The Kondo Effect in Mechanical Break Junctions

CNF Project # 598-96
Principal Investigator: Daniel C. Ralph

Abstract:
Our group recently demonstrated the fabrication of single-molecule transistors that combine the ability to mechanically adjust the spacing between two electrodes and to shift the energy levels of a molecule with a gate electrode [1]. Here we use this device geometry to study the Kondo effect, a many-body phenomenon that arises from the coupling between a localized spin and a sea of conduction electrons. By mechanically adjusting the electrode spacing, we are able to tune the Kondo effect.

Summary:
Our devices are fabricated on thin (200 µm) silicon wafers, which are flexible and allow for mechanical bending. We begin with a thermal growth of silicon oxide, followed by a wet etch to form trenches, and a re-growth of thin oxide in the trenches over which gold bridges will later be suspended. Using a series of photolithography and electron-beam lithography steps, we define the bonding pads as well as the critical features, namely 50 nm-wide gold wires. These wires are then suspended 40 nm above the silicon substrate using a combination of dry and wet etches. To incorporate molecules in our devices, 50 microliters of a dilute solution of C₆₀ or [Co(Tpy-SH)₂]²⁺ are deposited onto a chip. These molecules are of interest because the Kondo effect has been previously observed with them [2,3]. The chip is subsequently cooled down to 1.5 K in a cryostat, and a nanoscale gap is created in the gold wires by electromigration [4]. After electromigration, we find that one or a few molecules can sometimes be found bridging this gap. Our cryostat is equipped with a motorized mechanism that enables controlled bending of the silicon substrate, which in turn allows us to adjust the inter-electrode gap with very fine precision. We measure transport characteristics of our devices as a function of bias voltage, magnetic field, electrode spacing, and temperature.

Measurements on individual C₆₀ and [Co(Tpy-SH)₂]²⁺ molecules in our device geometry show signatures of the Kondo effect in a quantum dot, namely a zero-bias peak in dI/dV which splits in a magnetic field and is suppressed as a function of increasing temperature. This zero-bias peak reflects the formation of a spin-singlet state between a localized unpaired electron on the molecule and the conduction electrons of the leads at the Fermi energy. We are able to tune both the height and width of the Kondo resonance in our devices as a function of electrode spacing. The former gives us information about the asymmetry in coupling of the molecule to each electrode, and the latter tells us about the Kondo temperature, which is the many-body energy scale characteristic of the binding energy of the spin-singlet state. While we have been able to tune the Kondo effect and modulate current flow through single molecule devices by varying electrode spacing, we have not yet identified the dominant mechanism for the observed changes. Varying the electrode spacing could both alter the molecule-lead coupling as well as induce a gating effect, either of which could modify the Kondo effect.

References:
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Figure 1, top left: Scanning electron micrograph of a gold bridge suspended 40 nm above a silicon substrate.

Figure 2, bottom left: Differential conductance traces for a C_{60} molecule in a gold break junction at various temperatures, after subtraction of a small background. Inset: A fit to theory yields a Kondo temperature of 23 K.

Figure 3, above: Differential conductance traces for the same device at several electrode spacings. Inset: Conductance map showing the evolution of the Kondo peak as the electrodes are pulled apart.