Supporting Information for:

Origins of Spectral Broadening in Iodated Vaska's Complex in Binary Solvent Mixtures

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Table S1. Fit parameters to CLS decays for BA and $CHCl_3$ binary mixtures in benzene-d_6.

mole fraction	A_1 (norm.)	τ_1 (ps)	A_2 (norm.)	τ_2 (ps)	A_0 (norm.)	A_{total} (norm.)
			$CHCl_3$ in benzene-d_6			
0	0.25 (0.08)	2.4 (0.5)	0.20 (0.08)	7.0 (1.6)	0.017 (0.002)	0.46 (0.11)
0.1	0.19 (0.07)	2.0 (0.5)	0.31 (0.07)	6.8 (1.0)	0.010 (0.002)	0.51 (0.10)
0.3	0.22 (0.03)	1.5 (0.2)	0.46 (0.03)	8.0 (0.5)	0.009 (0.002)	0.69 (0.04)
0.5	0.16 (0.03)	1.4 (0.4)	0.65 (0.03)	8.6 (0.5)	0.009 (0.003)	0.82 (0.05)
0.7	0.08 (0.05)	1.6 (1.0)	0.69 (0.05)	8.1 (0.6)	0.019 (0.004)	0.775 (0.007)
0.9	0.08 (0.05)	1.6 (1.3)	0.75 (0.05)	9.3 (0.7)	0.009 (0.005)	0.821 (0.006)
1			0.808 (0.004)	7.7 (0.1)		0.808 (0.004)
			BA in benzene-d_6			
0	0.25 (0.08)	2.4 (0.5)	0.20 (0.08)	7.0 (1.6)	0.017 (0.002)	0.46 (0.11)
0.05	0.22 (0.12)	1.8 (0.7)	0.42 (0.12)	5.9 (1.1)	0.027 (0.004)	0.67 (0.17)
0.1	0.36 (0.07)	2.2 (0.4)	0.31 (0.07)	9.6 (1.8)	0.036 (0.005)	0.71 (0.10)
0.2	0.16 (0.05)	1.4 (0.6)	0.52 (0.06)	7.6 (0.8)	0.045 (0.005)	0.73 (0.08)
0.3	0.32 (0.09)	3.6 (0.8)	0.37 (0.09)	12.3 (2.1)	0.035 (0.004)	0.718 (0.13)
0.4	0.38 (0.07)	4.2 (0.6)	0.33 (0.06)	15.9 (2.4)	0.052 (0.004)	0.763 (0.09)
0.5	0.23 (0.03)	2.6 (0.5)	0.49 (0.03)	16.8 (1.5)	0.054 (0.005)	0.774 (0.05)
0.7	0.32 (0.05)	5.6 (0.9)	0.43 (0.05)	26.8 (3.6)	0.069 (0.008)	0.822 (0.07)
1	0.34 (0.04)	7.5 (1.0)	0.38 (0.03)	45.4 (9.3)	0.028 (0.019)	0.756 (0.06)

Table S2. Fit parameters from linear response function to reproduce linear lineshape.

mole fract.	Δ_1 (rad/ps)	τ_1 (ps)[a]	Δ_2 (rad/ps)	τ_2 (ps)[a]	Δ_0 (rad/ps)	T_2 (ps)	ν_{CO} (cm^{-1})	Γ (cm^{-1})
$CHCl_3$ in benzene-d_6								
0	0.47 (0.02/0.12)	2.4	0.41 (0.01/0.12)	7.0	0.12 (0.03/0.05)	4.6 (0.3/0.2)	2068.6	2.3 (0.1/0.1)
0.1	0.44 (0.02/0.10)	2.0	0.57 (0.01/0.09)	6.8	0.10 (0.03/0.03)	5.7 (0.4/0.3)	2068.8	1.9 (0.1/0.1)
0.3	0.49 (0.04/0.04)	1.5	0.68 (0.01/0.04)	8.0	0.10 (0.07/0.02)	4.5 (0.4/0.3)	2069.1	2.3 (0.2/0.1)
0.5	0.42 (0.10/0.04)	1.4	0.86 (0.02/0.03)	8.6	0.10 (0.15/0.02)	4.8 (1.1/0.5)	2069.3	2.2 (0.5/0.2)
0.7	0.36 (0.27/0.07)	1.6	1.03 (0.05/0.04)	8.1	0.17 (0.26/0.02)	5.6 (4.6/0.7)	2069.4	1.9 (1.6/0.2)
0.9	0.38 (0.42/0.07)	1.6	1.20 (0.10/0.03)	9.3	0.13 (0.84/0.02)	5 (6.1/0.3)	2069.6	2.1 (2.6/0.1)
1			1.34 (0.11/0.002)	7.7		4.9 (0.8/0.6)	2069.7	2.2 (0.3/0.3)
BA in benzene-d_6								
0	0.47 (0.02/0.12)	2.4	0.41 (0.01/0.12)	7.0	0.12 (0.03/0.05)	4.6 (0.3/0.2)	2068.6	2.3 (0.1/0.1)
0.05	0.39 (0.02/0.19)	1.8	0.53 (0.01/0.18)	5.9	0.14 (0.02/0.07)	4.3 (0.2/0.2)	2068.7	2.5 (0.1/0.1)
0.1	0.5 (0.01/0.1)	2.2	0.47 (0.01/0.1)	9.6	0.16 (0.02/0.04)	4.9 (0.3/0.2)	2068.8	2.2 (0.1/0.1)
0.2	0.35 (0.03/0.09)	1.4	0.62 (0.01/0.07)	7.6	0.18 (0.02/0.03)	5.5 (0.4/0.3)	2068.9	1.9 (0.1/0.1)
0.3	0.52 (0.01/0.12)	3.6	0.56 (0.01/0.12)	12.3	0.17 (0.03/0.05)	6.5 (0.6/0.5)	2068.9	1.6 (0.2/0.1)
0.4	0.56 (0.01/0.08)	4.2	0.53 (0.01/0.08)	15.9	0.21 (0.02/0.03)	6.7 (0.7/0.6)	2069.0	1.6 (0.2/0.1)
0.5	0.45 (0.02/0.04)	2.6	0.66 (0.01/0.04)	16.8	0.22 (0.02/0.02)	6.9 (0.7/0.6)	2069.1	1.5 (0.1/0.1)
0.7	0.53 (0.01/0.05)	5.6	0.62 (0.01/0.06)	26.8	0.25 (0.02/0.03)	11.6 (1.9/1.4)	2069.1	0.9 (0.1/0.1)
1	0.62 (0.01/0.05)	7.5	0.65 (0.01/0.04)	45.4	0.18 (0.03/0.06)	14.3 (2.7/2.0)	2069.2	0.7 (0.1/0.1)

a. τ values are the same as Table S1 as they were not an adjustable fitting parameter here.

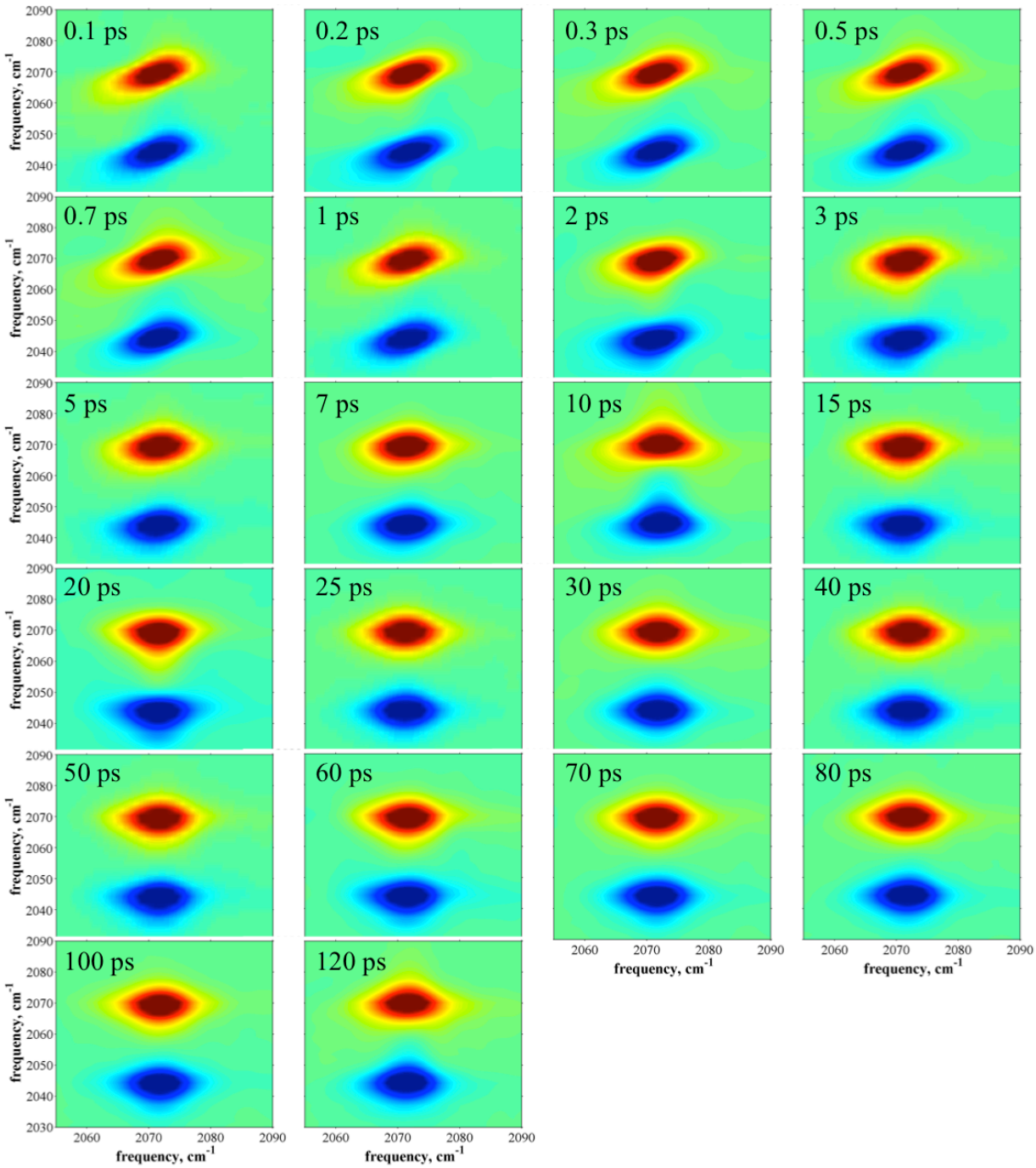

Figure S1. 2D-IR spectra for all T_w values for VC-I_2 in neat d_6-benzene.

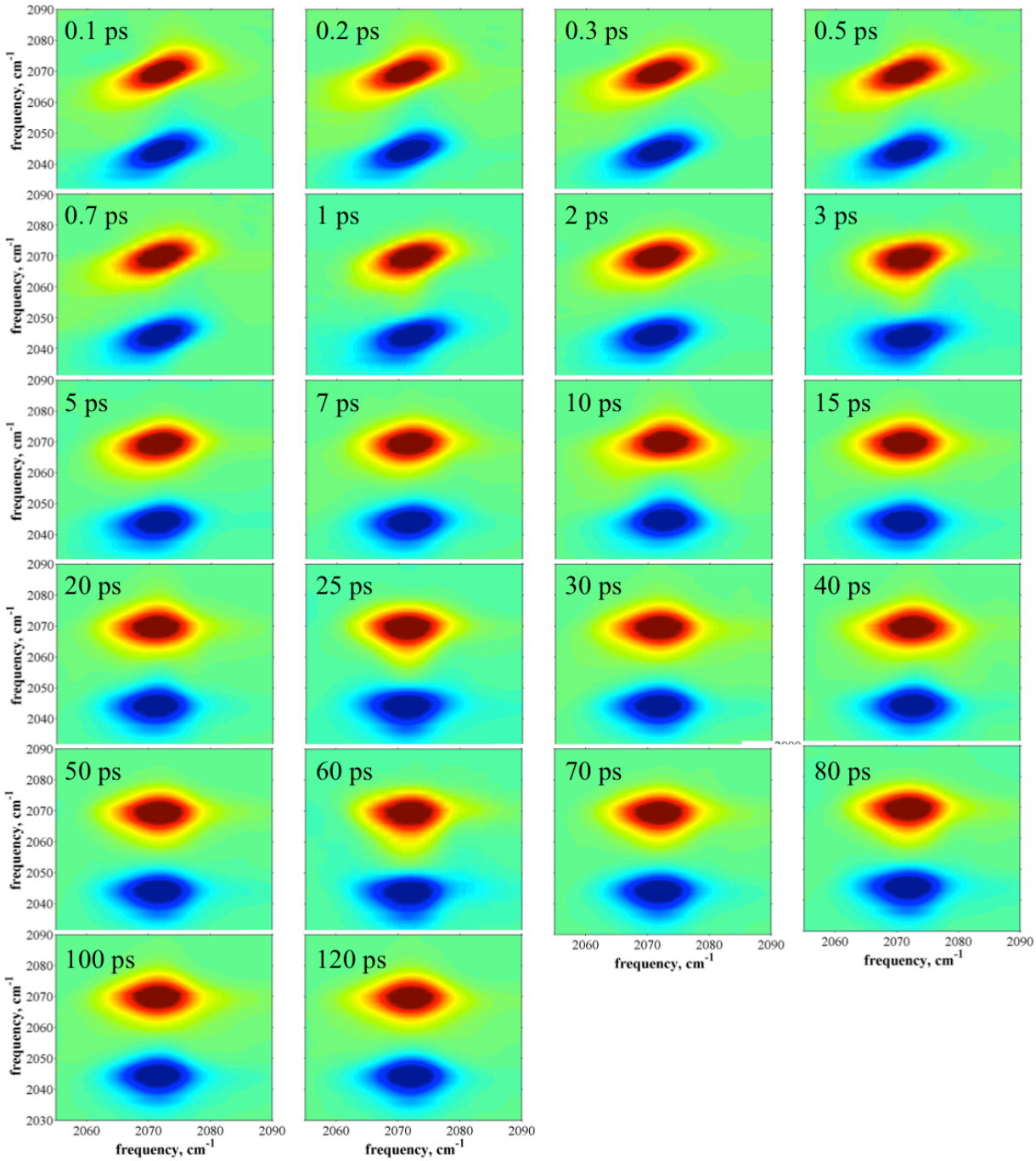

Figure S2. 2D-IR spectra for all T_w values for VC-I_2 in 0.1 mole fraction $CHCl_3$ in d_6-benzene.

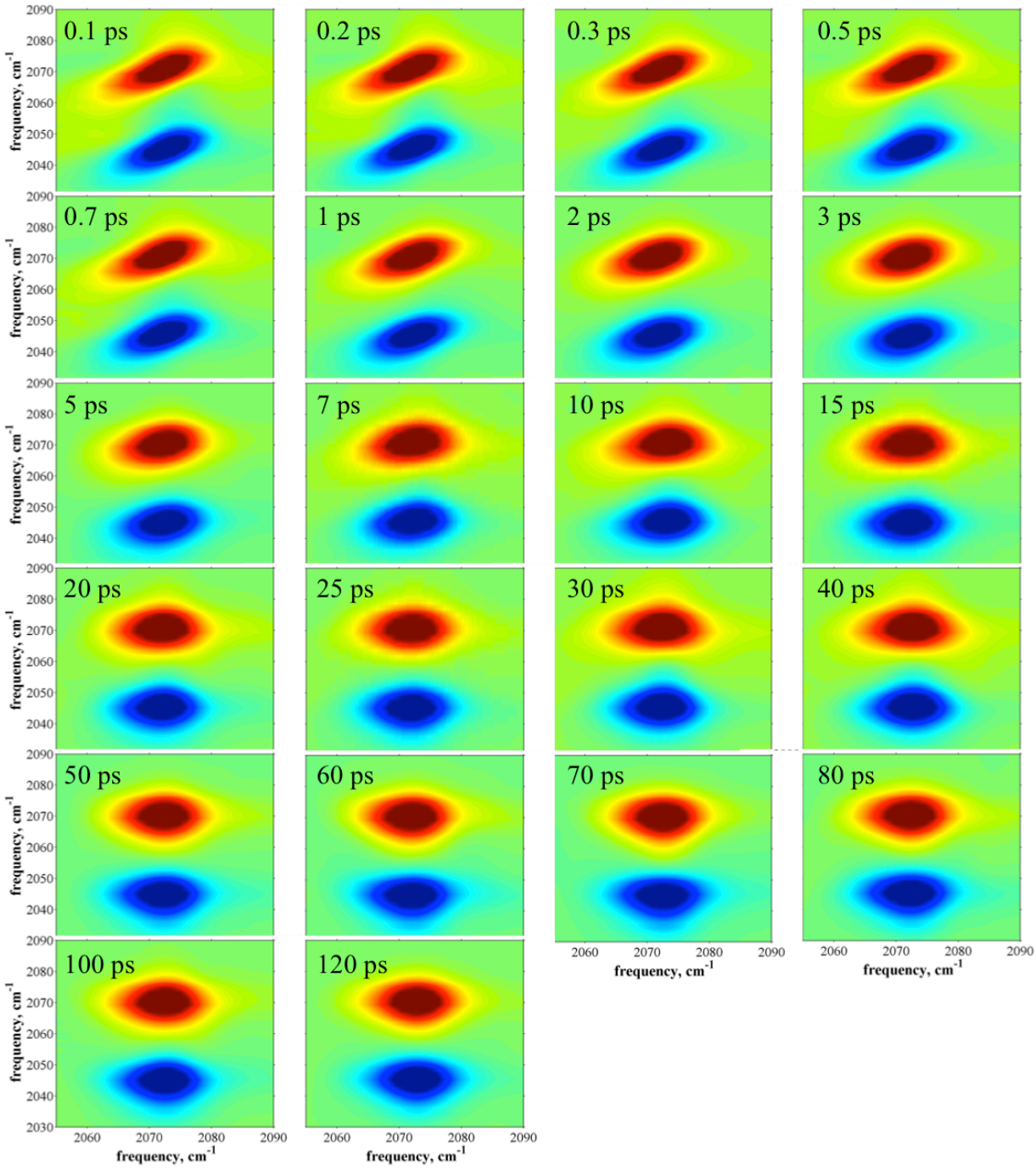

Figure S3. 2D-IR spectra for all T_w values for VC-I_2 in 0.3 mole fraction $CHCl_3$ in d_6-benzene.

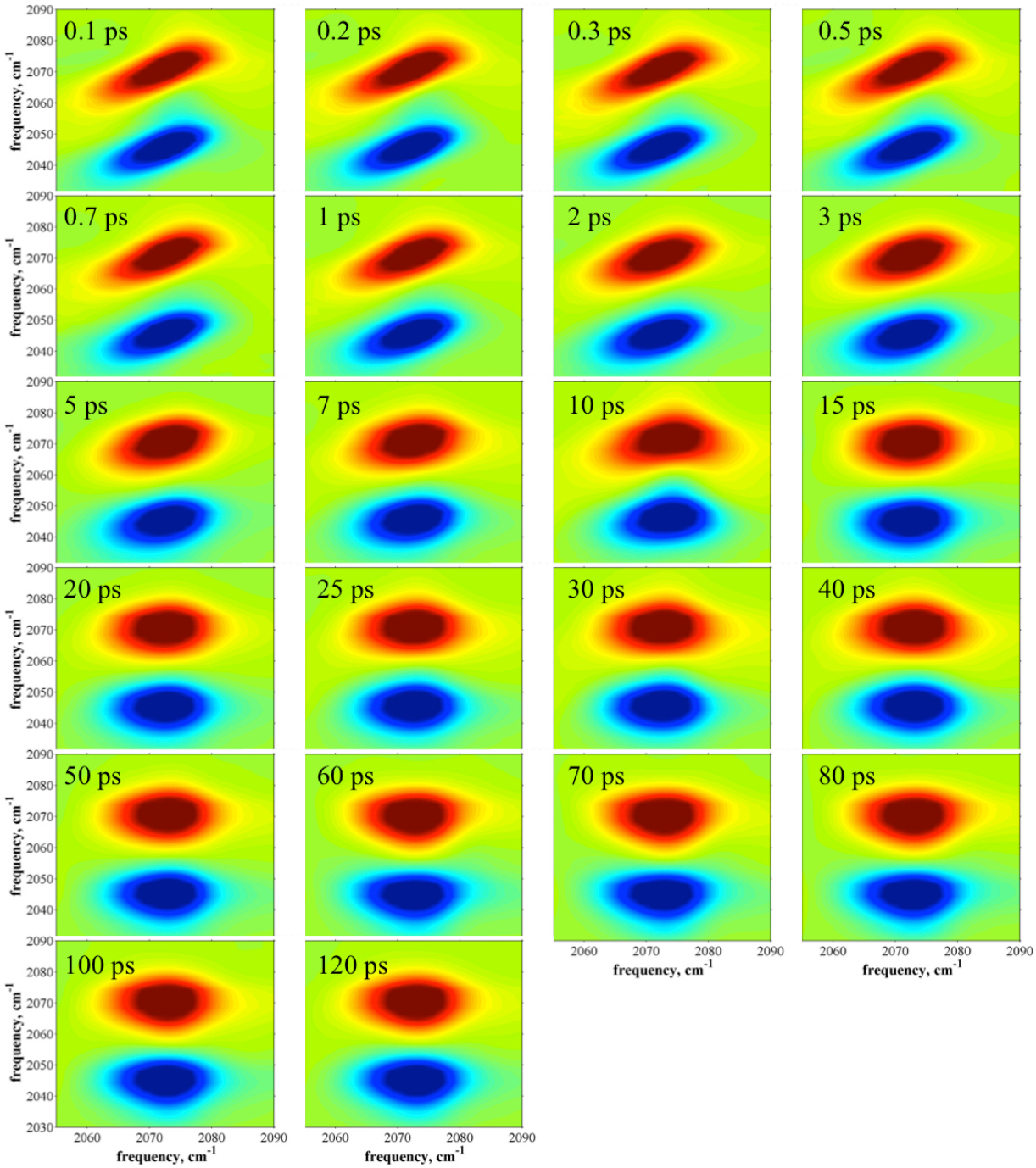

Figure S4. 2D-IR spectra for all T_w values for VC-I_2 in 0.5 mole fraction $CHCl_3$ in d_6-benzene.

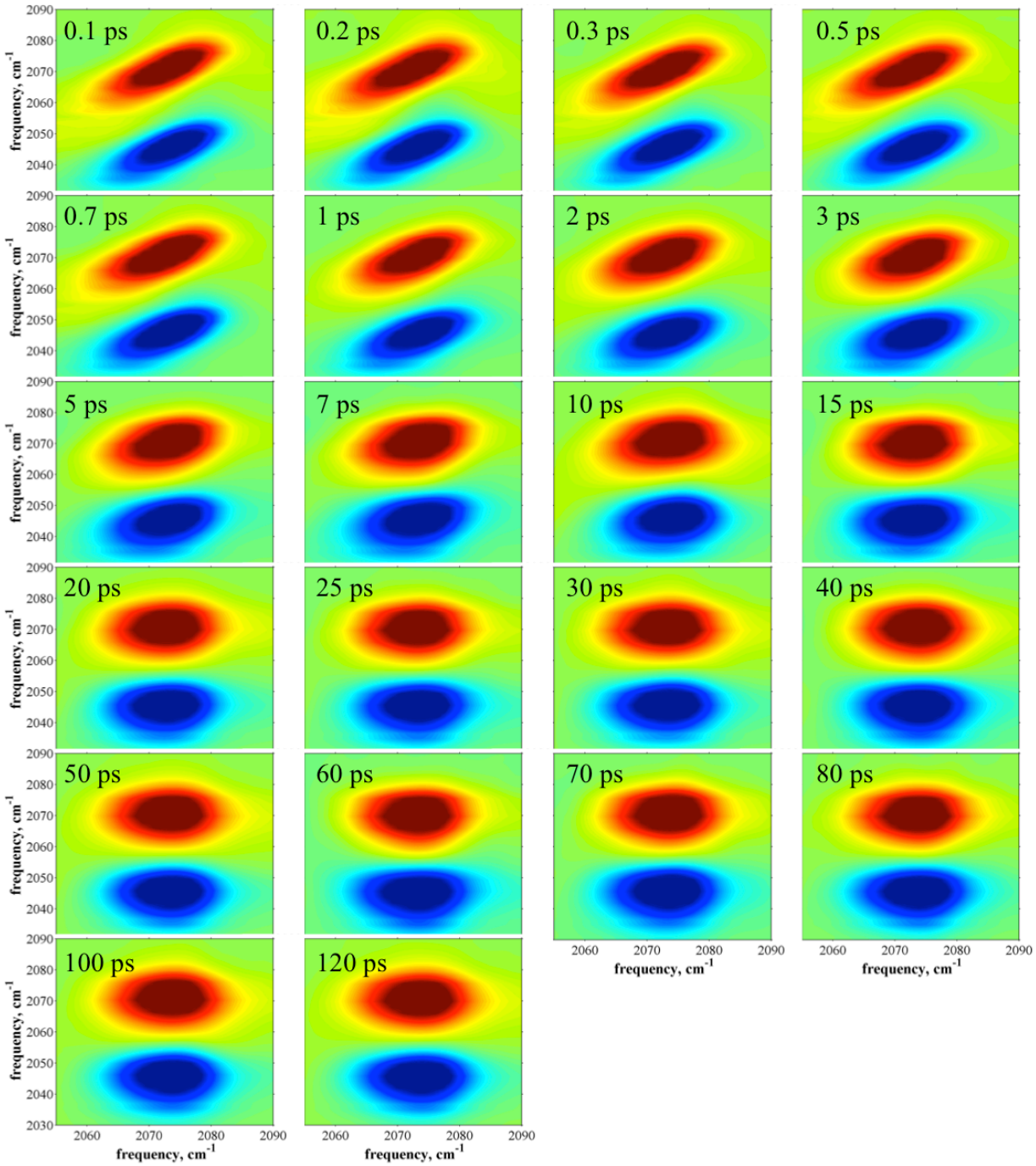

Figure S5. 2D-IR spectra for all T_w values for VC-I_2 in 0.7 mole fraction $CHCl_3$ in d_6-benzene.

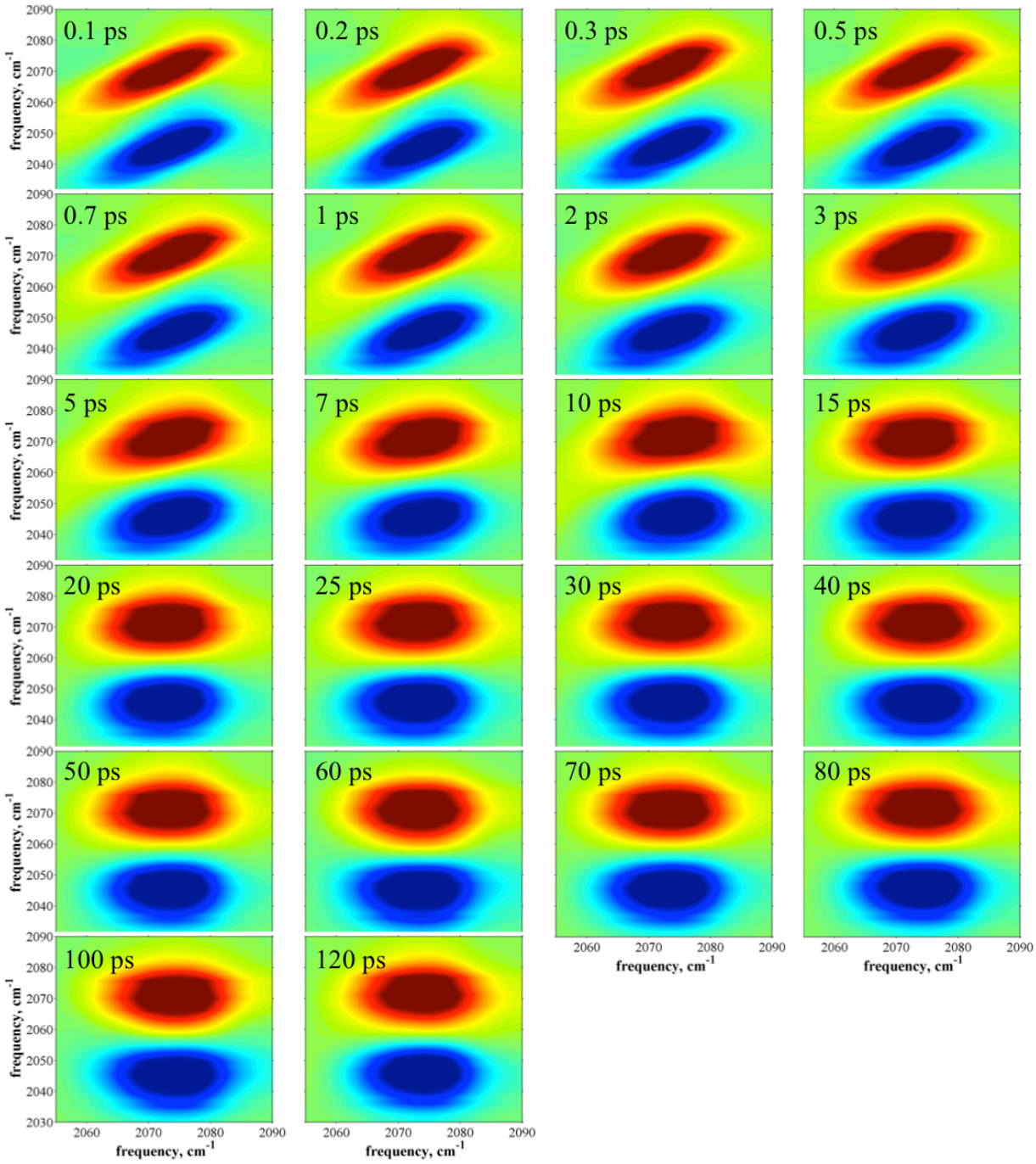

Figure S6. 2D-IR spectra for all T_w values for VC-I_2 in 0.9 mole fraction $CHCl_3$ in d_6-benzene.

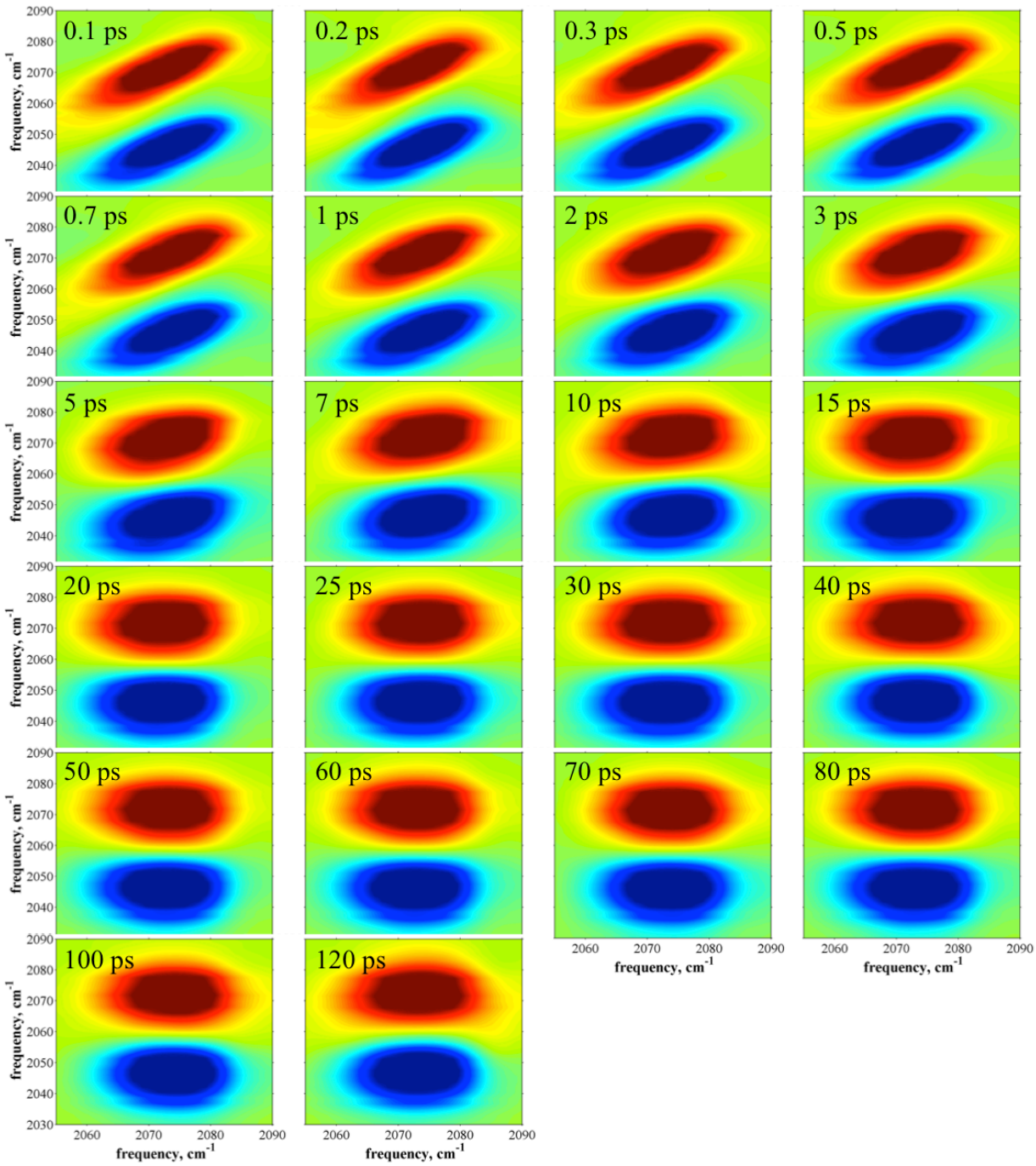

Figure S7. 2D-IR spectra for all T_w values for VC-I_2 in neat $CHCl_3$.

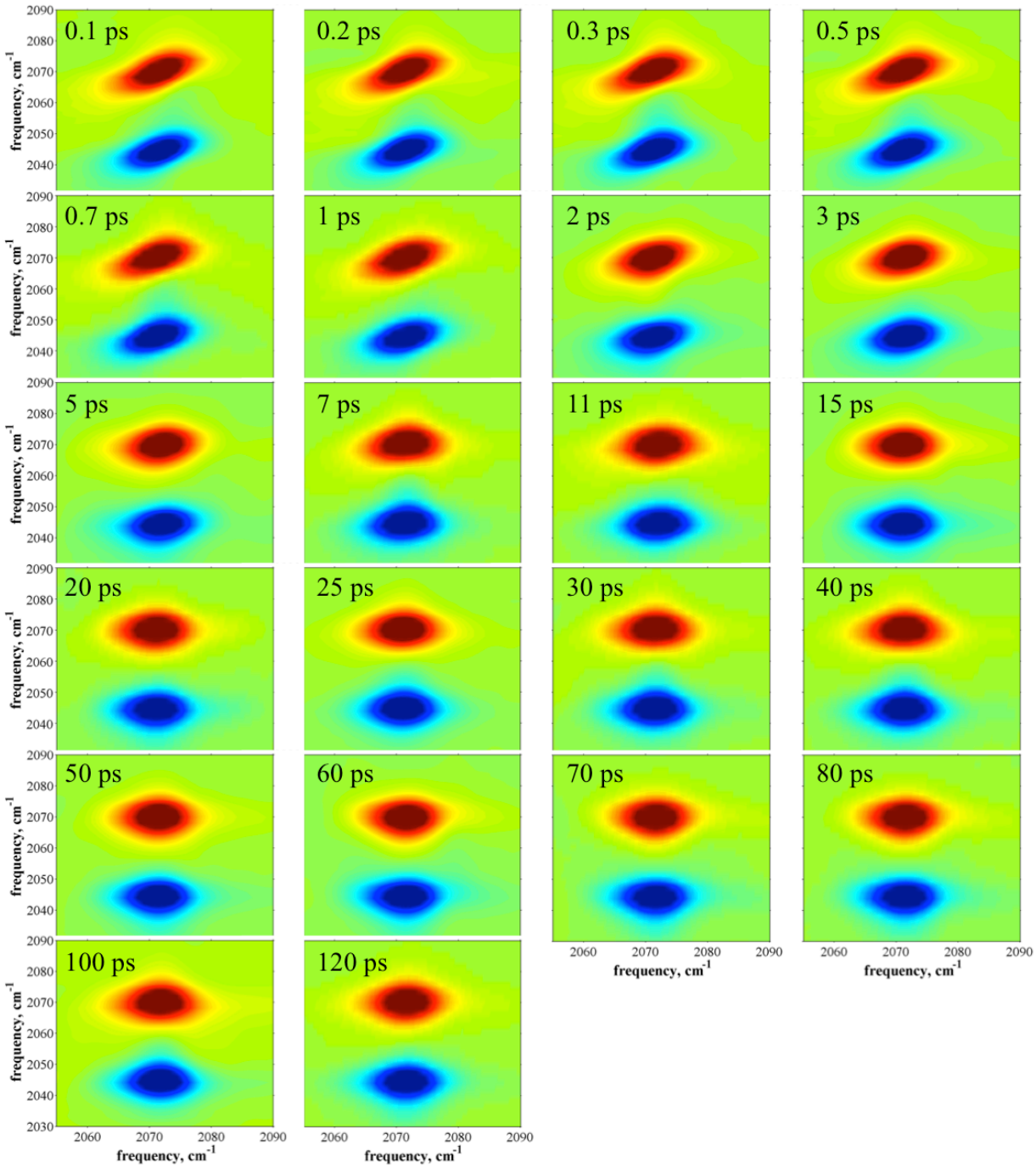

Figure S8. 2D-IR spectra for all T_w values for VC-I_2 in 0.05 mole fraction BA in d_6-benzene.

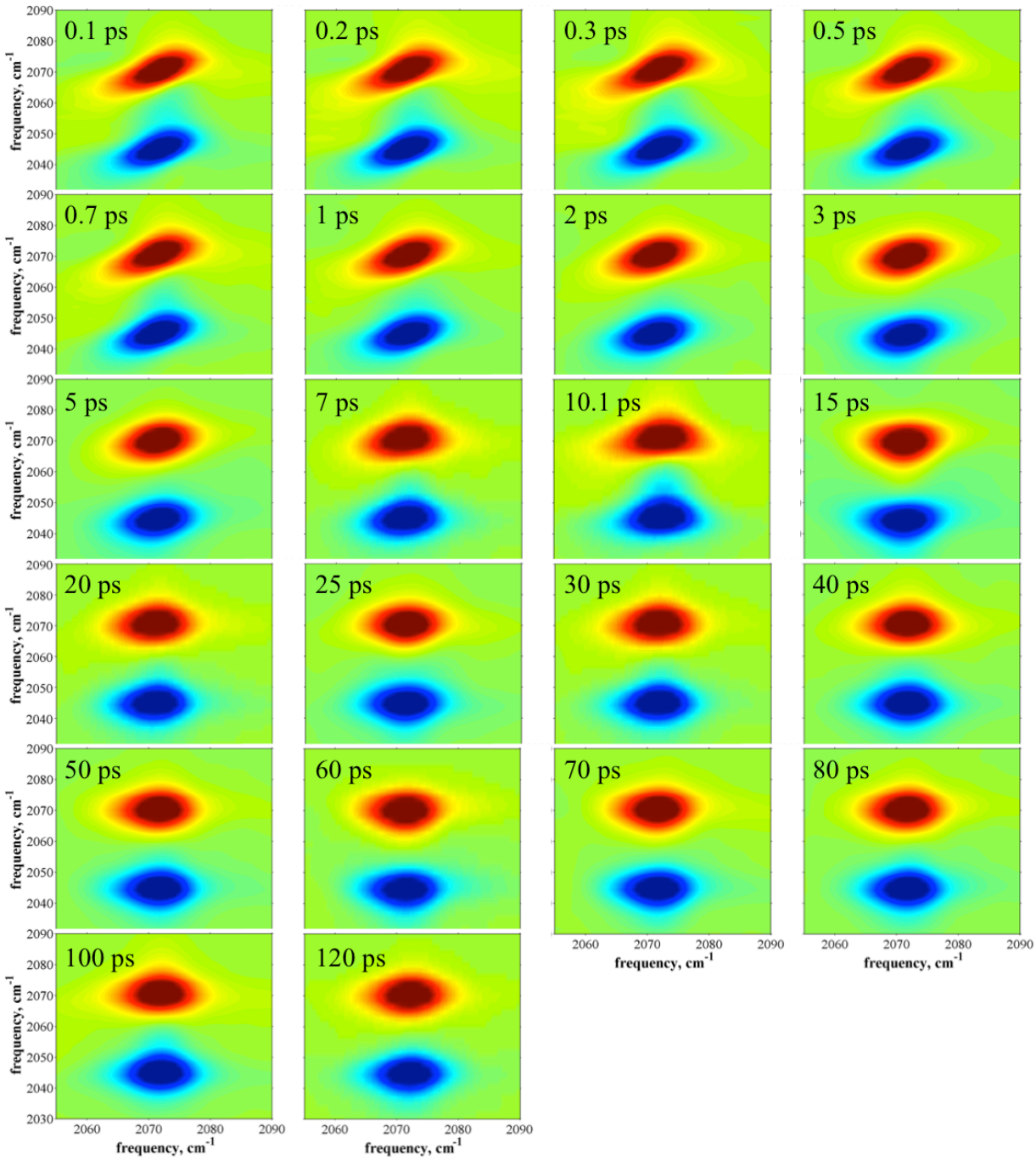

Figure S9. 2D-IR spectra for all T_w values for VC-I_2 in 0.1 mole fraction BA in d_6-benzene.

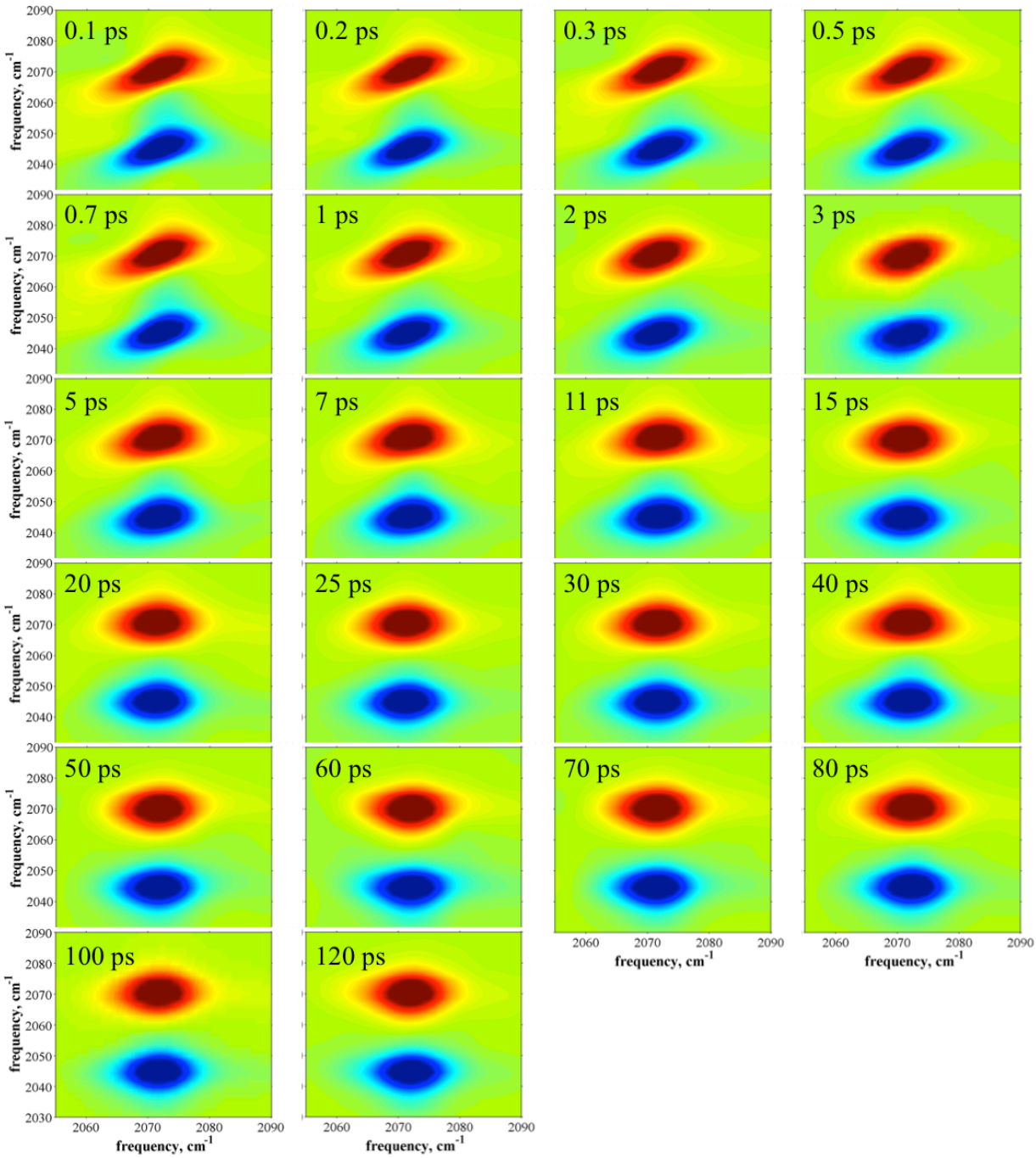

Figure S10. 2D-IR spectra for all T_w values for VC-I_2 in 0.2 mole fraction BA in d_6-benzene.

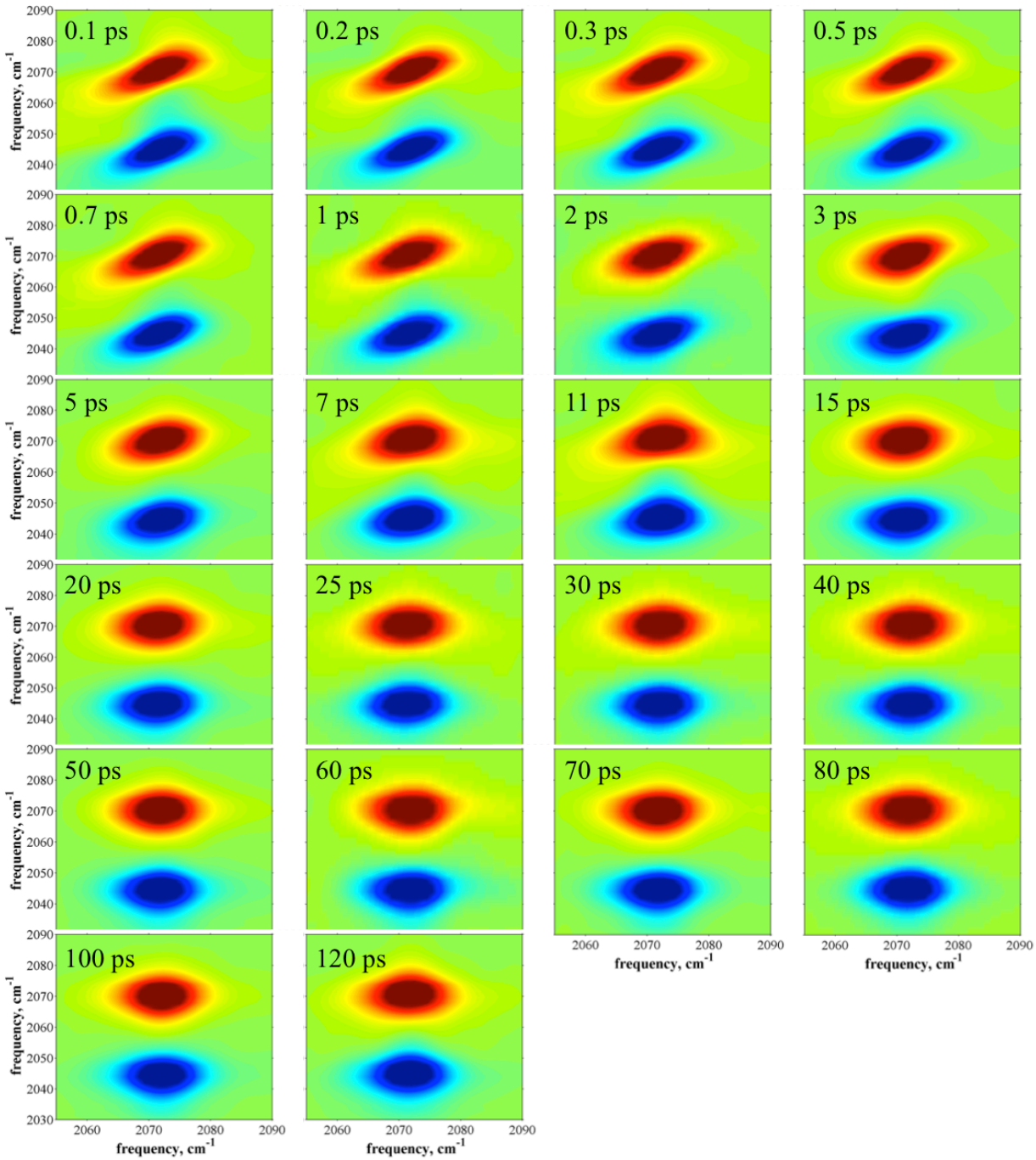

Figure S11. 2D-IR spectra for all T_w values for VC-I_2 in 0.3 mole fraction BA in d_6-benzene.

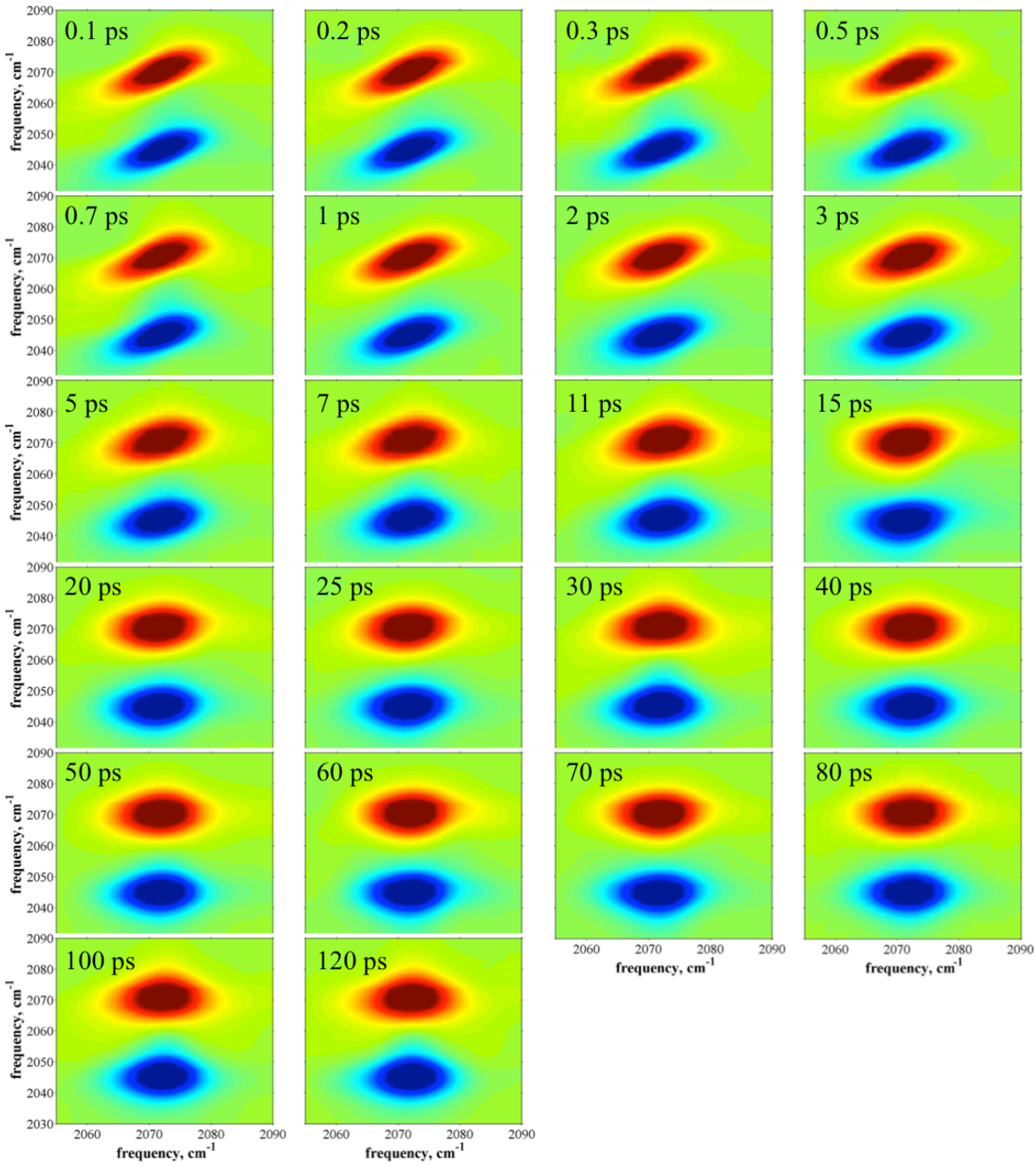

Figure S12. 2D-IR spectra for all T_w values for VC-I_2 in 0.4 mole fraction BA in d_6-benzene.

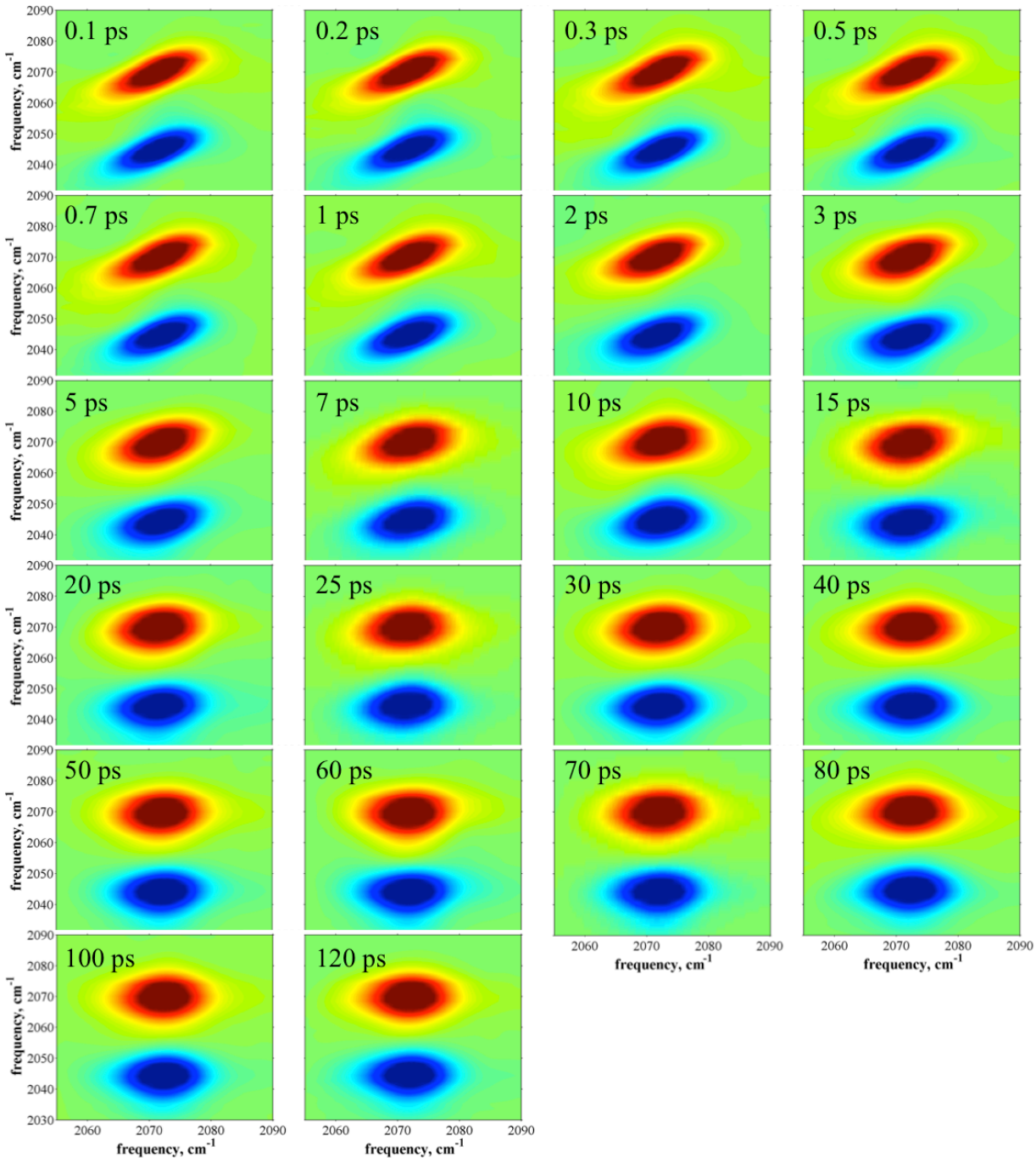

Figure S13. 2D-IR spectra for all T_w values for VC-I_2 in 0.5 mole fraction BA in d_6-benzene.

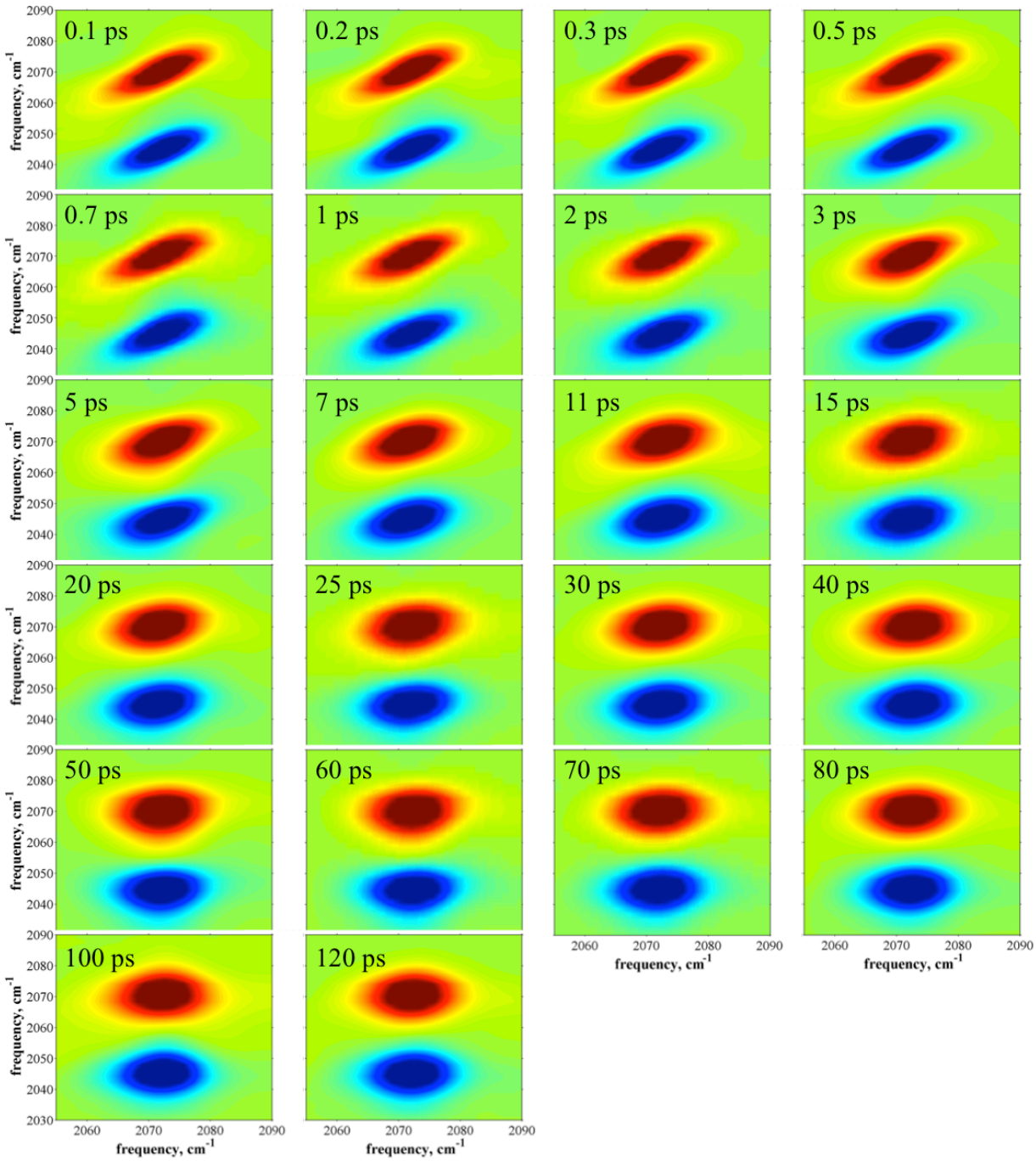

Figure S14. 2D-IR spectra for all T_w values for VC-I_2 in 0.7 mole fraction BA in d_6-benzene.

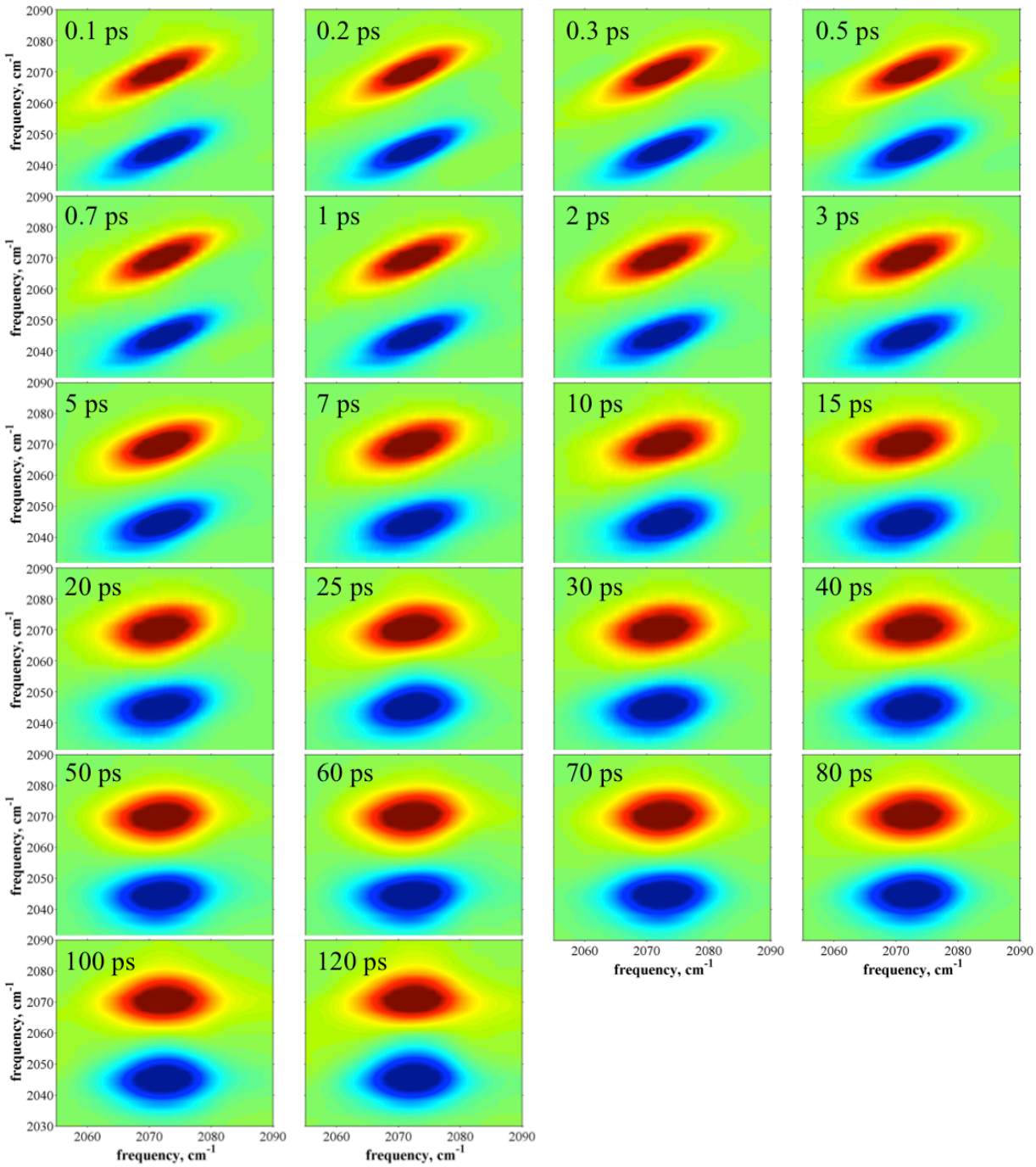

Figure S15. 2D-IR spectra for all T_w values for VC-I_2 in neat BA.

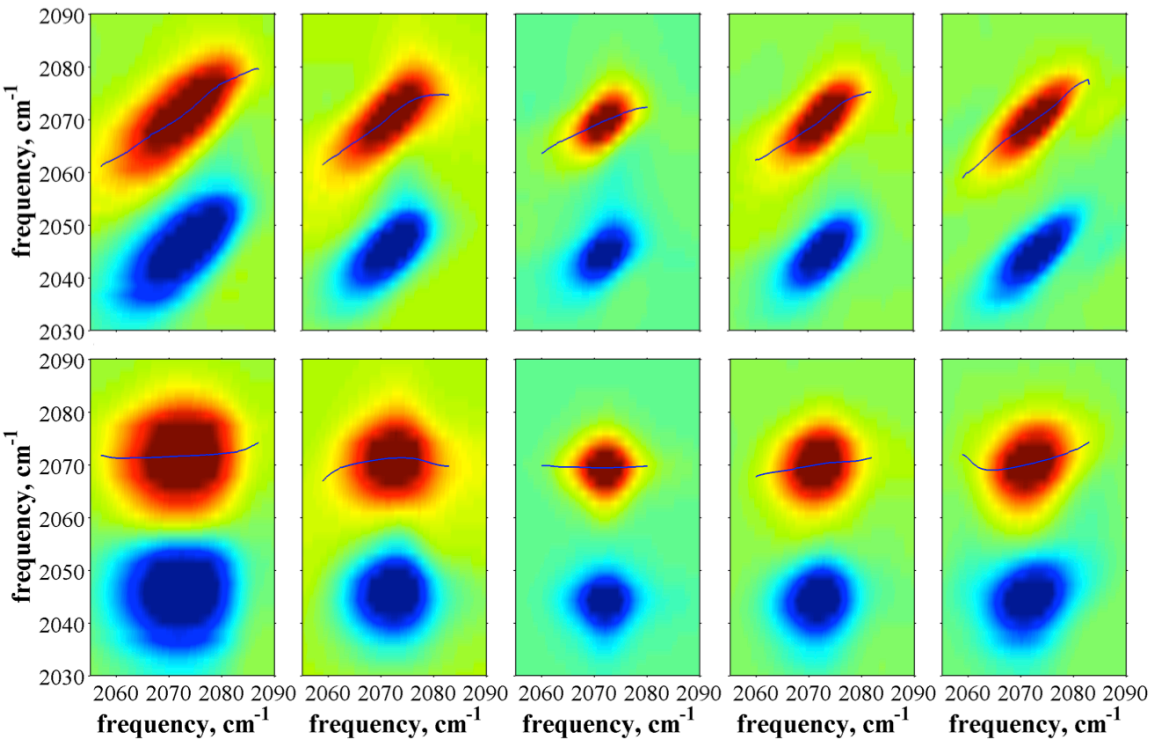

Figure S16. 2D-IR spectra collected at T_w = 0.3 ps (top row) and 30 ps (bottom row) for a) and b) neat $CHCl_3$, c) and d) 0.5 mole fraction $CHCl_3$ in d_6-benzene, e) and f) neat d_6-benzene, g and h) 0.5 mole fraction BA in d_6-benzene, and i and j) neat BA. Overlaid on the 0-1 transitions in each frame is the maximum of a slice taken parallel to ω_m axis. These maxima form the centerline whose slope will be analyzed in the CLS method. It can be seen that across the darkest portion of the linewidth the centerline slope is nearly constant.