Infrared absorption spectroscopy of a single trapped polyatomic molecular ion

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Quantum logic spectroscopy (QLS) has been established for high-precision spectroscopy of diatomic molecular ions co-trapped with atomic ions, contributing to fundamental physics and quantum information applications [1–4]. However, extending this protocol to larger polyatomic species is challenging because of their more complex internal structure. Furthermore, QLS is not directly applicable to studies using broadband ultrashort laser pulses to probe fast molecular processes such as intramolecular vibrational redistribution, which is important in chemical and biological processes. Using an adapted QLS method, we investigate polyatomic molecular ions by probing their photon-absorption signal. This is achieved by preparing the shared motional mode of the trapped molecular and atomic ions in a non-classical state sensitive to momentum recoil from photon absorption [5]. We demonstrate this scheme by measuring the infrared absorption spectrum of the O–H stretch mode of a $CaOH^+$ ion co-trapped with a Ca^+ ion using femtosecond laser pulses, in good agreement with ab initio theoretical predictions [6]. This suggests that recoil detection has potential for studying more complex polyatomic molecular ions at the single-particle level, including pump-probe experiments of ultrafast intramolecular dynamics [7]. Furthermore, improved signal-to-noise ratio could enable single-shot readout, allowing measurement-based rotational state preparation in polyatomic molecules.

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