Electrically tunable graphene anti-dot array terahertz plasmonic crystals exhibiting multi-band resonances: supplementary material

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This document provides supplementary information to “Electrically tunable graphene anti-dot array terahertz plasmonic crystals exhibiting multi-band resonances,” <http://dx.doi.org/10.1364/optica.2.000135>. Included contents are calculation of the surface-plasmon-polariton band-structure of square-lattice graphene periodic anti-dot arrays employing the empty lattice approximation, details for device fabrication and numerical simulation methods, electrical properties of the graphene material, characterization and simulation results of several additional investigated graphene periodic anti-dot arrays. © 2015 Optical Society of America

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1. Calculation of the SPP band-structure of square-lattice graphene anti-dot arrays employing the empty lattice approximation

The band-structure of the SPP dispersion curves associated with a graphene periodic anti-dot array with infinitely small apertures can be analytically calculated employing the so-called empty lattice approximation. The SPP dispersion relation for an intact graphene sheet in the THz frequency range is approximately:

$$\omega_{SP} = \sqrt{\frac{e^2 E_F}{2\pi\hbar^2 \epsilon_0 \epsilon_{avg}}} k_{SP} \quad (S1)$$

where ϵ_0 is the vacuum permittivity, ϵ_{avg} is the average dielectric constant of the environment surrounding the graphene sheet, and E_F is the Fermi energy which satisfies $E_F \gg \hbar\omega_{SP}$. In a graphene periodic anti-dot array with negligible aperture size, the periodic lattice structure introduces Bragg scatterings to propagating SPP waves, resulting in folding of the single SPP dispersion curve described in Eq. (S1) into the first Brillouin zone by different orders of reciprocal lattice vectors and thus forming a SPP band-structure. Such a SPP band-structure can be analytically calculated by simply performing such a zone-folding process using Eq. (S1), and an example is given in Fig. S1. As a consequence of the symmetry of the periodic lattice structure, multiple dispersion curves of such an empty lattice intercept at the high symmetry points (e.g. the Γ -point) of the first Brillouin zone. However, finite-sized apertures would introduce modifications to the band-structure of an empty lattice and lift certain degeneracies at these high symmetry points. Although the accurate band-structure must be computed numerically, the band-structure calculated based on the empty lattice approximation nevertheless provides a first-order estimate of the expected frequency range of dominant

optical responses (such as the coupled SPP modes) of a graphene periodic anti-dot array.

Fig. S1. Band-structure of the SPP dispersion curves of a square-lattice graphene periodic anti-dot array with a lattice constant of 4 μm calculated employing the empty lattice approximation ($E_F = 0.45$ eV). The dashed lines represent the light line. The reciprocal lattice vectors responsible for the zone-folding of different branches of SPP dispersion curves to the highly degenerate Γ -point are indicated. Note that all the equivalent reciprocal lattice vectors are indicated with one index in the plot, e.g. [1,0] represents in total 4 equivalent reciprocal lattice vectors which are [1,0], [-1,0], [0,1] and [0,-1], respectively.

2. Methods

Device fabrication

The patterning of all the large-area graphene periodic anti-dot array structures is accomplished with deep-UV photolithography (wavelength ~ 220 nm) with a ~ 200 nm thick layer of poly(methyl methacrylate) (PMMA) of 950k molecular weight as the

photoresist. After the exposure, the PMMA layer is developed in a mixture of 3:1 isopropanol:methyl isobutyl ketone (MIBK) for 60 s. Oxygen plasma (40 mTorr and 50 W) etching for 30 s is then employed to transfer the PMMA pattern onto the graphene sheet. Subsequently, to define the pattern for the metal electrodes, a second step of deep-UV photolithography is conducted with a double-layer photoresist consisting of a poly(methyl methacrylate-co-methacrylic acid) (P(MMA-MAA)) copolymer bottom layer and a PMMA top layer, utilized to facilitate the lift-off process. Deposition of 5 nm Ti and 70 nm Au is carried out with an electron-beam evaporator and followed by a lift-off process in warm acetone. Finally, the samples are annealed at 350 °C in N₂ ambient for 600 s to reduce the chemical residues adhered to the graphene sheet as well as to improve the metal-graphene contacts.

Full-wave simulation

Full-wave simulations of the transmission spectra are performed for all the studied graphene anti-dot arrays using finite-element frequency domain methods (CST Microwave Studio). The graphene sheet is modeled as a 0.3 nm thin conducting layer with a frequency dependent surface conductivity described within the Drude model [1,2] as:

$$\sigma(\omega) = \frac{e^2 E_F}{\pi \hbar^2} \frac{i}{\omega + i\tau^{-1}} \quad (S2)$$

where E_F is the Fermi energy and τ is the carrier relaxation time. In the frame of this work ($k_{SP} \ll k_F$, $\hbar\omega \ll E_F$, $k_B T \ll E_F$, where k_{SP} is the wave vector of the graphene SPP and k_F is the Fermi wave vector), Eq. (S2) is an accurate approximation to the graphene conductivity derived within the more general random phase approximation (RPA) formalism [1,2]. Measured diameters of the etched apertures are used in the simulation for higher accuracy. The frequency dependent dielectric constants of SiO₂ and Si are taken from a widely used reference [3]. The excitation source is set as a linearly polarized normal-incident plane wave from free space and the output port is positioned in the Si substrate as an open boundary (thus no additional reflection).

3. Carrier mobility of unpatterned graphene samples

The carrier mobility of the unpatterned graphene samples is extracted from their current-voltage characteristics when operated in a field-effect transistor (FET) configuration. Figure S2(a) shows the measured source-drain resistance as a function of the back-gate voltage offset with respect to the charge neutrality point (CNP) of a typical sample. The back-gate voltage to reach the CNP is about +90 V for this particular sample and it varies within a range of ± 15 V for different samples. The carrier mobility plotted in Fig. S2(b) is calculated based on the data in Fig. S2(a) as $\mu = 1/n\rho_S$ where ρ_S is the sheet resistivity, n is the sheet carrier density and its value is approximated with the simple parallel capacitor model as $n = (V_G - V_{G,CNP})C_{OX}$ (C_{OX} is the capacitance per unit area of the capacitor formed between the graphene sheet and the Si substrate with the ~ 300 nm thick SiO₂ layer as the dielectric medium). This relatively simple approximation of the sheet carrier density is rather accurate for the relatively high carrier density range ($> 10^{12}$ cm⁻²) relevant to the experimental observations of the distinct graphene SPP resonances in the transmission spectroscopy characterizations. The typical average carrier (hole) mobility at room temperature is ~ 1000 cm²/Vs within this carrier density range.

Fig. S2. (a) Source-drain resistance vs. back-gate voltage offset with respect to the CNP of an unpatterned graphene sample (the surface dimension of the graphene field is 3 mm $\times$ 3 mm) fabricated in a FET device configuration. (b) Carrier mobility extracted from the data in (a). An average hole mobility of ~ 1000 cm²/Vs is obtained for the carrier density range relevant to the observations of graphene SPP resonances in the transmission spectroscopy characterizations.

4. Transmission spectroscopy characterizations and simulation results for the graphene periodic anti-dot array samples SQ5 to SQ7

The transmission extinction spectra at various back-gate voltages for the graphene periodic anti-dot array samples SQ5 to SQ7 are shown in Fig. S3, in which a single relatively broad resonance peak is observed for each sample.

Fig. S3. Transmission extinction spectra at different back-gate voltages for the graphene periodic anti-dot array samples (a) SQ5, (b) SQ6, and (c) SQ7.

Full-wave simulations are conducted for all the structures in the same way as described in the main text. The simulated transmission extinction spectra for the graphene periodic anti-dot array samples SQ5 to SQ7 are shown in Fig. S4 together with the corresponding experimental results at the highest achieved carrier density. The simulation results reveal that each of these experimentally observed broad resonance peaks originates from a superposition of two or three SPP resonances that are not individually resolved.

Fig. S4. Simulated transmission extinction spectra for the graphene periodic anti-dot array samples (a) SQ5, (b) SQ6, and (c) SQ7, assuming two different values of the carrier relaxation time τ and in comparison with the experimental results.

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