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Quantum Logic Control of a Single Molecular Ion

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An amazing level of quantum control is routinely reached in modern experiments with atoms, but similar control over molecules has been an elusive goal. A method based on quantum logic spectroscopy [1] can address this challenge for a wide class of molecular ions [2,3]. We have now realized many basic aspects of this proposal.

In our demonstrations, we trap a calcium ion together with a calcium hydride ion (CaH+) that is a convenient stand-in for more general molecular ions. We laser-cool the two-ion crystal to its motional ground state and then drive Raman, mm-wave or vibrational overtone transitions in the molecular ion. Laser-based transitions in the molecule can deposit a single quantum of excitation
in the motion of the ion pair when a motional "sideband" is driven. We can efficiently detect this single quantum of excitation with the calcium ion, which projects the molecule into the final state of the sideband transition, a known, pure quantum state.

The molecule can be coherently manipulated after preparation by a first projection, and after attempting a transition, the resulting molecular state can be read out by another quantum logic state detection. We demonstrate this by driving Rabi oscillations between different rotational states [4, 5, 6] and by entangling the molecular ion with the logic ion [7]. Transitions in the
molecule are either driven by a single, far off-resonant continuous-wave laser, by a far-off resonant frequency comb or a frequency comb resonant with a certain vibrational overtone transition. This makes the approach suitable for quantum control and precision measurement of a large class of molecular ions. Controlled transitions to excited vibrational levels open avenues to precise characterization of the electronic ground state potential surface and to coherent dissociation along a specific bond.

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