Supplementary Information: Highly Resonant Graphene Plasmon Hotspots in Sub-Wavelength Geometries

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1. KPFM measurement of constriction geometries

Typically, the size and shape of a graphene nanoresonator is determined by atomic force microscopy (AFM). However, in structures composed of single-layer (1LG) and bi-layer (2LG) graphene, the Ångstrom-scale height differences (see Figure 2(d) of the main text) make it challenging to accurately determine map boundaries between them. Here we use Kelvin probe force microscopy (KPFM) instead, as it offers much greater contrast between 1LG and 2LG due to the significant differences in their local work functions[1].

This is clearly seen in Figure S1, which shows the various geometric approximations used to predict the behaviour plasmon nanoresonator hotspots in the main text. The errors in the nanoresonator critical lengths, L_c, are due to the ~50 nm resolution of frequency-modulated KPFM[1].

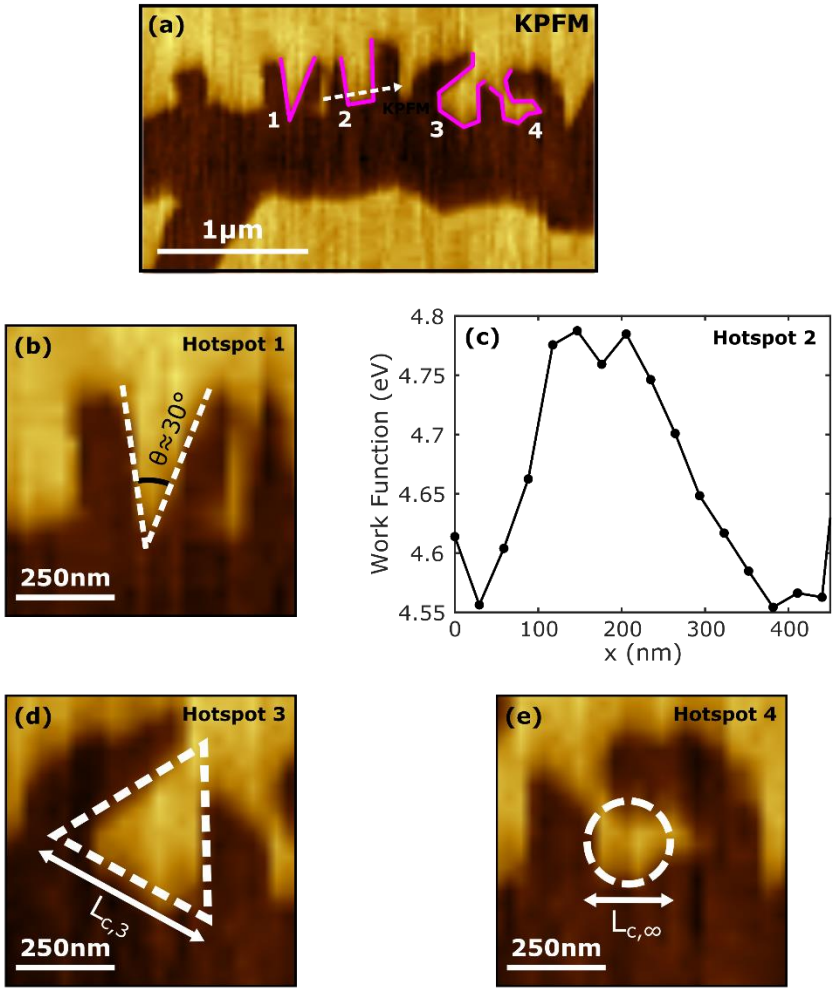


Figure S1. KPFM measurements of the geometries of the hotspot regions in (a). (b) Region 1 is approximated as a 30° wedge. The dashed white line in (a) indicates the scan of (c) the line profile used to measure region 2, which comprises a "1D" cavity of width $L_c = 200 \pm 50$ nm connected at the bottom by a straight edge. (d) An equilateral triangle of side length $L_{c,3} \approx 450$ nm approximates the straight edges of region 3. (e) A simple circle with diameter $L_{c,\infty} \approx 240$ nm is used for region 4.

2. Measurement of wave model parameters

The wave model introduced in the main text requires three parameters to be extracted from interference fringes caused by plasmons reflected from a simple straight. For the 1LG/2LG boundaries studied in this work, the first parameter, $m = 2$, is extracted from Figures 4(b) and

4(d), as discussed in the main text. The remaining two parameters, $F_1 = 0.14$ and $\rho \equiv \frac{F_2}{F_1} = 0.23$, are extracted from the s-SNOM line profile in Figure S2.

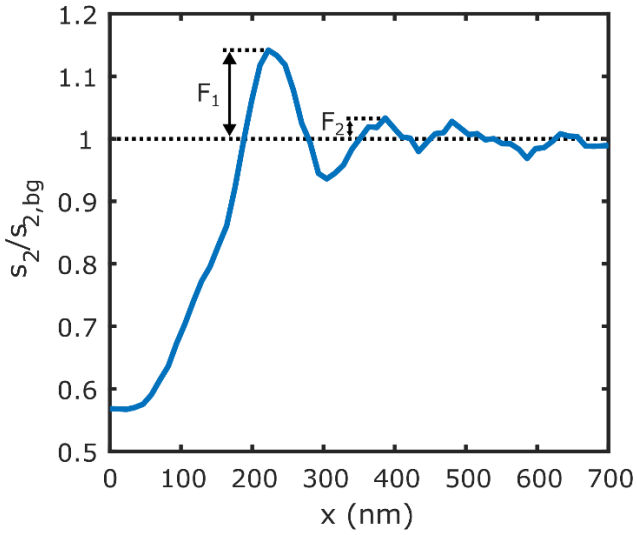


Figure S2. Normalised s-SNOM amplitude, $s_2/s_{2,bg}$, line profile of plasmon interference fringes caused by reflection from the straight 1LG/2LG boundary indicated by the cyan rectangle in Figure 4(a) of the main text. The measured values of the first and second fringe strengths are $F_1 = 0.142$ and $F_2 = 0.033$, respectively.

3. Comparison of plasmon reflection from 2LG islands and boundaries

As an extra test of the nature of the "islands" referred to in the main text (blue arrows in Figure 2(c)), we compare the form of plasmon reflection fringes from 1LG/Island boundaries with Raman-confirmed 1LG/2LG boundaries and SiC/1LG boundaries in Figure S3. The line profiles of near-field scattering amplitude, s_2, in Figure S3(b) clearly demonstrate that both the relative strength and form of the 1LG/Island fringes are in excellent agreement with the known 1LG/2LG fringes, while in disagreement with SiC/1LG fringes. This adds strong evidence that the islands are composed of 2LG, as opposed to exposed SiC.

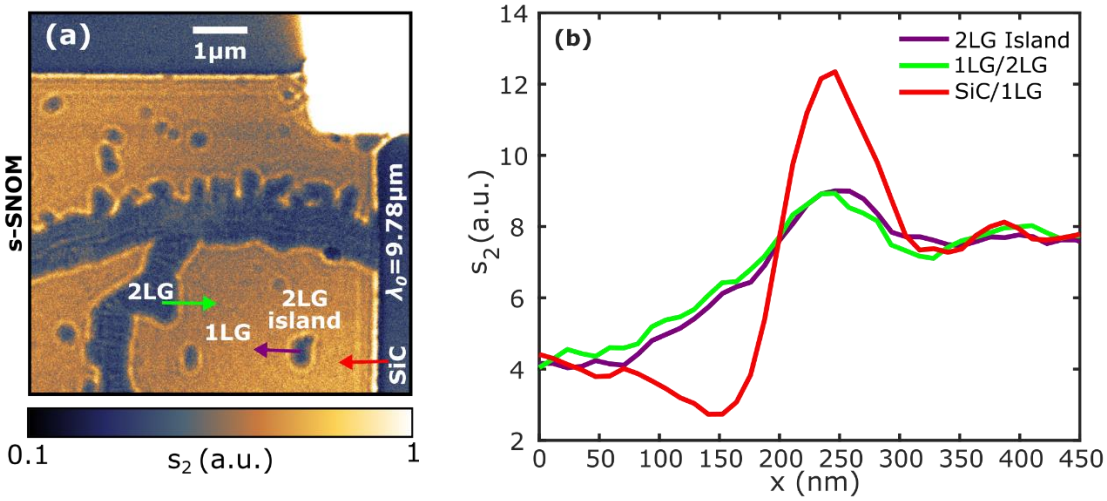


Figure S3. (a) Near-field scattered amplitude, s_2, showing the locations of 1LG, 2LG and a 2LG island. The coloured arrows correspond to (b) the line profiles of plasmon reflection fringes, which show that the island fringes are in strong agreement with 1LG/2LG fringes.

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References

[1] Panchal V, Pearce R, Yakimova R, Tzalenchuk A and Kazakova O 2013 Standardization of surface potential measurements of graphene domains *Sci. Rep.* **3** 2597