HIGH-RESOLUTION ROTATIONAL SPECTROSCOPY AND COHERENT CONTROL OF CaH^+

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We demonstrate methods for precision spectroscopy and coherent quantum state manipulation of a molecular ion, based on quantum-logic spectroscopy [1-3]. In thermal equilibrium with room temperature blackbody radiation, the electronic and vibrational degrees of freedom of the proof-of-principle CaH^+ molecule are in their ground states. We laser cool the coupled translational motion of a co-trapped Ca^+ atom and the molecule to near its ground state [4]. Subsequently, we coherently drive rotational Raman transitions using two beams derived from a single frequency laser that is far off-resonance from any molecular transition. Information regarding the molecular states is transferred to the atomic ion using the coupled harmonic motion as an information bus [1-3] and read out via state-dependent fluorescence detection without disturbing the molecular state. In this way, we initialize the molecular ion in a pure quantum state in a probabilistic but heralded fashion [3]. Following preparation, we can drive further rotational transitions up to the THz range with two beams derived from a single, far-off-resonant frequency comb [5, 6]. The final states of the transitions are detected, enabling unambiguous assignment of the observed signals to the corresponding transitions. For CaH^+, we have measured the frequency of THz rotational transitions with sub-kHz resolution, and improvement to the sub-Hz level seems feasible [7]. This protocol can be extended to investigate coherent rotational-vibrational transitions of a large class of diatomic and polyatomic molecules in the optical and infrared domains. [1] P. O. Schmidt et al., Science 309, 749 (2005). [2] F. Wolf et al., Nature 530, 457 (2016). [3] C. W. Chou et al., Nature 545, 203 (2017). [4] M. D. Barrett et al., Phys. Rev. A 68, 042302 (2003). [5] D. Leibfried, New J. Phys. 14, 023029 (2012). [6] S. Ding and D. N. Matsukevich, New J. Phys. 14, 023028 (2012). [7] A. Bartels et al., Opt. Lett. 29, 1081 (2004).