Abstract:
We are studying the effect of light on single molecule transistors. These three-terminal devices are fabricated using standard nanolithography techniques followed by electromigration. We have two experimental set-ups for illuminating the devices with UV/VIS/NIR radiation at cryogenic temperatures. Light-induced changes in conductance can be studied as a function of the gate and bias voltages.

Summary:
We are extending the study of single molecule transistors [1, 2] to include light-induced effects. Optically active molecules may be electronically excited, undergo a structural change, make and break chemical bonds, and/or exhibit photoconductivity. Such effects could show up as a change in conductance of the device, which can be studied as a function of bias voltage, gate voltage, and illumination wavelength. Because the nature of the response should be molecule-specific, light studies should also provide clear confirmation that the device contains the molecule of interest, rather than other species which may produce artifacts.

To make a single molecule transistor, a molecule must bridge a gap of comparable dimension between two conducting electrodes. Gaps of this size cannot be made with conventional lithography techniques, so we fabricate a continuous wire, then create the gap using electromigration [3]. The fabrication is done on an insulating SiO₂ layer on Si. Thin Al gate electrodes are deposited where the devices will be, then a native oxide is allowed to form. Au or Pt wires (150 nm wide, 500 nm long, and 10 nm thick) are fabricated on top of the gate electrodes with e-beam lithography and liftoff. These wires are contacted by larger Au leads.

A chip with 30 devices is cleaned in an oxygen plasma, then molecules are deposited. The sample is loaded into the measurement apparatus which is typically cooled to cryogenic temperatures to improve energy resolution. The wires are broken in situ by ramping the voltage bias until the wire fails, leaving a gap of order 1 nm between the resulting electrodes. In some of the devices, a molecule will bridge this gap. The presence of a molecule can be detected by Coulomb blockade, in which current turns on above a gate dependent threshold bias voltage.

We have two set-ups for measuring single molecule transistors under illumination. In the first, the devices are mounted at the bottom of a cryostat that is placed in a liquid helium storage dewar. An optical fiber is positioned to illuminate the devices with an external light source. The second set-up, currently being built, provides much higher photon intensities. The sample is placed close to a transparent window in a small cryostat which sits under a microscope. Laser light is focused onto the device of interest with a microscope objective. Light emitted from a device could also be detected in this set-up.

Thus far we have not seen reproducible changes in conductance of these devices with light. We have studied C₆₀ and an organic complex with a Ru atom at the center. However, it is likely that the illumination intensities were not high enough, and several improvements are being made to overcome this issue, including the second set-up mentioned above. We also plan to optimize the electrode geometry to maximize the field enhancement at the gap between the antenna-like electrodes.

References:
Purpose:
Study conductance changes of single molecule transistors when exposed to light.

Figure 2, below left:
Sample and optical fiber at base of cryostat.

Figure 3, below right:
Coulomb blockage in a [Ru(tpy-py)$_2$]$^{2+}$ single molecule transistor (T = 6 K). I-V curves as a function of gate voltage from -455 mV to -415 mV in steps of 5 mV. We have not yet achieved reproducible changes with illumination.

Figure 1, above:
SEM images of (a) an unbroken wire and (b) a wire broken by electromigration. In some devices a molecule deposited on the surface bridges the gap, creating a single molecule transistor.