DIRECT OBSERVATION OF RYDBERG-RYDBERG TRANSITIONS VIA CPMMW SPECTROSCOPY

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Rydberg-Rydberg transitions of Ca atoms are *directly* observed by chirped-pulse millimeter-wave spectroscopy, which is a form of broadband, high-resolution, free induction decay-detected (FID) spectroscopy with accurate relative intensities. A new setup, a 20 K Neon buffer gas cooled molecular beam system, has been constructed and tested in our lab. The number density of our target molecules, BaF, is increased by a factor of $>$100 relative to a Smalley-type laser ablation supersonic beam source. In addition, the laboratory frame velocity is decreased by factor 10, which improves our spectroscopic resolution to better than 50 kHz FWHM at 100 GHz. The improved molecular beam source opens the door to an extension of the CPmmW spectroscopy from atomic Rydberg states to molecular Rydberg states. I expect to present preliminary data from "pure electronic" spectra of BaF Rydberg molecules. We expect to produce 10^8 state-selected core-nonpenetrating Rydberg molecules in a single pulse of a laser-laser-mm-wave triple resonance excitation sequence.