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Graphene-covered 1D photonic crystals enabling TE-polarized graphene modes

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ABSTRACT

We present the design of a photonic crystal-based multilayer structure that allows to experimentally demonstrate, using attenuated total reflectance experiments, the existence of the predicted transverse electric (TE) polarized excitation in graphene. We show that this mode can be excited in a single layer of graphene, even at room temperature. Furthermore, we prove that the observed mode in the reflection spectra corresponds to the TE-polarized graphene excitation and not the Bloch Surface Wave of the photonic crystal experiencing graphene-induced loss. Finally, we point out that adding an extra layer of dielectric material on top of the structure would ensure the unambiguous identification of the TE graphene mode even in the presence of fabrication errors.

Keywords: Graphene, photonic crystals, plasmonics

1. INTRODUCTION

Because of its novel electronic and optical properties, graphene has generated much interest over the past decade.\footnote{The fact that these properties can be tuned by changing the graphene’s chemical potential $\mu$ - by means of electrical gating or chemical doping - opens up a whole variety of possible graphene-based applications.} The fact that these properties can be tuned by changing the graphene’s chemical potential $\mu$ - by means of electrical gating or chemical doping - opens up a whole variety of possible graphene-based applications.\footnote{Whereas intrinsic graphene with chemical potential $\mu = 0$ absorbs all the wavelengths from the far IR to the deep-UV with the same efficiency due to its linear electron dispersion, tuning $\mu$ to a value different from zero eliminates interband absorption for photon energies $h\omega < 2|\mu|$. In the latter regime, graphene obtains relatively low optical losses, making it an interesting building block for applications where strong but low-loss light-matter interactions are desired, such as in the field of plasmonics. For $h\omega < 1.667|\mu|$, the graphene conductivity $\sigma$ has a positive imaginary part $\sigma''$ and thus acts as a metal that supports transverse-magnetic (TM) propagating surface plasmon-polaritons (TM-SPPs) with very high field confinement. On the other hand, $\sigma''$ becomes negative in the narrow spectral region of $1.667|\mu| < h\omega < 2|\mu|$, thus indicating that for these optical wavelengths, graphene acts as a thin dielectric layer supporting low-loss transverse electric (TE) surface plasmon-polaritons (TE-SPPs).}

While the TM-SPPs have been characterised theoretically and experimentally, so far the existence of the TE-SPPs has been shown to exist only theoretically.\footnote{The use of an Otto configuration to excite the TE-SPPs in single and multi-layer graphene at cryogenic temperatures and room temperature respectively has been suggested by Mason et al. However, one should have sufficient control over the gap between the graphene sheet(s) and the prism to detect the excitation and it would be advantageous to be able to observe the TE-SPP at room temperature in a single layer of graphene.}

We therefore propose a novel way to overcome the problems that trouble the Otto configuration experiments: placing the graphene sheet(s) directly on a carefully designed photonic crystal (PC) and exciting the mode from the substrate below with a laser source. The proceeding is organised as follows: in Section 2, we detail the design strategy of a PC with which the TE-SPP can be excited in a single layer of graphene at room temperature and show that the mode can be clearly identified in the reflection spectra from attenuated total reflectance (ATR).
experiments. In Section 3, we prove that the excitation observed in the ATR spectra corresponds to a true TE-SPP rather than the Bloch Surface Wave (BSW) excitation that also exists in the multilayer. For this proof, we rely on the band diagram of the PC and the dispersion relations of all the modes involved. In Section 4, we show that the deposition of an extra layer of dielectric material on top would further facilitate the experimental observation of the graphene TE-SPP at room temperature. We also point out that the extra layer would make the structure more robust to fabrication errors. Finally, in Section 5, we conclude.

2. DESIGN OF THE STRUCTURE

To design a structure to excite TE-SPPs, we first recall from waveguide theory that optical modes in a thin dielectric slab can only survive if the difference between the material refractive indices of the cladding above and substrate below the dielectric slab is very small. This also applies to the TE-SPPs in a graphene sheet as they are of the same physical nature as the TE-polarized modes in the thin dielectric slab. To identify what substrate indices are allowed for a certain cladding, we calculate the maximal permissible optical asymmetry between an air cladding and a dielectric substrate with refractive indices \( n_0 \) and \( n_1 \geq n_0 \) respectively. The graphene conductivity can be written as \( \sigma = \sigma' + i\sigma'' \), and is dependent on the excitation wavelength \( \lambda \), the temperature \( T \), the chemical potential \( \mu \), and the scattering relaxation times \( \tau_{\text{intra}} \) and \( \tau_{\text{inter}} \) for the intra-band and interband scattering respectively. We take \( \tau_{\text{intra}} \) and \( \tau_{\text{inter}} \) equal to 10 fs and 1200 fs as reported by Gu et al.\(^\text{10}\) The graphene conductivity can then be calculated using expressions presented earlier.\(^\text{7,8,11}\)

For lossless graphene, \((\sigma' \to 0)\), the dispersion relation of the TE-SPPs in a single graphene sheet can be written as:\(^\text{7}\)

\[
\sigma'' = -\alpha_0 \left( \sqrt{n_{\text{eff}}^2 - n_0^2} + \sqrt{n_{\text{eff}}^2 - n_1^2} \right)
\]  

(1)

with the effective index \( n_{\text{eff}} = k_x \lambda/(2\pi) \) real and with \( k_x \) the wave number of the TE-SPP projected on \( \hat{x} \) (see Fig. 1). Since TE-SPPs are propagating along the graphene sheet, i.e. the fields must be evanescent in the substrate and cladding, we have \( n_{\text{eff}} > n_1 \geq n_0 \), and \( \sigma'' < 0 \). To find out how large a \( |\sigma''| \) is required for a certain asymmetry, we take the lower limit for \( n_{\text{eff}} \), namely \( n_{\text{eff}} = n_1 \) in eq. (1), so that we obtain \( |\sigma''| > \alpha_0 \sqrt{n_1^2 - n_0^2} \). For a symmetric environment, a TE-SPP can be realised for any \( |\sigma''| > 0 \), but introducing a slight asymmetry will require larger \( |\sigma''|\)-values that might not be achievable in fabricated graphene layers. As
Figure 2. $|R_N|^2$ at $\lambda = 1.31 \mu m$ as a function of the incident angle $\theta$ for the setup without graphene coverage (dotted lines) and with graphene coverage (full lines). The blue and red color indicate the absence or the presence of an additional PMMA layer respectively. The inset also indicates the angles $\theta_{TIR}$ and $\theta_{TE-SPP} = \theta_{min}$.

A case study, we consider the scenario of a graphene sheet with a relatively high chemical potential of $\mu = 0.5$ eV. Choosing the wavelength $\lambda = 1.31 \mu m$ in the region where the TE-SPPs can propagate, we obtain a value of $\sigma = (0.310 - i0.457) \sigma_0$ at room temperature ($T = 300$ K) with $\sigma_0 = e^2/4\hbar$ being the intrinsic conductivity of graphene. For this wavelength, the refractive index of air is $n_0 = 1.000274$. Neglecting $\sigma'$, the largest possible refractive index difference that is able to sustain TE-SPPs in the graphene sheet is given by $n_1 - n_0 = 5.5 \times 10^{-5}$. It is virtually impossible to find a solid substrate material with a refractive index $n_s$ that close to the $n_0$ of air. Including the loss term $\sigma'$ will even further lower the maximally allowed asymmetry between the cladding and substrate.

A solution to this problem is to use a PC, the optical response of which 'mimics' that of the cladding around the excitation wavelength $\lambda$ and effective index $n_{eff}$ of the TE-SPP in a graphene sheet fully embedded in a symmetric environment. Such a strategy, which has already been employed for detecting long range surface plasmons in thin metal films, yields the desired effective symmetry required to sustain TE-SPPs. The latter can then be detected by exciting the structure with light at a tuneable wavelength or tuneable incident angle through a substrate (see Fig. 1), and measuring the dips in the reflection spectrum.

We again consider the single sheet of graphene described above, for which the TE-SPP in the symmetric environment has a complex effective index of $n_{eff}^{TE-SPP} = n_0 + (0.7 + i1.9) \times 10^{-5}$. As shown in Fig. 1, we consider placing the graphene sheet on a (finite) PC with unit cells consisting of 2 different layers with thicknesses $d_a$ and $d_b$ and refractive indices $n_a$ and $n_b$, made of SiO$_2$ and TiO$_2$ respectively, a commonly used combination for the design of optical filters. We take the number of PC unit cells $N_{pc} = 5$. Finally, the PC is placed on top of a SiO$_2$ substrate. Next, we use a T-matrix algorithm and the values for $n_a$ and $n_b$ given in Ref. 12 to calculate the values of $d_a$ and $d_b$ where the optical symmetry that the TE-SPP experiences is maximised and where the TE-SPP can be clearly identified in the ATR spectra around $\Re \left\{ n_{eff}^{TE-SPP} \right\} = n_0 + 0.7 \times 10^{-5}$, corresponding to an angle of incidence in the substrate of $\theta_{TE-SPP} = \sin^{-1} \left( \Re \left\{ n_{eff}^{TE-SPP} \right\} / n_s \right) = 43.7389^\circ \approx 43.74^\circ$, where $n_s$ is the refractive index of the substrate.
The optical symmetry is achieved at the values in \((d_a, d_b)\)-space for which the reflection of an incident laser beam from the cladding on the PC vanishes, while the TE-SPP in the ATR spectra is most visible when the dips in the ATR spectra go to \(|R_{N1}|^2 = 0\), corresponding to maximal coupling towards the TE-SPP of light incident from the substrate. We note that these two conditions cannot both be perfectly satisfied due to the finiteness of the PC and the coupling with the substrate, so we find an optimal balance between the two conditions and calculate \(d_a\) and \(d_b\). For \(N_{per} = 5\), we end up with a PC unit cell with the following dimensions: \(d_a = 212.0\, \text{nm}\) and \(d_b = 206.1\, \text{nm}\).

We now plot the reflectivity of light incident from the substrate, \(|R_{N1}|^2\), in Fig. 2 as a function of the incident angle at the fixed wavelength of \(\lambda = 1.31\, \mu\text{m}\) in the cases where the graphene sheet is absent or present on the designed PC. We observe a dip in the \(|R_{N1}|^2\) spectrum when the graphene sheet is present, indicating the excitation of the TE-SPP, contrary to the case where the graphene sheet is absent. From the inset in Fig. 2, we can observe that the reflectivity minimum at \(\theta_{min}\) nicely overlaps with the angle where the TE-SPP is excited, \(\theta_{TE-SPP}\), showing that the device’s operation is indeed optimised around \(\Re \left\{ n_{eff}^{TE-SPP} \right\} \). Rather than performing an angular sweep, one could also observe the \(|R_{N1}|^2\) spectrum at the fixed angle \(\theta_{min} \approx 43.74^\circ\), as a function of the wavelength (see Fig. 3). Once more, we see that the device is optimized around the design wavelength of \(\lambda = 1.31\, \mu\text{m}\).

3. PHYSICAL NATURE OF THE EXCITATION

One might question whether the observed excitation is truly a graphene TE-SPP. Indeed, the excitation can also correspond to a BSW, a surface excitation that can exist without graphene coverage.\(^{17}\) If the BSW were present, then the reflectivity dip would merely correspond to a BSW that experiences the graphene loss, rather than a graphene TE-SPP interacting with the underlying multilayer.

To obtain more insight into the physics behind the excitation with which we are dealing, we calculate the \((E, k_x)\)-band diagrams (with \(E = hc/\lambda\) the photon energy) of the PC. To be sure our results are not affected
Figure 4. (A) Band diagram of the infinite PC with practically the same unit cell as the finite structure. The yellow and white regions indicate the band-pass and bandgap regions respectively. The lines $E_{\text{air}}$ (dashed dark line), $E_{\text{SiO}_2}$ (dashed orange line), $E_{\text{BSW}}$ (green line) and $E_{\text{TE-SPP}_\text{is}}$ (grey dot) are also indicated in (A). The results in the region around $(E, k_x)_{\text{design}}$ (black dot) are rescaled in (B), where the $E_i$-lines are shown relative to the $E_{\text{air}}$ line (i.e. for every value of $k_x$, the corresponding value of $E_{\text{air}}$ is subtracted from $E_i$). The dispersion relations $E_{\text{TE-SPP}_\text{PC}}$ (blue line) and $E_{\text{TE-SPP}_\text{is}}$ (grey line) as well as $(E, k_x)_{\text{BSW}}$ (green dot) and $(E, k_x)_{\text{TE-SPP}_\text{PC}}$ (blue dot) can now be clearly visualised.
by loss mechanisms and to obtain an exact solution for real $k_x$, we consider an infinite PC such that we avoid radiation losses in the substrate, and neglect the graphene loss by putting $\sigma = 0$ for each value of photon energy $E$. In this lossless case, some parameters will slightly change compared to the previous section: for a fixed $d_a = 212.0\,\text{nm}$, $n_{\text{eff}}^{TE-SPP}$ and $d_b$ now become $n_{\text{eff}}^{TE-SPP} = n_0 + 1.6 \times 10^{-5}$ and $d_b = 206.0\,\text{nm}$ respectively.

The band diagram is plotted in Fig. 4(A) where the yellow and white regions represent the band-pass and bandgap regions respectively. We also indicate the light lines for air (labeled $E_{\text{air}}$) and SiO$_2$ (labeled $E_{\text{SiO}_2}$). The dispersion relation of the BSW guided by the bare PC (labeled $E_{\text{BSW}}$) can clearly be seen, but many of the other excitations are indistinguishable from one another. Hence, in inset (B) of Fig. 4, we zoom in on the working point (labeled $(E, k_x)_{\text{design}}$ in the figure caption) corresponding to $n_{\text{eff}}^{TE-SPP} = n_0 + 1.4 \times 10^{-5}$ and $\lambda = 1.31\,\mu\text{m}$, and now can distinguish the dispersion relations of the TE-SPP in an isolated graphene sheet with air on both sides (labeled $E_{\text{TE-SPP},\text{a}}$) and the TE-SPP guided by the PC covered with the graphene sheet (labeled $E_{\text{TE-SPP}_{\text{PC}}}$) as well as the $(E, k_x)$-values where the $E_{\text{BSW}}$ and $E_{\text{TE-SPP}_{\text{PC}}}$ lines start (labeled $(E, k_x)_{\text{BSW}}$ and $(E, k_x)_{\text{TE-SPP}_{\text{PC}}}$ respectively). Finally, since the $E_{\text{TE-SPP}_{\text{a}}}$ does not exist for all $(E, k_x)$-values, we also indicate its starting point, labeled $E_{\text{TE-SPP}_{\text{a}}}'$ on Fig. 4(A). We see from (A) that $(E, k_x)_{\text{design}}$ is located in the bandgap, which means that the fields decay in the PC, just as they would were the PC replaced by an air layer, corresponding to the TE – SPP where the graphene is isolated in air. We see from inset (B) of Fig. 4 that the $E_{\text{TE-SPP}_{\text{PC}}}$ line crosses the $E_{\text{TE-SPP}_{\text{a}}}$ line exactly at $(E, k_x)_{\text{design}}$. This clearly shows that for this value of $(E, k_x)$, the PC perfectly mimics the air cladding. In inset (B), we also see that at $(E, k_x)_{\text{design}}$, no BSW exists at constant photon energy and that for larger $(E, k_x)$-values, the $E_{\text{TE-SPP}_{\text{PC}}}$ line resembles the $E_{\text{BSW}}$ line, except that the latter is shifted towards higher $k_x$-values. This behavior once more confirms the dielectric response of the TE – SPP in the graphene sheet corresponding to negative $\sigma'$. This conclusion is further confirmed by the maximum field enhancement (defined as the ratio between the intensity of the field at the graphene layer and the intensity of the incident field in the substrate) being present at $n_{\text{eff}}^{TE-SPP}$, and the field profile showing that the maximum of the mode is located at the graphene sheet at this $n_{\text{eff}}^{TE-SPP}$.

4. DEPOSITION OF AN EXTRA DIELECTRIC LAYER ON GRAPHENE

Although we have shown that the reflectivity dips in Figs. 2-3 do indeed correspond to the TE-SPPs, we see from Fig. 2 that these modes are located extremely close to the light line of air where total internal reflection (TIR) takes place between substrate and cladding. This means that the unavoidable surface roughness at the PC top layer in fabricated samples would make the detection of the TE-SPPs difficult. Furthermore, there is a dip in the $|R_{N1}|^2$ spectrum even from the structure without graphene coverage (in Fig. 3), arising from the wavelength dependence of the TIR condition. Finally, even if we assume that the experiment would not be affected by these problems, observing the reflectivity dip - or finding the angle where the dip takes place in case $|R_{N1}|^2$ is measured as a function of wavelength - would require a very high angular resolution, due to the fact that the dip is extremely narrow (in the order of 0.0001° - 0.001°).

These issues can be overcome by depositing an extra dielectric layer such as PMMA on top of the graphene. To meet the symmetry condition in this case, we recalculate the $n_{\text{eff}}^{TE-SPP}$ of the TE-SPP in a graphene sheet sandwiched between 2 identical PMMA layers and search for the values in $(d_a, d_b)$-space for which the reflection of a laser, incident from the air cladding on the bare PC, matches with the reflection of a laser on a PMMA layer with a certain thickness embedded in air. As an illustration, we imagine that a 20 nm thick layer of PMMA (refractive index taken to be $n_2 = 1.481^{12,18}$) will be deposited on the graphene, for which the corresponding TE-SPP in the graphene sandwiched between the 2 identical PMMA layers will have an effective index of $\Re \left\{ n_{\text{eff}}^{TE-SPP} \right\} = n_0 + 0.007$. Taking into account the symmetry condition at this $n_{\text{eff}}^{TE-SPP}$ and maximising the coupling from the substrate towards the TE-SPP in case we keep $N_{\text{per}} = 5$, the PC unit cell thicknesses end up to be $d_a = 216.0\,\text{nm}$ and $d_b = 205.2\,\text{nm}$. We then recalculate the $|R_{N1}|^2$ spectra and plot the results together with the other spectra in Figs. 2-3. Fig. 2 clearly shows that the dip, and the associated excitation, is shifted to higher $\theta$-values, further away from $\theta_{\text{TIR}}$, such that the optical response of this new configuration would be far less affected by the surface roughness at the PC top layer. In addition, we see that the dip is much wider and can therefore be located more easily during the measurements. Furthermore, for the wavelength measurements, the absence of a dip in the $|R_{N1}|^2$ spectrum in the case of no graphene present (see Fig. 3) shows that the TIR condition is now always met.
Finally, adding a PMMA layer will make sure that the mode is still excited under the influence of fabrication errors, such as thickness and refractive index variations of the PC layers. To illustrate this, we recalculate the $|R_{N1}|^2$ spectra when a maximal variation of $\Delta d = \pm 1$ nm on the layer thicknesses and of $\Delta n = \pm 10^{-3}$ on the refractive indices is added to the PC layers. The results are shown in Fig. 5. We can see that the fabrication errors deteriorate the optical response of the PC near $\theta_{TIR}$, meaning that in some cases we are not able to excite the TE-SPP at all. This is clearly not the case when the PMMA layer is present where the reflectivity dip will always survive - as the angle at which the TE-SPP appears when having PMMA on top is sufficiently far from $\theta_{TIR}$ - although it is shifted from its position in the absence of fabrication errors.

5. CONCLUSIONS

In conclusion, we have shown that we can excite the graphene TE-SPP in a PC based multilayer efficiently. Matching the optical response of the PC with that of the cladding, such that the graphene sheet experiences a 'symmetric environment', will enable the excitation of the TE-SPPs in a graphene sheet even at room temperature. We have also shown that depositing an extra dielectric layer such as PMMA on top of the structure will shift the TE-SPP sufficiently far from the TIR line that fabrication errors and other parasitic effects should not be problematic, thus enabling the excitation of the TE-SPPs even in non-ideal circumstances.

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