Unconventional chemistry of polyatomic molecules with optical cycling transitions

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Optical cycling centers (OCCs) is a recently coined term to indicate two states within a small or complex quantum object that can repeatedly experience laser excitation and spontaneous decay, while being well isolated from its environment. Stringent cycling conditions in atoms have led to the successful creation of a novel generation of atomic clocks, matter-wave interferometry, and the development of other highly-sensitive sensors. Extremely low temperatures have also allowed the confinement of di-atomic molecules, built from laser-cooled atoms, in electric, magnetic, and optical traps, where they are isolated from their environment and can be carefully studied.

Isolating cycling transitions in molecules, however, is problematic due to the complexity of their internal ro-vibrational structure. Nevertheless, direct laser-cycling transitions and cooling has been demonstrated for SrF, CaH, and YO dimers, made possible by diagonal Frank-Condon factors between the vibrational modes of the optical transition. This was followed by successful laser cooling on a OCC transition of the polyatomic SrOH molecule to microKelvin temperatures by Dr. Doyle's group at Harvard University.

The objective of our theoretical research is to gain a deep understanding of the electronic, vibrational, and rotational structure of polyatomic molecules that possess optical cycling transitions. In particular, we begin by studying the properties of strontium monoxide molecule, as these molecules have already been laser cooled. These simulations include electronic structure determination of the equilibrium configuration of both the ground and excited, multi-dimensional potential energy surfaces and the evaluation of vibrational and bending modes and corresponding Franck-Condon factors.

We also explore the idea of engineering closed optical cycling transitions and thus enabling Doppler cooling in families of related polyatomic molecules. This can be achieved by chemically bonding atomic and diatomic OCCs to ligand molecules thereby bridging the gap between controllability and tunability of cold atomic systems and the general complexity of large molecules.