Visible quantum plasmons in highly-doped few-layer graphene

Sharmila N. Shirodkar,1,* Marios Mattheakis,1,‡ Paul Cazeaux,2
Prineha Narang,1,¶ Marin Soljačić,3 and Efthimios Kaxiras1

1John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA
2School of Mathematics, University of Minnesota, Minneapolis, Minnesota 55455, USA
3Department of Physics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA

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Abstract

Doped graphene can support plasmon modes, but the limited range of doping achieved by gating prevents their frequency from reaching the visible to infrared range. Here we show, through the use of first-principles calculations, that the high levels of doping achieved by lithium intercalation in bilayer and trilayer graphene shift the plasmon frequencies into the visible range. To obtain physically meaningful results, we introduce a correction of the effect of plasmon interaction across the vacuum in a periodic cell by employing transparent boundary conditions in the direction perpendicular to the layers, a significant improvement over the Exact Coulomb cutoff technique employed in earlier works. We describe the features of these quantum plasmons, including the dispersion relation, losses and field localization. Our findings point to a strategy for fine-tuning the plasmon frequencies in graphene and other two dimensional materials.
After more than a decade of intensive scientific exploration, new plasmonic phenomena continue to be discovered, including quantum interference of plasmons, observation of quantum coupling of plasmons to single particle excitations, and quantum confinement of plasmons in single-nm scale plasmonic particles and materials. Further, plasmonic nanostuctures find widening applications in integrated nanophotonics [1], biosensing [2–4], photovoltaic devices [5–7], single photon transistors [8], single molecule spectroscopy [9] and even metamaterials [10, 11]. The current interest in quantum nanophotonics and plasmonics is in part driven by new materials, particularly low dimensional materials, that allow access to new regimes. The reduced dimensionality of plasmons in two-dimensional (2D) materials provides ultra-subwavelength confinement with phase velocities several orders of magnitude lower than the speed of light [12]. The origin of 2D plasmons is related to the local field effects and the non-local response of the material to external fields [13]. Hence, a study of these waves demands a fully quantum mechanical description of the material properties which allows us to refer to them as ‘quantum’ 2D plasmons. We effectively capture this 2D and quantum nature of these plasmons through our high-accuracy first-principles calculations, characterized by (a) our in-house methodology which correctly confines plasmons in two dimensions, and (b) a realistic estimate of carrier lifetime — the crucial factor that determines plasmon losses.

Graphene is quite special for 2D plasmonics [14] as it exhibits intriguing properties such as extremely high electrical mobility [15] and easily tunable electron and hole doping concentrations, $n$, through gating [15,16]. Graphene plasmon frequencies are easily tunable via doping [14], where typical doping concentration values achieved by gating are $\approx 10^{11}$ cm$^{-2}$, and the heaviest doping reached is $n_h > 10^{13}$ cm$^{-2}$ with hole concentration [17]. Plasmons in gate doped graphene typically emerge in the infrared to THz ranges, and seldom in the mid- or near-infrared range [4, 17, 18]. 2D plasmons in the visible range would be crucial for application in optoelectronic devices. One can try going beyond graphene to achieve plasmons in optical frequencies but plasmons in other 2D materials such as transition metal dichalcogenides (TMDCs), are expected to appear at THz frequencies [19, 20]. Also, plasmon modes on Be(0001) [14] observed in the visible range [21] may not be interpreted as a true 2D plasmon, since it has finite penetration depth into the underlying bulk material. Very recent report by Huang et al. [22] predicts that triangular polymorph of 2D boron sheet exhibits visible frequency plasmons. However, free-standing triangular 2D boron is dynam-
ically unstable \cite{23} and its experimental synthesis quite difficult, which makes it unsuitable for device applications.

On the other hand, atypical methods can dope graphene to levels beyond what is achievable through gating. Experiments have proved the feasibility of inserting metal atoms like lithium (Li) between layers of 2D materials \cite{24, 25} resulting in heavy doping. Inspired by these facts, we use a theoretical approach based on first-principles electronic structure calculations to explore the possibility of observing quantum plasmons in the visible range for Li-intercalated few-layer graphene. Using highly accurate first-principles calculations accompanied with appropriate boundary conditions and realistic estimate of carrier lifetimes; we show that this effect is indeed possible, opening up pathways for fine-tuning a wide range of plasmon frequencies including visible range, in 2D structures, which can be accomplished by varying the concentration and type of intercalants.

Our first-principles calculations are based on density functional theory (DFT) as implemented in the GPAW package \cite{26, 27}. The interaction between ionic cores and valence electrons is described by the projector augmented wave method \cite{28, 29}. A vacuum of 25 Å is included to minimize the interaction between periodic images along the direction perpendicular to the plane of the sheets (z direction). The Kohn-Sham wavefunctions are represented using a plane wave basis with energy cutoff of 340 eV, and the exchange correlation energy of electrons is described using Local Density Approximated (LDA) functional. For the linear response calculations, used to estimate the dielectric functions \cite{30}, we sample the Brillouin zone with a $256 \times 256 \times 1$ grid of k-points to include an accurate description of intraband transitions. For the dielectric response calculations we use a plane wave energy cutoff of 30 eV. All the other parameters were converged to within 0.05 eV of the plasmon energies, using the methodology developed by Andersen et al. \cite{19, 31} for calculating the quantum plasmon modes.

The potential $\phi(\mathbf{r}, \omega)$ and charge density $\rho(\mathbf{r}, \omega)$ of the quantum plasmon modes, are obtained as left and right eigenfunctions (which satisfy the Poisson equation) of the dielectric operator $\hat{\epsilon}(\omega)$, diagonalized in the plane wave basis:

$$\hat{\epsilon}(\omega) \phi_n(\omega) = [\hat{1} - \hat{\chi}^0(\omega)] \phi_n(\omega) = \lambda_n(\omega) \phi_n(\omega),$$ \hspace{1cm} (1)

where $\omega$ and $\mathbf{r}$ denote the frequency and in-plane spatial vector, respectively. Here, $\hat{\epsilon}(\omega)$ expressed in terms of the noninteracting linear response operator $\chi^0(\omega)$ and the Coulomb
interaction operator $\hat{v} = 1/|\mathbf{r} - \mathbf{r}'|$. The condition for observing a plasmon at frequency $\omega_p$ is $\Re[\lambda_n(\omega)] = 0$ or equivalently a peak in the loss function, $-\Im[\lambda_n(\omega)]/|\lambda_n(\omega)|^2$.

A key ingredient in obtaining the plasmon dispersion and losses is the carrier lifetime, $\tau$. To obtain reliable values of $\tau$, we use the DFT results for the energies and matrix elements of both electrons and phonons (see Supplemental Material [32]). This takes into account the detailed electronic structure effects such as inter-layer interactions and response of electrons far from the Dirac point, as well as scattering against both acoustic and optical phonons including Umklapp and inter-valley processes [33–36]. Changing the Fermi level, $E_F$, changes the equilibrium electron occupation factors in the Boltzmann equation as well as in the Fermi Golden rule; we account for this by explicitly evaluating these quantities for several different values of $E_F$ ranging from the neutral (undoped) value to 1.5 eV above it. Interestingly, our results show that the extremely large $\tau \approx 1$ ps for free standing undoped graphene drops to $\approx 29$ fs in doped graphene.

The standard approach for eliminating spurious effects due to finite size of vacuum [37] is inadequate for plasmons with small in-plane wavevectors ($\mathbf{q}$), and increasing the size of the vacuum region until these effects become negligibly small requires very expensive calculations. A significant methodological contribution of the present work is the formulation and implementation of transparent boundary conditions which overcome the drawbacks of the Coulomb cutoff method and offer a more accurate description of the quantum plasmon fields.

Let $z_-, z_+$ be the bounds of the super-cell (simulation box) along the $z$ direction (vacuum region) with $(x, y)$ plane being periodic. We apply a one-dimensional Fourier transform in the $z$ direction to obtain a real space representation in this coordinate. The response operator under random phase approximation (RPA) then has the form:

$$
\hat{\chi}^0 \phi(z, G_{xy}, q, \omega) = \int_{z_-}^{z_+} \sum_{G'_{xy}} \chi^0_{G_{xy}, G'_{xy}}(z, z', q, \omega) \phi(z', G'_{xy}, q, \omega) dz',
$$

where $G_{xy}$, $G'_{xy}$ are vectors of the in-plane reciprocal lattice. For values of $z, z'$ inside the super-cell, $z_- < z, z' < z_+$, the kernel $\chi^0_{G_{xy}, G'_{xy}}(z, z')$ is deduced from $\chi^0_{G, G'}$ by Fourier transform. The kernel is extended with zero values for $z$ or $z'$ that lie in the vacuum region outside this cell. We observe that Eq. (1) can be reformulated as the generalized eigenvalue problem [32]:

$$
\hat{\chi}^0 \phi_n(z, G_{xy}, q, \omega) = \frac{1 - \lambda_n}{4\pi} \left( |q + G_{xy}|^2 - \frac{\partial^2}{\partial z^2} \right) \phi_n(z, G_{xy}, q, \omega),
$$

where $\lambda_n$ is the $n$th eigenvalue of the problem.
with additional constraint that $|\phi_n| \to 0$ as $z \to \pm \infty$ so the problem is well-posed. The left-hand side vanishes in the vacuum region and Eq. (3) reduces to the one-dimensional Poisson equation. For any nonzero value of $|\mathbf{q} + \mathbf{G}_{xy}|$, we thus obtain an explicit solution

$$
\phi_n(z, \mathbf{G}_{xy}, \mathbf{q}, \omega) = \phi_n(z_\pm, \mathbf{G}_{xy}, \mathbf{q}, \omega)e^{-|\mathbf{q} + \mathbf{G}_{xy}| |z_\pm - z|},
$$

for $z \leq z_-$ and $z \geq z_+$. The continuity of $\phi_n$ and its first derivative with respect to $z$ leads to the transparent boundary conditions at $z = z_\pm$:

$$
\frac{\partial \phi_n}{\partial z}(\mathbf{q}, \mathbf{G}_{xy}, z_\pm, \omega) = \mp |\mathbf{q} + \mathbf{G}_{xy}| \phi_n(\mathbf{q}, \mathbf{G}_{xy}, z_\pm, \omega),
$$

which implies that the charge density and potential do not see the periodic boundary along the $z$ direction for any value of $\mathbf{q}$, and hence decay to zero as $z \to \pm \infty$. We solve numerically by finite differences the eigenvalue problem of Eq. (3) restricted to the finite band $z_- \leq z \leq z_+$, with the boundary conditions of Eq. (4) (see Supplemental Material for details [32]).

We model Li intercalated graphene (G) multilayers with an in-plane $\sqrt{3} \times \sqrt{3}$ multiple of the primitive unit cell of graphene, with the G/Li/G (bilayer) and G/Li/G/Li/G (trilayer) structures. There is one Li atom per unit cell between each pair of layers (see Fig. 1) [38, 39]. For the trilayer, we consider the structure with the two Li atoms at the same hollow site but between two different pairs of graphene layers, as this is the most stable configuration [39]. Li intercalation makes AA stacking energetically more preferable [38] and hence both bilayer and trilayer structures are inversion symmetric. The separation between the layers increases by 0.14 Å and 0.11 Å relative to its value in the AA stacked graphene bilayer (3.52 Å), for the bilayer and trilayer, respectively. Due to band folding in the $\sqrt{3} \times \sqrt{3}$ unit cell, the high symmetry K point and hence the Dirac point of primitive graphene cell folds onto $\Gamma$ point in the Brillouin zone (BZ) in our simulations (see Fig. 1). AA stacking preserves the sublattice symmetry of the layers and the linear dispersion of the electron bands at the Dirac point, unlike AB stacking where the bands are parabolic [40]. Intercalation also leads to charge transfer from Li to the graphene layers, and renders the system metallic (see Fig. 1) with $\approx 0.84e$ and $0.87e$ charge transferred from Li to bilayer and trilayer graphene (determined using Bader analysis), which corresponds to $n = 5 \times 10^{14}$ and $n = 10^{15}$, respectively. Subsequently, shifting the Fermi level from the Dirac point into the conduction band by 1.35 eV and by 1.51 eV for the bilayer and trilayer, respectively, as seen in Fig. 1.

Since we consider metallic multilayers, more than one plasmon modes emerge [19, 31, 40]. Depending on the phase of the charge density and potential fields, we differentiate them as
symmetric and antisymmetric plasmonic modes [see Fig. 2(a) and (d)]. For small \( q \), the decay length of 2D plasmons extends beyond the vacuum region giving rise to interactions with periodic images, and hence, spurious fields and pseudo charges at the vacuum edge. On the other hand, our transparent boundary conditions correct these periodic interactions and make the plasmon tails invisible to one another for the same vacuum length. The charge density with (solid lines) and without (dotted lines) transparent boundary conditions is shown in Fig. 2(a) and (d) for G/Li/G and G/Li/G/Li/G, respectively. We also note that the charge transferred from Li is equally distributed in the unoccupied \( \pi^* \) orbitals, which is confirmed from the charge density distribution of the plasmon modes [see in Fig. 2(a) and (d)], where the intensity of the fields is equal and reaches the maximum/minimum values away from the layers, consistent with the fact that the \( \pi^* \) orbitals of graphene extend away from the layers.

We plot the plasmon dispersion along \( \Gamma-M \) (the \( \Gamma-K \) direction is not as interesting in the band structure) with the magnitude of the real part of \( q \) ranging from \( |q| = 0.007 \ \text{Å}^{-1} \) to \( 0.21 \ \text{Å}^{-1} \), since both plasmon modes become very weak above \( q = 0.21 \ \text{Å}^{-1} \). The symmetric mode is more dispersive than antisymmetric, and varies as \( \sqrt{q} \) at small \( q \), corresponding to classical plasmon with Drude behavior due to intraband transitions. Whereas the antisymmetric mode varies almost linearly with \( q \) (has finite frequency at \( q=0 \)) and relates to interband transitions between perfectly nested bands of the two layers [40]. In G/Li/G the plasmon frequencies are between 0.8 eV to 2.2 eV for \( q \geq 0.007 \ \text{Å}^{-1} \); the antisymmetric mode is in the optical frequency range even at low \( q \), whereas the symmetric mode enters into this range at higher \( q \) values. The symmetric mode is always lower in energy than the antisymmetric mode due to finite coupling [40].

We quantify the plasmon losses from the ratio of real to imaginary component of wavenumber, \( \text{Re}[q]/\text{Im}[q] \) [41], which corresponds to the number of plasmon wavelengths that propagate before it loses most of its energy [see Fig. 2(c)]. For the doping in G/Li/G (\( E_F = 1.35 \) eV), a \( \tau \approx 29 \) fs was calculated using our methodology discussed above, which is much shorter in comparison with \( \tau \approx 135 \) fs for \( E_F = 0.135 \) eV [41] . We only give the ratio for the symmetric (intraband) mode in Fig. 2(c). Due to its linear dispersion, antisymmetric mode shows less variation in \( \text{Re}[q]/\text{Im}[q] \) as compared with symmetric mode (see Supplemental Fig. S1 [32]). The in-plane propagation length of the plasmons varies directly with this ratio, with the symmetric plasmons propagating longer at longer wavelengths (\( \lambda_{\text{air}} \)).
We also calculate the wave “shrinkage” or the field localization of the plasmons, shown in Fig. 2(c); this corresponds to the ratio by which the plasmon wavelength ($\lambda_p$) is smaller than that in vacuum, and is approximately 100 times for bilayer graphene.

There are three important decay modes that lead to plasmon damping: a) Landau damping due to intraband losses when $\omega<\omega_{\text{intra}}$, b) interband losses (single particle excitations, SPEs, identified as poles of the response function [40, 42]) when $\omega>\omega_{\text{SPE}}$, and c) decay via optical phonons in graphene for $\omega>\omega_{\text{ph}}$ ($\omega_{\text{ph}}=0.2$ eV or 6.2 $\mu$m) [41]. In case of G/Li/G, since the optical phonon $\omega_{\text{ph}}=1400$ cm$^{-1}$ $\equiv$ 0.17 eV [39, 43] is much smaller than the symmetric or antisymmetric plasmon frequencies (0.8 eV to 2.2 eV for $q \geq 0.007$ Å$^{-1}$), only multiple scatterings by phonons (which are less likely) will scatter plasmons into the damping regions. On the other hand, plasmons within frequency range $\omega_{\text{SPE}}-\omega_{\text{ph}}$ to $\omega_{\text{SPE}}$ can get scattered by phonons into Landau/interband scattering regions. Therefore making $\omega>\omega_{\text{SPE}}-\omega_{\text{ph}}$ the region where plasmons are damped by interband transitions and optical phonons. The SPEs at $q=0$ were identified at 0 eV, 0.6 eV and 2.4 eV originating from the intraband, low energy interband and the electron-hole interband transitions in G/Li/G. The damping regions are defined by $E_{\text{SPE}}=\pm hv_Fq \pm \hbar\omega_{\text{ph}}$ (including scattering by optical phonons), where $v_F$ is the Fermi velocity and $E_{\text{SPE}}$ is the single particle excitation energy [42, 44] [see gray shaded areas in Fig. 2(b)]. Heavy doping by lithium pushes the electron-hole interband threshold for the bilayer to $\omega_{\text{inter}} \approx 1.77$ eV ($\lambda = 0.7 \mu$m). Since the optical frequency range ($\omega_{\text{op}}$) is between 1.59 eV to 3.26 eV ($\lambda = 0.38 \mu$m to 0.78 $\mu$m) and $\omega_{\text{inter}} < \omega_{\text{op}}$, most of the symmetric and antisymmetric plasmon modes in this range are not damped by the interband transitions, indicated by the shaded regions in Fig. 2(b) and 2(e). Only for $q \geq 0.06$ Å$^{-1}$ are the symmetric and antisymmetric modes damped.

To push the interband threshold frequency, and hence the plasmon frequencies, higher into the optical range ($>2$ eV), the Fermi level needs to be moved farther into the conduction bands. Since the maximum possible intercalation in bilayer graphene corresponds to composition $C_{12}$Li, additional Li can be incorporated only by having more than two graphene layers. We therefore explore trilayer graphene since it can accommodate two Li layers, with a composition $Li_2C_{18}$, which increases the doping level to $E_F = 1.51$ eV. There are three modes in the trilayer structure in the 1.2 – 2.8 eV frequency range along the $\Gamma$-M direction for $q \geq 0.007$ Å$^{-1}$, two of which are symmetric and one antisymmetric, shown in Fig. 2(d). The third (second symmetric) mode emerges due to the third graphene layer which brings
in additional nesting of the bands. Similar to the bilayer case, the first symmetric mode due
to intraband excitations exhibits $\sqrt{q}$ dependence and the other two modes disperse linearly,
see Fig. 2(e). The loss function shows larger variations in the peak positions for the first
symmetric mode due to $\sqrt{q}$ behavior at low $q$ as compared to the antisymmetric mode (see
Supplemental Fig. S2 for details [32]). More interestingly, the first symmetric and antisym-
metric bands in the dispersion spectrum [red and blue curves in Fig. 2(e)] intersect and the
symmetric and antisymmetric modes are degenerate for $q > 0.067$ Å$^{-1}$ along $\Gamma$-M. The rea-
son behind this unusual degeneracy is the fine nesting between the bands and consequently
the absence of coupling between the two modes [40].

The higher doping concentration pushes the interband threshold frequency ($\omega_{\text{inter}}$) to
$\approx 2.0$ eV (0.62 $\mu$m) for the first symmetric and antisymmetric modes in G/Li/G/Li/G.
The poles at 0 eV, 0.64 eV, 0.93 eV and 2.5 eV correspond to the three damping regions
associated with intraband, low energy interband, and higher energy electron-hole interband
transitions. Hence, for $1.59$ eV $< \omega < 2.0$ eV (0.62 $\mu$m$< \lambda_{\text{air}} < 0.78$ $\mu$m) the first symmetric
and antisymmetric modes are undamped. More importantly, the second symmetric mode
gets damped at a higher frequency ($\omega > 2.2$ eV), so all three plasmon modes are undamped
and emerge in the optical range for $q < 0.05$ Å$^{-1}$. The $\tau$ in graphene for such high doping
concentration ($E_F = 1.51$ eV) is quite small $\approx 19$ fs. From the Re$[q]$/Im$[q]$ in Fig. 2(f),
we find that the first symmetric mode can be observed further into the mid-infrared range
(from extrapolation) ($\lambda_{\text{air}} > 3$ $\mu$m), whereas the other two modes have shorter wavelengths
($\lambda_{\text{air}} < 0.62$ $\mu$m). $\lambda_p$ is also shrunk by approximately 100 times, Fig. 2(f), as in the case for
bilayer graphene, in agreement with previous reports [41]. We only plot the ratio for the first
symmetric (intraband) mode in Fig. 2(f). Since the antisymmetric and second symmetric
modes disperse linearly, the variation in the Re$[q]$/Im$[q]$ is small. These plasmons exhibit
similar “shrinkage” as that of the symmetric mode (refer to Supplemental Fig. S2 for further
details [32]).

Controlling the number of layers and the concentration of intercalated Li atoms appears to
be a feasible method for engineering the properties of visible plasmons for applications. For
example, the mid-infrared region plasmons in both the bilayer and trilayer Li-intercalated
structures, can be used for plasmonic biosensing [4, 17]. We caution that certain technical
aspects of the calculations reported here, like the choice of exchange correlation functional
for the electronic structure, can affect the electronic spectrum and can shift the plasmon
energies to slightly different values than what we reported; such shifts could change the precise values of the damped plasmon frequencies but we do not expect them to alter the overall picture. Damping due to the presence of defects and substrate phonons, features that were not included in the model of the physical system considered here, can also influence the existence of undamped 2D plasmons in the visible frequency range. A detailed analysis of these parameters will constitute the future scope of this work. Our work can be easily extended to explore other multilayers of other 2D materials (such as black phosphorus, transition metal dichalcogenides) with different dopants and/or intercalants (K, Mg, Na etc), opening up new pathways for fine tuning the plasmon dispersion either by varying the number and type of layers, and/or by varying the concentration and type of intercalant atoms.

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FIG. 1. Atomic structures (side and top views) and electronic structures of: (a) the bilayer Li-intercalated graphene (G/Li/G, left) and (b) the trilayer Li-intercalated graphene (G/Li/G/Li/G, right). The shaded regions in (a) and (b) denote the occupied states, and the dashed black lines the Dirac point / Fermi level in undoped layers.
FIG. 2. Plasmon features for: (a)-(c), the G/Li/G system, and (d)-(f), the G/Li/G/Li/G system. (a) and (d) Plasmon charge density $\rho(\mathbf{r})$ at $q = 0.007 \text{ Å}^{-1}$ for the symmetric modes (blue and green lines) and the antisymmetric mode (red lines); solid lines (thicker and lighter shade) are for results with transparent boundary conditions, dashed lines (thinner and darker shade) for periodic boundary conditions with Coulomb cutoff (see text). (b) and (e) Dispersion relation of plasmons along the $\Gamma$ to $M$ direction; the diameter of the circles is proportional to the strength of the resonance [19]. Shaded areas represent regions of inter- and intra-band losses (including damping by optical phonon). (c) and (f) Re$\{q\}/\text{Im}\{q\}$ (left axis, solid line in blue), and field localization (right axis, dashed line in magenta), or “shrinkage”, of the lowest symmetric mode. $\tau$ is $\approx 29$ fs and 19 fs for the G/Li/G and the G/Li/G/Li/G systems, respectively. The grey shaded areas denote the region of inter-band losses, and the yellow shaded (hatched) areas denote the visible frequency range, calculated with the Fermi velocity of graphene.
Present address: Department of Materials Science and NanoEngineering, Rice University, Houston, TX 77005, USA; sns8@rice.edu
† Department of Physics, University of Crete, PO, Box 2208, 71003 Heraklion, Greece
‡ Faculty of Arts and Sciences, Harvard University, Cambridge MA 02138, USA


[32] Supplemental Material at [URL will be inserted by publisher] for detailed discussion on transparent boundary conditions, calculation of carrier lifetimes and figures for other modes.


