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### **Quantum-Logic Control and High-Resolution Spectroscopy of a Single Molecular Ion**

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We demonstrate coherent quantum state manipulation and precision spectroscopy of a molecular ion, based on quantum-logic spectroscopy [1-5]. Information regarding the states of a  $\text{CaH}^+$  ion is transferred to a co-trapped  $\text{Ca}^+$  ion using the coupled harmonic motion as an information bus and read out via state-dependent fluorescence detection without disturbing the molecular state. We can thus initialize the molecular ion in a pure quantum state in a probabilistic but heralded fashion [2, 3, 5]. The THz rotational transitions between states with different principal rotational quantum number  $J$  are directly probed with a frequency comb [2, 3] with sub-500 Hz spectroscopic linewidths, and improvement to the sub-Hz level seems feasible [6]. Coherent Rabi flopping is observed between different rotational  $J$ -manifolds. The initial and final states of the transitions, separated by  $J = 2$ , can both be nondestructively detected [2-5], which facilitates unambiguous assignment of the observed signal to the corresponding rotational transitions. We have also started exploring entanglement of a molecular ion with an atomic ion, with possible applications in quantum information science. We implement quantum logic operations to produce an entangled state where states of  $\text{CaH}^+$ , either in the same or different rotational manifolds, are entangled with magnetic sublevels of the  $S\ 1/2$  and  $D\ 5/2$  states of  $\text{Ca}^+$ . All of our methods can be extended to investigate and exploit coherent rotational-vibrational transitions of a large class of diatomic and polyatomic molecules in the optical and infrared domains. In collaboration with Y. Lin, A. Collopy, C. Kurz, T. Fortier, S. Diddams, D. Leibfried, and D. Leibbrandt. [1] P. O. Schmidt et al., *Science* 309, 749 (2005). [2] D. Leibfried, *New J. Phys.* 14, 023029 (2012). [3] S. Ding and D. N. Matsukevich, *New J. Phys.* 14, 023028 (2012). [4] F. Wolf et al., *Nature* 530, 457 (2016). [5] C. W. Chou et al., *Nature* 545, 203 (2017). [6] A. Bartels et al., *Opt. Lett.* 29, 1081 (2004).