CONTROL OF COHERENT LIGHT AND ITS APPLICATIONS TO MOLECULAR SPECTROSCOPY

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Recent progress in precise phase control of ultrafast lasers has led to a wide range of applications in both frequency and time domains, including measurement of optical frequencies, precision laser spectroscopy, optical atomic clocks, optical frequency synthesis, as well as pulse timing stabilization, coherent synthesis of optical pulses, and phase-sensitive extreme nonlinear optics. New capabilities for atomic and molecular spectroscopy are emerging.

The combination of precision femtosecond lasers and ultracold atoms have enabled us to enter a qualitatively new regime of ultrahigh resolution spectroscopy. We utilize a phase-coherent wide-bandwidth optical comb to induce the desired multi-path quantum interference effect for the resonantly enhanced two-photon transition (from 5S to 5D states) in the cold Rb atoms. The large bandwidth of the femtosecond laser comb allows us to recover high precision information on hyperfine intervals (RF) and optical transition frequencies among *all* related states. In addition to structural information, we can also obtain information regarding dynamic population transfer among all hyperfine states by varying the interaction time between the femtosecond laser pulses and the cold atoms.

Understanding of molecular structure and dynamics often involves detailed spectral analysis over a broad wavelength range. Such a task can now be accomplished with a desired level of accuracy uniformly across all relevant spectral windows, allowing precise investigations of minute changes in the molecular structure over a large dynamic range. Furthermore, molecular samples can now be prepared in a well-controlled environment and in a well-defined internal state. In our current experiments we accelerate/decelerate a supersonic beam of the Hydroxyl free radicals (OH) to a mean speed adjustable between 500 m/s to rest, with a translational temperature tunable from 1mK to 1K. These velocity-manipulated stable "bunches" contain 10^{6-9} molecules (depending on the translational temperature) at a density of $\approx 10^{7-10}$ cm^{-3} in the beam and $\approx 10^{9}$ cm^{-3} in the trap. These cold samples can be held in an electrostatic trap for precise measurements.

Simultaneous control of timing jitter and carrier-envelope phase is used to phase coherently superpose a collection of successive pulses from a mode-locked laser. Such a passive pulse "amplifier", along with the synchronization technique we developed for pulse synthesis, has led to significant improvements in experimental sensitivity and spatial resolution for nonlinear-optics based spectroscopy and imaging of bio-molecular systems.