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Simultaneous electronic transport and Raman spectroscopy in single-molecule devices[1]

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Over the last decade several techniques have been developed to examine electronic transport through individual small molecules. These include scanned probe methods, mechanical break junctions, and electromigrated junctions. One recurring challenge is the need to confirm that current flow is, indeed, through the molecule of interest rather than a contaminant. We recently discovered (D. R. Ward *et al.*, Nano Lett. **7**, 1396 (2007)) that the same electromigrated Au source and drain electrodes used for transport are tremendously effective optical antennas in the near infrared. Surface plasmon modes localized to the nm-scale interelectrode gap lead to large enhancements of the local electric field relative to that from incident radiation. The result is that these nanoscale gaps are tremendous "hot spots" for surface-enhanced Raman scattering (SERS). We perform simultaneous measurements of electronic transport and SERS in junctions incorporating molecules of interest, and find in 10-15% of devices that the conductance and SERS emission are strongly correlated in time (D. R. Ward *et al.*, Nano Lett. **8**, 919 (2008)). Since the conductance mechanism is tunneling and therefore dominated by a volume comparable to that of a single molecule, this strongly implies that the SERS emission comes from that same molecule. The distinctive SERS spectra allow us to confirm that conduction in these devices is through the molecule of interest. Furthermore, these devices open up many opportunities, including studies of electron-vibrational couplings and dissipation at the single-molecule level.

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