

Cavity-enhanced two-dimensional infrared spectroscopy of gas-phase molecules: solution to the problem of optical coherence diffusion

The goal of this project is to acquire fundamentally new experimental data leading to the elucidation of the problem of optical coherence diffusion in molecules in the gas phase. Molecules absorb electromagnetic waves with frequencies corresponding to energy differences between the energy states of the molecule. In the jargon of quantum mechanics, the mathematical object representing such optical excitation, the oscillation of a molecule between two states, is called optical coherence. What happens to the optical coherence after it is created determines what the radiation spectrum of a given molecule will look like in detail. Specifically: after what time does this excitation disappear naturally or under the influence of collisions? How will the motion of the molecule and therefore the motion of the excitation in space change the spectrum, and how will changes in the trajectory of this motion under the influence of collisions change the spectrum? The answer to this question is important for atmospheric studies and for astronomical observations, because just as by knowing the answer to these questions we can predict the shape of the spectrum, so by knowing the spectra we can determine the amount of greenhouse gases in the atmosphere, the temperature of a distant planet or its period of oscillation around the nearest star.

In this project, we will try to answer this question by developing a new experimental technique that combines ultrafast measurements of correlations between different excitations in a molecule with very high sensitivity and resolution. The new technique will allow us to directly observe how the velocities of excited molecules change over time on the pico- and nanosecond scale, and how quickly they converge to the velocity distribution of unexcited molecules. This will bypass the problems faced by other methods that require complex data analysis and are thus subject to systematic errors of several tens of percent. As important as solving this particular molecular physics or physical chemistry problem will be, it will open the way for further research. In particular, increased sensitivity will make it possible to study the dynamics of isolated clusters of water and other molecules in the future, circumventing the interpretive difficulties of liquid-phase measurements.