Surface Characterization of Colloidal Silica Nanoparticles by Second Harmonic Scattering: Quantifying the Surface Potential and Interfacial Water Order

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Shorthand notations for independent tensor components

Table S1: Overview of the shorthand notation for independent tensor components used in this work.

| Indexed component | Tensor component |
|-------------------|------------------|
| \( \chi_1^{(2)} \) | \( \chi_{||\perp}^{(2)} - \chi_{||\perp}^{(2)} - \chi_{\perp\perp}^{(2)} \) |
| \( \chi_2^{(2)} \) | \( \chi_{||\perp}^{(2)} \) |
| \( \chi_3^{(2)} \) | \( \chi_{\perp\perp}^{(2)} \) |
| \( \chi_4^{(2)} \) | \( \chi_{||\perp}^{(2)} \) |
Parameters used for surface potential fitting

**Table S2**: Parameters used for the AR-SHS fits of NaOH series

| NaOH addition | pH 5.7 | pH 10 | pH 11 |
|---------------|--------|-------|-------|
| Radius (nm)   | 145    | 148   | 142   |
| Second harmonic wavelength (nm) | 515   | 515   | 515   |
| Refractive index of the particle (silica) | 1.46 | 1.46 | 1.46 |
| Refractive index of the solvent (water) | 1.33 | 1.33 | 1.33 |
| Temperature (°C) | 23    | 23    | 23    |
| Number of particles (particles/ml) | $3.50 \times 10^{10}$ | $3.50 \times 10^{10}$ | $3.50 \times 10^{10}$ |
| Concentration of ions (M) | $4.02 \times 10^{-6}$ | 0.0001 | 0.001 |

**Table S3**: Parameters used for the AR-SHS fits of NaCl series

| NaCl addition | pH 10 | pH 10 + 0.1 mM NaCl | pH 10 + 1 mM NaCl | pH 10 + 10 mM NaCl |
|---------------|-------|---------------------|-------------------|-------------------|
| Radius (nm)   | 147   | 146                 | 142               | 143               |
| Second harmonic wavelength (nm) | 515 | 515 | 515 | 515 |
| Refractive index of the particle (silica) | 1.46 | 1.46 | 1.46 | 1.46 |
| Refractive index of the solvent (water) | 1.33 | 1.33 | 1.33 | 1.33 |
| Temperature (°C) | 23    | 23                 | 23                | 23                |
| Number of particles (particles/ml) | $3.50 \times 10^{10}$ | $3.50 \times 10^{10}$ | $3.50 \times 10^{10}$ | $3.50 \times 10^{10}$ |
| Concentration of ions (M) | $1 \times 10^{-4}$ | $2 \times 10^{-4}$ | 0.0011 | 0.0101 |