MILLIMETER WAVE SPECTROSCOPY OF RYDBERG STATES OF MOLECULES IN THE REGION OF 260-295 GHz

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Free induction decay detected chirped pulse millimeter wave spectroscopy of Rydberg-Rydberg transitions in atoms and molecules is a powerful and flexible method for characterizing the electronic structure of Rydberg states and determining the structure and dynamics of the ion-core. Complicating the use of this technique are the difficulties in reliably and repeatedly accessing not just the most information rich core-nonpenetrating states, but also the low-ℓ core-penetrating Rydberg states in the area of principal quantum number $n^* > 35$. Small transition moments and narrow linewidths for transitions between valence electronic states and high Rydberg states are the primary limiting factor. We demonstrate a simple method to avoid the problem entirely by using chirped pulse technology operating in the frequency range of 260-295 GHz, which allows us to sample a lower range of n^* values than before with comparable frequency resolution and accuracy as our previous W-band experiments. Further improvements to our experiment in order to accurately capture details of Stark demolition, a technique that provides rapid differentiation between core-penetrating and core-nonpenetrating states, will also be discussed.