

Supporting Information:

Transition in the Acid-Base Component of
Surface Free Energy of Ice upon the
Premelting of its Second Molecular Bilayer

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Additional SFG Spectra

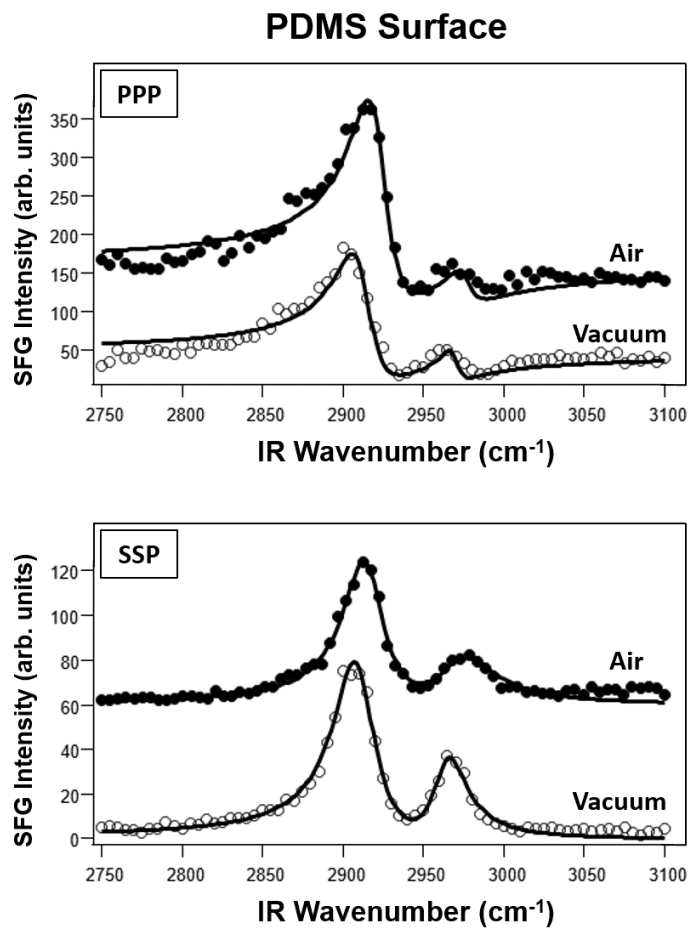


Figure S1: SFG spectra of the PDMS-air and PDMS-vacuum (base pressure 35 Torr) interfaces at 298 K in the PPP and SSP polarizations. The spectra are offset for visual clarity, but are not scaled. The solid lines are Lorentzian peak fits.

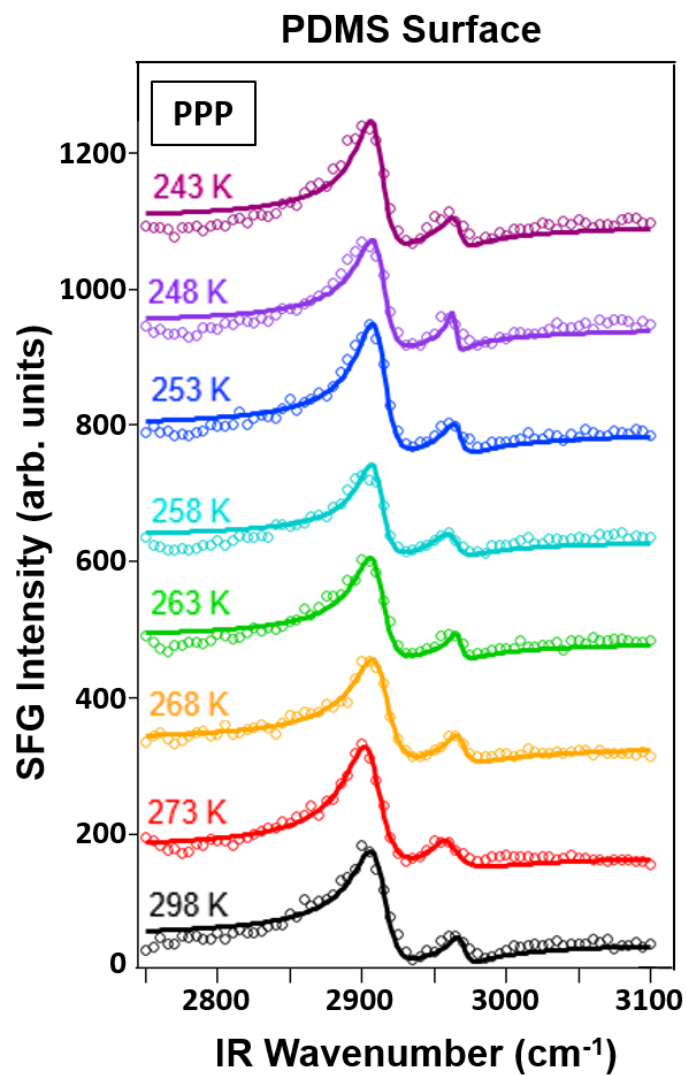


Figure S2: SFG spectra of the PDMS surface (PDMS-vacuum interface, base pressure 35 Torr) in the PPP polarization. The spectra are offset for visual clarity, but are not scaled. The solid lines are Lorentzian peak fits.

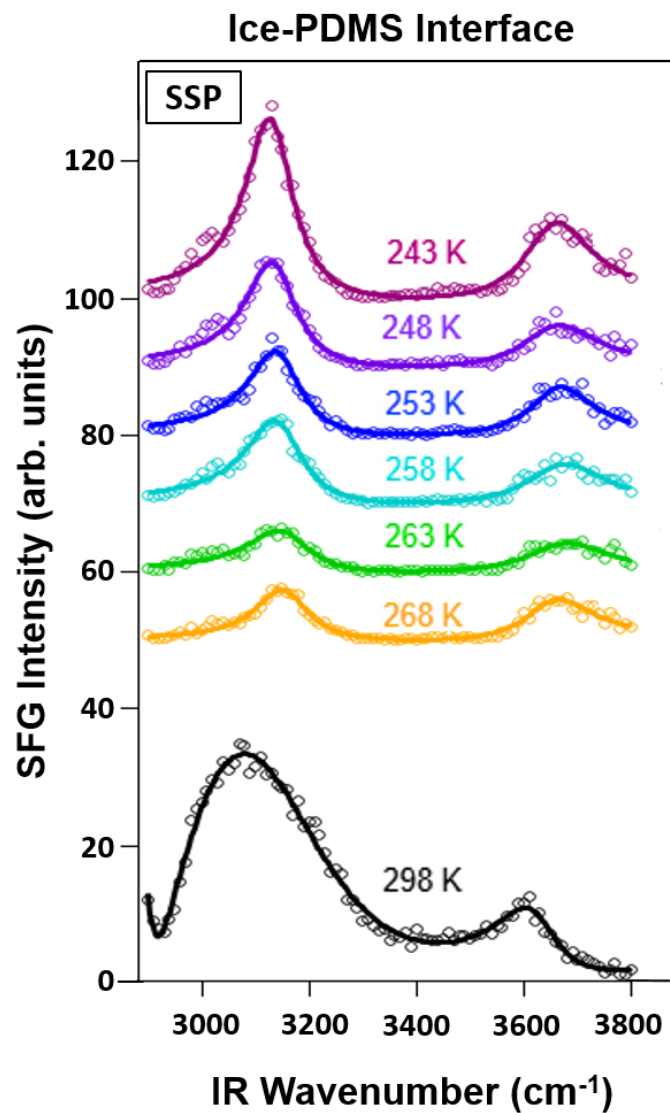


Figure S3: SFG spectra of the ice-PDMS interface in the SSP polarization. The spectra are offset for visual clarity, but are not scaled. The solid lines are Lorentzian peak fits.

SFG Fitting

All parameters in Equation 5 from the main text are allowed to vary freely in order to obtain the best empirical fit of the SFG spectra to the Lorentzian peak functions.

Table S1: Fitting parameters for the representative SSP PDMS-vacuum SFG spectra given in the main text.

T (K)	ω_1	A_1	Γ_1	ω_2	A_2	Γ_2	χ_{NR}
243	2909.4 ± 0.5	130 ± 5	15.0 ± 0.6	2963.2 ± 0.8	73 ± 6	14 ± 1	-0.3 ± 0.1
248	2911.3 ± 0.6	126 ± 5	15.2 ± 0.6	2965.6 ± 0.8	62 ± 6	13 ± 1	-0.2 ± 0.1
253	2910.5 ± 0.5	110 ± 4	15.0 ± 0.6	2964.4 ± 0.7	54 ± 5	12 ± 1	-0.2 ± 0.1
258	2915.2 ± 0.5	110 ± 5	14.5 ± 0.6	2968 ± 1	68 ± 6	16 ± 2	-0.5 ± 0.1
263	2912.3 ± 0.5	136 ± 5	16.7 ± 0.6	2967.2 ± 0.8	70 ± 6	15 ± 1	-0.5 ± 0.1
268	2912.5 ± 0.3	128 ± 4	14.5 ± 0.4	2965.9 ± 0.6	67 ± 4	12 ± 0.8	-0.92 ± 0.09
273	2911.5 ± 0.3	110 ± 3	14.1 ± 0.4	2964.3 ± 0.5	64 ± 3	11.8 ± 0.7	-0.90 ± 0.08
298	2909.7 ± 0.5	134 ± 5	16.1 ± 0.5	2964.3 ± 0.7	65 ± 5	13 ± 1	-0.6 ± 0.1

Table S2: Fitting parameters for the representative PPP PDMS-vacuum SFG spectra given in the Supporting Information.

T (K)	ω_1	A_1	Γ_1	ω_2	A_2	Γ_2	χ_{NR}
243	2911.5 ± 0.8	109 ± 10	11 ± 1	2967 ± 3	26 ± 10	7 ± 3	-7.0 ± 0.2
248	2912.7 ± 0.8	104 ± 10	11 ± 1	2964 ± 1	16 ± 8	3 ± 4	-6.8 ± 0.2
253	2912.8 ± 0.6	121 ± 8	11.5 ± 0.8	2967 ± 2	28 ± 8	7 ± 2	-6.6 ± 0.1
258	2911.5 ± 0.9	86 ± 9	10 ± 1	2964.0 ± 0.6	28 ± 7	8 ± 3	-5.8 ± 0.2
263	2911.5 ± 0.7	111 ± 9	12 ± 1	2967.6 ± 2	21 ± 8	5 ± 3	-5.9 ± 0.1
268	2913.8 ± 0.4	128 ± 6	13.9 ± 0.7	2968 ± 1	35 ± 6	8 ± 2	-5.73 ± 0.09
273	2907.4 ± 0.5	157 ± 10	14.6 ± 0.8	2960 ± 2	57 ± 10	11 ± 3	-4.8 ± 0.1
298	2912.7 ± 0.7	122 ± 9	13 ± 1	2970 ± 2	22 ± 8	6 ± 3	-6.7 ± 0.1

Table S3: Fitting parameters for the representative PPP ice-PDMS SFG spectra given in the main text.

T (K)	ω_1	A_1	Γ_1	ω_2	A_2	Γ_2
243	3133 ± 6	689 ± 57	61 ± 4	3413 ± 9	1618 ± 70	212 ± 11
248	3133 ± 8	657 ± 67	70 ± 5	3418 ± 8	1598 ± 64	222 ± 22
253	3132 ± 5	597 ± 40	72 ± 4	3421 ± 8	1251 ± 62	210 ± 19
258	3130 ± 5	586 ± 51	58 ± 6	3427 ± 9	1196 ± 67	205 ± 17
263	3139 ± 7	501 ± 68	72 ± 6	3416 ± 10	1204 ± 74	217 ± 26
268	3141 ± 5	454 ± 63	78 ± 8	3418 ± 11	1029 ± 58	218 ± 20
	ω_3	A_3	Γ_3	χ_{NR}		
	3704 ± 9	106 ± 41	44 ± 18	1.1 ± 0.2		
	3699 ± 6	91 ± 29	37 ± 11	1.8 ± 0.3		
	3697 ± 6	119 ± 28	46 ± 11	1.8 ± 0.2		
	3701 ± 4	39 ± 14	18 ± 7	1.4 ± 0.3		
	3703 ± 6	85 ± 27	38 ± 12	1.2 ± 0.2		
	3701 ± 5	71 ± 18	35 ± 10	0.9 ± 0.2		

Table S4: Fitting parameters for the representative SSP ice-PDMS SFG spectra given in the Supporting Information.

T (K)	ω_1	A_1	Γ_1	ω_2	A_2	Γ_2	χ_{NR}
243	3131 ± 2	274 ± 7	55 ± 2	3654 ± 5	246 ± 14	76 ± 5	-0.08 ± 0.08
248	3136 ± 3	219 ± 8	58 ± 2	3657 ± 7	202 ± 15	85 ± 8	-0.09 ± 0.08
253	3144 ± 3	202 ± 8	60 ± 3	3664 ± 5	189 ± 11	73 ± 5	-0.15 ± 0.08
258	3144 ± 3	216 ± 8	65 ± 3	3670 ± 6	208 ± 13	89 ± 7	-0.15 ± 0.08
263	3155 ± 5	163 ± 10	70 ± 5	3673 ± 8	184 ± 13	92 ± 8	-0.11 ± 0.09
268	3155 ± 3	159 ± 6	60 ± 3	3653 ± 4	185 ± 9	79 ± 5	0.10 ± 0.06

Table S5: Fitting parameters for the representative PPP (main text) and SSP (Supporting Information) water-PDMS SFG spectra at 298 K.

Polarization	ω_1	A_1	Γ_1	ω_2	A_2	Γ_2	
PPP	2887 ± 3	56 ± 13	22 ± 5	3045 ± 9	1214 ± 88	178 ± 12	
SSP	—	—	—	3111 ± 15	1227 ± 125	235 ± 11	
	ω_3	A_3	Γ_3	ω_4	A_4	Γ_4	χ_{NR}
	3398 ± 12	337 ± 116	144 ± 31	3684 ± 3	107 ± 11	33 ± 5	1.3 ± 0.1
	—	—	—	3626 ± 5	141 ± 21	72 ± 9	-3.8 ± 0.2