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Surface-enhanced Raman detection of a vibrational Stark effect in C60-containing molecular junctions YAJING LI, Department of Physics and Astronomy, MS 61, Rice University, PETER DOAK, JEFFREY NEATON, Molecular Foundry, Lawrence Berkeley National Laboratory, LEEOR KRONIC, Weizmann Institute of Science, Rehovot, Israel, DOUGLAS NATELSON, Department of Physics and Astronomy, MS 61, Rice University — Understanding the interplay of local electric fields and molecular vibrational degrees of freedom is of considerable interest. One nontrivial consequence of this coupling is the vibrational Stark effect, in which vibrational energies are altered through coupling to externally applied electric fields. We investigate this physics through nanoscale Au bowtie structures functioning as surface enhanced Raman(SERS) substrates. Following electromigration, these metal nanostructures possess nanometer-scale interelectrode gaps that support highly localized surface plasmon resonances, resulting in SERS electromagnetic enhancements sufficient for single-molecule studies. These structures have also proven suitable for simultaneous single-molecule electronic transport experiments, in which we observed the vibrational modes of the molecules shift systematically as a function of applied bias. We will present measurements of the electrically driven vibrational energy shifts of C60 in such junctions and compare those with theoretical expectations obtained from DFT calculations.

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