Supporting Information for es049066a:

Table 1. SFG fitting parameters

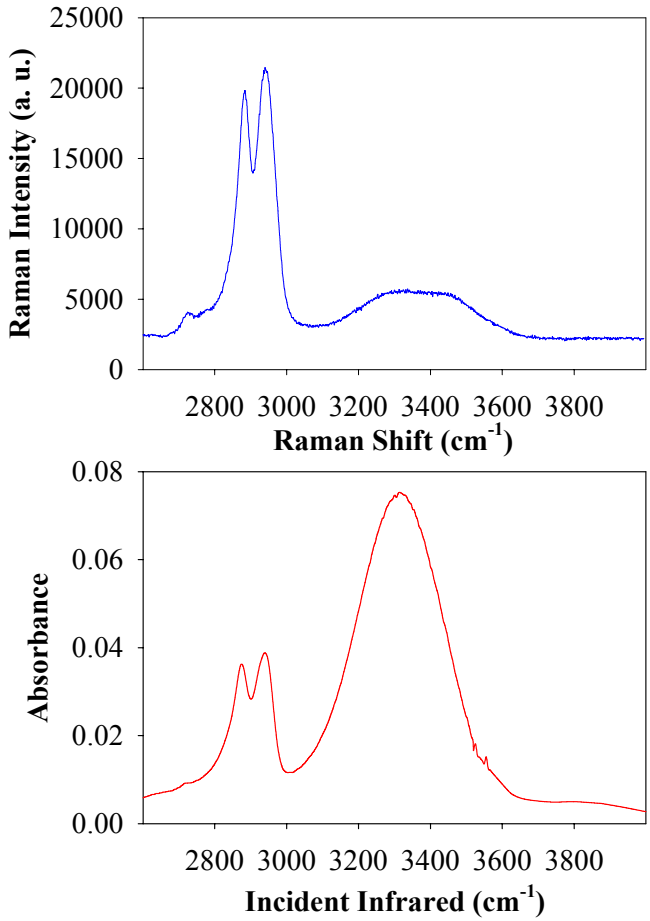
	Amplitude (A_v)	Position (ω_v)	HWHM (Γ_v)	Nonresonance Real	Nonresonance Imaginary	*χ^2		Amplitude (A_v)	Position (ω_v)	HWHM (Γ_v)	Nonresonance Real	Nonresonance Imaginary	*χ^2
H_2O	0.0	2857.8	10.1	-0.98	0.11	23.01	0.1x EG	28.6	2858.1	11.3	-0.35	0.65	48.93
	0.0	2883.1	11.9					31.1	2882.8	10.1			
	0.0	2938.4	9.9					17.2	2944.8	10.0			
	0.0	2954.3	22.5					29.3	2960.8	17.8			
	29.0	3229.6	85.9					76.7	3229.0	85.9			
	119.7	3446.3	115.1					106.8	3446.0	115.1			
	20.1	3533.1	51.4					19.7	3553.0	51.4			
	-35.9	3706.5	13.2					6.9	3710.5	25.6			
0.001x EG	0.5	2857.8	10.1	-0.88	0.45	33.86	0.33x EG	114.0	2871.9	29.3	-1.82	0.78	54.82
	2.3	2883.1	11.9					47.2	2882.8	10.1			
	0.2	2938.4	9.9					36.7	2944.8	10.0			
	1.0	2954.3	22.5					49.2	2957.8	18.2			
	48.5	3229.6	85.9					62.9	3229.6	85.6			
	101.5	3446.3	115.1					116.7	3446.3	115.1			
	5.0	3533.1	51.4					15.9	3533.0	51.4			
	-40.3	3706.5	13.2					-5.5	3709.0	13.0			
0.01x EG	2.8	2857.8	10.1	-0.93	0.27	41.84	0.5x EG	85.5	2875.1	22.2	-2.06	0.86	66.40
	11.8	2883.1	11.9					46.0	2882.8	10.1			
	7.7	2938.4	9.9					34.1	2944.8	10.0			
	-7.7	2954.3	22.5					57.9	2957.8	18.2			
	25.7	3229.0	85.9					84.4	3229.6	85.6			
	115.7	3446.0	115.1					98.3	3446.3	115.1			
	32.1	3533.0	54.0					-6.9	3533.0	51.4			
	-31.0	3706.5	13.2					-8.1	3709.0	13.0			
0.05x EG	20.7	2857.8	10.1	-0.32	0.60	50.81	1.0x EG	106.8	2878.1	22.2	-1.72	0.02	47.76
	25.4	2883.1	11.9					40.1	2882.8	10.1			
	10.3	2950.4	9.9					38.9	2944.8	10.0			
	22.0	2974.3	22.5					62.5	2957.8	18.2			
	78.9	3229.0	85.9					77.1	3229.6	85.6			
	148.5	3446.0	115.1					197.2	3446.3	115.1			
	2.3	3533.0	54.0					-26.4	3533.0	51.4			
	6.5	3713.2	13.0					0.2	3709.0	13.0			
EG + SiO_2	27.5	2878.8	17.0	-1.43	-0.09	43.63	EG + $Al2O_3$	7.9	2846.2	9.5	-1.74	1.13	27.61
	21.6	2883.3	17.3					32.1	2870.4	15.9			
	7.8	2945.8	19.5					11.4	2928.0	16.8			
	42.1	2965.8	20.1					4.5	2932.2	17.7			

*: χ^2 (not the macroscopic susceptibility) describes the goodness of fit and is defined as

$$\chi^2 = \sum \left(\frac{y - y_i}{W_i} \right)^2$$

where y is a fitted value for a given point, y_i is the original data value for the point, and w_i is the standard error for the point. The weighting wave provides the w_i values. The values in the

weighting wave can be either $1/\sigma_i$ or simply σ_i, where σ_i is the standard deviation for each data value. χ^2 is a measure of the goodness of fit. It has absolute meaning only if one has specified a weighting wave containing the reciprocal of the standard error for each data point.



Supporting Figure 1. Raman and infrared spectra of neat ethylene glycol.

Raman

The Raman experimental setup consists of a 532 nm CW laser (Spectra-Physics, Millennia II), a 5 mm focusing Raman Probe (InPhotonics, RP 532-05-15-FC.), a 500-mm monochromator (Acton Research, SpectraPro SP-500) using a 600 g/mm grating and a back-illuminated CCD (Roper Scientific, LN400EB, 1340 x 400 pixel array and deep depletion). Raman spectra were collected using a fiber optic, which was coupled to the entrance slit of the monochromator through a fiber optic imaging coupler (Acton Research, FC-446-030). SpectraSense software (Acton Research version 4.1.9) was used for data collection and display. The power of the 532 nm beam for sample illumination was 20 mW. Before data collection, the Raman system was calibrated by using the 435.83 nm line of a fluorescence lamp and was verified by comparison to the Raman spectrum of naphthalene. The Raman spectral resolution and acquisition temperature were 0.8 cm^{-1} and ~23ºC. The sample was placed in a glass vial.

The sample glass vial was placed in a home-built sample holder, which integrates the sample vial and the Raman probe. The alignment of the Raman probe in the sample holder can affect the detected Raman intensity to some extent. The intensity variation can be normalized by taking a Raman spectrum of a reference sample (i.e. neat ethylene glycol in this study) when comparison of the Raman intensity between different samples is necessary.

FTIR

A Thermo Nicolet FT-IR spectrometer (Avatar^{TM} 370, Thermo Electron Corporation) was employed in the FTIR spectroscopy experiments. The spectrometer is equipped with DTGS KBr detector and purged with hydrocarbon-H_2O-CO_2-free air. Spectra were collected with a spectral resolution of 4 cm^{-1} and 128 scans at a temperature of ~ 24ºC. A demountable IR cell equipped with a pair of CaF_2 windows was utilized. The second derivative method is used to obtain the positions of the overlapped peaks in the FTIR spectrum as a resolution enhancement technique.