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Fast single-laser optical cooling of molecular rotations[1]

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Although ion-ion sympathetic cooling is now commonly used to reduce speeds of trapped molecular ions to milliKelvin temperatures, cooling of the decoupled internal molecular modes remains a challenge. Here, we demonstrate that a single spectrally filtered femtosecond laser, tuned to the electronic A-X transition of trapped AlH^+, can efficiently cool rotations from room temperature to the ground state. Since this single-laser technique simultaneously drives rotationally de-exciting electronic excitations from all initially populated states, each parity can be cooled as quickly as 100 μs. In our current implementation, full population collection into the $N = 0$ positive-parity level relies on vibrational relaxation on the timescale of 100 ms. Since this technique cools by step-wise rotational pumping, it is most readily applicable to "alkali-like" molecular species, such as AlH^+, with semi-closed cycling transitions. Although we currently use a destructive two-photon state readout technique, these species are also promising candidates for single-molecule fluorescence detection. The ability to quickly cool molecular rotations is expected to be important for precision spectroscopy experiments, quantum information processing applications, and for ultracold chemistry studies.

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