CAVITY RING-DOWN SPECTROSCOPY MEASUREMENTS OF RESONANCE-ENHANCED TWO-PHOTON ABSORPTION BY N_2O

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As recently discussed by K.K. Lehmann in a theoretical analysis of two-photon absorption (TPA) [1], the combination of high circulating power and counterpropagating fields associated with the cavity ring-down spectroscopy (CRDS) technique can yield relatively prominent, two-photon, Doppler-free absorption features for near-resonant, three-level rovibrational systems. Here, we present CRDS measurements of TPA by N_2O near λ =4.5 μm. The experiment comprises a quantum cascade laser which is frequency-coupled to a high-finesse optical resonator through controlled optical feedback. Consistent with Lehmann's model, we observe CRDS decay signals that depend upon two characteristic decay rates: a constant single-photon value and an intracavity-power-dependent two-photon absorption rate. The resulting spectra exhibit strong Doppler-free features, saturation effects at low pressure, and TPA cross-section and collisional broadening coefficient consistent with theoretical predictions.

1. K.K. Lehmann, "Resonance enhanced two-photon cavity ring-down spectroscopy of vibrational overtone bands: A proposal," J Chem Phys **151**, 144201 (2019).