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Laser Cooling of Polyatomic Molecules

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14. ABSTRACT
During this grant we achieved and characterized efficient Sisyphus laser cooling of a polyatomic molecule from 50 mK to below 1 mK in 1 dimension. We demonstrated transverse cooling (and heating) of a SrOH beam using two different electronic transitions, studied loss channels to vibrational states (including the bending mode), and highlighted extensions to more complex metal monoalkoxides with six and more atoms. Our results were published in Physical Review Letters and selected as Editor’s suggestion and featured in Physics Viewpoint. To explore laser cooling of more complex molecules, we performed detailed theoretical calculations of Franck-Condon factors and vibrational branching ratios for SrOCH3 and CaOCH3, which are isoelectronic to triatomic SrOH and CaOH. We also created a 1-D MOT of CaOH, a significant step to a MOT of CaOCH3. In addition, in collaboration with Tim Steimle of ASU, we performed dispersed fluorescence of CaOCH3, our target molecule for laser cooling in the future. We determined experimentally the Franck-Condon factors for the X-A and X-B transitions, finding both suitable for laser cooling (although the X-A transition is simpler).

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Compared to atoms, the additional rotational and vibrational degrees of freedom in molecules give rise to a wide variety of potential and implemented scientific applications, including quantum computation, precision measurements, and quantum simulation. While ultracold diatomic molecules will be extremely valuable in opening novel research frontiers, molecules with three or more atoms have unique capabilities enabled by their significantly more complicated structure for advancing fundamental physics, chemistry, and quantum technologies. Cooling molecular degrees of freedom significantly aids in realizing such applications. Yet, the desired quantum complexity that molecules provide also leads to challenges for control, detection, and cooling.

Cooling of the external motion of neutral atoms from above room temperature into the sub-millikelvin range (leading to, e.g., Bose-Einstein condensation) commonly relies on the use of velocity-dependent optical forces. Laser cooling requires reasonably closed and strong optical electronic transitions, so its use for molecules has been severely limited. Recently, laser cooling and magneto-optical trapping has been achieved for diatomic molecules SrF, CaF and YO. However, since SrOH had 3 distinct vibrational modes, including a doubly-degenerate bending mode absent in diatomics, the question of direct laser cooling for polyatomic molecules remained widely open.

Leading up to the past year of activities, we achieved and characterized efficient Sisyphus laser cooling of a polyatomic molecule from 50 mK to below 1 mK in 1 dimension. We demonstrated transverse cooling (and heating) of a SrOH beam using two different electronic transitions, studied loss channels to vibrational states (including the bending mode), and highlighted extensions to more complex metal monoalkoxides with six and more atoms. Our results were published in Physical Review Letters and selected as Editor's suggestion and featured in *Physics* Viewpoint. To explore laser cooling of more complex molecules, we performed detailed theoretical calculations of Franck-Condon factors and vibrational branching ratios for $SrOCH_3$ and $CaOCH_3$, which are isoelectronic to triatomic SrOH and CaOH.

In the past year we have accomplished experimental and theoretical work geared toward laser cooling of symmetric top molecules, which is our proposed work under our AFOSR renewal. Symmetric top molecules (STMs) like $MOCH_3$ (M = Ca, Sr) have first-order linear Stark shifts in small fields. We have been working on detailed analysis of a quantum computing platform based on STMs, including detailed simulations. We will submit our thorough analysis this month (October). On the experimental side, we built over the past year the infrastructure for a MOT of

STM molecules. In addition, in collaboration with Tim Steimle of ASU, we performed dispersed fluorescence of $CaOCH_3$, our target molecules for laser cooling. We determined experimentally the Franck-Condon factors for the X-A and X-B transitions, finding both suitable for laser cooling (although the X-A transition is simpler). This included, of course, production of the molecule. We are therefore in the position to laser cool this species and are currently continuing our experimental build-up to that end.