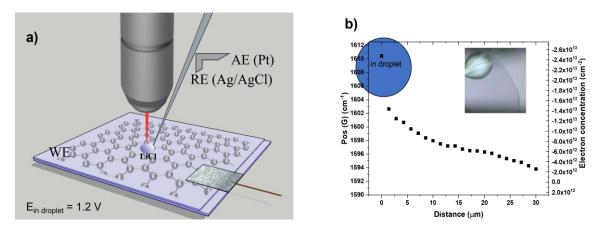
## Exploring spatial charge carrier distribution in locally gated graphene using Raman µ-droplet spectroelectrochemistry

## Iryna Ivanko

Martin Jindra, Otakar Frank, Matěj Velický; J. Heyrovský Institute of Physical Chemistry of the CAS, v. v. i. Dolejškova 2155/3, 182 23 Prague 8, Czech Republic;

iryna.ivanko@jh-inst.cas.cz

Graphene, with its exceptional electronic properties, continues to captivate researchers for applications in next-generation electronic devices [1]. The precise control and monitoring of charge-doping levels is pivotal for tailoring graphene's electronic characteristics. This work focusess on monitoring lateral charge doping of single-layer graphene (SLG) induced by localized electrochemical top gating. In situ Raman spectroelectrochemistry in a microdroplet electrochemical cell (Fig. 1 a) was used to investigate the extent of lateral doping beyond the biased area (droplet). Raman spectra were acquired in line profiles with gradually increasing distance from the droplet, revealing subtle softening of the G band (Fig. 2 b) and related changes of the 2D band, corresponding to the decay of doping levels outside the droplet. Potential of 1.2V applied to the  $\mu$ -droplet is reflected as 2.5x10<sup>-13</sup> cm<sup>-2</sup> hole carrier concentration (Fig. 2 b). Analysis of Raman spectra along the line scan reveals decay of the charge carrier's concentration reaching up to 30  $\mu$ m from the electrochemically gated area (Fig. 2 b). This might imply that graphene does not behave as a conductor but rather as a semiconductor. It is worth mentioning that the lateral distribution of charge is notably contingent upon the spatial positioning of the contact point. To disentangle the overlapping effects of changes in the SLG electronic and structural properties induced by doping and strain, vector analysis of the G and 2D band positions was done [2]. The correlation plot of  $\omega_{2D}$  and  $\omega_{G}$  modes reveals a quasi-linearity ( $\Delta\omega_{2D}$  / $\Delta\omega_{G}$  = 0.7), attributing lateral decay the charge carriers induced by doping rather than strain.



**Figure 1:** a) Schematic of the *in situ* microdroplet Raman spectroelectrochemistry setup; b) Position of the G band and charge concentration as a function of distance from the droplet.

## References

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- [2] Lee, J. E., Ahn, G., Shim, J., Lee, Y. S., Ryu, S. Nature communications, 3, (2012), 1024.

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