TWO-COLOR, INTRACAVITY PUMP-PROBE, CAVITY RINGDOWN SPECTROSCOPY

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We report a proof-of-principle demonstration of intracavity pump-probe, cavity ringdown (CRD) detection in a three-mirror, traveling-wave cavity. With both cavity-enhanced pump $power$ and probe absorption $pathlength$, the technique is a generalized, high-sensitivity, high-selectivity trace detection method. In our experiments, the pump radiation is switched off during every other probe ringdown, which allows uncorrelated measurements of analyte and background cavity decay rates. The net, two-color signal from the difference between the pump-on and pump-off decay rates is immune to empty cavity ringdown fluctuations and spectral overlaps from non-target molecular transitions. The immunity to the ringdown fluctuations allows longer signal averaging, and thus higher detection sensitivity. The ability to compensate for the unwanted, background molecular absorption greatly enhances the detection selectivity in spectrally congested regions. While its application is not limited to a specific spectral range, the technique is well-suited for trace detection in the mid-IR region, where pump-probe schemes based on strong ro-vibrational transitions can be applied. In this work, our two-color CRD detection is implemented on a ladder-type, three-level system based on the N_2O, $\nu_3 = 1 \leftarrow 0$, P(19) (pump) and $\nu_3 = 2 \leftarrow 1$, R(18) (probe) ro-vibrational transitions. By frequency-locking two quantum cascade lasers to the p-polarization (pump, Finesse = 5280) and s-polarization (probe, Finesse = 67700) modes of the ring-cavity, we achieve high intracavity pump power (36 W) and high probe ringdown rates (>2 kHz). The observed two-color CRD spectra are simulated based on a density-matrix, three-level-system model which is solved under the constraints of the cavity resonance conditions. In addition to its background compensation capability, experimental flexibilities in the selection of pump-probe schemes and signal insensitivity to intracavity laser power are further features which enhance the general utility of our technique for mid-IR trace detection.