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A flexible control on electromagnetic behaviors of graphene oligomer by tuning chemical potential

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Abstract

In this work, we demonstrate that the electromagnetic properties of graphene oligomer can be drastically modified by locally modifications of the chemical potentials. The chemical potential variations of different positions in graphene oligomer have different impacts on both extinction spectra and electromagnetic fields. The flexible tailoring of the localizations of the electromagnetic fields can be achieved by precisely adjusting the chemical potentials of the graphene nanodisks at corresponding positions. The proposed nanostructures in this work lead to the practical applications of graphene-based plasmonic devices such as nanosensing, light trapping and photodetection.

Keywords: Graphene oligomer, Surface plasmon, Electromagnetic tunability, Optical absorption

Introduction

Recently, an increasing number of subwavelength components and structures have been designed and manufactured based on metamaterials (MMs) which get spotlight by the versatility of controlling the electromagnetic (EM) behaviors [1]. MMs support unique phenomena which cannot exist in nature including negative refractive index [2], extraordinary optical transmission [3], and electromagnetically induced transparency [4]. Due to unique properties of MMs, the nanodevices composed of MMs have more prominent advantages that the nanodevices possess pronounced and flexible ability to regulate and control EM behaviors, which leads the development of nanodevices toward high quality and integrability. Plasmonic MMs is one kind of metamaterials that exploits surface plasmons (SPs) to achieve novel optoelectric properties [5, 6]. SPs are the oscillations of free electrons in metal, originating from the interaction of light with metal-dielectric materials. Under certain circumstances, the interaction of incident light with the surface plasmons is able to produce self-sustaining, propagating electromagnetic waves known as surface plasmon polaritons (SPPs) which

propagate along the metal-dielectric interface [7]. The SPPs are much shorter than incident light in wavelength, which is suitable for nanostructures with subwavelength footprint [8]. Light hitting the plasmonic MMs is transformed into SPPs, leading to the appearance of strong field localization in these structures at the resonance frequencies. The EM properties of plasmonic structures are primarily controlled by their geometry, making it possible to optimize the electric and magnetic behaviors over a broad range [9–12]. In practice, electron-beam lithography and focused-ion beam milling are two common methods to fabricate plasmonic structures on planar substrates. The excellent EM behaviors stem from the unique features of plasmonic structures with characteristics smaller than the wavelength of light separated by subwavelength distances, revealing a striking way to design applications in nanoscale such as sensing [13], surface-enhanced spectroscopies [14], and nonlinear optics [15]. Most common plasmonic MMs are composed of gold and silver which exhibit negative real permittivity [16]. However, noble metals have relatively large ohmic loss and low flexibility that once the structure is fixed, the EM behaviors cannot be optimized further, which restricts the development of nanodevices based on plasmonic structures [17, 18].

Graphene is one two-dimensional material made up of sp^2 hybridization of carbon atoms in the honeycomb lattice structure. Due to the surpassing behaviors in

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electronics as well as photonics of graphene, various research groups investigate graphene with different methods to create plasmonic structures that exhibit lower losses, higher confinement and tunability of the EM fields [19–23]. Graphene is able to accommodate SPPs in large range from terahertz to mid-infrared frequencies [24–26]. Graphene has great potential for improving light-matter interactions in a two-dimensional regime because of SPs with strong light confinement [27]. Graphene oligomers constitute plasmonic molecules (PMs) via interactions between components, where the EM fields with strong field enhancements follow symmetries analogous to the coupling of atoms in chemical molecules [28]. By changing the chemical potential of graphene, the graphene PMs can reach high quality and flexibility [29]. However, there are more adjustable structure parameters such as chemical potential of graphene in different positions for graphene nanostructures to control the EM behaviors. Most of reported graphene nanostructures concentrate on changing chemical potential of the whole structure, which is short of elucidation of the relationship between chemical potential of graphene in different position and the EM behaviors of graphene nanostructure. The proposed graphene nanostructures may stimulate more surpassing EM properties and will affect a wide range of plasmonic applications.

To verify the effect mechanisms of PMs based on graphene, a numerical study on the graphene oligomer consisting of 13 equal-sized graphene nanodisks has been systemically conducted by intentionally varying chemical potential of partial graphene in this work. The graphene oligomer with D_{12h} symmetry is able to sustain two plasmonic modes in calculated range. Further utilization of graphene oligomer relies on the precise control of local chemical potential of graphene. By selectively varying chemical potentials of graphene oligomer, the two innate plasmonic modes are profoundly modulated. Tuning the chemical potential of prominent graphene nanodisks in two plasmonic modes respectively has different influence on two plasmonic modes. The change of chemical potential of intersection part between the two plasmonic modes intensifies both two plasmonic resonances and leads to degeneration of plasmonic modes. In addition, the change of chemical potential of central graphene nanodisk also significantly affects the EM properties of graphene oligomer. The simulated results show that the graphene oligomer possesses high tunability and flexibility, and provides new degrees of freedom for designing plasmonic nanodevices capable of tailoring two-dimensional light confinement.

Simulated methods and models

In our model, the graphene is treated as one thin film with one atom layer thickness Δ and modeled by a complex permittivity ϵ [22].

$$\epsilon = 1 + \frac{i\sigma_g \eta_0}{k_0 \Delta}, \quad (1)$$

where $\Delta = 0.334$ nm, σ_g is the complex surface conductivity of graphene, $\eta_0 = 377$ Ω stands for the impedance of the free space, and $k_0 = 2\pi/\lambda$ is the wave number of the light in air. The complex surface conductivity σ_g of graphene monolayer is modeled by Kubo's formulation, which consists of contributions from both intraband electron-photon scattering σ_{intra} and interband electron-electron transition σ_{inter} [30],

$$\sigma_g = \sigma_{intra} + \sigma_{inter}, \quad (2)$$

where

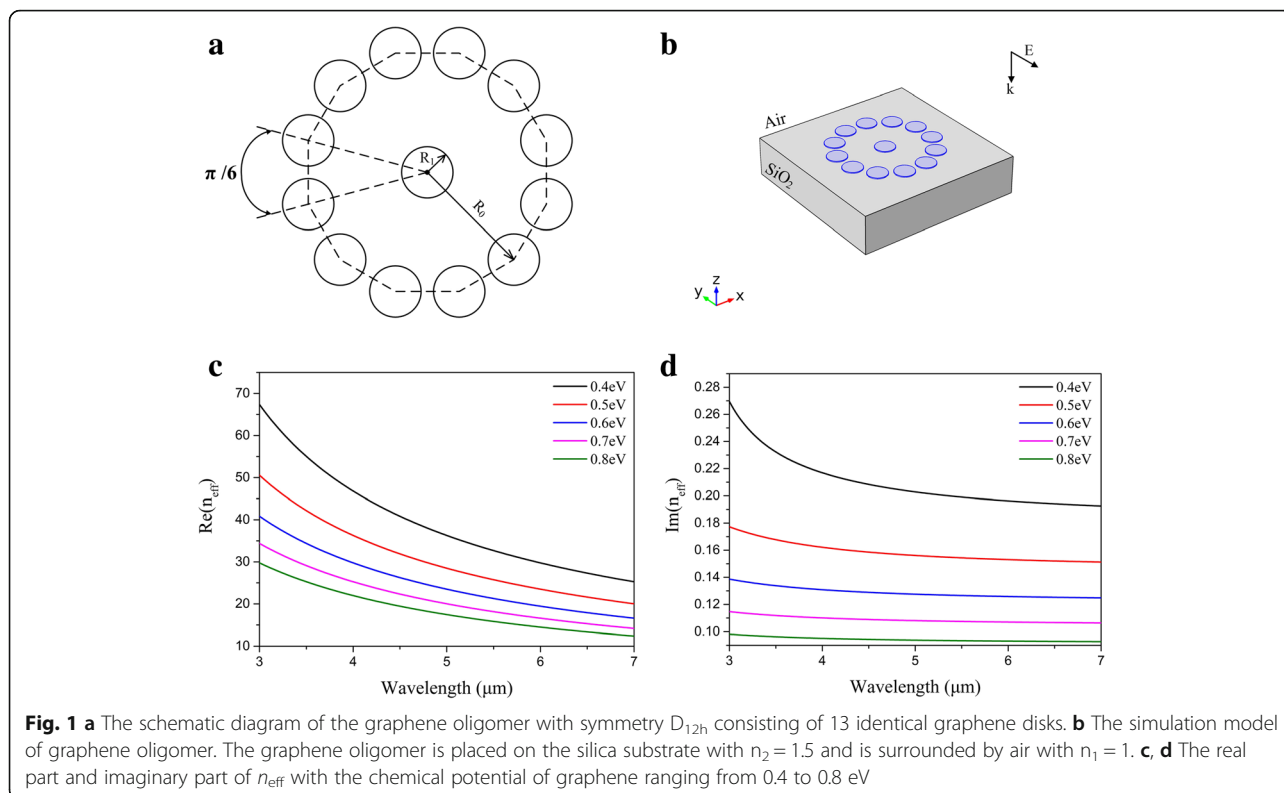
$$\begin{aligned} \sigma_{intra} &= \frac{2e^2 k_B T}{\pi \hbar^2} \cdot \frac{i}{\omega + i\tau^{-1}} \left[\ln \left(2 \cosh \left(\frac{\mu_c}{k_B T} \right) \right) \right], \quad (3) \\ \sigma_{inter} &= \frac{e^2}{4\hbar} \left[\frac{\sinh \left(\frac{\hbar\omega}{2k_B T} \right)}{\cosh \left(\frac{\mu_c}{k_B T} \right) + \cosh \left(\frac{\hbar\omega}{2k_B T} \right)} \right. \\ &\quad \left. - \frac{i}{2\pi} \ln \frac{(\hbar\omega + 2\mu_c)^2}{(\hbar\omega - 2\mu_c)^2 + (2k_B T)^2} \right]. \quad (4) \end{aligned}$$

In these equations, e is the charge of an electron, \hbar is the reduced Planck constant, k_B is the Boltzmann constant, T is the temperature set as 300 K, τ is the momentum relaxation time set as 0.5 ps, ω is the radian frequency, and μ_c is the chemical potential of graphene.

We incorporate graphene nanodisk arrays into a graphene oligomer with D_{12h} symmetry (Fig. 1a) to investigate the EM behaviors. The graphene oligomer consists of 13 graphene nanodisks of equal size, where one nanodisk is placed in the center and the others surround it with dodecagon symmetry. The radius of the addendum concentric circle R_0 is 240 nm and the radius of individual nanodisks R_1 is 50 nm. The graphene oligomer composed of large number of graphene nanodisks has advantage in flexible selections to change chemical potentials. As shown in Fig. 1b, the graphene oligomer is surrounded with air described by a refractive index $n_1 = 1$ and adheres to a silica substrate with a refractive index $n_2 = 1.5$. The incident light is vertical to graphene oligomer and the polarization is along y axis. Theoretically, the effective refractive index of graphene is described by

$$n_{eff} = \frac{2i\epsilon_{eff}\epsilon_0 c}{\sigma_g}. \quad (5)$$

where ϵ_{eff} is the effective permittivity of the environment



media, ϵ_0 is the vacuum permittivity and c is the speed of light in vacuum. According to the equations (2, 3, 4, and 5), it is seen that n_{eff} is a function of μ_c and the relationship is plotted in Fig. 1c and d, meaning that the resonance of our proposed structure can be expediently modified via manipulating the chemical potential of graphene. It should be pointed out that $|Im(n_{eff})|/|Re(n_{eff})|$ is significantly small. So the real part of n_{eff} mainly effect the calculate results and the imaginary part of n_{eff} has little effect on our model with chemical potential changing. We therefore neglect the effect of imaginary part of n_{eff} in this study.

The electric fields and extinction spectra of graphene oligomer are calculated in the commercial finite element method (FEM) software, COMSOL Multi-Physics, RF Module. The extinction cross-section σ_{ext} is obtained as $\sigma_{ext} = \sigma_{sc} + \sigma_{abs}$, where σ_{sc} corresponds to the scattering cross-section

$$\sigma_{sc} = \frac{1}{I_0} \int \int (\vec{n} \cdot \vec{S}_{sc}) dS, \quad (6)$$

and the absorption cross-section σ_{abs} , is determined by

$$\sigma_{abs} = \frac{1}{I_0} \int \int \int Q dV. \quad (7)$$

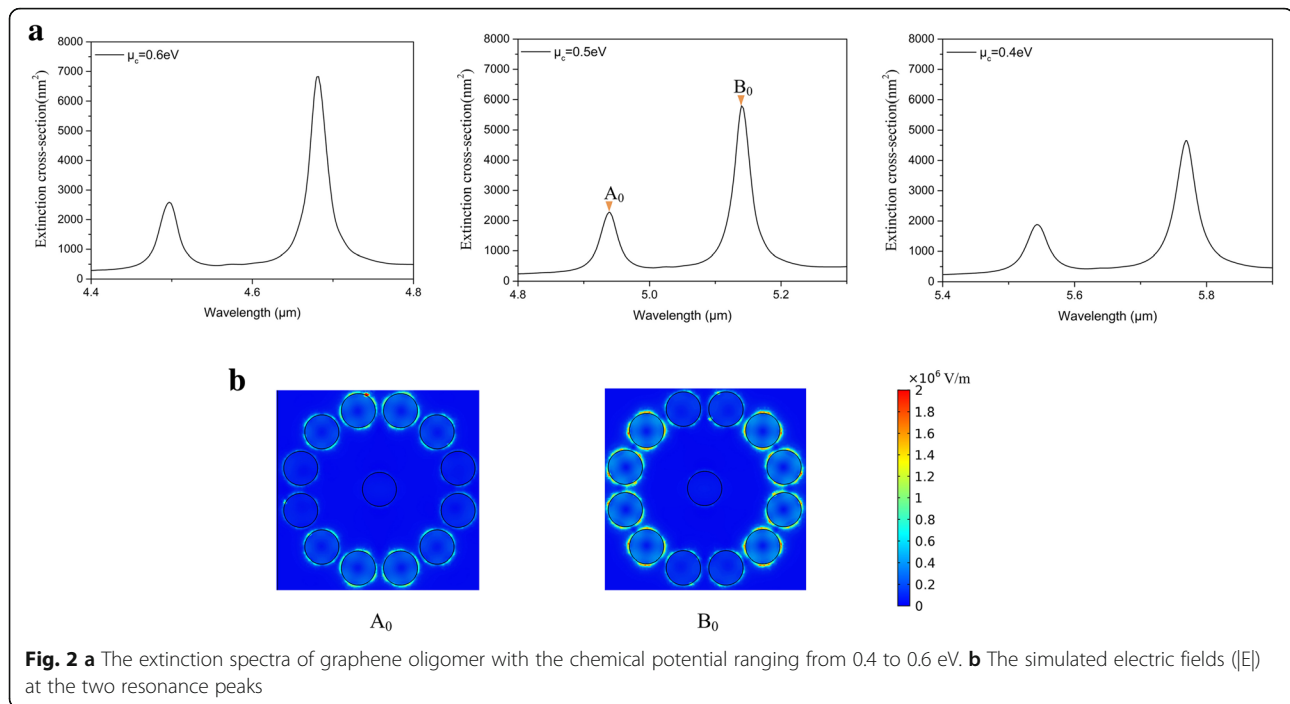
In these equations, I_0 is the incident intensity. \vec{n} stands for the normal vector pointing outwards from the

plasmonic nanocluster, \vec{S}_{sc} indicates the Poynting vector for the scattered field. The integral in Equation (6) is taken over the closed surface of the scatter. Q is the power loss density in the oligomer. The integral in equation (7) is taken over its volume. The extinction spectra are calculated in the selected wavelength range of mid-infrared. The Perfectly Matched Layer (PML) is applied around the proposed nanostructure to avoid the reflected light fields. The thickness of the graphene is meshed at least five layers to guarantee simulation accuracy.

Simulation results and discussions

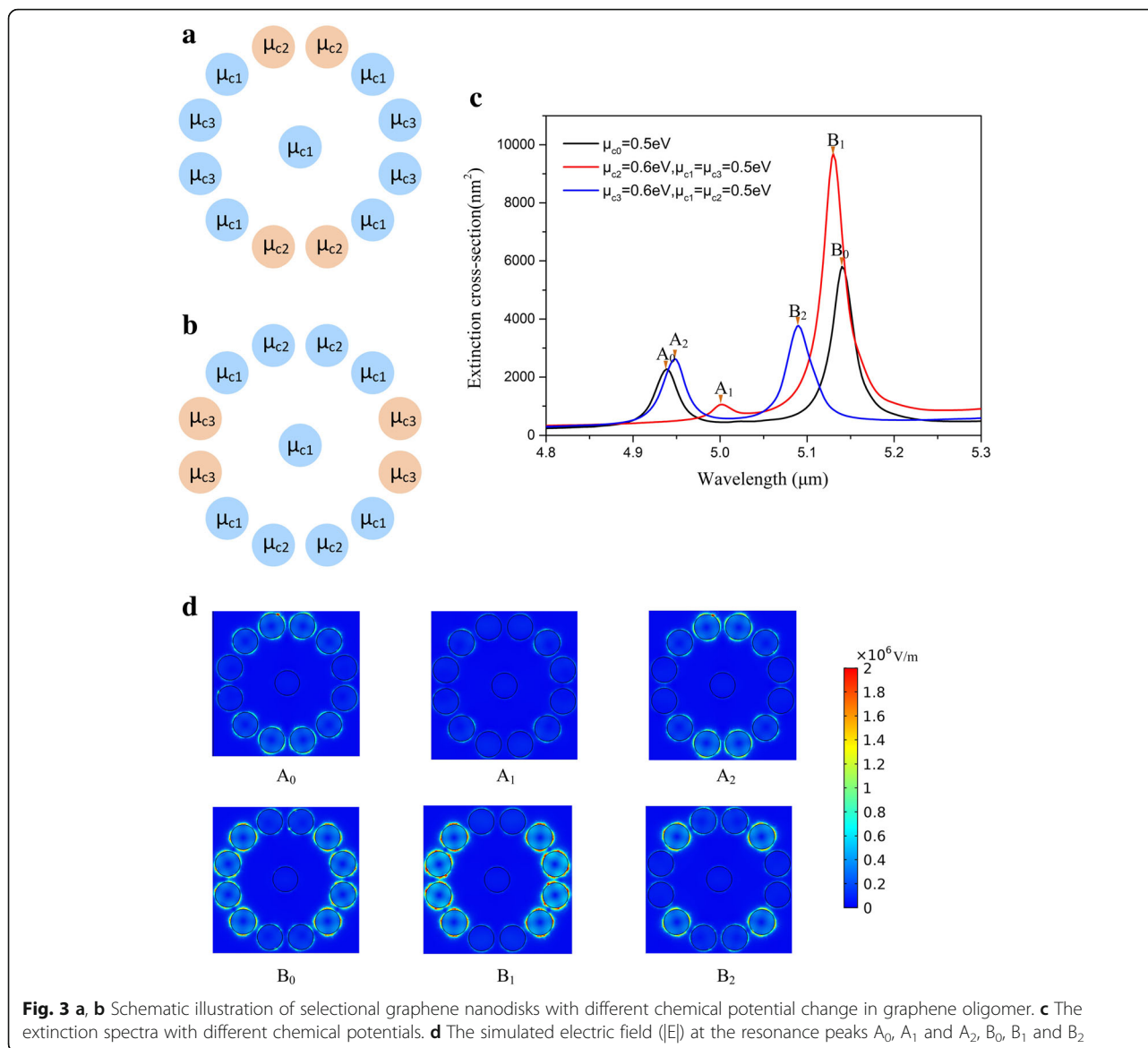
The effect of local chemical potential change of graphene nanodisks in plasmonic modes

For the proposed structure, the extinction spectra (Fig. 2) exhibit two prominent resonances associated with the excitation of plasmons in the graphene oligomer. The graphene oligomer is able to sustain two plasmonic modes, which are both sensitive to the graphene chemical potential μ_c . By varying μ_c of the whole graphene oligomer from 0.4 eV to 0.6 eV, both two plasmonic resonances become intense, and the positions move to higher frequency range simultaneously. The distinct enhancement of absorption in graphene oligomer is ascribed to the promotion of carrier density with increasing μ_c , which creates an optical gap where



plasmons avoid being quenched through coupling to electron-hole pairs (Landau damping). The increase of virtual electron-hole pair transitions allowed gives rise to the significant interaction of coherently coupled graphene nanodisks which intensifies extinction maximum [21]. We choose the extinction spectrum with $\mu_c = 0.5\text{eV}$ as the benchmark and the two peaks labeled by A_0 and B_0 represent two different plasmonic modes and the corresponding electric fields are presented in Fig. 2b. Strong concentrated electric fields appear as the nanoscale electromagnetic hot spot and lead to extinction enhancement. For peak A_0 , the hot spots mainly concentrate on the eight nanodisks on the top and bottom, and especially focus on the four nanodisks on the highest and lowest positions in the nanostructure. For peak B_0 , the hot spots mainly concentrate on the eight nanodisks on the left side and right side, and the brightest four nanodisks are on the most left and right positions in the nanostructure, which is perpendicular to the mode of peak A_0 . Based on the different electric field distributions of peak A_0 and B_0 , we define the mode of peak A_0 as Y mode and the mode of peak B_0 as X mode for a clear expression. The four brightest graphene nanodisks in Y mode are extremely dark in X mode and vice versa. Another four graphene nanodisks composed of a square are relatively bright both in Y mode and X mode defined as the intersection part. We divide the peripheral graphene nanodisks into three parts with different chemical potentials μ_{c1} , μ_{c2} and μ_{c3} respectively (shown in Fig. 3a and b). The nanodisks with μ_{c2} or μ_{c3} are the brightest part in Y mode or X mode. The chemical potential of

intersection part and center μ_{c1} keeps 0.5 eV in next calculation. At first, μ_{c2} increases to 0.6 eV and others keep 0.5 eV (shown in Fig. 3a). Then μ_{c3} increases to 0.6 eV and others keep 0.5 eV (shown in Fig. 3b). By changing μ_{c2} or μ_{c3} to 0.6 eV respectively, a series of spectral variations visibly appear in Fig. 3c. We can see that by changing the chemical potential of sectional graphene nanodisks and leaving the other parameters constant, a flexible reconfiguration of the overall spectral shape is obtained, manifested by a systematic change in the height of two resonance peaks. In Fig. 3d, the electric fields of variant Y mode and X mode are plotted in detail. As shown in Fig. 1c, the real part of n_{eff} is inversely proportional to the chemical potential. Therewith when the chemical potential increases, the confinement of the incidence light becomes weak. The mechanism of local chemical potential change in graphene oligomer is that, the increase of chemical potential reduces the interaction between light and the graphene nanodisks, and pushes the hot spots to surrounding nanodisks. If the orientation of pushing is to the location of strong plasmonic resonance, the resonance is strikingly strengthened, otherwise it is reduced. This means that the effect of local chemical potential change relies on the electric field distributions of different modes. When μ_{c2} increases to 0.6 eV, peak A_0 significantly decreases and red shift to peak A_1 due to the weak confinement of the four brightest graphene nanodisks for the incidence light, where the hot spots mainly concentrate on the intersection part. Simultaneously, peak B_0 significantly increase and blue shift to peak B_1 , which is attributed to the fact



that the increase of μ_{c2} sufficiently enhance the X mode. For $\mu_{c3}=0.6$ eV, it is the other way round. Peak A_0 slightly increases and red shifts to peak A_2 arising from the enhancement of Y mode with μ_{c3} increasing. In the meantime, peak B_0 blue shifts to peak B_2 and decreases with the concentration of hot spots on the intersection part, which is in accord with peak A_1 .

These variations of Y mode and X mode give rise to the descent or enhancement in extinction spectra. A flexible control over the extinction curves is achieved by adjusting the EM behaviors of Y mode and X mode arising from selectively adding the chemical potentials of graphene nanodisks, which opens a novel path way for designing graphene nanodevices with different functions. For example, when $\mu_{c2} = 0.6$ eV, peak A_0 become lower while peak B_0 significantly intensifies, which makes the

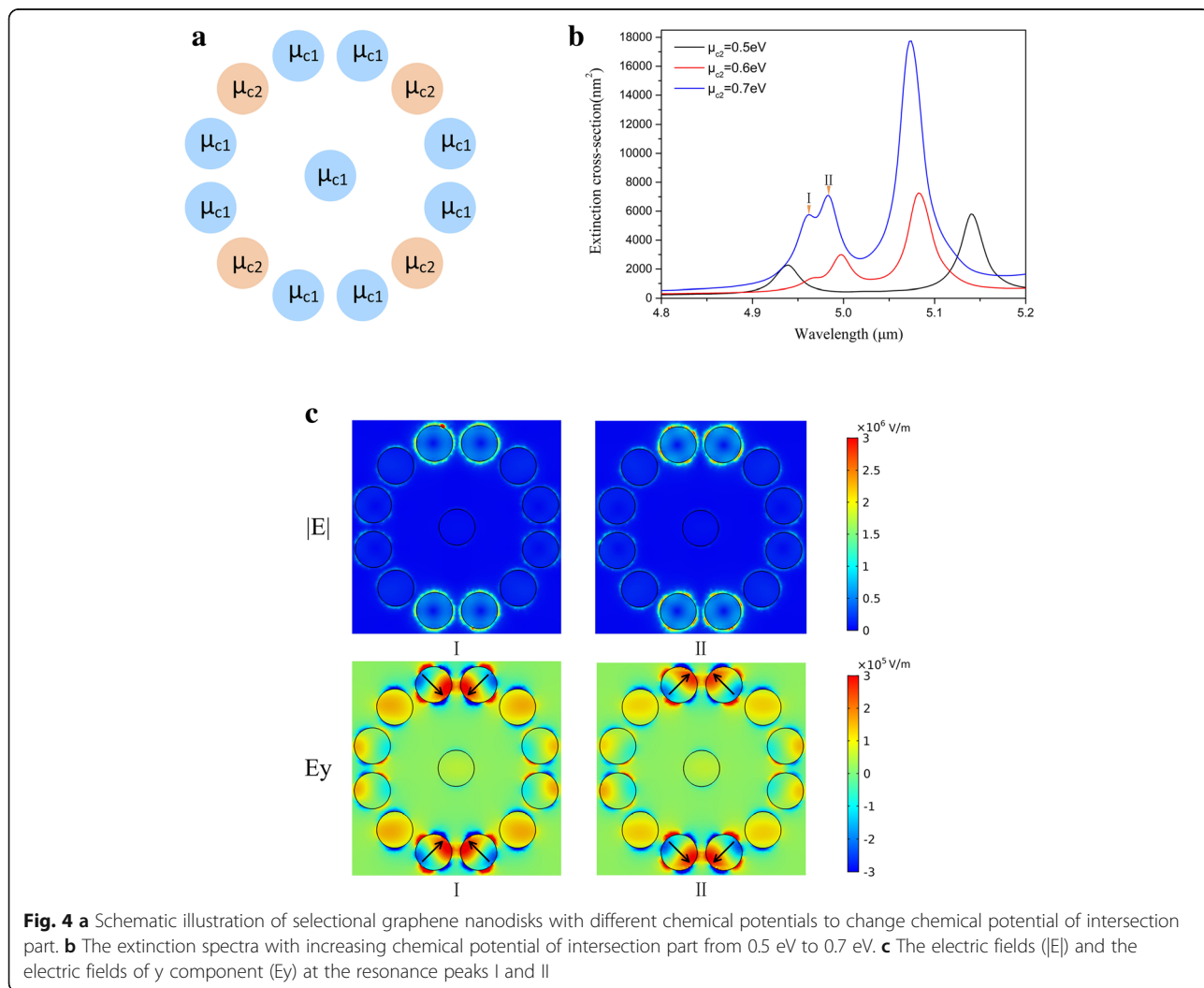
graphene oligomer suitable for high efficient absorbers. In the other way, when $\mu_{c3} = 0.6$ eV, the values of two peaks approach closely, which is convenient to design dual-band nanosensors.

The mode enhancement by increasing chemical potential of the intersection part

For the electromagnetic fields of two plasmonic modes, an intersection part composed of four graphene nanodisks between two plasmonic modes appears. As shown in Fig. 3d, the electric fields mainly concentrate on the four graphene nanodisks in the intersection part by locally changing the chemical potential. So we believe that the chemical potential of intersection part significantly influences the EM characteristics of graphene oligomer and profile of extinction spectra. We redistribute the chemical

potentials in graphene oligomer. The chemical potential of four graphene nanodisks in intersection part is set as μ_{c2} . The chemical potential of other nanodisks μ_{c1} keeps at 0.5 eV (shown in Fig. 4a). On the basis of the mechanisms of local chemical potential change, the increasing chemical potential of the intersection part intensifies both Y mode and X mode. As shown in Fig. 4b, with increasing μ_{c2} , the extinction spectrum is drastically modified. When μ_{c2} increases to 0.6 eV, both two resonance peaks have a promotion compared with $\mu_{c2}=0.5$ eV. It is noted that there is a new resonance peak appears around the resonance peak of Y mode. When the μ_{c2} further increases to 0.7 eV, the two resonance peaks become stronger and a new resonance peak obviously appears around the resonance peak of Y mode. The elucidation of strong enhancement of resonance peaks is that the increase of μ_{c2} efficiently intensifies the both Y mode and X mode. The increase of μ_{c2} facilitates the plasmonic oscillations of four graphene nanodisk in Y mode and X mode respectively.

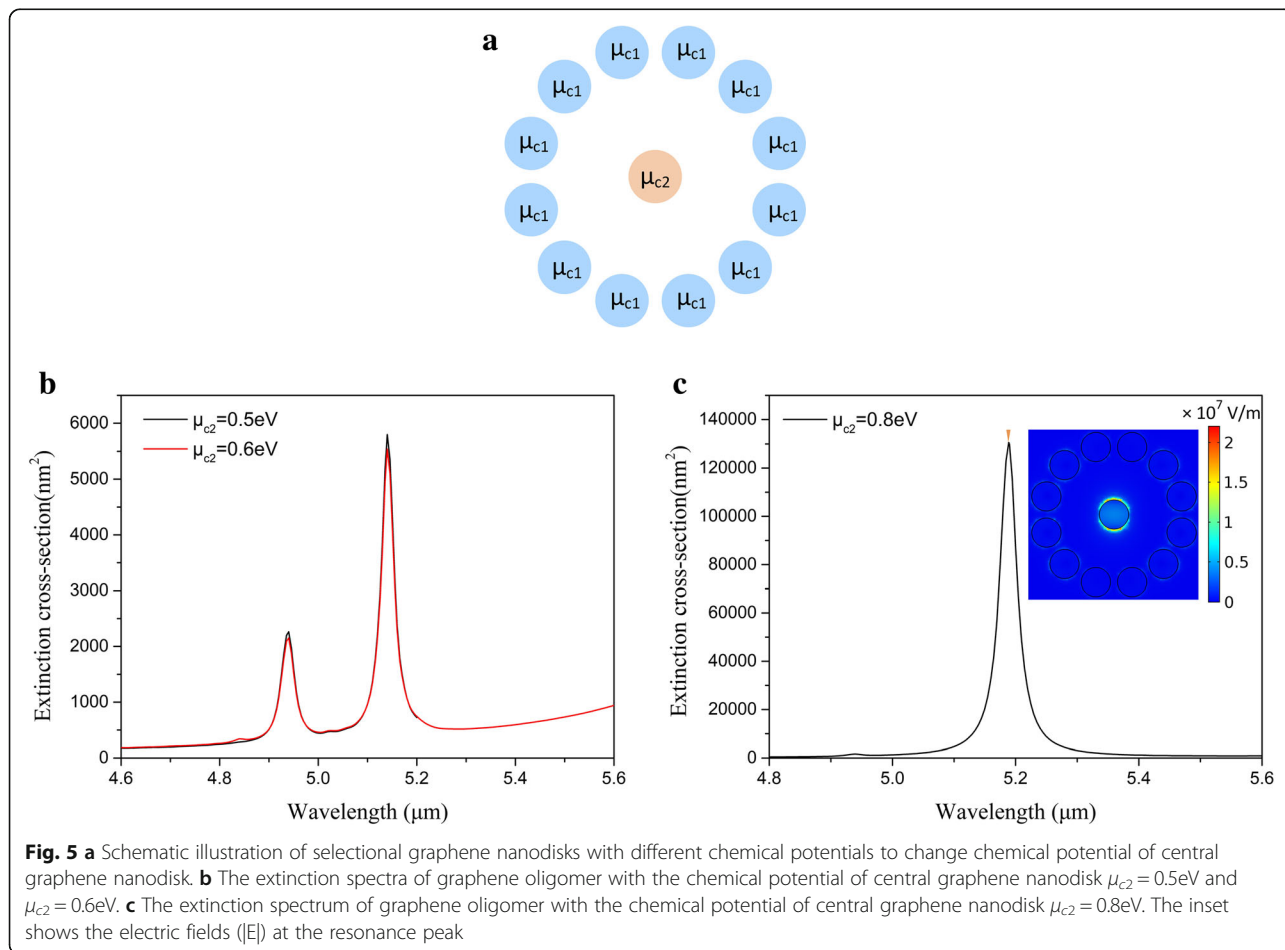
The resonance peak of Y mode splitting into two resonance peaks is a process of degeneration. As shown in Fig. 4c, the two resonance peaks labeled by I and II have same electric fields but the components of electric field are different. The directions of E_y of peak I and II are perpendicular to each other, which represent two plasmonic modes degenerating from Y mode. The two new plasmonic modes originally merge in Y mode, and the two modes begin to separate with μ_{c2} increasing. In addition, both two degenerate resonance peaks with $\mu_{c2} = 0.6$ eV are much larger than resonance peak with $\mu_{c2} = 0.5$ eV. In such a manner, by choosing the graphene nanodisks of intersection part to increase their chemical potential, one can improve all resonance peaks in extinction spectra. It is proposed to enhance the absorption of graphene nanodisks by selectively changing the chemical potentials of adaptive graphene nanodisks, which helps to design plasmonic nanodevices capable of light absorption with high efficiency.



The effect of chemical potential of central nanodisk

The central graphene nanodisks introduced into the graphene oligomer is aimed to enable the nanostructures possess more flexibility and further investigate the effect of local chemical change in different positions. Due to the large distance between the central graphene nanodisk and the peripheral graphene nanodisks, the central graphene nanodisk cannot couple with the other graphene nanodisks in two plasmonic modes. In this section, we set the chemical potential of central graphene nanodisk as μ_{c2} . Others are set as μ_{c1} keeping 0.5 eV (shown in Fig. 5a). Changing the chemical potential of central graphene nanodisk μ_{c2} is able to modify EM fields of the graphene oligomer without changing the geometry. The results by increasing μ_{c2} are shown in Fig. 5b and c. The increase of μ_{c2} enhances the plasmonic oscillations of central graphene nanodisks. However, when increase of μ_{c2} is relative small, the oscillator strength of central graphene nanodisk is not enough to support new plasmonic mode and influence the intrinsic modes, so the extinction spectrum $\mu_{c2} = 0.6\text{eV}$ has almost no change compared with $\mu_{c2} = 0.5\text{eV}$, where two resonance peaks still appear (shown in Fig. 5b). When μ_{c2} reaches to a large value (0.8 eV), a new

resonance peak appears obviously in the extinction spectrum (shown in Fig. 5c). The huge improvement of plasmonic oscillations profoundly changes the profile of extinction spectrum. The new resonance peak originates from the strong interaction between the incident light and the central graphene nanodisk, of which the EM fields mainly concentrate on the central graphene nanodisk, which is defined as the central mode. The resonance peak supported by the central mode is much larger than two intrinsic resonance peaks, while the two intrinsic resonance peaks are drastically suppressed and even disappear in the extinction spectrum. The effect of μ_{c2} is different from the effect discussed earlier, because the central graphene nanodisk is not contained in the innate plasmonic modes. The effect of μ_{c2} consists with changing the chemical potential of whole graphene oligomer which is discussed in the beginning. In such a manner, by increasing μ_{c2} , one can design the novel plasmonic device capable of absorbing incident light efficiently. Combining with aforementioned studies, the flexible tailor of the localizations of the electromagnetic field can be achieved by precisely adjusting the chemical potentials of the graphene nanodisk in different positions.



In practice, continuous atomic single-layer of graphene is first grown using an optimized chemical vapor deposition method with CH_4 as the carbon source. Then graphene film is determined to be monolayer by Raman measurements. The electron-beam lithography with poly(methyl methacrylate) (PMMA) as an electron beam resist is used to pattern the graphene film to produce the proposed nanostructures, and the exposed area is etched away by the oxygen plasma, remaining the pattern of graphene protected by a PMMA layer with subsequent lift-off using acetone. Then the device is ready for test. The chemical potential can be tuned via manipulating chemical and electrostatic doping. For chemical doping, local chemical potential change can be realized by exposing the required graphene nanodisks to HNO_3 vapor and simultaneously preventing the contact between other graphene nanodisks and HNO_3 vapor. For electrostatic doping, an appropriate top gate configuration may locally manipulate the chemical potential of graphene by supplying top gate voltage.

Conclusions

In conclusion, we have demonstrated the versatility of the graphene oligomer to modify the EM behaviors and spectral lineshape by varying chemical potential of graphene at the nanoscale. The characteristics are summarized from the electric fields and extinction spectra of the various chemical potentials. First, by changing the chemical potential of two graphene nanodisks in Y mode and X mode respectively, a flexible variation of two resonance peaks appears in extinction spectra. The two resonance peaks can be enhanced or reduced by changing the different chemical potentials of graphene oligomer. Second, increasing the chemical potential of intersection part intensifies the both two resonance peaks and gives rise to degeneration of Y mode. Third, high chemical potential of central graphene nanodisk is able to support a strong resonance peak and simultaneously restrain two in-nate resonance peaks. The dependence of the graphene oligomer on chemical potential suggests that one can modify the EM behaviors of the graphene nanostructure with chemical potential without modifying the geometry. The previous studies based on graphene nanostructures can only change one absorption peak by changing chemical potential of whole graphene [19–23], but the method of changing chemical potential of graphene in this paper can tune the spectra with additional flexibility, which brings out more surpassing EM phenomena. In the realm of practical applications, our studies provide a new degree of freedom for modifying the graphene plasmonics by tuning the chemical potential of the graphene nanostructures. The graphene nanostructures provide a facile platform to cultivate the EM behaviors with light in two dimensions, which pave a way for the design of graphene-based plasmonic nanodevices for nanosensing, light trapping and photodetection.

Abbreviations

EM: Electromagnetic; MMs: Metamaterials; PML: Perfectly Matched Layer; PMs: Plasmonic molecules; SPPs: Surface plasmon polaritons; SPs: Surface plasmons

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Availability of data and materials

The data supporting the conclusions of this article are included within this article.

Authors' contributions

WQ supervised this project, JR and WQ provided the original idea, JR, HC, and PQ created the models and run the simulations, JR, GW, WQ, PQ, ZL, J-XW, QK, and J-QP discussed and analyzed the results, JR and WQ wrote the paper. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no competing interests.

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