QUANTUM LOGIC CONTROL AND SPECTROSCOPY OF A SINGLE MOLECULAR ION

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Pure quantum states of charged molecules can now be prepared, coherently manipulated, and efficiently detected in high-resolution spectroscopy experiments by employing quantum-logic spectroscopy, an action spectroscopy at the single quantum level. The coupled harmonic motion of co-trapped ions can be utilized to transfer action information from a ”spectroscopy” molecular ion to an easily manipulated and detected atomic “logic” ion. With this technique and precise characterization of the systematic effects of the trap RF electric field on molecular levels, we demonstrate hertz-level resolution in terahertz-scale rotational spectroscopy of a single CaH^+ molecule using a Ca^+ logic ion. Precision measurements of molecular transitions can be used to determine molecular properties, and here we experimentally determine the CaH^+ dipole moment for the first time. In addition to detecting departure from the initial state, we can also detect arrival into the final state, facilitating confirmation of transition assignment in cases where the spectrum is congested or a priori information is limited. Because all of our laser operations on the molecule are driving stimulated Raman transitions with the lasers far detuned from molecular transitions, the techniques should be generalizable to a wide variety of charged molecules.

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