## **Optimizing the Multicycle Rotational Cooling of Diatomic Molecules**

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Cooling the rotational motion of molecules beyond the milikelvin regime is a task that is under extensive research during the last decade. This is usually achieved by controlled processes which involve the use of external electric fields. However, these coherent fields can induce only coherent, purity-conserved transformations, and cannot alter the system's temperature directly. The complementary component to induce cooling is spontaneous emission. This part of the process can create a change in the purity but is uncontrollable. To merge to two components into a single cooling device we employ a field optimized unitary transformation that will stir the system to a situation that will lead to spontaneous cooling. The optimization process is composed by two ingredients: (I) The ergodic theorem is combined with a classical optimization to find a unitary transformation that will lead that system into a pure stead-state after multiple excitation-emission cycles. (II) A quantum-optimal-control theory based algorithm is carried to find the correct field that will produce the desired unitary transformation defined by the former algorithm. The obtained optimal transformation is shown to be capable to cool the  $AlH^+$  ions to sub mili-kelvins, several orders of magnitude below the current experimental state-of-art work [1].

[1] Lien, Chien-Yu, Christopher M. Seck, Yen-Wei Lin, Jason HV Nguyen, David A. Tabor, and Brian C. Odom. "Broadband optical cooling of molecular rotors from room temperature to the ground state." Nature communications 5 (2014).