**Evaluating the electronic and vibrational contributions to second and third harmonic scattering responses**

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The second harmonic (βSHS) and third harmonic (γTHS) scattering hyperpolarizabilities and depolarization ratios of small molecules (water, carbon tetrachloride, chloroform, dichloromethane, chloromethane, and acetonitrile) have been evaluated at the coupled cluster response theory level of approximation[1,2] and compared to experimental data. Besides the static and dynamic βSHS and γTHS responses, the depolarization ratios, which reveal the nature of the responses, are calculated. For γTHS these constitute the first quantum chemical investigations[3,4], following two recent publications on their measurements[5,6].

This contribution will address i) the choice of a suitable atomic basis set, ii) the importance of electron correlation contribution as a function of the chemical composition, iii) the frequency dispersion of the responses, iv) the amplitude of the vibrational contributions (pure vibrational and zero-point vibrational average[7]) in comparison to their electronic counterpart, and v) the relationships between the hyperpolarizabilities/depolarization ratios and the chemical composition.

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