Coupled cluster evaluation of the second and third harmonic scattering responses of small molecules

Flash poster

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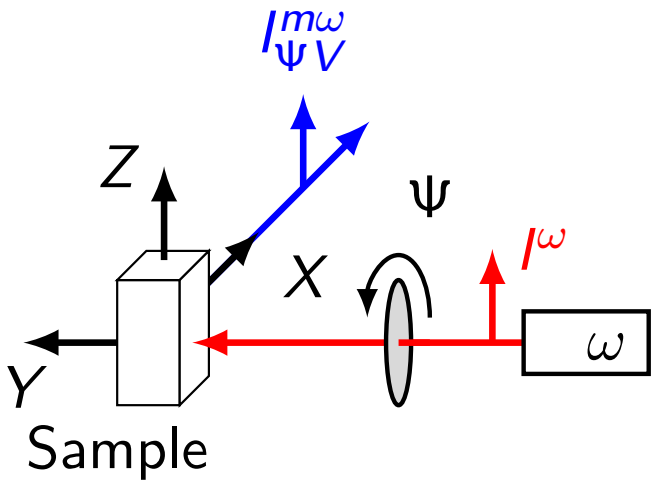
EDT CHIM Meeting (May, 23rd, 2019)

- Interaction between $\vec{F}$ and a molecule (nuclei and electrons) induce charge reorganization (**electronic response**).
- Phenomenological description of the induced $\Delta\vec{\mu}$:

$$\Delta\vec{\mu}(\vec{F}) = \alpha \cdot \vec{F} + \frac{1}{2!}\beta : \vec{F}^2 + \frac{1}{3!}\gamma \vdots \vec{F}^3 + \dots$$

- Experimental measurement rely on **relative** rather than absolute measurement which requires **precise knowledge** of the response of these reference compounds.
- **Quantum chemistry** comes in handy ! β was already investigated for different systems, but there are only **two recent** experimental studies for γ.

Castet, F. *et al. J. Chem. Phys.* **136**, 024506 (2012) ; Van Steerteghem, N. *et al. Anal. Chem.* **89**, 2964 (2017); Rodriguez, V. *J. Chem. Phys. C* **121**, 8510 (2017).



- For a non-polarized incident light in laboratory frame, intensity of scattered light is proportional to β_{SHS} ($m = 2$) or γ_{THS} ($m = 3$). Depolarization ratio (DR) gives information about the geometry.
- Goals of this work are: definition of an approximate level of approximation and reference values as well as extracting structure-activity relationships.

$\Rightarrow$ See poster 23.4

Beaujean, P. and Champagne, B. *Theo. CHem. Acc.* **137**, 50 (2018).