

Ultrafast lithium diffusion in bilayer graphene

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Solid mixed conductors with significant ionic as well as electronic conduction play a pivotal role for mass transfer and storage as required in battery electrodes. Single-phase materials with simultaneously high electronic and ionic conductivity at room temperature are hard to come by and therefore multi-phase systems with separate ion and electron channels have been put forward instead [1]. Here, we explore bilayer graphene as a true single phase mixed conductor and demonstrate ultrafast lithium diffusion exceeding diffusion in bulk graphite by an order of magnitude and even surpassing diffusion of sodium chloride in liquid water [2].

To this end, an innovative electrochemical cell architecture has been developed (see Fig. 1), where the redox-reaction forcing lithium intercalation is localized at a protrusion of the device only. Its remainder consists of pristine bilayer graphene unperturbed by an electrolyte. The geometry lends itself to the use of magnetotransport machinery known from mesoscopic low-dimensional physics. Time dependent Hall measurements (as in Fig. 2) across spatially displaced Hall probes deliver a direct view on the in-plane diffusion kinetics. The device layout with a perimeteral electrochemical cell is transferable to other 2D materials as well as thin films and may promote a paradigm shift on the use of electrolytes in on-chip experiments.

References

- [1] C.-C. Chen, L. Fu & J. Maier, *Nature* 536 (2016) 159
- [2] M. Kühne, F. Paolucci, J. Popovic, P. M. Ostrovsky, J. Maier & J. H. Smet, arXiv:1701.02399 [cond-mat.mes-hall]

Figures

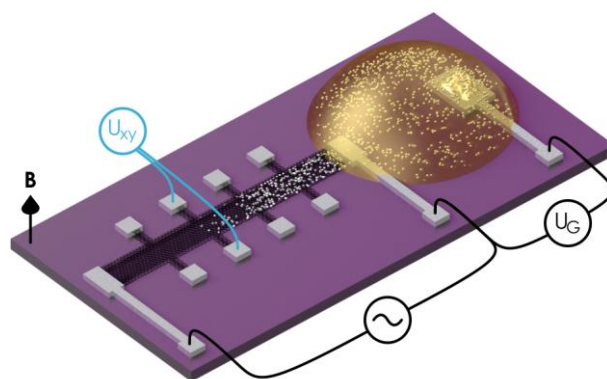


Figure 1: Schematic electrochemical device design with bilayer graphene (black) on a SiO₂-terminated silicon substrate (violet). Electrodes (grey) either enable electronic transport measurements or serve as a counter electrode to control the lithiation (via the gate voltage U_G). The electrolyte (yellow) covers the bilayer only at one end. Li-ions are represented by white spheres.

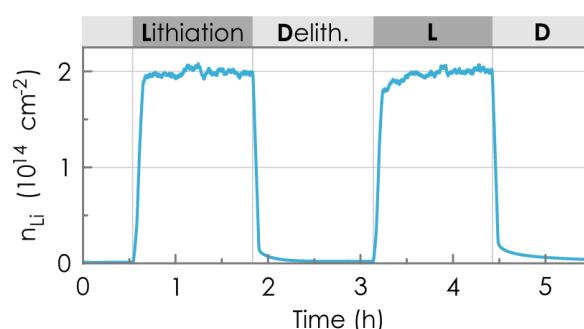


Figure 2: Reversible electrochemical lithiation of a bilayer graphene device. Lithiation (L) and delithiation (D) induces a measurable increase and decrease of the lithium concentration n_{Li} , respectively, as extracted from *in-situ* Hall measurements outside the electrolyte-covered region.