MODELING THE OPTICAL-OPTICAL DOUBLE RESONANCE LINESHAPES IN CH_4

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Optical-optical double resonance spectroscopy is a powerful tool to unravel and assign complex molecular spectra, such as found when exciting the overtone transitions of methane. When essentially monochromatic radiation is used for both pump and probe waves, such spectra are often described as "Doppler-free", but it is more accurate to describe them as having much reduced Doppler broadening. Common treatments describe the lineshape of individual transitions as homogeneously broadened, however, they have widths multiple times the power-broadened homogeneous broadening of the levels involved, even when treated using simplified Bloch equations of three coupled levels. The spatial variation of pump field intensity, both transverse to the propagation vector and due to pump absorption, as well as the M_J dependence or transition dipole moments, further complicate the lineshape but are easily accounted for. More difficult to account for is the presence of inelastic scattering, which changes the Doppler shifts in the probe spectrum, and requires a full collision Kernel to properly model. This talk will present the results of an ongoing attempt to quantitatively model the double resonance spectra we observe using a 3.3 μ continuous wave pump and a 1.67 μm comb probe, in particular their dependence on pump power and sample pressure.