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Ultracold atoms have been successfully used in quantum simulations and precision measurements. Molecules possess a richer internal structure promising new applications. However, only relatively simple molecules have been produced and employed at ultralow temperatures. This project aims to understand and harness the increasing complexity of ultracold polyatomic molecules to probe the fundamentals of chemistry and physics.

We will extend the range of ultracold polyatomic molecules and their applications in controlled chemistry and precision spectroscopy. We will propose and theoretically investigate two paths: 1) association of ultracold deeply-bound diatomic molecules into ultracold weakly-bound polyatomic molecules and 2) direct cooling deeply-bound polyatomic molecules carefully selected and manipulated with electromagnetic fields. The first approach will build on established atomic techniques, which we will extend to molecular systems. The second one will employ strong fields, short pulses, and structural modifications to engineer closed transitions suitable for laser cooling. We will combine and develop novel electronic structure and quantum scattering methods enhanced by machine learning and high-performance computing. Next, we will study new applications exploiting features emerging from single- molecule and coherent control, conical intersections, and non-trivial electronic states and geometries absent in simpler systems. Applications will range from quantum-controlled chemical reactions and molecular dynamics to precision measurements of fundamental constants and their spatio-temporal variation.

The realization of the project will push cold chemistry into the quantum realm and bring unprecedented complexity to ultracold physics, thus, give new insights into the physical basis of chemistry and the fundamental laws of nature. A unique experience of the PI in both quantum chemistry and ultracold atomic physics will be instrumental in achieving these goals.