Determining graphene's induced band gap with magnetic and electric emitters

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We present numerical and analytical results for the lifetime of emitters in close proximity to graphene sheets. Specifically, we analyze the contributions from different physical channels that participate in the decay processes. Our results demonstrate that measuring the emitters’ decay rates provides an efficient route for sensing graphene’s optoelectronic properties, notably the existence and size of a potential band gap in its electronic bandstructure.

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Driven by its successful isolation, graphene has not stopped fascinating the research community. Although, this allotropic form of carbon had been theoretically investigated for decades, experimental access to graphene has offered new perspectives as well as novel directions for fundamental research and technological applications [1, 2]. Graphene’s exotic properties [3] have lead to the investigation of a wide range of phenomena such as ballistic transport [4], the quantum Hall effect [1, 5], and thermal [6] as well as electrical conductivity [7, 8]. Developing a detailed understanding, followed by appropriate engineering of these properties, lies at the heart of future graphene-based technologies. For this, an accurate determination of graphene’s properties in realistic experimental settings and the detailed validation of various theoretical models (cf. Ref [7, 9–11]) is indispensable. Promising designs where the semi-metal will play an important role, aim at combining condensed-matter with atomic systems. Such hybrid devices are geared towards reaping the best of the two worlds for advanced high-performance devices.

In this work, we demonstrate how the high degree of control and accuracy available in quantum systems like cold atoms and Si- and NV-centers in nano-diamonds, can be employed for detailed investigations of graphene’s optoelectronic properties [12–15]. Specifically, we focus on modifications in the life times of emitters held in close proximity of graphene layers and show that these allow for direct experimental access to features like band gaps as well as plasmons and/or plasmon-like resonances. In graphene, a band gap $\Delta$ (cf. Fig. 1) is created (i) when the atomically thin material is deposited on a substrate [16, 17], (ii) when strain is applied, (iii) when impurities are present, and (iv) in cases where graphene bilayers instead of a single layer are considered. Values for $\Delta$ of the order of tens of meV have been predicted [16, 17], thus triggering corresponding experimental investigations. These band gaps and the features connected with them are still the subject of discussions [18, 19] so that reliable experimental means for their analysis are highly desirable.

For planar geometries the decay rate of an emitter is a functional of the system’s optical scattering coefficients. We model a monoatomic graphene layer in terms of a 2+1-dimensional Dirac fluid [10, 20, 21] and embed it in a non dispersive and non dissipative dielectric medium with permittivity $\varepsilon_m$. As a result, the graphene layer is characterized by an induced band gap and a chemical potential $\mu = 0$ (cf. Fig. 1) while the corresponding electromagnetic reflection coefficients for transverse magnetic (TM) and transverse electric (TE) waves are [20, 21]

$$r_{\text{TM}} = -\frac{\alpha \Phi(y)}{y\varepsilon_m/\kappa_m - \alpha \Phi(y)} , \quad r_{\text{TE}} = -\frac{\alpha \Phi(y)}{\kappa_m + \alpha \Phi(y)} . \quad (1)$$

where $\alpha = 137^{-1}$ is the fine structure constant and

$$\Phi(y) = 1 - (\sqrt{y} + 1/\sqrt{y}) \arctanh (\sqrt{y}) , \quad (2)$$

with $y = \omega^2 - v_F^2 k^2$. Further, $\kappa_m = \sqrt{k^2 - \varepsilon_m \omega^2}$ and $k = \sqrt{k_x^2 + k_y^2}$ denote, respectively, the moduli of the

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FIG. 1: Schematic of the physical situation considered in this work. An emitter (red sphere) is positioned at distance $z_0$ from a graphene sheet. Graphene’s bandstructure is approximated by $E_{\pm} = \pm \sqrt{\Delta^2 + \varepsilon_m k^2}$ (see inset) and the chemical potential is chosen as $\mu = 0$ (yellow: filled band).
out-of-plane and in-plane wave vectors in the dielectric medium. In addition, we use dimensionless variables, which amounts to the replacements $\hbar \omega / 2\Delta \rightarrow \omega$, $\hbar k / 2\Delta \rightarrow k$, and $v_F / c \rightarrow v_F$ ($\approx 300^{-1}$ for graphene). Life time modifications are usually associated with the strength of scattering processes. Owing to its minute thickness (few Å), the optical response of a single graphene layer is rather small ($\sim 2\%$ reflection [27]). Thus, for emitters near a graphene layer, small life time modifications might naively be expected. However, graphene’s exotic properties introduce additional features that affect the emitters’ dynamics, such as TE plasmons and single (SPE)- and multiple (MPE)-particle excitations.

Different frequencies are associated with the different physical processes: propagating fields occur for $0 \leq k < \omega / \sqrt{\varepsilon_m}$ and evanescent fields are characterized by $k > \omega / \sqrt{\varepsilon_m}$. Further, we identify another regime where $k < \omega^2 / 2\varepsilon$ and only exists if $\omega > 1$, i.e., if the radiation frequency exceeds that associated with the band gap. In this regime, the propagating regime of Dirac fluid model of graphene features the creation of electron-hole pairs. In the propagating regime, the scattering process in graphene systems is very similar to that in ordinary thin films. This similarity, however, already breaks down for evanescent waves, for which the scattering process is associated with surface plasmons or plasmon-like phenomena. While in ordinary materials these resonances are usually present only in TM polarization, graphene is associated with surface plasmons or plasmon-like phenomena: While in ordinary materials these resonances are dominated by the magnetic field. Conversely, TE plasmons result from resonances in the motion of the current density so that they are dominated by the electric field. Conversely, TE plasmons result from resonances in the motion of the current density so that they are dominated by the magnetic field.

Mathematically, these phenomena are related to the matrix elements that only exist if $\omega > 1$, i.e., if the radiation frequency exceeds that associated with the band gap. In this regime, the propagating regime of Dirac fluid model of graphene features the creation of electron-hole pairs. In the propagating regime, the scattering process in graphene systems is very similar to that in ordinary thin films. This similarity, however, already breaks down for evanescent waves, for which the scattering process is associated with surface plasmons or plasmon-like phenomena. While in ordinary materials these resonances are usually present only in TM polarization, graphene is associated with surface plasmons or plasmon-like phenomena: While in ordinary materials these resonances are dominated by the magnetic field. Conversely, TE plasmons result from resonances in the motion of the current density so that they are dominated by the electric field.

In order to analyze the above terms in more detail, we will first discuss the case of magnetic decay keeping in mind that a magnetic emitter ought to be more sensitive to the magnetic field associated with plasmonic TE resonances. The emitter has a transition frequency $\omega_0$ and is located at $z = z_0 > 0$ above the graphene layer at $z = 0$ (see Fig. 1). Within second-order perturbation theory [30, 31] the modification of the decay rate can be written as

$$\Gamma_{\text{TM}} = \frac{3}{4} \int_{-\infty}^{\infty} dy \text{Im} \left[ \frac{r_{\text{TM}}}{k_s[y]} \right] \frac{k_s[y]}{2k_0 v_F} \exp \left( -2d K_s[y] / v_F \right),$$

$$\Gamma_{\text{TE}} = \frac{3}{2} \int_{-\infty}^{\infty} dy \text{Im} \left[ K_s[y] \right] \left( \frac{2d K_s[y]}{k_0 v_F} \right) \exp \left( -2d K_s[y] / v_F \right).$$

Here, $k_0 = \omega_0 \sqrt{\varepsilon_m}$, $d = 2z_0 \Delta / \hbar c$. We have also defined $k_s[y] = \sqrt{\omega_0^2 / \varepsilon_m - y}$ and $K_s[y] = v_F k_s[y] = \sqrt{\omega_0^2 / \varepsilon_m - y}$. In Eqs. (4), the evanescent contribution is associated with the range $-\infty \leq y \leq (\omega_0 / \omega_k)^2$, while the $(\omega_0 / \omega_k)^2 \leq$
Therefore, the integrands can be expanded around $y = \omega_0^2 (\ll 1)$ corresponds to the propagating region. The SPE range corresponds to $1 < y < \omega_0^2$.

We first consider the contribution to the decay rate from the evanescent range, imputable only to the resonance in the reflection coefficients. In view of the above discussion of the dispersion relation, Eqs. (3), this contribution features two different regimes. For $\omega_0 < \omega_k$, i.e., when the dispersion curve is very close to the light cone, the resonance is located at $y_p \approx (\omega_0/\omega_k)^2 [1 - (4\epsilon v_F / 3)^2 (\omega_0/\omega_k)^2]$. The leading terms of Eqs. (4) are then

$$\Gamma_\parallel \approx 16\alpha^3 \pi \omega_0^3 \exp(-d/d_0), \quad \Gamma_\perp \approx 2\pi \alpha \omega_0 \sqrt{\epsilon_m} \omega_k^2 \exp(-d/d_0). \quad (5)$$

Given the rather large characteristic decay length $d_0 = [3\epsilon_\omega \omega_k^2/(8\alpha)] k_0^{-2}$, these contributions exhibit weak distance-dependencies for experimentally relevant emitter-graphene separations of a few microns. For $\omega_0 > \omega_k$, the resonance is instead located close to the boundary of the SPE region, $y_p \approx 1 - 2 \exp[-(1 + K_s[1]/(\alpha v_F))]$ and we obtain

$$\Gamma_\parallel \approx 3\pi K_s[1] e^{-\left(\frac{(1+K_s[1])}{\alpha v_F k_0^3}\right)} \exp(-2d K_s[1]/v_F), \quad (6a)$$

$$\Gamma_\perp \approx 3\pi K_s[1] e^{-\left(\frac{(1+K_s[1])}{\alpha v_F k_0^3}\right)} \exp(-2d K_s[1]/v_F). \quad (6b)$$

Due to the small values of $v_F$ and $\alpha$, the above terms are strongly suppressed in graphene unless $K_s[1] \sim 0$, which only occurs when $\omega_0 \sim \omega_k \gg 1$.

For the same parameters, the propagating regime corresponds to a rather small integration range in Eqs. (4). Therefore, the integrands can be expanded around $y = \omega_0^2$ and after some rearrangements we obtain

$$\Gamma_\parallel \approx \frac{\alpha (\epsilon_m + 1)}{2\epsilon_m d} \left[ \frac{4\alpha k_0 (\epsilon_m + 1)}{9\epsilon_m (\epsilon_m + 1)} \sin(2k_0 d) + \frac{\sin(2k_0 d)}{2k_0 d} - \cos(2k_0 d) \right], \quad (7a)$$

$$\Gamma_\perp \approx -\frac{\omega_k^2 d}{2d_0} \int_0^1 d\zeta \left[ 1 - \frac{\zeta}{2k_0 d_0} \right]^2 \left[ \frac{\zeta \sin^2(\zeta)}{2k_0 d} + \frac{\omega_k^2 \cos^2(\zeta)}{2k_0 d_0} \right]$$

$$d \ll k_0^{-1} \approx -\frac{\pi \alpha}{\sqrt{\epsilon_m}} \omega_0 \left( 1 + \frac{8k_0 d}{3\pi} \right). \quad (7b)$$

Interestingly, because of the overall minus sign of $\Gamma_\perp$, this contribution tends to increase the emitter’s life time, suppressing the decay process relative to $\gamma_0$. In addition, since $k_0^{-1} \ll d_0/\omega_0^2$, due to the dephasing between the propagating waves, $\Gamma_\perp$ exponentially decays for distances $d \gtrsim d_0/\omega_0^2$. It follows a behavior similar to the TE plasma but with characteristic decay length $d_0/\omega_0^2$. Therefore, since $\omega_0 \sim 1$, $\Gamma_\perp$ is almost exactly canceled by $\Gamma_\parallel$ (see Fig. 3(b)). For even larger distances ($d \gg d_0/\omega_0^2$, not shown), due to the interference between incoming and scattered waves, $\Gamma_\perp$ oscillates in space like $\Gamma_\parallel$ with a frequency $2k_0$ (see Fig. 3(c)).

Finally, we consider the modification of the decay rate that stems from the SPE region. This contribution only occurs when the emitter’s transition frequency becomes larger than the electronic band gap ($\omega_0 > 1$). Although, the total SPE region includes both evanescent and propagating contributions, the non-radiative part dominates at short distances and, as in the previous case, is almost constant for $d \ll k_0^{-1}$. Again, since $\alpha, v_F \ll 1$, in this limit we can write

$$\Gamma_{SPE} \approx \frac{\alpha \pi}{4 v_F^2 \epsilon_1 \omega_k^3} \left[ 1 + 3 \left( \frac{\omega_k}{\omega_0} \right)^2 - 4 \left( \frac{\omega_k}{\omega_0} \right)^3 \right], \quad (8)$$
This demonstrates that $\Gamma^\perp_{\text{SPE}}$ varies non-monotonously with frequency and exhibits a maximum for $\omega_0 = 2\omega_g$, where it takes the value $\Gamma^\perp_{\text{SPE}} \approx 645$. At intermediate distances, the total (evanescent and propagating) SPE contribution decays as a power law, $\Gamma^\perp_{\text{SPE}} + \Gamma^\parallel_{\text{SPE}} \approx 2(\Gamma^\perp_{\text{SPE}} + \Gamma^\parallel_{\text{SPE}}) \approx \alpha \pi \omega g (\omega_0^2 + 1)(6\omega_0d)^{-2}$ (see Fig. 3(d)). For $d \gg k_0^{-1}$, the propagating waves induce once again spatial oscillations with frequency $2k_0$ (not shown).

In Fig. 4(a) we present the frequency dependence of all the above-discussed contributions to the decay rate at a fixed distance $d = (3 \cdot 10^8)^{-1}$ from the graphene layer (corresponding to $z_0 = 1 \mu m$ for an emitter with transition frequency of 1 MHz). As discussed above, for emitters with transition frequencies smaller than graphene’s electronic band gap ($\omega_0 < 1$), the two main decay channels are the TE plasmonic resonance and the radiative decay. Their relative importance differs, depending on the spatial orientation of the dipole-matrix elements. We see that in $\Gamma^\perp$ the plasmonic TE resonance provides an enhancement while the radiative contribution leads to a suppression. Also, $\Gamma^\perp \approx -2\Gamma^\parallel_{\text{SPE}}$ over a very large range of frequencies. For $\Gamma^\parallel$, the radiative contribution dominates and leads to an enhancement of the decay rate. In this case, the plasmonic TE resonance, due to its proportionality to $\omega_0^3$, represents a subleading contribution. For $\omega_0 > 1$, the dominant contribution for both $\Gamma^\perp$ and $\Gamma^\parallel$ stems from the SPE contribution (see Fig. 4(a), inset) and leads to an enhancement of the decay rate by three orders of magnitude. Note, that the increase of the decay rate occurs quite abruptly as the frequency of the emitter moves across the band gap and, for larger frequencies, takes on a weakly frequency-dependent value around $\alpha \pi / (4r_F \omega_0^3 m^2)^{3/2} \approx 10^9$. In both $\Gamma^\perp, \Gamma^\parallel$, the TE contributions are dominant and lead to the non-monotonic behavior discussed above.

Most of the above-described characteristics also qualitatively apply to the case of an electric dipole emitter (see Fig. 4(b)). Indeed, the relevant expressions can be easily obtained by swapping the reflection coefficients in Eqs. (4) [30, 32]. For brevity we will only mention that, as a consequence of the replacement $r^\text{TM} \leftrightarrow r^\text{TE}$, some features are found in $\Gamma^\parallel$ instead of $\Gamma^\perp$. Curiously, for $\omega_0 < \omega_g$, due to the proximity of the TE plasmonic dispersion relation to the light cone, its contribution to the decay rate is of the same order of magnitude for both emitters and for all distances, i.e., $\Gamma^\parallel \approx 2\Gamma_{\text{magn}}^\text{magn} / \sqrt{\varepsilon_m}$. More importantly, the SPE channel still provides a large enhancement of the decay rate for $\omega_0 > 1$, featuring again a quite abrupt jump for frequencies near the electronic band gap of graphene. However, for the electric emitter both $\Gamma^\parallel, \Gamma^\perp$ exhibit a monotonous frequency dependence.

In conclusion, the above results suggest atomic or atom-like emitters as sensitive quantum probes to determine the physical properties of graphene and, in particular, to investigate a band gap in its electronic bandstructure. Using these systems allows for an accurate analysis of this quantity, especially in complex (but relevant for graphene-based technologies) situations where it is no longer spatially homogenous: This occurs, e.g., when the sheet (i) is exposed to mechanical stress [33], (ii) is positioned on an inhomogeneous substrate or (iii) absorbs impurities (in a controlled [34] or uncontrolled fashion). In our approach, the emitter non-invasively probes graphene’s properties in different physical regimes, enabling experimental investigation of unusual graphene properties such as TE surface resonances (see also [12, 13]) and providing results complementary to those accessible when using other procedures. In addition, the possibility to engineer different internal quantum states of the emitter and study their lifetimes can also offer new opportunities which are presently not accessible with other techniques. As a concrete experimental approach, we suggest to extending the known use of microtrapped Bose-Einstein condensates [35, 36] to map the local band gap structure of graphene sheets with micron resolution. One would detect the spin flip rate by measuring the spatially dependent spin population after a known time since its preparation as a spin-polarized gas. For enhanced sensitivity, it will be advantageous to employ an optical dipole trap, ideally configured as a light sheet, tuned to a frequency below the main atomic transition. Fluorescence imaging following selective resonant excitation of the emitter decay target state will en-
able the measurement of even very slow decay rates down
to a few events per time across the ensemble of typically 10^5 atoms. The high temporal resolution of this tech-
tique can offer an important advantage in analyzing the
different (relatively slow) processes cited above.

In addition to atomic quantum gases other very well suited candidates are Si- and NV-centers in nano-
diamonds. They do not only show tunable magnetic and
electric transitions from the MHz to the THz frequency
range but also simultaneously allow for high position res-
olution [37]. Small band gaps can be investigated by cooling
the system to the mK regime, such that magnetically
tunable Zeeman [38] or hyperfine transitions [39] can be
utilized. Our work can open additional pathways to en-
tangle Zeeman [38] or hyperfine transitions [39] can be
ing the system to the mK regime, such that magnetically

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