Preparation and coherent manipulation of pure quantum states of a single molecular ion\textsuperscript{1} CHRISTOPH KURZ, CHIN-WEN CHOU, DAVID B. HUME, Time and Frequency Division, National Institute of Standards and Technology, Boulder, Colorado 80305, USA, PHILIPP N. PLESSOW, Institute of Catalysis Research and Technology, Karlsruhe Institute of Technology, Karlsruhe, Germany, DAVID R. LEIBRANDT, DIETRICH LEIBFRIED, Time and Frequency Division, National Institute of Standards and Technology, Boulder, Colorado 80305, USA — We demonstrate control of individual molecules based on quantum-logic spectroscopy [1, 2]. In our experiment, we drive the motional sidebands of Raman transitions in a molecular ion and probe the secular motion with a co-trapped atomic ion. Detection of motional excitation projects the molecule into a pure internal state. The state of the molecule can subsequently be coherently manipulated, as demonstrated by Rabi oscillations between magnetic sublevels of rotational states. We need only one far off-resonant continuous-wave laser to manipulate the molecule. This makes our approach applicable to coherent control and precision measurement of a vast class of molecular ions.

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