

Optical cycling in polyatomic molecules

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An electronically excited state decays after a characteristic lifetime to any of the available molecular states of lower energy. The decay populates various electronic, vibrational, rotational, and spin states. Some molecules however, feature an excited state that decays almost exclusively back to the ground state. In that case a repeated laser excitation followed by a spontaneous decay forms a closed optical cycle. Optical cycling on a bright transition enables rapid transfer of momentum from photons to molecules forming a basis for laser cooling. Laser-cooled molecules find applications as probes in test of fundamental physics, hosts of a high fidelity qubits, or platforms for studying quantum controlled chemistry.¹⁻⁴ The first demonstration of molecular laser cooling was achieved in 2010.⁵ The breakthrough fueled more efforts in the field resulting in laser cooling of three- and six-atomic molecules in respectively 2017 and 2020 and making a promise of achieving optical cycling in even larger polyatomic molecules.^{6,7} I will present the recent work from our group that focuses on extending the domain of molecules with optical cycling centers (OCCs) and new applications stemming from functionalizing molecules with OCCs.⁸⁻¹⁰

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