THEORETICAL INVESTIGATION OF THE HIGH-RESOLUTION DOPPLER-FREE TWO-PHOTON CAVITY RING-DOWN LASER ABSORPTION SPECTROSCOPY OF ALKALI RYDBERG STATES

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Due to the phase-coherent relationship of intra-cavity pulse reflections leading to the narrow frequency-selectivity of the cavity, Cavity Ring-Down Laser Absorption Spectroscopy (CRLAS) is a promising method to study Doppler-free two-photon absorption at resolutions at least two orders of magnitude better than the pulse bandwidth of even Fourier Transform Limited nsec pulsed lasers. The nonlinear absorption will lead to a nonexponential cavity decay with can be used to discriminate it from both linear absorption and intrinsic cavity loss. In this study, several issues have been examined in order to access the feasibility of obtaining the high-resolution two-photon absorption spectra of the Rydberg states of alkali dimers by this technique. Using atomic transition dipoles as an approximation, the laser intensities needed to elicit reasonable excitation were estimated, and found to be well within presently available levels. The importance of the various linewidth-limiting mechanisms was also examined. The relative magnitudes of pressure broadening, AC Stark broadening, cavity stability, and time-of-flight broadening lead us to expect a resolution of ~ 1 MHz or better.