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**Evolution of Single-Molecule Vibrational Modes from Tunneling** to Quantum Point Contact<sup>1</sup> W. MAR, W. KO, C. R. MOON, B. K. FOSTER, L. S. MATTOS, H. C. MANOHARAN, Stanford University — A detailed understanding of how molecular junctions form and evolve is vital for emerging fields such as molecular electronics. We present high-precision scanning tunneling microscopy studies tracing the evolution of molecular junctions from the tunneling regime to quantum point contact. We employ a model system of CO molecules on Cu(111)and are able to extend inelastic spectroscopy into the point contact regime, thus following the energy shifts of specific vibrational modes as the molecular contact is formed. We observe surprising non-monotonic shifts, confirmed by simultaneous noise measurements traceable to molecular motion. In point contact, we also observe a novel "nucleonic gating" effect in which the carbon nucleus controls a measurable dc molecular conductance shift. This shows that the electrical properties of molecular wires can be profoundly altered by their isotopic makeup. We extend these measurements to geometries where the three-dimensional approach vector of the tip relative to the target molecule is finely controlled, a technique not possible in break junction measurements.

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Warren Mar Stanford University

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