Second harmonic scattering: access to the orientational structure of water.

Julien Duboisset[1] et Pierre-François Brevet[2]

[1]Aix-Marseille Université, CNRS, Centrale Marseille, Institut Fresnel
[2]Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière

Recently, a new optical type of measurement has shown promising results for water/electrolytes organization [1-2]. The technique is based on the nonlinear optical process of second harmonic scattering (SHS), a nonlinear optical phenomenon involving the conversion of two photons at the fundamental frequency ω into one photon at the harmonic frequency 2ω. This second order process is forbidden within the dipole approximation, in centrosymmetric media such as liquids. But, owing to the orientational fluctuations of the non-centrosymmetric water molecules, an incoherent intensity at the harmonic frequency is allowed, which corresponds to the sum of the intensity scattered by each single molecule. Though, when molecules orientations are correlated, the scattered photons emitted by each individual molecule interfere and the scattering pattern is modified. Though tiny, this modification can be detected and attributed to orientational correlations and can be connected to rotational invariant of the orientational pair correlation function derived with integral equation. Based on this principle, the role of electrolytes on water organization can be explored. However, the total magnitude of the SHS signal used in these studies is not fully understood.

[1] Salt Induced long-to-short range orientational transition in water, *Phys. Rev. Lett*. 120, 263001 (2018)

[2] Long-Range Orientational Organization of Dipolar and Steric Liquids, *J. Phys. Chem. Lett.* 2020, 11, 22, 9869–9875