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Coherent anti-Stokes Raman spectroscopy in the presence of strong resonant signal from background molecules MARTIN BITTER, VALERY MILNER, University of British Columbia — Laser spectroscopy based on femtosecond coherent anti-Stokes Raman scattering (fs CARS) often involves simultaneous excitation of multiple resonances covered by the broad spectral bandwidth of ultrashort pulses. Determining the chemical composition of a mixture of molecular species with close vibrational frequencies typically requires Fourier analysis of the detected time-resolved fs CARS signal. Here we propose and demonstrate an alternative method of separating vibrational responses from two molecular species with neighboring vibrational modes (here, oxygen and carbon dioxide). We utilize ro-vibrational coupling as a mechanism of suppressing the strong vibrational response from the dominating molecular species (O_2). Coherent ro-vibrational dynamics leads to long "silence windows" of zero CARS signal from oxygen molecules. In these silence windows, the detected signal stems solely from the minority species (CO_2) enabling background-free detection and characterization of the O_2/CO_2 mixing ratio. In comparison to a Fourier analysis, our technique does not require femtosecond time resolution or time-delay scanning.

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