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Observation of vibrational overtones by single molecule resonant photodissociation GANG SHU, Georgia Inst of Tech, NCAMISO KHANYILE, Georgia Inst of Tech & University of Stellenbosch/Center for Scientific and Industrial Research, KENNETH BROWN, Georgia Inst of Tech — Molecular ions sympathetically cooled by a chain of laser-cooled atomic ions are ideal for performing high-precision molecular spectroscopy with applications in astrochemistry and fundamental physics. The same system can be coupled with a broadband laser to perform survey spectroscopy and discover new molecular transitions. Here we present our results using three-ion chains of Ca⁺ and CaH⁺ to observe vibrational transitions via resonance enhanced multiphoton dissociation detected by Ca⁺ fluorescence [1]. Based on theoretical calculations, the observed peaks are assigned to two vibrational overtones corresponding to transitions from the ground vibrational state, ν = 0, to the excited vibrational states, ν = 9 and ν =10. Our method allows us to track single molecular events, and it can be extended to handle any molecule by monitoring normal mode frequency shifts to detect the dissociation. [1] N. B. Khanyile, G. Shu, and K. R. Brown, Nat. Comm. 6, 7825 (2015)

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