The excitement following the isolation of graphene [1] is due in part to the remarkable optoelectronic properties the material possesses [2, 3]. In addition to a high electron mobility [4] and fast optical response [5], graphene and its bilayer (BLG) exhibit tunable broadband optical absorption [3–9], suggesting applicability to such devices as optical modulators [9, 10], photodetectors [11, 12], and possibly all-optical switches [13–16].

Recent studies have revealed that graphene has a large third-order susceptibility [17, 18], leading to a strong two-photon absorption (2PA) coefficient [19], which is stronger yet in the bilayer system [20] due to the nested manifold of bands present at the K-point of the Brillouin zone. As a result of this K-point band-commensuration and the larger number of absorptive pathways, the 2PA coefficient for the ungapped bilayer system is several orders of magnitude higher than that of monolayer graphene (MLG) in certain frequency ranges [20].

Despite the attractive optoelectronic properties of undoped and gapless graphene systems, the absence of a gap at the K-point precludes MLG from use in many device applications. Multilayer graphene systems, however, do exhibit a band gap when chemically doped or externally gated, permitting a wide range of control over the conductivity. In fact, by simultaneous use of top and bottom gating, the carrier density and gap size can be independently tuned [21], providing a degree of dynamical control of the optical response not available in many optoelectronic materials. Below, we demonstrate that the 2PA of bilayer graphene displays a rich structure as a function of these two parameters. Moreover, as the doping concentration of graphene is very sensitive [22] to synthesis methodology and conditions, a characterization of doped, gapped graphene systems will prove paramount to experimental characterization and successful device design.

At present, a theoretical characterization of the 2PA strength in doped, gapped BLG has yet to be reported, and calculations of the 2PA coefficient [20] in the ungapped system are incomplete since they do not encompass all possible intermediate...
states. Here, we compute using a perturbative approach the full one- and two-photon absorption coefficients for a graphene bilayer with a band gap and a tunable chemical potential. The physical scenario we discuss is a back-gated graphene bilayer placed underneath a transparent top gate (see, for example, [23]), providing simultaneous and independent control over the chemical potential, $\mu$, and the asymmetry gap, $\Delta$. The one-photon absorption (1PA) spectrum for the gapped system has been computed previously by Nicol and Carbotte [24], which we reproduce for comparison to the 2PA spectrum.

An electric field oriented perpendicular to a Bernal-stacked graphene bilayer gives rise to an asymmetry gap [25], for which the tight-binding Hamiltonian in the $K$-valley using the basis (A1, B2, A2, B1) in the sublattice space is

$$\mathcal{H}_{\text{TB}} = \begin{bmatrix} -\Delta/2 & 0 & 0 & \gamma_1 \rho e^{-i\phi} \\ 0 & \Delta/2 & \gamma_1 \rho e^{i\phi} & 0 \\ 0 & \gamma_1 \rho e^{i\phi} & -\Delta/2 & \gamma_1 \\ \gamma_1 \rho e^{-i\phi} & 0 & \gamma_1 & -\Delta/2 \end{bmatrix}$$

We use the following notation: $\phi = \tan^{-1}(p_y/p_x)$ is the polar angle of the in-plane momentum, $\rho \equiv v_F p/\gamma_1$ is the reduced momentum; $\gamma_1 = 0.4$ eV is the A2-B1 interlayer coupling; and $v_F \sim 10^6$ ms$^{-1}$ is the Fermi velocity. The corresponding exact eigenvalues and eigenvectors are

$$\varepsilon_j^\pm = \pm \left( \gamma_1^2 \rho^2 + \frac{\Delta^2}{4} + \frac{\gamma_1^2}{2} + \gamma_1(-1)^j \sqrt{\rho^2(\Delta^2 + \gamma_1^2) + \frac{\gamma_1^2}{4}} \right)^{1/2}$$

and

$$|\psi_j^\pm(\rho)| = \frac{1}{N_j} \begin{bmatrix} 2\gamma_1 \rho \exp(-i\phi) \\ 2\gamma_1^2 + \Delta \\ 2\gamma_1 \rho \exp(i\phi) \\ C_j - 2\gamma_1^2 \Delta \end{bmatrix}$$

where $j = 1(2)$ denotes the low-energy (high-energy) bands split by $\Delta$ at the $K$-point; $N_j$ is a normalization factor; and $C_j \equiv \Delta^2 + \gamma_1^2 + (-1)^j \sqrt{4\Delta^2 \rho^2 + 4\gamma_1^2 \rho^2 + \gamma_1^2}$. For an
The interaction Hamiltonian \[26\] is given by

\[ \hat{H}_{\text{int}} = \frac{e^2}{2mc} \mathbf{A} \cdot \hat{p} = \frac{e^2}{2c} \left[ \mathbf{E} \cdot \hat{v} \right] \hat{v} = \frac{e^2}{2c} \mathbf{E} \frac{\partial \hat{H}_{\text{TB}}}{\partial p_i}. \]  

Assuming a relative permittivity \( \varepsilon_r \approx 9 \) for BLG \[27\], and an incident irradiance \( I = \mathcal{E}_0 \varepsilon_0 \sqrt{\varepsilon_r/2} \), perturbation theory gives the one- and two-photon absorption coefficients \[28, 29\], which are, respectively,

\[ \beta_1 = \frac{2\hbar \omega}{I} 2\pi \sum_{i,f} \left| \langle f | \hat{H}_{\text{int}} | i \rangle \right|^2 \delta(\varepsilon_f - \varepsilon_i - h\omega), \]

and

\[ \beta_2 = \frac{4\hbar \omega}{I} 2\pi \sum_{i,f} \sum_{I} \left| \langle f | \hat{H}_{\text{int}} | I \rangle \langle I | \hat{H}_{\text{int}} | i \rangle \right|^2 \delta(\varepsilon_f - \varepsilon_i - 2h\omega), \]

where \( \beta_2 = 2n\hbar \omega W_n \gamma_n \) is the \( n \)-photon absorption coefficient; \( W_n \) represents the \( n \)-th-order interband transition probability rate per unit area; \( g = 4 \) is a factor accounting for spin and valley degeneracy; and the subscripts label the initial (\( i \)), intermediate (\( I \)) and final (\( f \)) states.

The value of \( \varepsilon_r \) for BLG will vary somewhat according to the choice of substrate, which affects only the irradiance prefactor, \( I/\hbar \), in the \( n \)-photon absorption coefficient, \( \beta_n \).

Figure 1 shows the band structure at the \( K \)-point for a gapped graphene bilayer. The interband 1PA and 2PA transitions are shown, each of which must satisfy the constraints of energy and momentum conservation imposed by the \( \delta \)-functions in the expressions for \( \beta_n \). For the case of 1PA, the thick black arrows in figures 1(a)–(d) denote the four one-photon pathways (T1–T4) permitted from \( \varepsilon_i \rightarrow \varepsilon_f \) for a neutral chemical potential (\( \mu = 0 \)) at a temperature of \( T = 0 \) K. The thick arrow in the fifth panel, figure 1(e), illustrates T5, the transition allowed from \( \varepsilon_i^e \rightarrow \varepsilon_f^e \) when \( \mu > 0 \).

Associated with each 1PA transition is a family of possible 2PA pathways \[30\], which are illustrated in figure 1 by thin red arrows. In the case of 2PA, two photons are absorbed simultaneously via two energy-nonconserving transitions of \( |i\rangle \rightarrow |I\rangle \) and \( |I\rangle \rightarrow |f\rangle \), for which energy is conserved over \( |i\rangle \rightarrow |f\rangle \). For each of the initial and final state combinations, four pathways are possible, all of which are indicated in figure 1. The transitions T3 and T4 displayed in figures 1(c) and (d) are degenerate. A determination of the criteria imposed on T1–T5 by energy conservation requires the calculation of several important energy values, which are provided for reference in figure 1(f) using the nomenclature of Nicol and Carbotte \[24\] and we shall refer to these values below. We confine our analysis to the case \( \mu \geq 0 \) since electron-hole symmetry implies that the response is identical for \( \mu < 0 \).

The diagrams in figure 1 and the denominator of equation (6) allow for several immediate insights into the frequency response of \( \beta_2 \) when \( \Delta = 0 \) and \( T = 0 \). The transitions taking part in the 2PA process for which \( |i\rangle \rightarrow |I = i\rangle \) or \( |I\rangle \rightarrow |f = I\rangle \), denoted by circular arrows, produce a
singularity in $\beta_2$ at $\hbar\omega = 0$. When $\Delta$ becomes large compared to the temperature, this resonance disappears since there are no pairs of initial and final states which satisfy the $\delta$-function. Similarly, contributions for which $\varepsilon_f - \varepsilon_i = 2\hbar\omega = 2\gamma_I$ produce a singularity at $\gamma_I$, and 2PA transitions for which $\varepsilon_f - \varepsilon_i = 2\hbar\omega = \gamma_I$ give rise to a singularity at $\gamma_I/2$. For 1PA when $\mu \geq 0$ and $T > 0$, a singularity exists only due to $T5$, which generates an intense, narrow peak at $\hbar\omega = \gamma_I$ when bands $\varepsilon_I^+$ and $\varepsilon_2^+$ are perfectly nested. This peak will exist for $T > 0$ even when $\mu = 0$ due to thermal population of conduction band $\varepsilon_I^+(\rho)$ and depopulation of valence band $\varepsilon_I^-(\rho)$.

When an asymmetry gap $\Delta > 0$ is present, the conditions imposed by energy conservation become more complex [24] due to the so-called ‘sombrero’ structure [25] of the low-energy bands $\varepsilon_I^\pm$. The bands are not perfectly nested as they are for $\Delta = 0$, leading to the rich absorptive structure displayed in figure 2. Figure 2 shows color plots of both the 1PA and 2PA coefficients for a wide range of gap values. Note that the highest value of $\Delta = 1.5$ is probably unattainable using current experimental techniques, but we wish to display the effects when $\Delta \geq 1$ since several spectral features change. A transformation of $\omega \rightarrow \omega + \Gamma/\hbar$ accounts for thermal broadening and we take $\Gamma = 5.4 \times 10^{-3}\gamma_I$, which corresponds to a temperature of $T = 25$ K.

The thermal broadening leads to the emergence in figure 2(a) of an intense, narrow peak at $\hbar\omega = \gamma_I$ due to $T5$ for (see footnote $^5$) $\Delta = 0$. At $T = 25$ K, band $\varepsilon_I^+$ receives spectral weight due to thermal population, giving rise to $T5$ transitions of $\varepsilon_I^+ \rightarrow \varepsilon_I^\pm$. Similarly, in the $\beta_2$ spectrum for $\Delta = 0$ in figure 1(b), the thermal smearing of $\mu$ leads to a peak at $\hbar\omega = \gamma_I/2$, as anticipated during the analysis of figure 1. When $\Delta$ is small, the $1/\Omega$ divergence in the 2PA is caused by the resonance near the center of the Brillouin zone. This is not present in the 1PA because it is cancelled by the absorption matrix element. When $\Delta$ increases, the $T5$ component of both

Figure 3. 1PA (left column) and 2PA (right column) for the indicated value of the gap, $\Delta$. The solid, dotted, and dashed curves correspond to the identical lines superimposed on the dispersion curves in the insets in the right-hand column, indicating the absorption for chemical potentials of $\mu = 0$ (continuous black line), $\mu = (E_{g1} + E_{g0})/2$ (dotted red line), and $\mu = (E_{g1} + E_{g0})/2$ (dashed blue line).

For plots labeled $\Delta = 0$, we employed a gap value of $\Delta = 10^{-5}\gamma_I$ in the corresponding computations.
$\beta_1$ and $\beta_2$ broadens into the cleaver-shaped regions outlined in green for $\Delta = \gamma_1$ in figures 2(a) and (b). The asymmetric broadening of the T5 region is due to the sombrero shape of $\epsilon_1^\pm$ and the loss of spectral weight when $E_0^2 + \mu > 0$. The panels corresponding to $\Delta = \gamma_1$ in figures 2(a) and (b) identify the energy thresholds mapped in figure 1(f), illustrating regions where the chemical potential either blocks or permits absorptive pathways. For $\Delta > 0$, the band gap means that there is no absorption at $\omega < \Omega_1$ and the increased density of states at the band edge produces regions of increasing 1PA and 2PA near the T1 cutoff. The other spectrally empty region that emerges with increasing $\Delta$ occurs between the T3/T4 and T5 pockets.

In contrast with the $\epsilon_1 - \omega = \hbar \omega$ requirement of 1PA, 2PA requires $\omega_i - \omega_f = 2 \hbar \omega$, which compresses and shifts the T1–T5 regions in $\beta_2$ relative to those $\beta_1$. Also, equation (6) shows that the resonances will be stronger in the 2PA due to the additional factor of $(\hbar \omega - \epsilon_f - \epsilon_i)^2$ in the sum over intermediate states. The spectra for $\beta_2$ shown in figure 2(b) contain a prominent resonance not present in $\beta_1$. This $\beta_2$ resonance, arising at $\Omega = 1$ when $\Delta = 0$, is due to transitions T3 and T4, which contain denominators of $\hbar \omega - \gamma_1$ and give rise to a singularity. As $\Delta$ increases, this resonance shifts toward higher $\Omega$ due to the increasing size of the band gap. At this T3/T4 resonance, the BLG 2PA strength is several orders of magnitude larger for bilayer graphene than for monolayer graphene [20].

Figure 3 presents 1PA and 2PA curves extracted from the color maps of figure 2, grouped by gap value. The left-hand column shows $\beta_1(\Omega, \mu)$, and the right-hand column shows $\beta_2(\Omega, \mu)$. For each plot of $\beta_1$ and $\beta_2$, several values of $\mu$ are chosen, each of which corresponds to an identical horizontal line on the accompanying dispersion shown in the inset to the $\beta_2$ plots. The solid black curves are for $\mu = 0$; the dotted red is for $\mu = (E_{g1} + E_{g2})/2$ which is where the chemical potential is in the middle of the sombrero region; the dashed blue line is for $\mu = (E_{g1} + E_{g2})/2$, and lies halfway between $\epsilon_1^+$ and $\epsilon_2^+$ at $\mu = 0$. When $\Delta = 0$, the dotted red slice is equivalent to that of $\mu = 0$, so that the solid black and dotted red lines are identical. As $\Omega \to \infty$, the contributions to $\beta_2$ of transitions T1–T4 tend to $1/\Omega^2$.

Figure 4 reveals an intense, narrow peak in $\beta_2$ that emerges when $\Delta \geq \gamma_1/\sqrt{2}$, resulting from two $|i\rangle \to |f\rangle$ routes within the family of 33 and 44 pathways. In particular, routes $\epsilon_1^+ \to \epsilon_2^+$ and $\epsilon_1^- \to \epsilon_2^-$ lead to a vanishing denominator $\epsilon_f - \epsilon_i - \hbar \omega$ in equation (6) at values of $\Omega_4$ at $\omega_i = \epsilon_2^+$, $\omega_f = \epsilon_1^+$, and $\omega = \epsilon_2^- - \epsilon_1^- - \hbar \omega$ in equation (6) at values of $\Omega_4$ at $\omega_i = \epsilon_2^+$, $\omega_f = \epsilon_1^+$, and $\omega = \epsilon_2^- - \epsilon_1^- - \hbar \omega$.
When $\Omega = \Omega_0$ and $\Delta \geq \gamma_0 \sqrt{2}$ for $\Gamma = 0$, the position of the peak, $\Omega_0$, eclipses the T3/T4 cutoff, causing the peak to appear when the photon energy $\hbar \omega = \Omega_0$ coincides with $\epsilon_1^0 - \epsilon_1^0 \geq (E_0) + (E_Q)/2$ and $\epsilon_2^0 - \epsilon_2^0 \geq (E_0) + (E_Q)/2$.

Figures 4(a) and (b) show $\beta_1$ and $\beta_2$ as a function of $\Omega$ and $\Delta$ for $T = 25$ K and $\mu = 0$. As $\Delta$ increases from 0, certain regions of increasing intensity materialize in both 1PA and 2PA spectra, including the peak in $\beta_2$ at $\Omega_0$, which is outlined in $\mu_0$. The inset in figure 4(b) provides a zoomed-in view of the boxed region, in which the dotted black line indicates the T3/T4 cutoff, and the dashed blue line shows the onset of the emerging peak. Thus far, the assumed thermal broadening corresponds to a temperature of 25 K—a value large enough to reveal the T5 resonance when $\mu = 0$, yet small enough to avoid the smearing of finite features. Figures 4(c) and (d) illustrate the effect of thermal broadening on $\beta_1$ and $\beta_2$ for $\Delta = 0.2 \gamma_0$ at neutral doping ($\mu = 0$). The most pronounced impact of increasing temperature is the broadening of $\beta_1$ and $\beta_2$ into the gap region. Once $\Gamma$ reaches room temperature, the otherwise abrupt T3/T4 cutoff just beyond $\Omega = 1$ spreads into a more diffuse spectral bulge. Similarly, the sharp $\beta_2$ resonance residing near $\Omega = 1$ at $T = 10$ K suffers appreciably within the temperature range examined, dropping in intensity by more than two orders of magnitude when $T = 300$ K.

We have demonstrated that when an electric field is applied perpendicular to a graphene bilayer, the resulting asymmetry gap gives rise to complex linear and nonlinear optical absorption. This is experimentally verifiable in a simple photoabsorption measurement using transparent gates. For the gapped bilayer system with a tunable chemical potential, we have calculated both one- and two-photon absorption spectra over an expansive range of the gap and chemical potential, taking into account all possible absorption pathways in the calculation of $\beta_2(\Omega, \mu)$. We analyze the 2PA resonances that emerge in the gapped, doped bilayer system, and examine the evolution of these resonances as a function of $\Delta$ and $\mu$. The effects of thermal broadening are incorporated into the computations, providing insight into the degradation of optical performance at or near room temperature. As graphene-based optical architectures mature, the absorption spectra calculated above will prove important for optimizing and enhancing device performance.

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