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Abstract The interaction between doped graphene nanoislands connected by narrow junctions constitutes an ideal testbed to probe quantum effects in plasmonic systems. Here, the interaction between graphene plasmons in neighboring nanoislands is predicted to be extremely sensitive to the size and shape of the junctions. The reported ab initio calculations reveal three different regimes of interaction: (1) for narrow bridges (<4 carbon-atom rows), the conductance of the junction is too low to allow electron transport and the optical response is dominated by a characteristic bonding dipolar dimer mode that also appears in a classical description; (2) for wider junctions (4-8 carbon rows), a strong charge polarization is induced across the junction, which gives rise to a novel junction plasmon that has no counterpart in a classical description; (3) for even wider junctions (≥ 8 rows), their conductance is sufficiently large to allow charge transport between the two graphene islands, resulting in a pronounced charge-transfer plasmon, which can also be described classically. This work opens a new path for the investigation of intrinsic plasmon quantum effects.



Quantum junction plasmons in graphene dimers

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1. Introduction

A major advantage of plasmons -the collective excitations of conduction electrons in metals- is that they can produce high levels of light confinement and optical field enhancement [1], which enable them to strongly interact with objects that have relatively low optical cross-sections, such as atoms and simple molecules in solid-state environments [2]. Additionally, confined plasmons can display intense dipoles through which they couple to external radiation. Therefore, plasmons can act as intermediaries (antennas) that boost the interaction of light with molecules (e.g., in techniques as useful as surface-enhance Raman scattering (SERS) [3,4] and surface-enhanced infrared resonant absorption (SEIRA) [5], and also in promising applications to tumor removal [6], drug delivery [7], improved photovoltaics [8], and catalysis [9]). An interesting scenario is found when a plasmon is supported by a single molecular structure rather than an extended nanoparticle. Many molecules have been found to exhibit plasmons [10–14], including the C_{60} molecule and carbon nanotubes [15, 16]. Like other carbon allotropes, they display plasmon bands at energies around \sim 5 eV (π plasmons) and \sim 15–20 eV (σ plasmons). However, these are conventional plasmons, in the sense that they are not too sensitive to the charge state of the molecules, similar to those found in noble metal nanoparticles. Instead, we are here concerned with plasmons hosted by doped graphene molecular nanostructures, which in contrast to noble-metal plasmons, only occur when they are electrically charged. These plasmons emerge at lower energies for typical levels of doping.

Graphene has recently emerged as an attractive electrically tunable, optical material [17] displaying strong infrared plasmons, the frequencies of which scale roughly as $|n|^{1/4}$ with the doping charge density n [18–21]. Graphene plasmons provide unprecedented levels of light confinement and field enhancement [22], which are promising tools for accessing quantum-optics phenomena [23-26]. In this context, graphene offers the additional benefits of a robust structure and a simple and convenient electrical tunability for controlling quantum interactions [27]. The electrical modulation of graphene-plasmon-related phenomena also suggests potential application in optical waveguiding and signal processing [28-30], metamaterials [31, 32], quantum optics [22], and spectrometry [33]. Experimental evidence of electrostatic control over plasmonic features in the absorption spectra of graphene has been recently reported [33], followed by the observation and spatial mapping of confined plasmons in this material [34, 35]. Additionaly, graphene has been used to extrinsically modulate the plasmons of neighboring metallic structures [36, 37]. These advances have sparked considerable

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interest into the unique optical behavior of nanostructured graphene.

Here, we describe intrinsic quantum effects in the plasmonic response of bowtie graphene nanodimers bridged by thin junctions. Due to the two-dimensional character of this material, the addition of a small number of atoms (<10) is sufficient to dramatically modify the absorption spectrum of the entire dimer. Three distinct regimes of plasmonic behavior are predicted by our first-principles calculations. For separated nanotriangles bridged by narrow junctions (< 4 atomic rows wide), we find a characteristic hybridized bonding dipolar plasmon (BDP) mode [38]. In the limit of wide junctions (more than 8 atomic rows), a dramatic reshaping of the optical spectrum leads to a pronounced charge-transfer plasmon (CTP), which emerges at lower energies, similar to what happens when the gap vanishes in a conventional metallic nanoparticle dimer [39-42]. The plasmonic features in both of these regimes are also found within a classical electrodynamics description using the appropriate dielectric permittivity for the graphene, although the behavior of the CTP is substantially modified by quantum mechanical effects (see below). In contrast, for intermediate junction widths (4 - 8 atomic rows), a plasmonic feature shows up at intermediate energies, which is absent in classical-electrodynamics calculations. We refer to this mode as a junction plasmon (JP) because it is caused by quantum effects in the small *molecular* junction. We also predict a minor dependence on the length of the junction and a strong dependence on its width, thus confirming the importance of a quantum mechanical description of the optical properties of bridged graphene dimers.

2. Results and discussion

We simulate the optical response of nanoscale bowtie graphene structures using the random-phase approximation (RPA) and wave functions derived from a tight-binding description of the carbon sheet. Further details of the method are given elsewhere [43]. The triangular structures forming the bowties are oriented with respect to the graphene lattice in such a way that they have armchair edges, which prevent undesired losses typically observed in zigzag-edge structures, and caused by the presence of zero-energy electronic edge states [43]. Such states are not present in the structures under consideration, because they have the same number of atoms in both carbon sublattices [44]. For comparison, we include classical-electrodynamics calculations, which are performed by describing the graphene through its local-RPA surface conductivity [22] using a finite elements method (COMSOL). We fix the side length of the nanotriangles to 8 nm in all cases ($\sim 10^3$ atoms in each triangle). We set the Fermi energy of our structures to $E_F = 0.4 \,\mathrm{eV}$, and the intrinsic damping to $\hbar \tau^{-1} = 1.6$ meV, corresponding to a DC mobility of $10,000 \text{ cm}^2/(\text{Vs})$ [21].

Our main results are summarized in Fig. 1. We show details of the junction region in Fig. 1(a) for three characteristic values of the width (m = 0, 4, 8, where m is the

number of carbon-atom zigzag rows forming the junction, and m = 0 stands for the non-touching configuration). The corresponding spectra are shown in Fig. 1(b)-(d) for a range of junction lengths n = 2 - 10 (*n* is defined as the number of carbon hexagons that are needed to join the graphene triangles for m = 2, see Fig. 1(a)). These spectra show a clear trend from high to low energy features as the junction width is increased, irrespective of the length. This behavior is further examined in Fig. 1(e), where the plasmon features are arranged as a function of energy (vertical axis) and junction width (m, horizontal axis) for different values of the junction length n (see color code in the upper inset; full spectra are provided in the Supporting Information (SI) [45]). The area of the circles is made proportional to the intensity of the plasmon mode defined as the area under the corresponding plasmon peaks in the extinction spectra (for light polarization along the gap). For narrow bridges, the spectra exhibit prominent plasmonic features centered around ~0.47 eV. In contrast, the spectra for the wide-junction limit are dominated by lower-energy plasmonic features around $\sim 0.22 \,\text{eV}$. At intermediate values of the junction width, there is a complex transition between these two regimes, with intermediate-energy features around ~0.35 eV showing up in the spectra. The transition is fast, but not singular. These conclusions seem to be rather independent of the length of the junctions n. As Fig. 1(e) illustrates, no significant spectral changes are induced when the plasmon energies are plotted as a function of junction length n. The three distinct behaviors of the plasmon energies noted above for narrow, intermediate, and wide junctions appear for all values of the junction length.

Further insight into the character of these plasmons is provided by examining their induced-charge distributions, which are plotted in Fig. 2(a)-(d) for representative structures along the transition from narrow to wide junction regimes. The polarization profile for the highenergy plasmon around ~0.47 eV in the non-touching dimer (m = 0, Fig. 2(a)) displays a dipole-dipole pattern that is not substantially modified when a narrow junction (m = 2, Fig. 2(b)) is present between the two triangular islands of the structure. This distinct polarization pattern is characteristic of a BDP. In contrast, the intermediate energy plasmon around ~0.35 eV (Fig. 1(c)) for a junction width m = 6shows a clear dipolar polarization pattern across the junction. This is the JP that is formed from the local electronic properties of the junction. A weak charge transfer between the two nanotriangles is also evident in the plot and corresponds to the CTP, which is delocalized over the entire graphene structure like an standing wave. The latter shows a consistent +- charge polarization that does not change with increasing m, as shown for m = 8 in Fig. 2(d). The CTP becomes the dominant feature once the junction is sufficiently wide. The signs superimposed on the density plots of Fig. 2(a)-(d) qualitatively correspond to the distribution of the plasmon induced charge as a function distance to the dimer center, which is shown in Fig. 2(e) (see also SI [45]). This analysis is rather independent of the length of the junction n. An in-depth inspection of bowties of



Figure 1 (online color at: www.lpr-journal.org) Quantum junction plasmons in graphene bowties. (a) Details of the junction region in the structures under study, with definitions of bridge length *n* and width *m*. (b-d) Spectra for selected bridge widths m = 0, 4, 8, and various lengths as indicated by different colors. The insets show the complete graphene structures for n = 2. (e) Plasmon resonances as a function of bridge width *m*. The color code for different lengths *n* is given in the upper inset. The area of the circles is proportional to the area under the extinction peak for each plasmon feature. Classical-electrodynamics plasmon energies are shown by dashed curves for n = 4 (red) and n = 8 (blue), flanked by shaded areas that represent the strength of the modes. (f) Plasmon resonances as a function of bridge length *n*. The graphene is doped to a Fermi energy $E_F = 0.4 \text{ eV}$ and has a mobility $\mu = 10,000 \text{ cm}^2/(\text{V s})$. The full length of the bowties is 8 nm.



Induced charge density (arb. units)

Figure 2 (online color at: www.lpr-journal.org) Plasmon induced-charge distributions for representative graphene bowties. (a-d) Density plots with the color of each atom indicating its induced charge for different junction widths m = 0, 2, 6, 8 and the same length n = 4. (e) Induced charge integrated along the horizontal direction and averaged over four nearest carbon-atom neighbors.

increasing length leads to results that are consistent with the above conclusions, including the emergence of quantum junction plasmons once the width of the junction and the length of the structure are sufficiently large (see SI [45] for more details).

A classical-electrodynamics description of these graphene nanostructures (see broken curves in Fig. 1(e) and SI [45]) does not completely reproduce the plasmonic behavior derived from first-principles. For instance, classical calculations miss the intermediate-energy junction plasmon that we observe in the quantum calculations for intermediate junction widths. Another discrepancy is that classical theory predicts a smooth fadeout of the dipoledipole mode exhibited by non-overlapping triangles when the junction width is increased, while the CTP shows a singular behavior, as it migrates towards zero energy in the limit of vanishing junction width. This is in striking contrast to the results from the quantum description, in which the CTP is rather dispersionless and fades out smoothly in that limit (see Fig. 1(e)). The dipole-dipole and charge-transfer



Figure 3 (online color at: www.lpr-journal.org) Electronic junction states and electron-hole transition strengths. (a,b) Electron density distribution of electronic states in n = 4 bowtie antennas for m = 0 ((a), separated triangles) and m = 6 ((b), bridged triangles). For each state of energy E > 0, there is a state of energy -E (not shown) with identical density distribution. Only low-order states close to the Dirac point (zero energy) are shown. The energies of these states are indicated by black lines right under the corresponding density plots. (c) Dipole matrix elements between electronic states of the same bowtie as in (b). The area of the circles is proportional to the dipole strength. We only show transitions between states separated by energy differences larger than $|E_i - E_j| > 0.01$ eV. The solid lines connect initial and final energies corresponding to 0.47 eV (BDP), 0.35 eV (JP), and 0.22 eV (CTP) excitations.

nature of the classically predicted plasmons is clear from their induced-charge distributions (see SI [45]).

The electronic states involved in the plasmon of separated nanotriangles (Fig. 3(a)) are nearly identical to those of individual triangular islands, but with a small amount of Coulombic interaction and hybridization. When a bridge of intermediate width is added to form a junction (Fig. 3(b)), the level of hybridization of these states increases, and new electronic junction states emerge. In particular, two new junction states are observed near the Dirac point (zero energy), which are expected due to the presence of carbon zigzag edges in the bridge [46,47]. Thus, the junction plasmon noted above must be supported by excitations involving electrons or holes in these electronic junction states. The strength of the electron-hole pair (e-h) dipole transitions in this system are represented in Fig. 3(c) as a function of initial and final energies (the strength is represented through the area of the circles). Clearly, the plasmon energies (solid curves) do not strongly overlap with the dominant e-h transitions. This energy mismatch shows that the optical transitions are not single-particle excitations and is a strong argument supporting our conclusion of collective plasmonic nature of the bowtie optical excitations under consideration. Similar results are observed for different widths of the junction (see SI [45]).

3. Outlook and conclusions

Graphene dimers provide advantageous systems compared to metallic dimers both because of the structural robustness of this material and because the carbon structure can be reliably imaged with atomic detail [48, 49], and therefore, a detailed correlation between atomic structure and optical response is within reach experimentally. This is not the case of gaps formed by closely spaced three-dimentional metallic particles or tips [50, 51], which suffer from inherent faceting and structural uncertainties. Planar graphene nanostructures are thus ideal systems on which to study the interplay between structural features (e.g., ripples and defects) and the plasmonic response (for example, the amount by which they increase the plasmon scattering and dephasing rate via electron-hole pair generation). Recent advances in atomically resolved imaging [52] and patterning [53] of graphene should facilitate this task.

The remarkably large variations here reported in the optical properties of plasmonic structures consisting of thousands of carbon atoms by just adding a few (< 10) extra atoms provides a handle for tuning plasmons in graphene. Together with the already established chemical [54] and electrical [55] tunability of this material, this adds a novel tuning parameter, which can be potentially exploited for ultrasensitive optical sensing (e.g., by analyzing the dependence on the presence of molecules near the junction, which can modify the local valence charge density).

Although for simplicity we only discuss above freestanding graphene with a Fermi energy $E_F = 0.4 \text{ eV}$, no qualitative changes are observed for different doping or when the graphene sheet is placed on a substrate (see SI). Quantum effects are found to be more significant when the level of doping is reduced (see results for $E_F = 0.2 \text{ eV}$ in the SI), as one expects for the smaller number of doping electrons participating in the plasmons, whereas the CTP energies are closer to the classical description when the doping increases (see $E_F = 0.8$ eV results in the SI).

In conclusion, using a fully quantum mechanical approach we have studied the optical properties of graphene dimers consisting of several thousand carbon atoms connected by a narrow graphene junction. The optical properties of the system are found to be strongly sensitive to the detailed structure of the junction, with the spectrum exhibiting drastic changes with the addition of a few carbon atoms in that region. In addition to the conventional hybridized and charge transfer plasmons observed for metallic dimers [39-42], graphene dimers display junction plasmons of a purely quantum mechanical origin, which does not seem to appear in conventional metallic junctions [56]. The plasmonic response of this graphene structure is found to be strongly influenced by quantum effects, making it of significant interest in the emerging field of quantum plasmonics as well as for novel nanophotonics and optoelectronics applications.

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