

Electrical Transport through a Large-Area Self-Assembled Monolayer of Molecules Coupled with Graphene for Scalable Electronic Applications

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Abstract : While it is challenging to fabricate electronic devices close to atomic dimensions in conventional top-down lithography, molecular electronics is promising to help maintain the exponential increase in component densities via using molecular building blocks to fabricate electronic components from the bottom up. It offers smaller, faster, and more energy-efficient electronic and photonic systems. A self-assembled monolayer (SAM) of molecules is a layer of molecules that self-assembles on a substrate. They are mechanically flexible, optically transparent, low-cost, and easy to fabricate. A large-area multi-layer structure has been designed and investigated by the team, where a SAM of designed molecules is sandwiched between graphene and gold electrodes. Each molecule can act as a quantum dot, with all molecules conducting in parallel. When a source-drain bias is applied, significant current flows only if a molecular orbital (HOMO or LUMO) lies within the source-drain energy window. If electrons tunnel sequentially on and off the molecule, the charge on the molecule is well-defined and the finite charging energy causes Coulomb blockade of transport until the molecular orbital comes within the energy window. This produces 'Coulomb diamonds' in the conductance vs source-drain and gate voltages. For different tunnel barriers at either end of the molecule, it is harder for electrons to tunnel out of the dot than in (or vice versa), resulting in the accumulation of two or more charges and a 'Coulomb staircase' in the current vs voltage. This nanostructure exhibits highly reproducible Coulomb-staircase patterns, together with additional oscillations, which are believed to be attributed to molecular vibrations. Molecules are more isolated than semiconductor dots, and so have a discrete phonon spectrum. When tunnelling into or out of a molecule, one or more vibronic states can be excited in the molecule, providing additional transport channels and resulting in additional peaks in the conductance. For useful molecular electronic devices, achieving the optimum orbital alignment of molecules to the Fermi energy in the leads is essential. To explore it, a drop of ionic liquid is employed on top of the graphene to establish an electric field at the graphene, which screens poorly, gating the molecules underneath. Results for various molecules with different alignments of Fermi energy to HOMO have shown highly reproducible Coulomb-diamond patterns, which agree reasonably with DFT calculations. In summary, this large-area SAM molecular junction is a promising candidate for future electronic circuits. (1) The small size (1-10nm) of the molecules and good flexibility of the SAM lead to the scalable assembly of ultra-high densities of functional molecules, with advantages in cost, efficiency, and power dissipation. (2) The contacting technique using graphene enables mass fabrication. (3) Its well-observed Coulomb blockade behaviour, narrow molecular resonances, and well-resolved vibronic states offer good tuneability for various functionalities, such as switches, thermoelectric generators, and memristors, etc.

Keywords : molecular electronics, Coulomb blockade, electron-phonon coupling, self-assembled monolayer

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