DOPPLER-FREE NONLINEAR ABSORPTION IN C_2H_4 USING CW CAVITY RING-DOWN SPECTROSCOPY

CHRISTINE R. BUCHER and KEVIN K. LEHMANN, *Department of Chemistry, Princeton University, Princeton, NJ 08544*; DAVID F. PLUSQUELLIC and GERALD T. FRASER, *Optical Technology Division, National Institute of Standards and Technology, Gaithersburg, MD 20899*.

We report the first systematic study of Doppler-free nonlinear absorption using cavity ring-down spectroscopy (CRDS). We have developed a variant of cw-CRDS for the mid-infrared region between 9-11 μm. A cw CO_2 laser with $\sim$1 mW of tunable microwave sidebands is used as the excitation source. The cavity consists of a Fabry-Perot etalon defined by two mirrors having a reflectivity coating of 99.5% at 10.6 μm. We scan the microwave frequencies to observe a Doppler-free Lamb dip in the ν_7 $11_{1,10} \leftarrow 11_{2,10}$ rovibrational transition of ethylene (C_2H_4) centered at 941.9 cm^{-1} at a total pressure of 10 mTorr (0.5% C_2H_4/He by volume). Power studies on the Lamb dip are presented to understand the effect of saturation on the Lamb dip linewidth, peak height and broadband absorption. The optical power dependence of the results are unexpected based upon the predicted saturation behavior for steady state, counter propagating free space beams, suggesting that a full theory of nonlinear CRDS will be needed to model the present results. A goal of the present experiments is to determine the IR transition moment from the saturation parameter for radicals and other unstable species where the integrated linestrength cannot be directly determined due to unkonwn concentration.