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High-Resolution Molecular Spectroscopy on Multiple Potential Energy Surfaces

ABSTRACT: The most precise and fundamental knowledge of molecules comes from high-resolution spectroscopy. Molecular spectroscopy provides quantitative insight into the structure and dynamics of molecules by studying their interaction with the electromagnetic wave. It has broad applications in both pure and applied sciences and industry. High-resolution spectra of molecules can be used as molecular "fingerprints" in combustion diagnostics, atmospheric chemistry, and astrochemistry. Therefore, molecular spectroscopy enables some indispensable methods for investigating chemical reaction kinetics. Furthermore, the complex interplay between the vibration and rotation of nuclei and motions of the electron, including its spin, can be unraveled through analyzing experimentally obtained spectra. In atomic, molecular, and optical (AMO) physics, high-precision laser spectroscopy measurements have been used to test fundamental physical laws and search for new physics beyond the Standard Model.

Research in our lab (https://sites.google.com/site/uofllaserlabs/) consists of spectroscopic studies of gas-phase molecules using state-of-the-art high-resolution laser systems and cutting-edge spectroscopy techniques. Our spectroscopy studies center on the detection and characterization of reactive chemical intermediates, e.g., free radicals and molecules in excited electronic states. The spectroscopic methods employed in our experiments include laser-induced fluorescence/dispersed fluorescence (LIF/DF) spectroscopy for alkoxy (RO·) radicals and cavity ring-down (CRD) spectroscopy for peroxy (ROO·) radicals. These two techniques are also used to study metal-containing molecules as candidates for direct laser cooling and the search for the origin of the matter-antimatter imbalance in the Universe. Recently, our group has started developing a novel cavity-enhanced two-photon double-resonance spectroscopy technique to investigate molecular "dark states". We are particularly interested in molecular species with the Jahn-Teller (JT) effect and the pseudo-Jahn-Teller (pJT) effect, vibronic (vibrational-electronic) interactions that cause spontaneous distortion of the symmetry of polyatomic molecules in degenerate or nearly degenerate electronic states. High-level quantum chemistry calculations are used to help understand the geometry, energy level structure, and dynamics of molecules on multiple potential energy surfaces. Our group also develops spectroscopic models and software to analyze, simulate, and fit the vibronic, rotational and fine structure in experimentally obtained spectra.