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Arbitrary internal state control of molecular ions through optical pumping[1] PATRICK STOLLENWERK, IVAN ANTONOV, SRUTHI VENKATARAMANABABU, BRIAN ODOM, Northwestern University — Despite the significant challenges associated with controlling the internal modes of molecules, a number of successful techniques for efficiently populating the rovibrational ground state have been demonstrated. Fewer techniques have been implemented for arbitrary state control and none have been used to sustain high energy rotations. Robust, sustainable, arbitrary state preparation is especially useful for trapped ions as they are capable of long trapping lifetimes. We demonstrate a technique relying on spectral pulse shaping of a single broadband laser for populating arbitrary target rotational states of SiO^+ up to rotational quanta exceeding N=65 including ground state preparation with sub-Kelvin internal temperatures. Additionally, the technique is extended to populating the first excited vibrational mode. In contrast to coherent manipulation or selective filtering methods, optical pumping represents a driven, dissipative method of control and therefore, neglecting chemical reactions, population in the target state can be sustained indefinitely. We also discuss some of the potential applications of control as well as our progress on direct fluorescent imaging of SiO^+.

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