Modeling Gate-Tunable Ionic Transport Using Atomically Thin Patterned Graphene Membrane

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INTRODUCTION: Atomically-thin two-dimensional (2D) materials emerges as the most promising next-generation membrane technology. Experimentally, large area patterned graphene membrane[1] exhibits salt-rejection behavior upon electrostatic gating even with pore size as large as 20±10 nm. Here we investigate the gate-tunable ionic transport using finite element methods in COMSOL®.

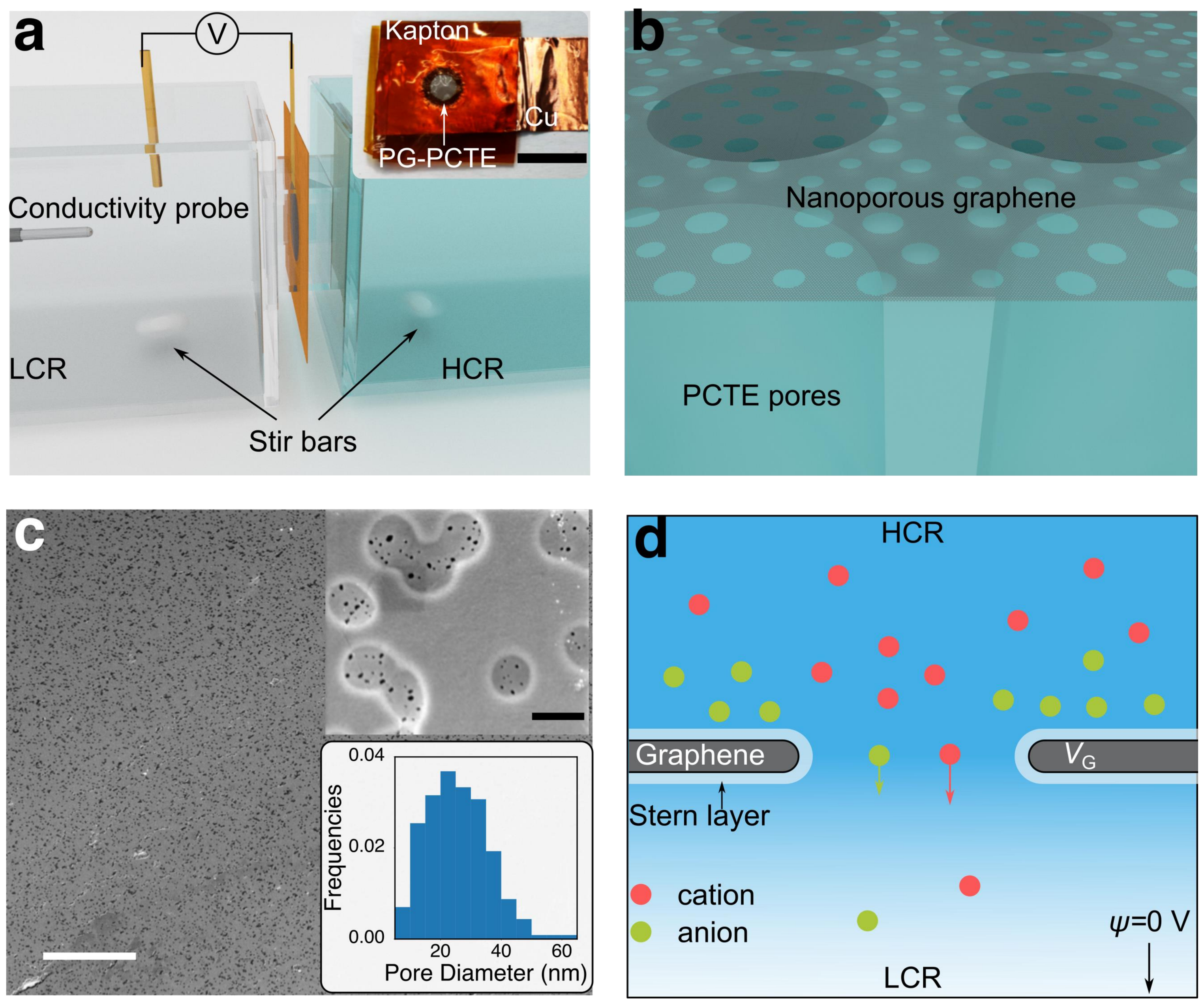

Figure 1. Experimental setup for the gate-tunable ionic transport through graphene nanopores.

COMPUTATIONAL METHODS: The transport of ions is modeled using the Poisson-Nernst-Planck model within the electrochemistry module of COMSOL®. The potential and charge on graphene is explicitly solved by the self-consistent equations for quantum capacitance of 2D Dirac electron gas[2,3]:

$$V_G = \Delta\phi_G + \psi_G \qquad \Delta\phi_G = \mathrm{sign}(\sigma_G)\frac{\hbar v_F}{e}\sqrt{\frac{\pi|\sigma_G|}{e}}$$

$$\sigma_G S_G + \sum_i \int_\Omega z_i c_i N_A e d^3\Omega = 0$$

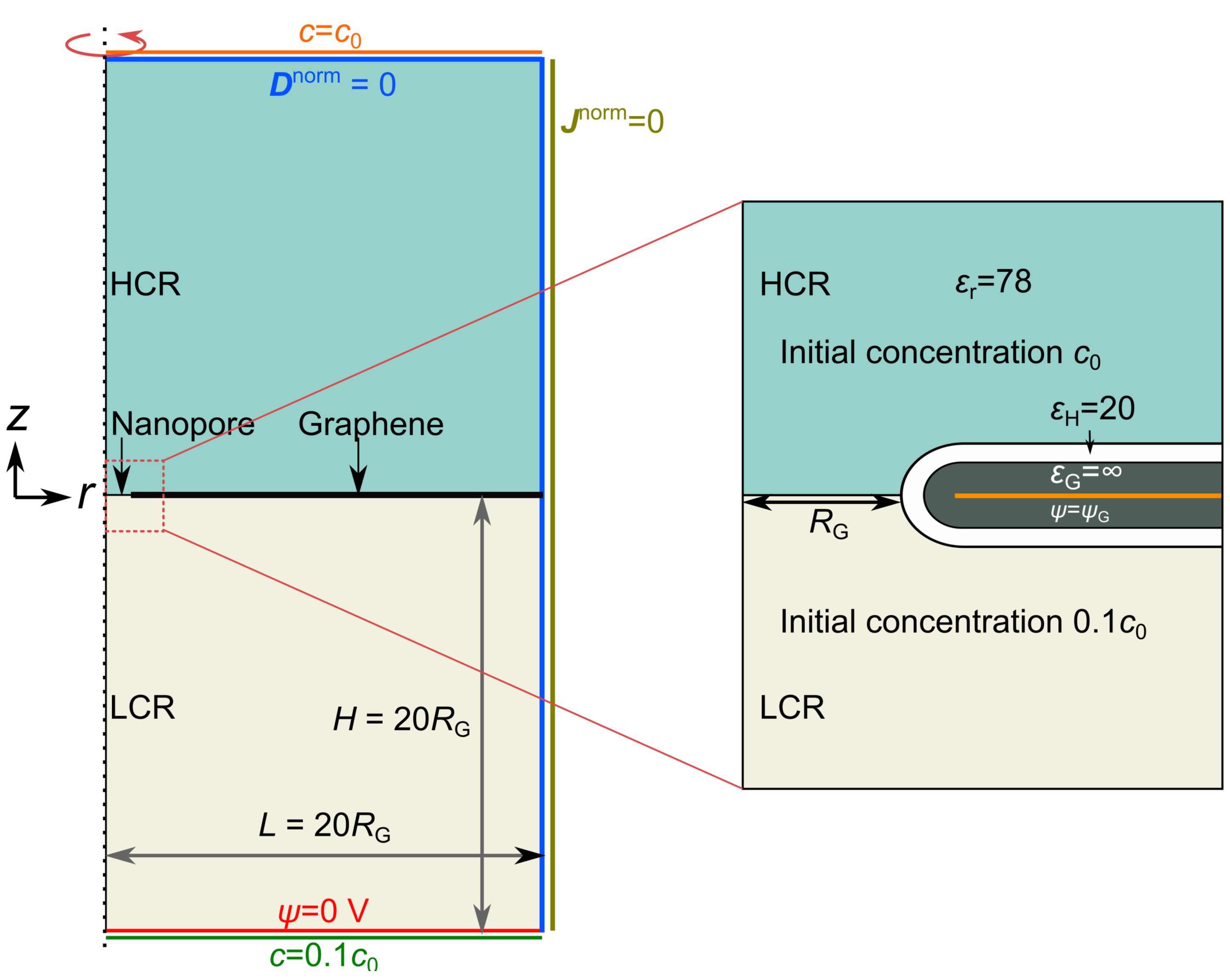

Figure 2. Geometry of the simulation domain with the boundary conditions.

RESULTS: We study the ionic transport and investigate the origin of ion rejection through graphene nanopores upon gating.

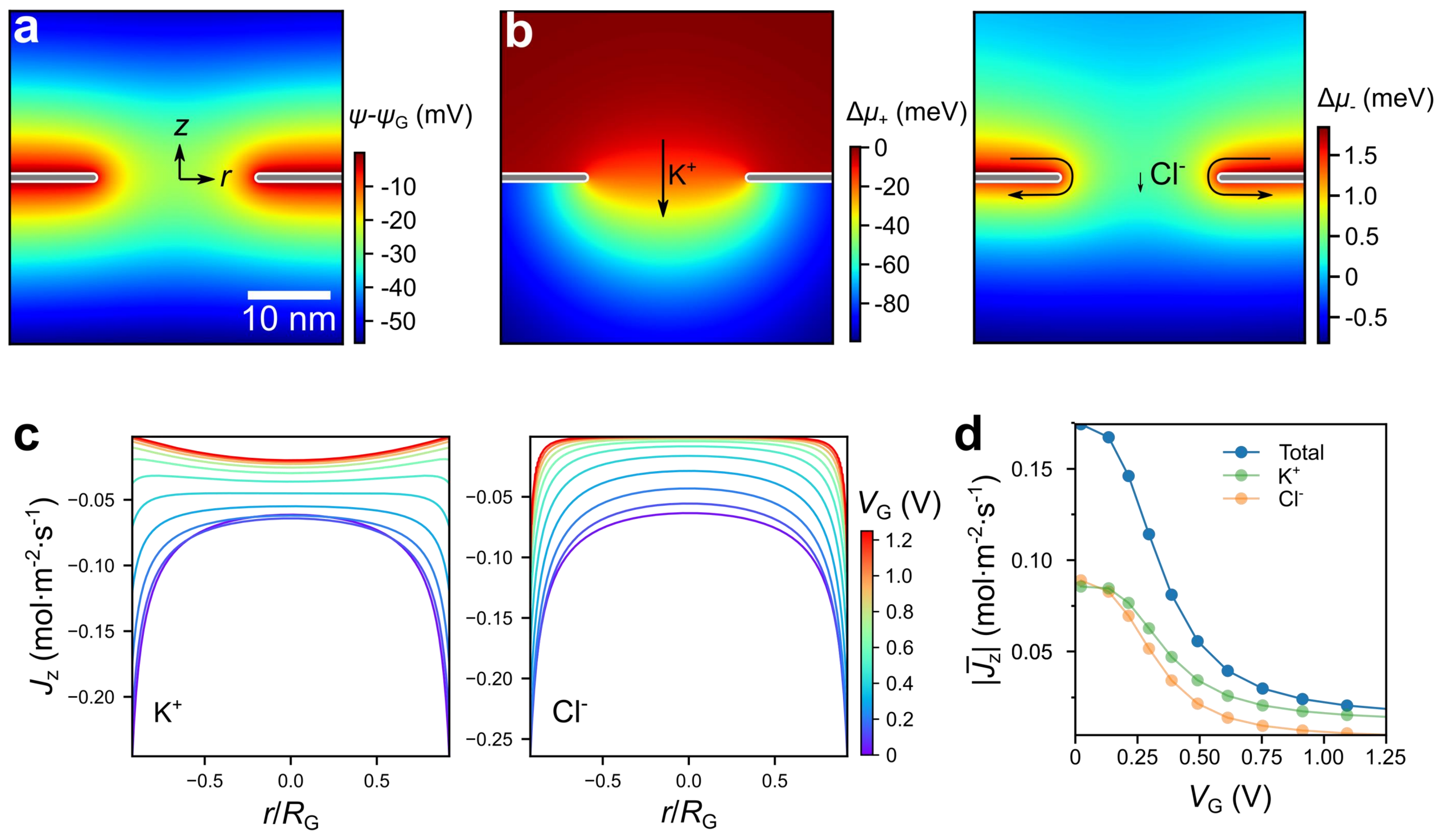

Figure 3. Simulated electrostatic and electrochemical potentials of the ions near a graphene nanopore.

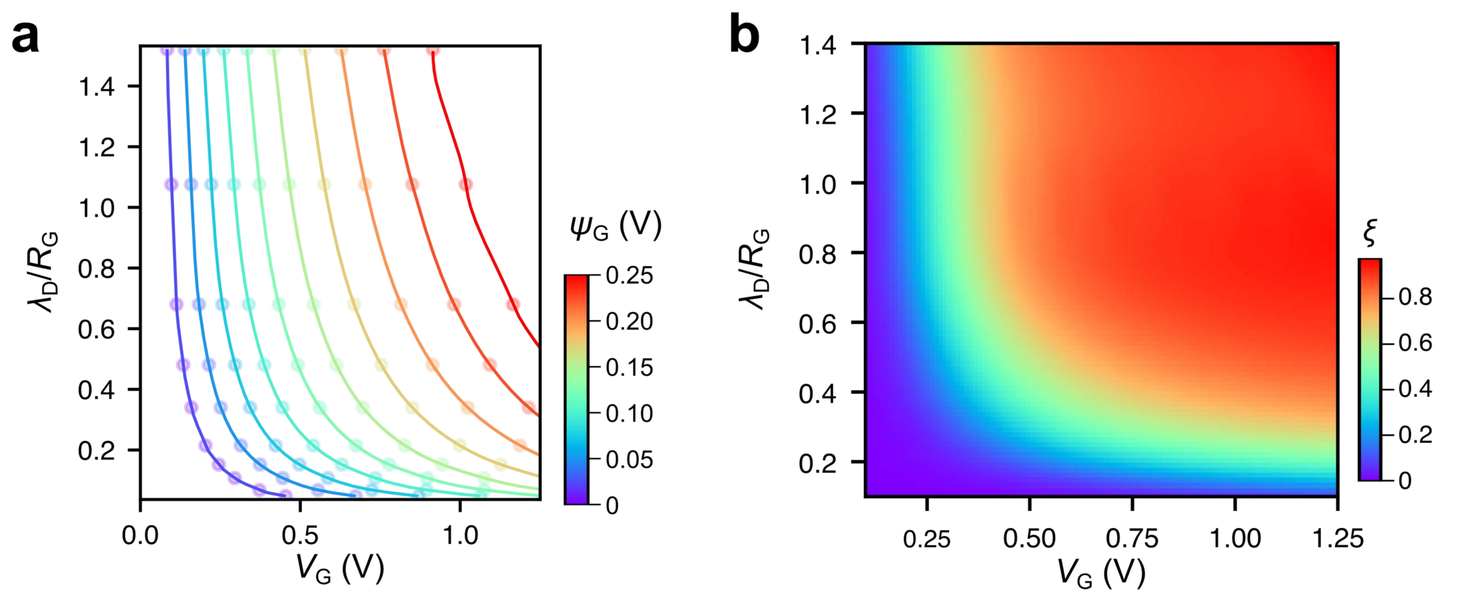

Figure 4. Simulated ionic rectification ξ for ionic transport as a function of Debye length λ_D and gate voltage V_G.

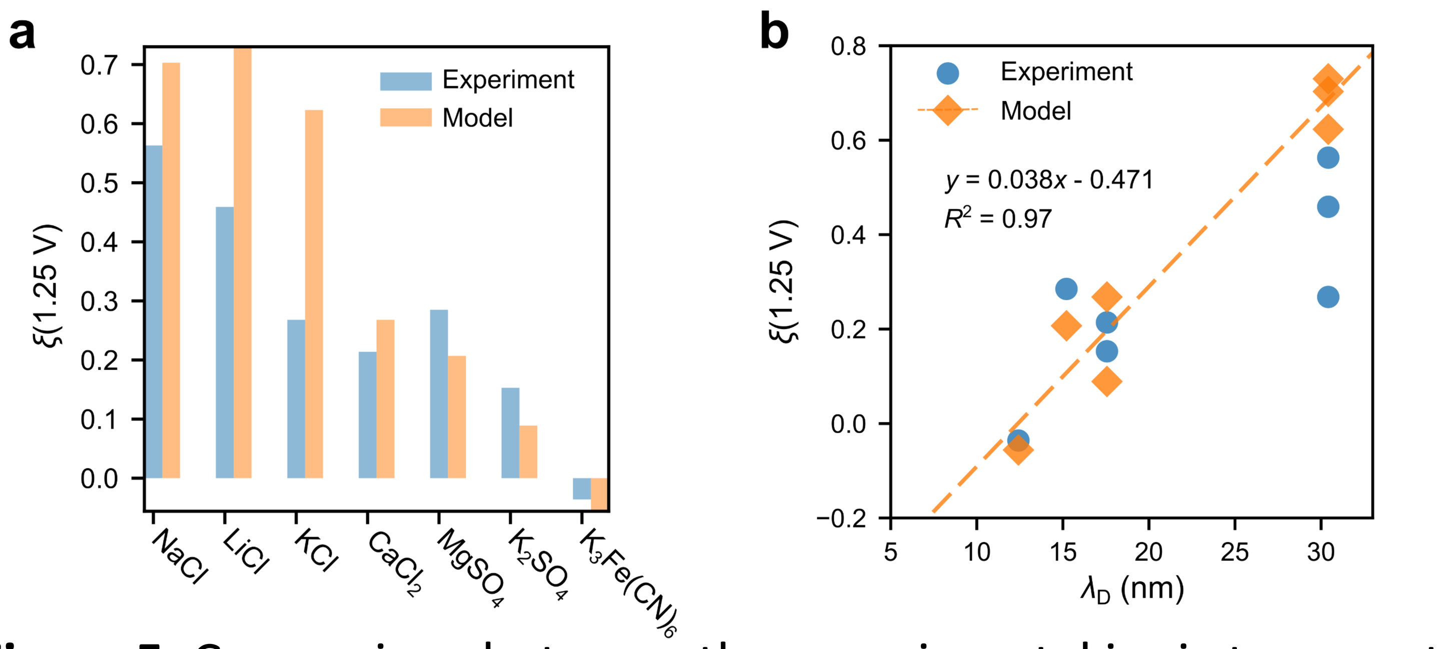

Figure 5. Comparison between the experimental ionic transport and the model.

CONCLUSIONS:

1. Ionic transport through graphene nanopores can be modulated by gating.
2. The performance of ionic diffusion rejection is highly related with both the Debye length and gate voltage.
3. Graphene's quantum capacitance greatly affects the surface potential at the liquid interface and needs to be explicitly modeled for 2D-material-based nanofluidic devices.
4. The FEM analysis can help the design and optimization of 2D-material-based nanoscale transport applications.

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