Supplementary materials

# Resonant Raman Scattering by Graphene: SERS and gap-mode TERS

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We demonstrate the gm-TERS map of the D mode intensity of graphene. As seen, the highest intensity of D mode is observed in the semi-ring area at the edge of the gold nanodisk and in the area of the nanofold (Fig.S1a). The gm-TERS spectra of graphene near the nanodisk are shown in (Fig.S1b) and the spectral range of mapping is noted by the blue rectangle.

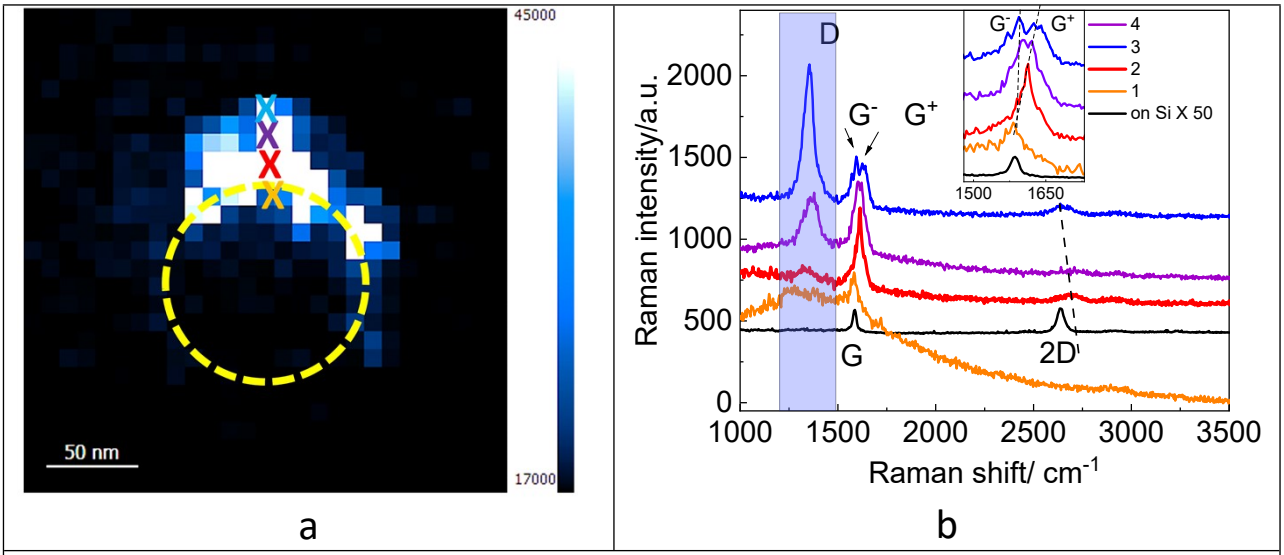


**Figure S1** Gm-TERS map of **D mode** intensity for a sample area of 250 × 250 nm2 A yellow dashed circle depicts the position of gold nanodisk (a) Gm-TERS spectra of graphene near a gold nanodisk measured from areas marked in Fig.S1a by crosses

To demonstrate the variety of gm-TERS images we present we present another map of the intensity of G-mode of graphene, measured in the area of a neighbour nanodisk (Fig.S2a). The map looks pretty much the same as the one shown in Fig. 4a. Nevertheless, local spectra extracted from nanofold area have some differences in the frequency positions of the peaks (Fig.S2b).

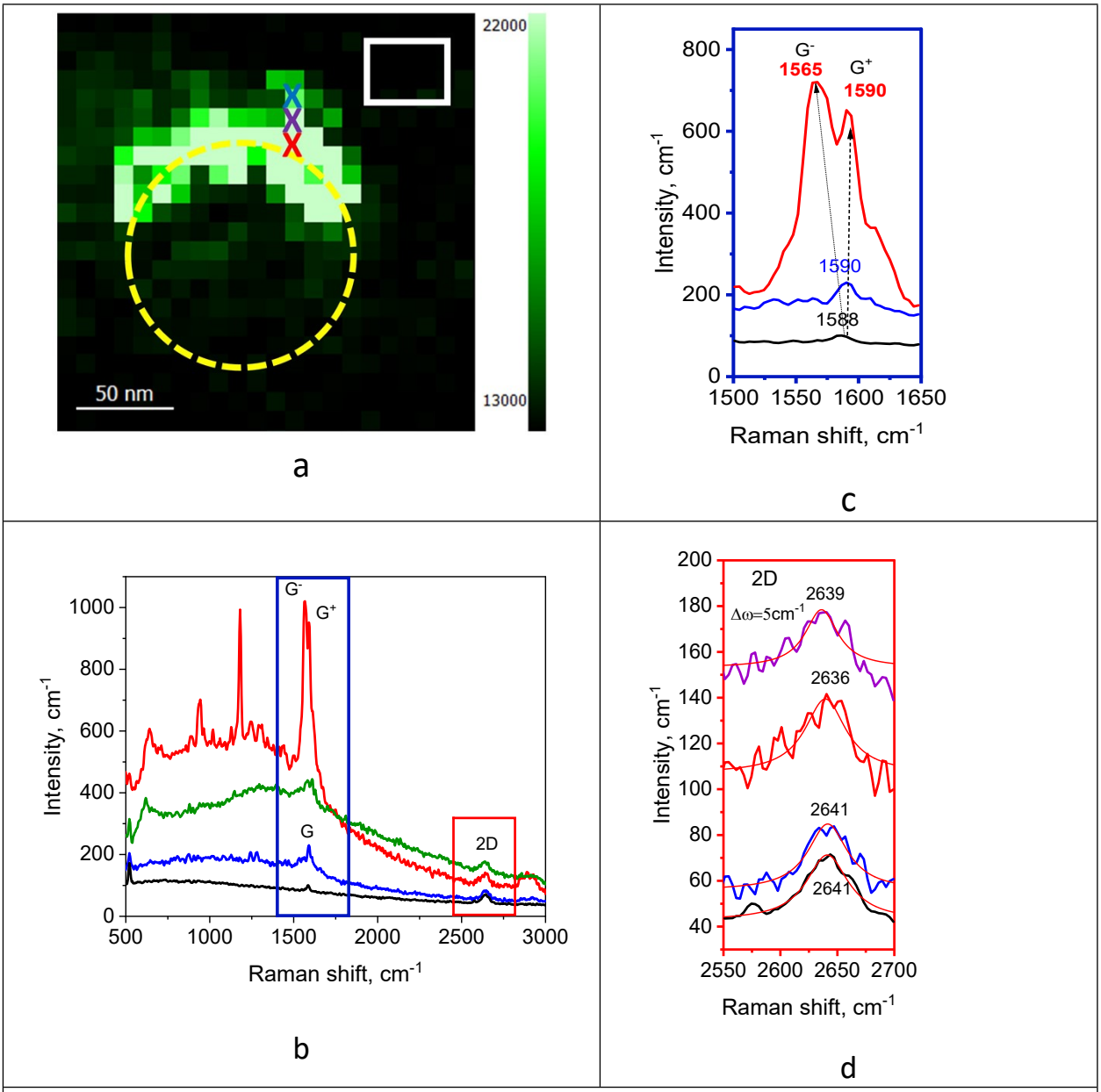


**Figure S2** Gm-TERS map of G mode intensity for a sample area of 250 × 250 nm$^2$. A yellow dashed circle depicts the position of gold nanodisk (a) Gm-TERS spectra of graphene near a gold nanodisk measured from areas marked in Fig.S2a by crosses (b) and the same spectra presented in the spectral ranges of 1500-1650 cm$^{-1}$ (c) and 2550-2700 cm$^{-1}$ (d). The TERS spectra and the areas indicated by crosses are shown by the same colors.

To make the plasmonic enhancement effect clear, we measured gm-TERS signal from bare gold nanodisks which are shown in (Fig.S3a). As expected, the intensive gap-mode plasmon is observed in the semi-ring area near the edges of the disks (Fig.S3b). The gm-TERS plasmon feature was compared with that of the plasmon, arising at the apex of the silver probe (Fig.S3c). As seen, in contact with the gold nanodisk, the plasmon energy changes due to formation of "collective" gap-mode plasmon with a notable increase in intensity.

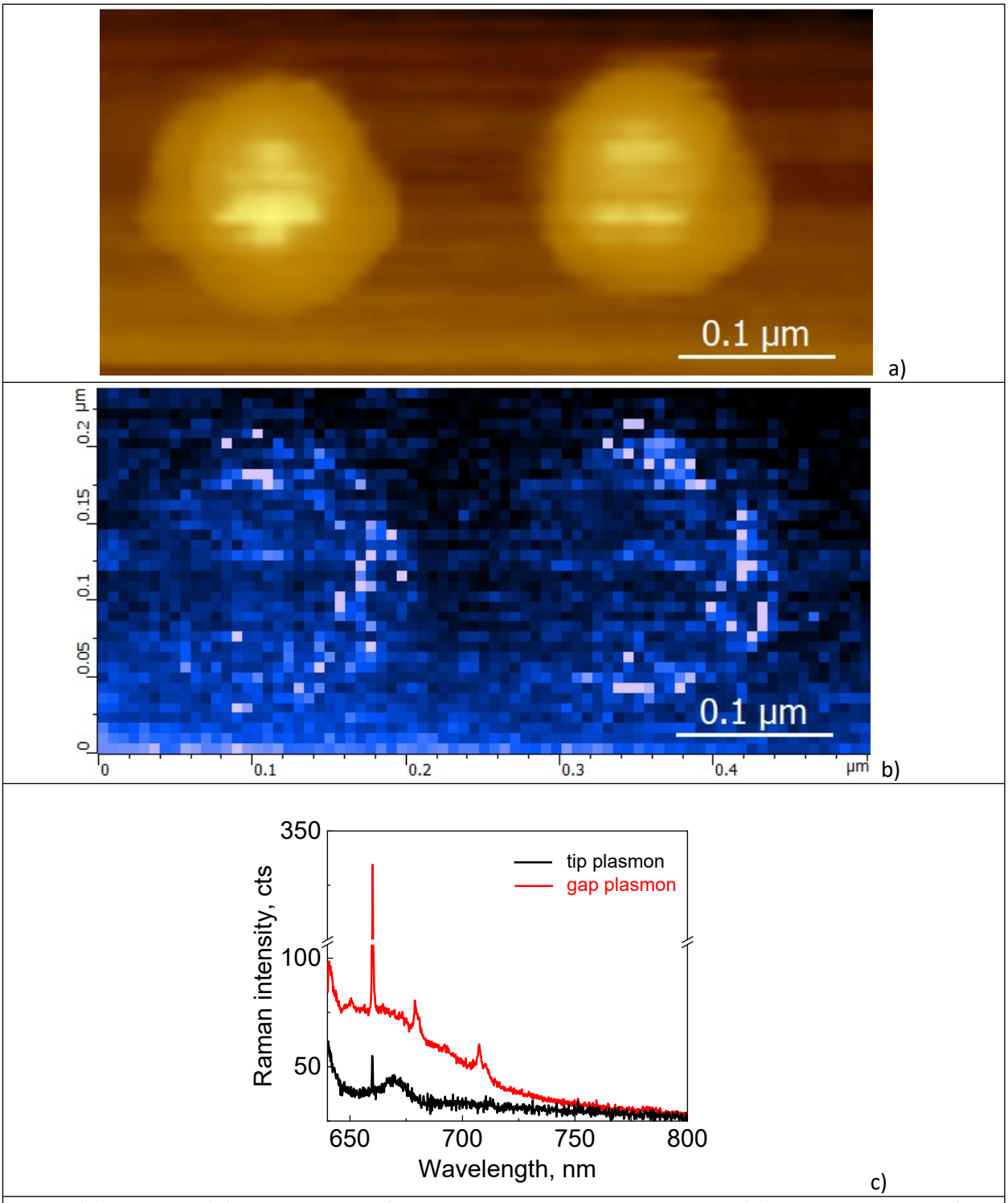


**Fig.S3** (a)- AFM and (b) TERS- images of bare nanodisks measured by Ag tip. (c) Raman spectrum of Ag tip apex and TERS spectrum Au nanoclusters measured in gm-TERS conditions