Signatures of Spin Transitions in an Endofullerene-Based Single-Molecule Transistor

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Abstract

A single-molecule transistor (SMT) is a three-terminal device which has as its active element a single organic molecule [1]. SMTs allow us to study the quantum-mechanical transport of electrons though single molecules as small as 1 nm. Our previous research on single-molecule transistors has involved work on the coupling of molecular vibrations to electron transport [2], as well as studies of Mn$_{12}$ (acetate) molecules having anisotropic magnetism [3]. In the experiment presented here, we examine SMTs based on a high-spin endofullerene molecule. We then manipulate the electron transport through these devices using an externally-applied magnetic field.

Summary of Research:

We have made single-molecule transistors based on the high-spin endofullerene, N@C$_{60}$, which consists of a magnetic nitrogen atom inside the cage of a C$_{60}$ molecule. We observe two uniquely-magnetic signatures in the electron tunneling spectra of the molecule. First, there is a change in slope of the ground-state conductance peaks when plotted against bias voltage and magnetic field (see Figure 1), which indicates a low-spin-to-high-spin transition in the ground state of the molecule as a function of applied magnetic field. This change in slope is not observed in the tunneling spectra of simple C$_{60}$ molecules (see Figure 2). Second, some excited-state conductance peaks terminate in other excited-state peaks (see Figure 3), as opposed to terminating in ground-state peaks as is observed in non-magnetic spectra.

Figure 1: Conductance vs. V and B in a N@C$_{60}$ single-molecule transistor.

Figure 2: Conductance vs. V and B in a C$_{60}$ single-molecule transistor.
This behavior is a signature of transitions originating from non-equilibrium spin states of the molecule. While similar changes in spin state are commonly observed in larger quantum-dot devices, the electronic energy-level spacing in small molecules is normally so large as to preclude spin changes at experimentally accessible fields. The spin changes observed in N@C$_{60}$ are made possible by the combination of the high symmetry of the molecule and the exchange interaction between the electrons on the nitrogen and those on the C$_{60}$ cage.

We prepared the single-molecule transistors following techniques developed previously in the CNF [2, 3]. We began by fabricating an Al gate electrode 16 nm thick and 2 μm wide and then exposing the Al to air to form a thin insulating oxide. On top of the gate electrode, we fabricated continuous Pt wires with widths of approximately 150 nm and thicknesses of 10 nm. The chips were cleaned with an oxygen plasma and immediately covered with 25 μL of either a 0.1 mM solution of N@C$_{60}$ for 2.5 minutes or a 0.5 mM solution of C$_{60}$ in toluene for 1 minute. Then the excess solution was blown off the chip with nitrogen, and the deposition process was repeated. This technique produced a convenient yield of single-molecule devices in the control C$_{60}$ samples. After the molecules were deposited, we cooled to cryogenic temperatures and broke the wires using electromigration, forming nanometer-scale gaps in which a molecule was sometimes trapped (see Figure 4).

The success rate for observing Coulomb blockade transport characteristics due to the presence of a molecule was 9/19 for N@C$_{60}$ devices and 17/59 for C$_{60}$ devices, while 0/39 control devices prepared using pure toluene instead of a fullerene solution showed Coulomb blockade.

References