

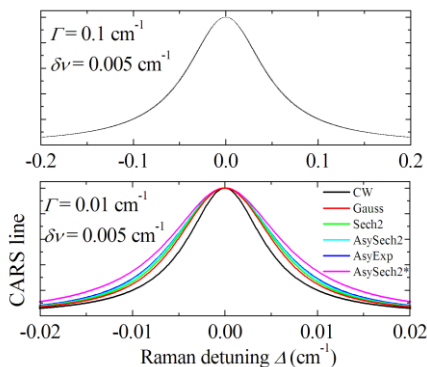
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We review the basic criteria behind the fitting procedure used in traditional CARS approaches for diagnostic application to gases that are probed with laser pulses showing time asymmetry. It appears that asymmetric laser pulses play a role in approaches where a high level of accuracy is requested to decipher vibrational spectra. In particular, the analysis suggests that the contribution to the linewidth of rotational transitions caused by the interaction of such laser pulses with the Raman molecule could be erroneously attributed to the intrinsic Raman width if the customary approach, based primarily on the Voigt lineshape, is assumed.

In the figure, CARS lineshapes are shown in relation to laser pump pulses having the same linewidth of 0.005 cm^{-1} and different time profiles (CW, Gauss, square of the hyperbolic secant, asymmetric square of the hyperbolic secant, asymmetric exponential and strongly asymmetric square of the hyperbolic secant). The upper and the lower panel refer respectively to the chosen extremes of 0.1 and 0.01 cm^{-1} for the Raman linewidth. Note that in the upper extreme, the lineshapes overlap.



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