Supplementary Information – Frustration *vs*. Prenucleation: Understanding the Surprising Stability of Supersaturated Sodium Thiosulfate Solutions

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FIT PARAMETERS FOR THE 1:5 AQUEOUS SODIUM THIOSULFATE SOLUTION

Table SI1. Fit Parameters for the Modified Havriliak-Negami Functions in the 1:5 Solution Spectra. [a]

		measurement temperature / °C				
		-25	**0**	**25**	**50**	**75**
CC	S_{OKE} / a.u.	1.12	0.45	1.09	1.20	1.57
	τ / ps	44.1×10^3	27.2	20.9	10.2	10.1
	Γ_{Rise} / THz	1.0	1.0	1.0	1.0	1.0
	Γ_{Decay} / THz	0.0	0.5×10^{-3}	0.02	0.03	0.03
	α	0.28	0.42	0.67	0.70	0.66
	β	1	1	1	1	1
D1	S_{OKE} / a.u.	-	-	0.48	0.53	0.59
	τ / ps	-	-	99.6	45.4	25.4
	Γ_{Rise} / THz	-	-	0.1	0.1	0.1
	Γ_{Decay} / THz	-	-	0.0	0.0	0.0
	α	-	-	1	1	1
	β	-	-	1	1	1
D2	S_{OKE} / a.u.	-	1.32	1.06	1.24	-
	τ / ps	-	5.1×10^3	510.5	210.0	-
	Γ_{Rise} / THz	-	0.1	0.1	0.1	-
	Γ_{Decay} / THz	-	0.0	0.0	0.0	-
	α	-	1	1	1	-
	β	-	0.56	1	1	-
D3	S_{OKE} / a.u.	-	-	1.90	1.67	2.16
	τ / ps	-	-	2.2×10^3	704.7	206.7
	Γ_{Rise} / THz	-	-	0.1	0.1	0.1
	Γ_{Decay} / THz	-	-	0.0	0.0	0.0
	α	-	-	1	1	1
	β	-	-	1	1	1

[a] Used to model the α- and β-relaxations of the temperature-dependent spectra.

Table SI2. Fit Parameters for the Anti-symmetrized Gaussian Functions in the 1:5 Solution Spectra.[a]

		measurement temperature / °C				
		-25	**0**	**25**	**50**	**75**
G1	S_{OKE} / a.u.	0.26	0.19	0.35	0.33	0.29
	ω_0 / THz	1.95	1.78	1.65	1.59	1.51
	γ / THz	1.35	1.34	1.41	1.45	1.43
G2	S_{OKE} / a.u.	0.13	0.16	0.28	0.26	0.33
	ω_0 / THz	4.45	3.96	3.61	3.52	3.44
	γ / THz	1.88	3.07	3.02	2.99	3.48

[a] Used to model the librations of temperature-dependent spectra.

Table SI3. Fit Parameters for the Brownian Oscillator Functions in the 1:5 Solution Spectra. [a]

		measurement temperature / °C				
		-25	**0**	**25**	**50**	**75**
rock	S_{OKE} / a.u.	0.025	0.038	0.051	0.047	0.080
	ω_0 / THz	10.24	10.24	10.24	10.24	10.24
	γ / THz	0.37	0.54	0.37	0.37	0.76
stretch	S_{OKE} / a.u.	0.035	0.038	0.060	0.054	0.056
	ω_0 / THz	13.43	13.43	13.43	13.43	13.43
	γ / THz	0.30	0.35	0.30	0.30	0.36
bend	S_{OKE} / a.u.	7.0×10^{-3}	0.011	0.015	0.014	0.016
	ω_0 / THz	16.24	16.24	16.24	16.24	16.24
	γ / THz	0.47	0.64	0.47	0.47	0.58

[a] Used to model the vibrations of the thiosulfate ion in the temperature-dependent spectra. The vibrations visible in these spectra are the S-O-S rock, the S-S stretch as well as the asymmetric S-O bend.

FIT PARAMETERS FOR THE 1:32.7 AQUEOUS SODIUM THIOSULFATE SOLUTION

Table SI4. Fit Parameters for the Anti-symmetrized Gaussian Functions in the 1:32.7 Solution Spectra. [a]

	S_{OKE} / a.u	ω_0 / THz	γ / THz
G1	0.082	1.30	1.38
G2	0.10	3.34	3.74
TA_{H2O}	0.057	0.76	1.74
LA_{H2O}	0.051	4.77	3.03

[a] Used to model the two librational modes of the thiosulfate anions as well as the acoustic phonon modes (transverse and longitudinal) of water.

Table SI5. Fit Parameters for the Modified Havriliak-Negami Functions in the 1:32.7 Solution Spectra. [a]

	S_{OKE} / a.u	τ / ps	Γ_{Rise} / THz	Γ_{Decay} / THz	α	β
CC	0.65	9.3	1	0.03	0.65	1
CD	0.48	87.0	0.5	0.0	1	0.67
CC_{H2O}	0.086	0.7	10.0	0.0	0.94	1

[a] Used to model the α- and β-relaxations of the thiosulfate anions. An additional Cole-Cole function was used to model the water contribution.

Table SI6. Fit Parameters for the Brownian Oscillator Functions in the 1:32.7 Solution Spectra. [a]

	S_{OKE} / a.u	ω_0 / THz	γ / THz
rock	0.021	10.24	0.85
stretch	0.012	13.43	0.42
bend	4.3×10^{-3}	16.24	0.66
lib_{H2O}	0.047	12.57	6.81

[a] Used to model the vibrations of the thiosulfate ion. The vibrations visible in these spectra are the S-O-S rock, the S-S stretch as well as the asymmetric S-O bend. Additionally, the libration of water was modeled with a Brownian oscillator function.

FITTED SPECTRA FOR THE 1:5 AQUEOUS SODIUM THIOSULFATE SOLUTION

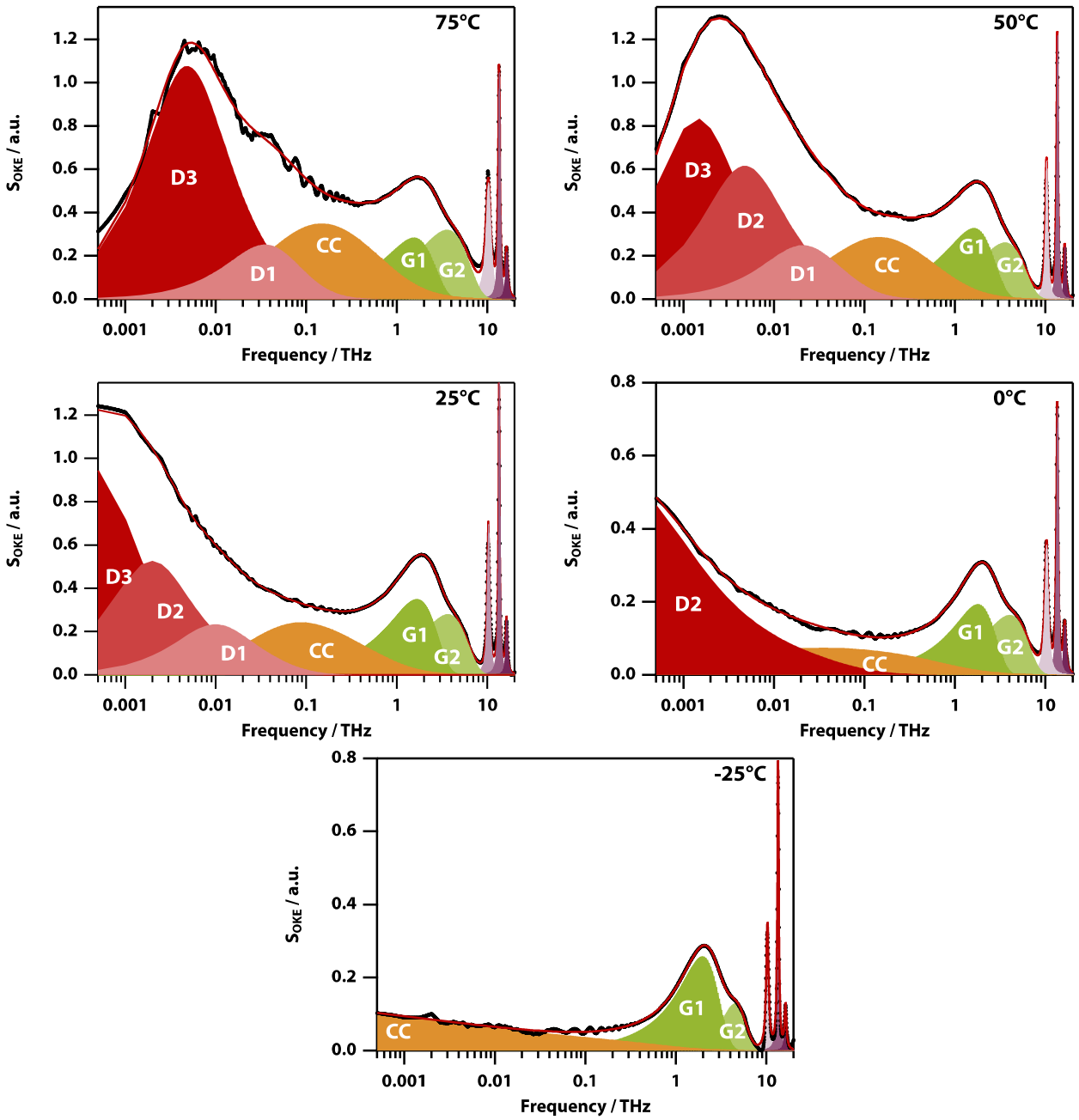

Figure SI1. Fits for the OKE Spectra of the 1:5 Solution at 75, 50, 25, 0, and -25°C. Shown are the vibrational modes (purple), the librational modes G1 and G2 (green), the β-relaxation CC (orange), and the α-relaxation (red), which is labelled as D1-D3.