Mechanism of the electrochemical hydrogenation of graphene

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Abstract

The electrochemical hydrogenation of graphene has been recently shown to induce a robust and reversible conductor-insulator transition, which is of strong interest in logic and memory applications. However, its mechanism remains unknown. Here we show that it proceeds as a reduction reaction in which proton adsorption competes with a process attributable to the formation of H2 molecules. Graphene's electrochemical hydrogenation is up to 6 orders of magnitude faster than alternative hydrogenation methods and is fully reversible via the oxidative desorption of protons. We demonstrate that the proton reduction rate in defect-free graphene can be enhanced by an order of magnitude by the introduction of nanoscale corrugations in its lattice and that the substitution of protons for deuterons results both in lower potentials for the hydrogenation process and in a more stable compound. Our results pave the way to investigating the chemisorption of ions in 2D materials at high electric fields, opening a new avenue to control these materials' electronic properties.

References

[1] Tong, J. et al, Nature, 630 (2024) 619-624
[2] Li, S. et al., Nature Electronics, 4 (2021) 254-260
[3] Elias, D. C. et al., Science, 323 (2009) 610-613.

Figures

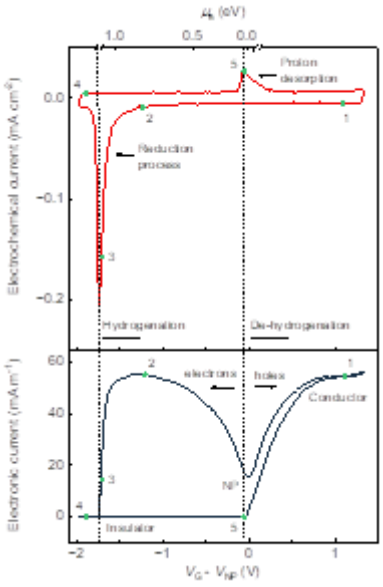

Figure 1: Current density vs VG from a typical device. Numbered green dots mark specific points during the voltage sweep. Dashed vertical lines, position of the conductor-insulation transition (hydrogenation) and its reversal (dehydrogenation). Top x-axis, Fermi energy vs neutrality point (NP).