## In-situ Raman study on CVD-grown graphene microbridge under high current density

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*In-situ* Raman spectroscopy was performed on microbridges of single-layer graphene (SLG) and double-layer graphene (DLG) under electrical current density up to ~10<sup>8</sup> A/cm<sup>2</sup> at room temperature in air. The graphenes were prepared by chemical vapor deposition (CVD) in high vacuum and the microbridges (5  $\mu$ m × 80  $\mu$ m) were fabricated by electron beam lithography and etching with O<sub>2</sub> plasma. The 2D phonon peak shifts to lower frequencies as the current density increases through the graphene microbridges, as well known by previous studies[1, 2]. The peak normally returns back to its original values upon cooling, when the current density is lower than 10<sup>7</sup> A/cm<sup>2</sup>. However, beyond the current density of 0.6 × 10<sup>8</sup> A/cm<sup>2</sup>, we find that the 2D and G peaks do not restore fully back to their initial values after switching off the current. The Raman peaks are found to be at higher frequencies than the initial values for both SLG and DLG microbridges. The up-shift of the 2D peaks, after switching off the electrical current, is believed to be due to p-doping of the graphene samples in air. Our findings suggest that the doping of graphene can be dependent on the current density.

## References

- [1] I. Calizo et al., Nano Lett., 7 (2007) 2645.
- [2] I. Calizo et al., Appl. Phys. Lett., 91 (2007) 071913.

## Figures



FIG. 1. (a) A schematic diagram of the experimental setup for *in-situ* Raman measurements under electrical current flow. (b) Optical microscopy images and Raman spectra of single- and double-layer graphene microbridges with a dimension of 5 x 80  $\mu$ m<sup>2</sup>.



FIG. 2. Evolution of Raman spectra of graphene as a function of the applied current. Raman spectra of (a) SLG under current from 0 to 2600  $\mu$ A, and (b) DLG under current from 0 and 4000  $\mu$ A. The blue lines are the Raman spectra of the graphene at the initial state (zero current), the black lines under the electrical current and the red lines off the current. Note that the peaks of the red spectra are not fully restored to those of the blue spectra. The blue dotted lines are the guides for the G and 2D peaks of the graphene at the initial state.



FIG. 3. (a) The frequency shifts of the Raman G and 2D peaks ( $\Delta \omega_{G, 2D}$ ) of SLG and DLG at various levels of applied current density on and off. (b) The down-shifts of Raman frequency correspond to Joule heating of the graphene microbridges under current. (c) The up-shifts of Raman frequency correspond to doping in air after the applied current is switched off.