Multiscale modeling of resistive switching in two-dimensional heterostructures

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Memristive operation is governed by coupled electronic and ionic processes spanning multiple spatial and temporal scales [1]. While Density Functional Theory (DFT) provides a rigorous baseline for understanding quantum transport and defect energetics, its high computational cost limits its utility for high-throughput screening and large-scale dynamic modeling. We propose a multiscale, data-driven framework that integrates DFT, machine-learned interatomic potentials (MLIPs), and kinetic Monte Carlo (kMC) to efficiently model candidate heterostructures. Starting with benchmark systems such as Au–MoS_2–Au, the methodology is systematically extended to unexplored configurations.

The computational workload is partitioned into hierarchical tracks to ensure physical validity while maximizing throughput. Initially, MACE-type MLIPs [2] implemented via LAMMPS [3] execute rapid structural relaxations, significantly reducing the overhead required for subsequent high-fidelity optimization in Siesta [4]. Following relaxation, Non-Equilibrium Green's Function (NEGF) methods (TranSIESTA/TBtrans) are employed to evaluate the electronic properties of static ON/OFF states, yielding zero-bias transmission spectra and local device density of states.

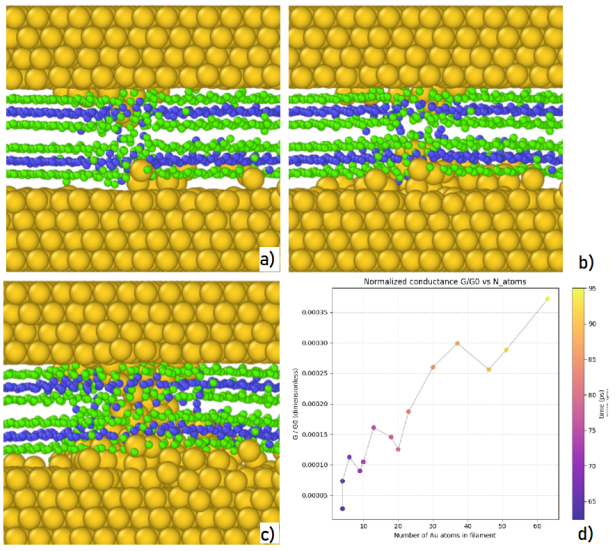

Fig. 1: The SET process at 40 V: a) t = 62.5 ps, b) t = 77.5 ps, c) t = 90 ps. d) Normalized conductance G/G_0 vs number of atoms in filament.

Parallel to the static quantum transport analysis, large-scale dynamic phenomena are modeled using classical and electrochemical molecular dynamics. Building upon established methodologies using ReaxFF [5] to simulate the formation of ionic filaments in Au–MoS_2–Au heterostructures, we employ LAMMPS to model voltage-driven vacancy kinetics and filament growth pathways beyond the size constraints of DFT (~ 5000 atoms in our model). Filament dynamics are simulated via ReaxFF in LAMMPS using a five-stage protocol: thermal equilibration (30–300 K), zero-bias stabilization at 300 K, and a 40 V SET process. Structural persistence is then tested at 0 V (retention), followed by a -20 V RESET phase performed at 2000 K; this elevated temperature serves as a thermally accelerated dynamics approach to resolve diffusion events within MD time-scale constraints.

For systems lacking validated reactive potentials, we plan to test approach using by MLIP-driven molecular dynamics to extract qualitative migration trends.

Finally, to bridge atomistic defect physics with macroscopic device operation, activation barriers and formation energies derived from static DFT and Nudged Elastic Band (NEB) calculations are used to parameterize kMC simulations. This approach resolves the temporal evolution of resistive switching, enabling the prediction of switching latencies, state retention times, and stochastic filament dynamics.

We expect that this multiscale approach provides a computationally efficient method to overcome the limitations of DFT calculations alone and provides sufficient resolution for systematic screening of new memristor materials and detailed studies of the stochastic dynamics of the filaments.

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