Silica/water EDL structure from SHG salt screening experiments

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Directly experimentally characterizing key physical parameters, like the dielectric constant, of the electrical double layer (EDL) at charged solid/liquid interfaces under environmentally relevant conditions in a nonperturbative way remains difficult. Here, we demonstrate using the non-resonant, interfacial potential sensitive variant of second harmonic generation (SHG) known as the Eisenthal or $\chi^{(3)}$ technique to probe EDL structure at the silica/water interface. By varying salt concentrations and utilizing the Gouy-Chapman model of interfacial potential, we are able to calculate key interfacial parameters such as the surface charge density and the dielectric constant of the interfacial layer. The latter calculation relies on a mathematical formalism of second harmonic generation from aqueous interfaces that explicitly accounts for the optical phase of the generated signal and theoretically predicts the initial increase and subsequent decrease of SHG signal that we experimentally observe from these interfaces. https://arxiv.org/abs/1702.02496