

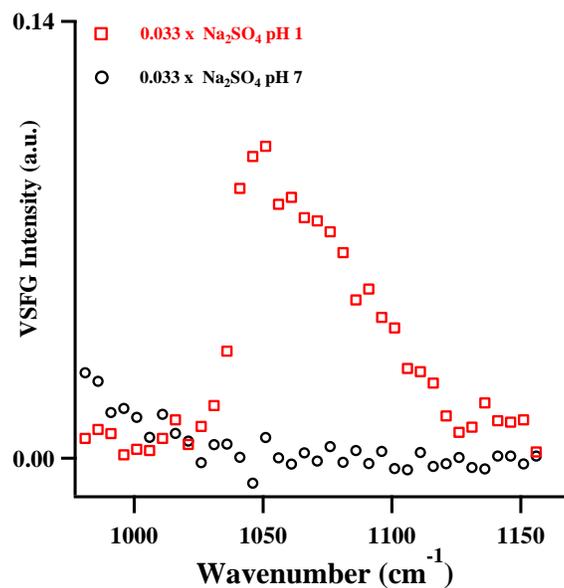
**Supporting Information for:**

“Bisulfate ( $\text{HSO}_4^-$ ) Dehydration at Air/Solution Interfaces Probed by Vibrational Sum Frequency Generation Spectroscopy”

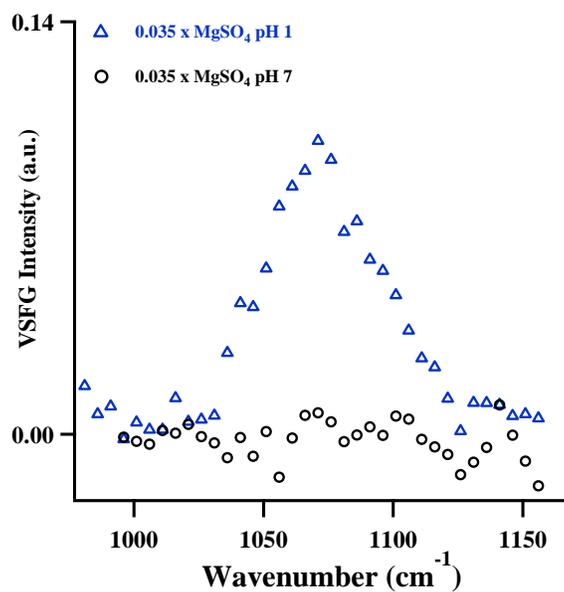
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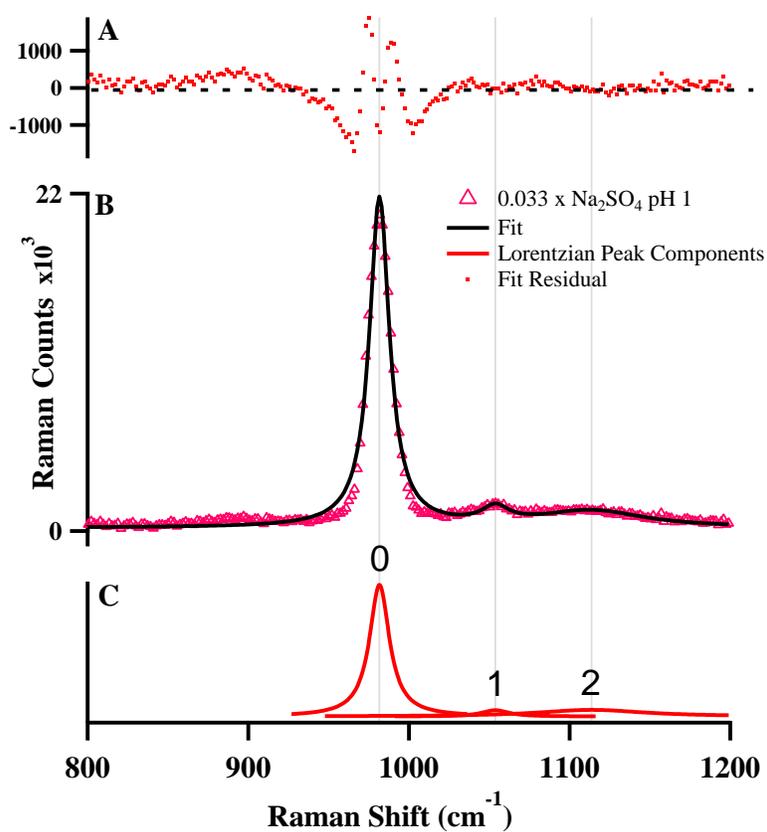
**Figure S1.** SSP VSFG spectra of pH 1 (*red squares*) and pH 7 (*black circles*) 0.033 x Na<sub>2</sub>SO<sub>4</sub> solutions.



**Figure S2.** SSP VSFG spectra of pH 1 (*blue triangles*) and pH 7 (*black circles*) 0.035 x MgSO<sub>4</sub> solutions.

$$I_{VSG} = \left| B_{NR} e^{i\phi} + \left( \sum_{\nu} \frac{A_{\nu}}{\omega_{\nu} - \omega_{IR} + i\Gamma_{\nu}} \right) \right|^2$$

**Equation S1.** Modified Lorentzian profile used to fit the VSGF spectra.



**Figure S3.** Fit of representative Raman spectra (B) with fit residuals (A) and Lorentzian peak components (C). Data (*pink triangles*) from 0.0033 x Na<sub>2</sub>SO<sub>4</sub> solution, fit (*black line*), fit residuals (*red dots*), and Lorentzian peak components (*red lines*) labeled 0-2 shown.

**Table S1.** Raman Spectra Fitting Results for  $\nu_{\text{SS-SO}_3}$  Peak  $\pm 1$  Standard Deviation

Solution	Concentration (x)	$\omega$ (cm <sup>-1</sup> )	I (counts)	FWHM (cm <sup>-1</sup> )	Area
H <sub>2</sub> SO <sub>4</sub>	0.4	1036 $\pm$ 0	3.7 ( $\pm$ 0.05) x 10 <sup>5</sup>	51 $\pm$ 1	3.0 ( $\pm$ 0.07) x 10 <sup>7</sup>
Na <sub>2</sub> SO <sub>4</sub>	0.009	1053 $\pm$ 1	711 $\pm$ 47	27 $\pm$ 3	29906 $\pm$ 2643
	0.017	1053 $\pm$ 2	880 $\pm$ 137	19 $\pm$ 5	26152 $\pm$ 6697
	0.033	1054 $\pm$ 2	1020 $\pm$ 215	20 $\pm$ 8	32705 $\pm$ 11947
MgSO <sub>4</sub>	0.009	1052 $\pm$ 2	475 $\pm$ 79	18 $\pm$ 5	13181 $\pm$ 3741
	0.017	1055 $\pm$ 3	427 $\pm$ 127	19 $\pm$ 10	12806 $\pm$ 6307
	0.035	1059 $\pm$ 6	402 $\pm$ 250	19 $\pm$ 22	12175 $\pm$ 12768