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Isotope-Resolved and Charge-Sensitive Force Imaging Using Scanned Single Molecules YAN SUN, DOMINIK RASTAWICKI, YANG LIU, WARREN MAR, HARI MANOHARAN, Stanford University, ANNA MIGLIO, SORIN MELINTE, JEAN-CHRISTOPHE CHARLIER, GIAN-MARCO RIGNANESE, Université catholique de Louvain, LIANHUA HE, FANG LIU, AIHUI ZHOU, Chinese Academy of Sciences — Originally conceived as surface imaging instruments, the scanning tunnelling microscope (STM) and the atomic force microscope (AFM) were recently used to probe molecular chemical bonds with exquisite sensitivity. Remarkably, molecule-functionalized scanning tips can also provide direct access to the inelastic electron tunneling spectrum (IETS) of the terminal molecule. Here we report atomic manipulation experiments addressing carbon monoxide (CO) isotopes at low temperatures. The unique and quantifiable dependence of the CO vibrational modes offers insight into tip-controlled force and charge sensing of surface adsorbates, subsurface defects, and quantum nanostructures. The specific behavior of the monitored vibrational modes originates from the interplay of interaction forces between the top electrode—a scanned tip functionalized with a single molecule—and the atomic scale force field surrounding the target atomically-assembled nanostructure. We also present density functional theory (DFT) computations that have been performed in order to scrutinize and visualize the vibrational spectroscopic fingerprints and local force fields.

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