Edge- and quantum confined states in atomically well-defined graphene quantum dots

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Despite the availability of good quality, large scale graphene layers, the realization of both the room-temperature graphene transistor as well as the more advanced theoretical ideas require well-defined samples, in particular in terms of the graphene edge structure. This level of control is currently not available through conventional lithographic techniques and there is a lack of experimental data on atomically well-defined graphene nanostructures. For example, opening a sufficient gap for room-temperature operation through quantum confinement requires structures in the size range of 10 nm. Furthermore, the electronic structure of graphene nanostructures is sensitive to the structure of the edges (e.g. zig-zag vs. armchair) [1]. There is a possibility of edge reconstructions and attachment of various functional groups, which further complicate the comparison between theory and experiment [2].

We use chemical vapour deposition (CVD) from ethylene on Ir(111) surface to grow small graphene flakes with well-defined edge structure [3]. We focus on the possible edge states and size-dependence of the quantum confined wavefunctions in these graphene quantum dots (GQDs) [4,5]. We explore the size-dependent electronic properties of the GQDs on Ir(111) using low-temperature, scanning tunneling microscopy (STM, Figure, top left). Graphene interacts only weakly with the underlying Ir(111) substrate and retains the electronic structure of isolated graphene [6].

The CVD growth yields a relatively broad distribution of different GQD sizes and shapes ranging from a couple of nanometers up to ca. 20 nm with a roughly hexagonal shape. All the flakes have edges in the zig-zag direction with a very small roughness (we see steps with a height of a single atomic row at the flake edges). We can readily access individual GQDs and measure their atomic structure using STM (Figure, top right). The local electronic properties can be probed by scanning tunneling spectroscopy (STS), which allows the measurement on the local density of states (LDOS, proportional to the d*I*/d*V* signal) with atomic spatial resolution. Figure (middle row) shows d*I*/d*V* maps (measured under constant-current STM feedback) at different bias voltages that show the presence of quantum confined states with different envelope wavefunction symmetries. We are able to reproduce the experimental results using tight-binding calculations of free-standing GQDs with the experimentally determined atomic structure (Figure, bottom row).

We have measured the size-dependence of the energy level spacing and the band gap of these GQDs. Our preliminary analysis shows that they both are approximately inversely dependent on the size of the GQD. In addition to these delocalized states, we are able to visualize edge states at energies close to the Dirac point. Our measurements show that CVD-grown graphene quantum dots form an ideal platform for a fundamental study of the electronic properties of atomically well-defined graphene nanostructures.

References

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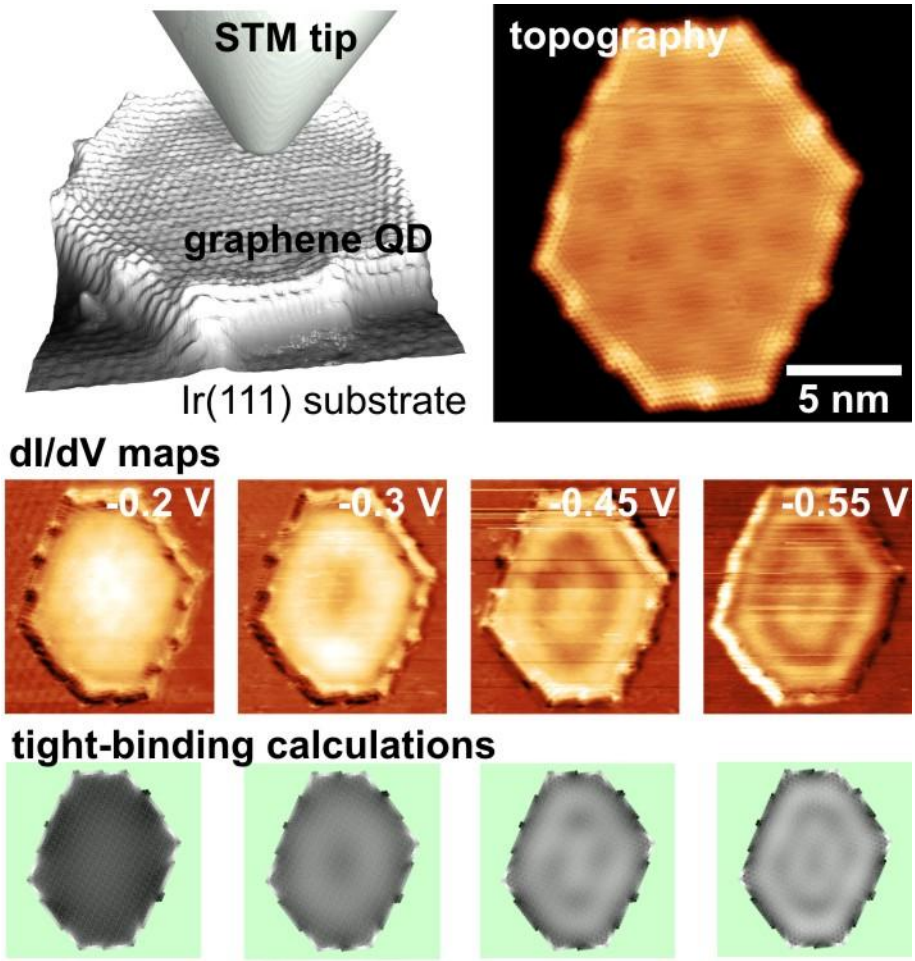
Figures



Top left: Schematic of the experimental setup. Graphene quantum dots with well-defined edges are grown epitaxially on Ir(111) and investigated by low-temperature STM. Top right: Typical atomically resolved STM image acquired at 0.5V / 100 pA. Middle row: dI/dV maps (proportional to local density of states) acquired under STM feedback at the bias voltages indicated in the figure. Bottom row: Local density of states at the energies corresponding to the experimental dI/dV images based on tight-binding calculations on a GQD with the experimentally determined atomic structure.