Scalable and Tunable Periodic Graphene Nanohole Arrays for Mid-Infrared Plasmonics

Gopalan, Kavitha K.; Paulillo, Bruno; Mackenzie, David; Rodrigo, Daniel; Bareza, Nestor; Whelan, Patrick; Rebsdorf; Shivayogimath, Abhay; Pruneri, Valerio

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Scalable and tunable periodic graphene nano-hole arrays for mid-infrared plasmonics


*ICFO-Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels, Spain

⊥Center for Nanostructured Graphene (CNG), Technical University of Denmark, DK-2800, Kgs, Lyngby, Denmark.

‡Department of Micro- and Nanotechnology (DTU Nanotech), Technical University of Denmark, DK-2800, Kgs, Lyngby, Denmark

#ICREA-Institució Catalana de Recerca i Estudis Avançats Passeig Lluís Companys, 23,08010 Barcelona, Spain

E-mail: valerio.pruneri@icfo.eu
A) Plasmonic linewidth evolution vs geometry in GNHAs

The linewidths of the plasmonic modes M1 and M2 have been extracted from the simulated spectra in Figure 2(b)-(c) to follow their evolution as a function of the geometric parameters. A multi-peak Lorentz fit was used to deconvolute the two modes and estimate their bandwidth. We observe that the linewidth scales inversely with geometric parameters P and D as the damping pathways of graphene plasmons, namely edge and phonon scattering, are modified. This has been thoroughly investigated in the paper by Yan et al. 1. For similar reasons, the linewidth increases as a function of the gate voltage for a fixed GNHA geometry, as one can see from the spectra in Figure 3 and 4 of the manuscript.

As a consequence, the M1 mode at large periods (e.g. 180 nm) shows Q-factors as high as 20 due to the interaction with SiO2 phonons, the mode spectral position being at the edge of SiO2 Reststrahlen band. Conversely, M2 mode is far enough from SiO2 phonons and it shows lower Q-factor (≤10). We finally note that the experimental plasmonic bandwidth for e-beam and nano-imprinted GNHA samples are similar.

B) Further measurements on nano-imprinted graphene nanohole arrays

The graphene nanohole arrays fabricated using nanoimprint lithography were also characterized by the following measurements.

**Electrical Measurements:**

Devices were electrical characterized in a Linkam LTS600-P probe station with a controlled dry N2 atmosphere. Prior to measurements, devices were thermally treated at 225 °C for 30 minutes in N2, with measurements performed at 30 °C. The gate-dependent sheet resistance $R_S$ was determined using the van der Pauw equation $e^{-\frac{\pi R_A}{R_S}} + e^{-\frac{\pi R_C}{R_S}} = 1$ 3 with $R_A$ and $R_C$ as defined in Figure 1 a). In addition, gate-dependent device uniformity defined by $\beta = R_A/R_C$ was simultaneously determined, as previously defined here 4.
Devices were measured after NIL processing to determine conductivity, doping level and back-gate integrity, with a typical result shown in Figure 1 a). Here we observe a sheet resistance of ≈ 15 kΩ at the charge neutrality point (CNP), a CNP of ≈ 1 V, and carrier mobility of ≈ 200 cm²/V·s. Devices showed CNPs of up to 20 V, consistent with previously measured NIL devices. The gate dependent homogeneity of the devices (Figure S2 a) inset) was found to be 1.5 at the CNP, which is consistent with previous large scale CVD devices, with only a small variation as a function of $V_G$, which suggests there is relatively low doping homogeneity across the device.

Figure S2. a) Gate-dependent electrical characteristic of a device post NIL processing, with definition of $R_A$ and $R_C$ shown in top right corner. Inset: Gate-dependent device homogeneity $\beta = R_A/R_C$. b) THz-TDS-derived conductivity map of a device before NIL processing. c) THz-TDS-derived conductivity map of the device from Figure 1 b after NIL processing.

**THz Time Domain Spectroscopy Conductivity Measurements**

THz-TDS is a rapid wireless technique for graphene conductivity mapping. THz-TDS was performed using a Picometrix T-ray 4000, with the THz conductivity ($\sigma_{\text{THz}}$) being derived as previously described, with the $\sigma_{\text{THz}}$ averaged over the 0.3-1.4 THz range, and with a step size of 200 µm. Devices were mapped both directly before the NIL processing (Figure S2b) and after resist removal (Figure S2c). Pixels where no graphene conductivity values were obtained (typically at the metal contacts) are set to black. An overall decrease in $\sigma_{\text{THz}}$ was observed as a result of the NIL process, which is consistent with previous comparison of NIL devices before and after imprinting. The uniformity across the 5 mm × 5 mm device area shows comparable variations in $\sigma$ to other graphene THz-TDS maps of the same sized area. The THz-TDS measurements were performed in ambient conditions in the presence of humidity an effect known to affect $\sigma_{\text{THz}}$, with the lower values of $\sigma_{\text{THz}}$ compared to $\sigma_{\text{vdP}}$ of Figure S2 a) being attributed to p-doping of the device by water.

**Raman Characterization of NIL fabricated GNHA**

Micro-Raman Spectroscopy was performed using a Thermo Scientific DXRxi using a 532 nm laser with power of 2 mW. A map was performed over the entire device with a stepsize of 100 µm. Each pixel used an exposure frequency of 180 Hz with 200 scans per pixel, giving 2500 spectra for each device. The graphene Raman peaks were with fitted with Lorentzians as previously described.
The ratio of the intensity of the G-peak ($\approx 1580 \text{ cm}^{-1}$) and the D-peak ($\approx 1350 \text{ cm}^{-1}$) are an indication of the defect density of graphene. The results of the Raman mapping for the device characterized in Figure S2 a) are shown in Figure 2. We observe a relatively low $I_D/I_G$ median of 0.25, as shown in Figure S3.

![Graph showing ratio of intensity of D-peak to G-peak](image)

Figure S3. Micro-Raman analysis of the device shown in Figure S2a for 2500 spectra performed after NIL processing. Ratio of the intensity of the D-peak to intensity of the G-peak ($I_D/I_G$).

**Raman Characterization of electron-beam lithography fabricated GNHA**

Figure S4 shows results from the Raman characterization of the EBL fabricated samples were carried out using a Renishaw inVia with a laser of 532 nm and power of 2mW over an area of 150 µm * 150 µm. The median of the ID/IG ratio is 0.15, which indicates a lower defect density of the EBL fabricated GNHA samples.

![Graph showing ratio of intensity of D-peak to G-peak](image)

Figure S4. Ratio of the intensity of the D-peak to the intensity of the G-peak ($I_D/I_G$) of the GNHA fabricated by electron beam lithography over a sample area of 150 µm x 150 µm.
References:


