

**Supporting Information to**  
**“Impact of Salt Purity on Interfacial Water Organization Revealed by Conventional and**  
**Heterodyne-Detected Sum Frequency Generation Spectroscopy”**

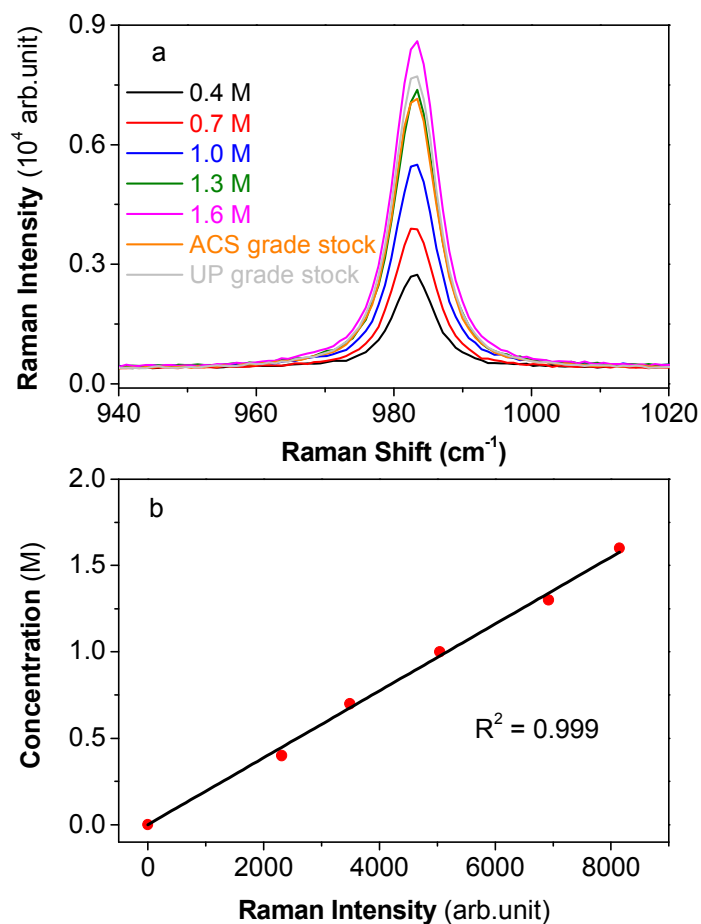
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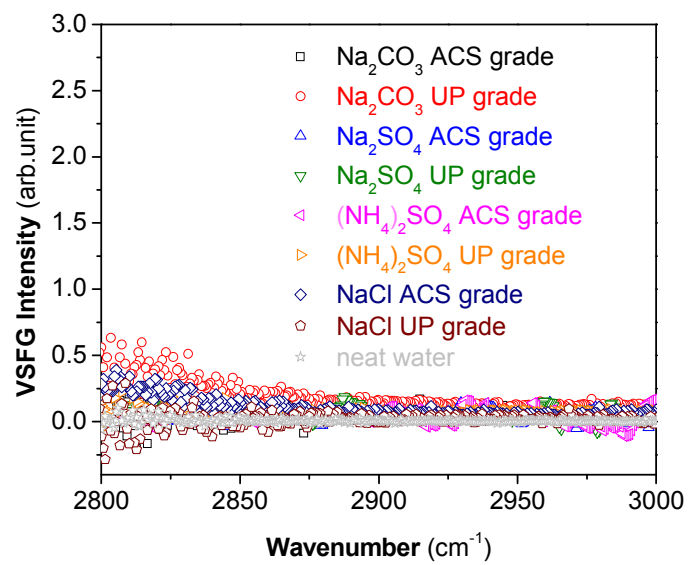
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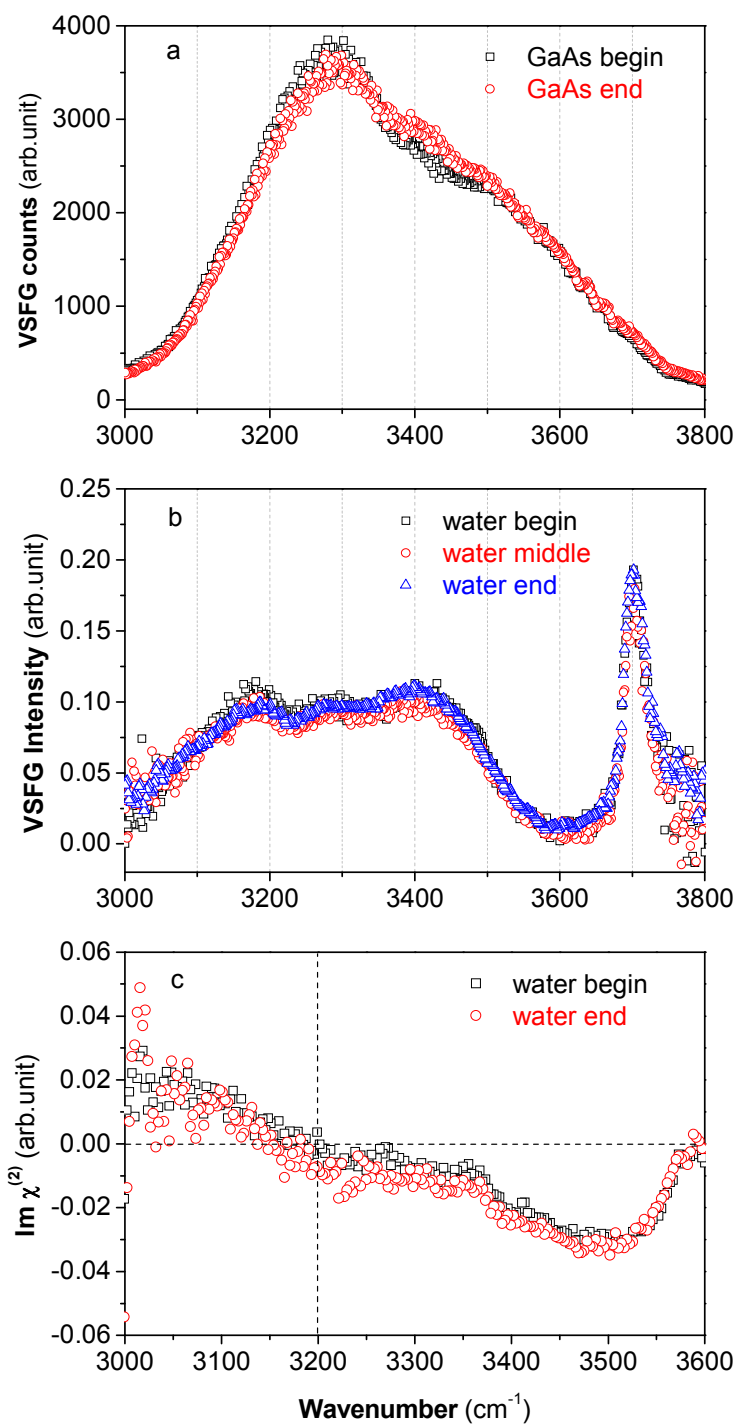
## SUPPLEMENTARY FIGURES



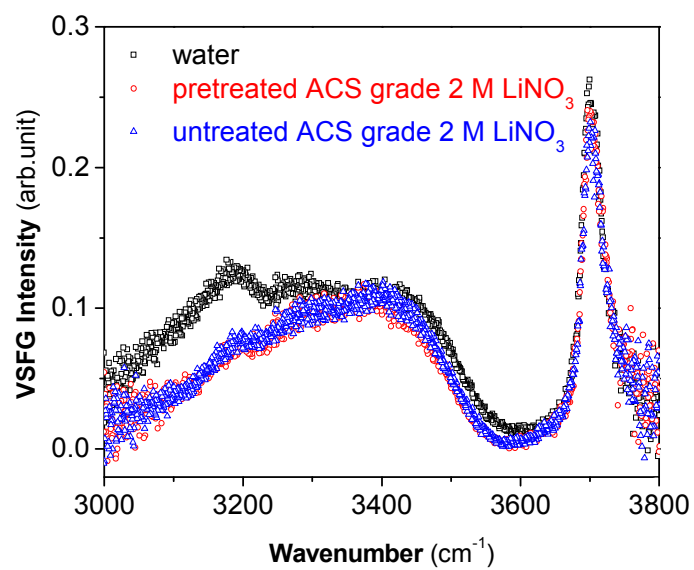
**Figure S1.** (a) Raman spectra of 0.4 M, 0.7 M, 1.0 M, 1.3 M, 1.6 M Na<sub>2</sub>SO<sub>4</sub>, as well as ACS and UP grade pretreated Na<sub>2</sub>SO<sub>4</sub> stock solutions, (b) Calibration curve of Na<sub>2</sub>SO<sub>4</sub> solutions using the height of the vibrational symmetric stretch ion mode of each individual Raman spectrum. The concentrations of other sulfate, carbonate, and nitrate stock solutions were determined in the same manner.



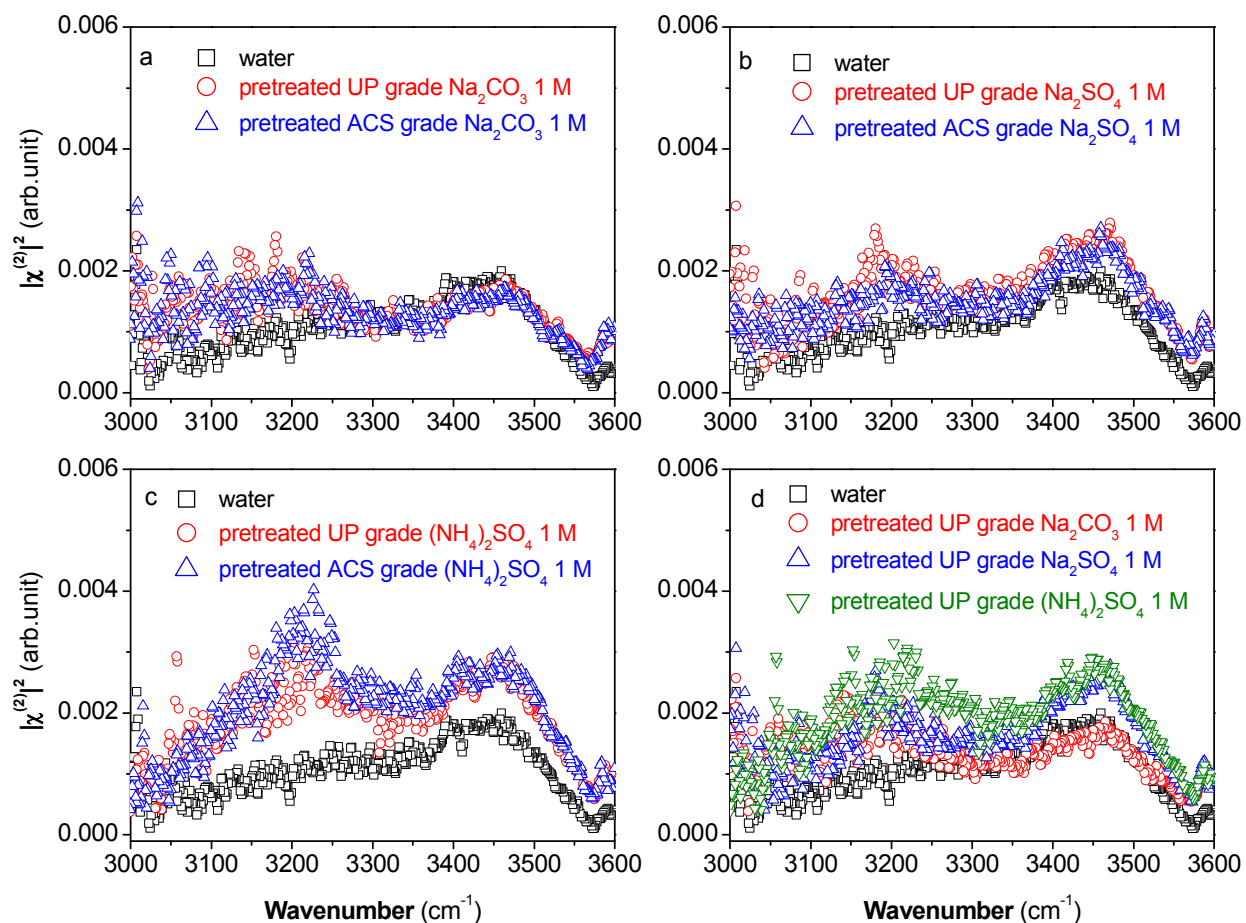
**Figure S2.** Conventional VSGF spectra in the CH stretching region (2800–3000 cm<sup>-1</sup>) of neat water and all stock salt solutions studied after filtering 2 to 4 times.



**Figure S3.** Stability of SFG system shown by the GaAs profile, conventional VSF and HD-VSF  $\text{Im } \chi^{(2)}$  spectra of neat water in the OH stretching region ( $3000 \square 3800 \text{ cm}^{-1}$ ). (a) GaAs profile and (b) conventional VSF spectra in the OH stretching region, and (c)  $\text{Im } \chi^{(2)}$  spectra in the OH stretching region from 3000 to 3600  $\text{cm}^{-1}$ .



**Figure S4.** Conventional VSFG spectra of neat water and ACS grade 2 M LiNO<sub>3</sub> solution in the OH stretching region (3000–3800 cm<sup>-1</sup>) before and after filtration.



**Figure S5.** Deduced power spectra  $|\chi^{(2)}|^2$  from HD-VSFG results of ACS and UP grade salt solutions in the OH stretching region ( $3000 \leq 3600 \text{ cm}^{-1}$ ). (a) 1 M  $\text{Na}_2\text{CO}_3$ , (b) 1 M  $\text{Na}_2\text{SO}_4$ , (c) 1 M  $(\text{NH}_4)_2\text{SO}_4$ , and (d) comparison of all 1 M salt solutions. Neat water spectra are shown as reference.