Simulation Predictions of Non-Linear Optics at Solid-Liquid Interfaces

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We report two applications of classical molecular dynamics simulations of solid-liquid interfaces linked with non-linear optics experiments with emphasis on interfaces between mineral particles and aqueous solutions:

1) Using non-polarizable flexible model of water we successfully predict phase-resolved vibrational sum frequency generation (vSFG) spectra of air/water, fluorite (111)/water and α-alumina (0001)/water interfaces. This strongly reduces computation cost compared to *ab initio* calculations [1] and opens the possibility to study larger systems for long periods of time.

2) Using the rigid model of water and analysing dipolar orientation of the interfacial water as a function of surface charge (pH) and ionic strength, we provide simulation evidence allowing for molecular interpretation of polarimetric angle-resolved second harmonic scattering (ARSHS) experiments carried out on SiO_2 and TiO_2 colloidal suspensions [2].

[1] S. Pezzotti, D. R. Galimberti, and M.-P. Gaigeot (2019) *Phys. Chem. Chem. Phys.* **21**, 22188-22202.
[2] A. Marchioro, M. Bischoff, C. Lütgebaucks, D. Biriukov, M. Předota and S. Roke (2019) *J. Phys. Chem. C* **123**, 20393−20404.