy) and 2) magneto-Coulomb binding, leading \( n \)-dependent velocity parameters associated with \( T_n \) or \( L_n \) transitions. The measured rise of \( v_F, n \) as \( B_n \) decreases is qualitatively consistent with our previous measurements.\(^{[15]}\) We should stress that MPR probe charge carriers at a fixed energy set by \( \hbar \omega_C = T_n \). However, in a one-electron picture, MPR occur at \( B_n \propto \sqrt{n+1} + \sqrt{n} \) for \( N = 1 \) (Equation (2)).

Previous investigations of \( L_n \) transitions in suspended graphene\(^{[5]}\) and \( T_n \) transitions in BN-encapsulated graphene\(^{[29]}\) indicate that at a given \( B \), \( v_F \) increases with \( n \), up to \( n = 3 \) (see also Figure 5b). Our data are in line with these results because \( v_{F,0} < v_{F,1} < v_{F,2} \) (Figure 2c). However, Russell et al.\(^{[20]}\) also report a slight decrease in \( v_F \) at fixed \( B \) for higher order \( T_n \) (with \( n = 3, 4, 5 \)). In the wake of recent studies of filling-factor-dependent many-body effects in graphene,\(^{[18,20,44,45]}\) further efforts are needed to separate the contributions of self-energy and magneto-Coulomb binding to the renormalization of \( v_F \). Remarkably, in keeping with our previous measurements of \( L_n \) transitions,\(^{[14]}\) many-body effects are essentially observed in monolayer graphene and nearly vanish for \( N \geq 2 \). Such observations are consistent with recent works, showing that the reduction of the exciton binding energy in an atomically thin semiconductor coupled to graphene layers is readily maximized with one graphene monolayer and does not increase further if a bilayer graphene is used instead.\(^{[46]}\) In addition, the parabolic dispersion of massive fermions in effective bilayers (Figure 1b,c) creates a finite density of states at the Dirac point, which may be sufficient to quench electron–electron interactions and the subsequent renormalization of electronic bands. Here also, additional investigations of many-body effects are required to account for the abrupt transition between the mono- and bilayer cases.\(^{[17,48]}\)

In conclusion, we have demonstrated that MPR provide invaluable fingerprints of the low-energy electronic dispersion of suspended graphene layers, which are in excellent quantitative agreement with a one-electron effective bilayer model for \( N \geq 2 \).
In the monolayer limit, electrons undergo minimal screening, leading to pronounced many-body effects which, to experimental accuracy, do not yield sizeable experimental signatures for \( N \gtrsim 2 \). Many-body effects in graphene and Dirac fermions in general continue to attract considerable interest, and future directions include the interplay between many-body effects and electronic lifetime in graphene and related systems.\(^{[149]}\)

**Experimental Section**

The suspended graphene samples investigated here were prepared by mechanical cleavage of Bernal-stacked natural graphite and dry transfer onto \( 8 \mu \text{m}\)-diameter pits etched in Si/SiO\(_2\) substrates as described in detail elsewhere.\(^{[13,14]}\) Such pristine, minimally screened graphene layers have been shown to exhibit negligible residual doping and charge inhomogeneity, making them ideally suited for MMRS studies.\(^{[15,14]}\) The samples were held in a home-built magneto-optical cryostat where MMRS measurements were performed in a back-scattering geometry under transverse magnetic fields up to 29 T. A continuous-wave (cw) laser beam at 514 nm was focused onto a \( \approx 2 \mu \text{m}\)-diameter spot. A cross-circular polarization configuration was implemented, where incoming and Raman-scattered photons have opposite helicity. In these conditions, the magneto-Raman spectra are dominated by MPR.\(^{[28,33]}\) We have used laser powers \( \approx 1 \mu \text{W} \) at our samples to minimize laser-induced heating effects while still having a sufficiently large MRRS signal. All measurements were performed at a base temperature of 4 K. The Raman G-mode spectra were fit to Lorentzian functions, and the peak frequencies and full width at half maximum are determined with an experimental error that is typically smaller than the symbol size.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**

electron–electron interactions, graphene, magneto-excitons, magneto-phonon resonance, magneto-Raman spectroscopy

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