

### HUBBARD's equations :

In the case of full anisotropic diffusion rotation, the reduced spectral densities can be expressed as :

$$\tilde{J}^{rr'}(\omega) = \sum_{k=-2}^2 a_k \frac{(2/\lambda_k)}{1 + \omega^2/\lambda_k^2}$$

where r and r' denote relaxation mechanisms (r=r': auto-correlation; r≠r': cross-correlation) and

$$\begin{aligned} \lambda_0 &= 4D_Z + 2D_+ \\ \lambda_{\pm 1} &= D_Z + 5D_+ \pm 3D_- \\ \lambda_{\pm 2} &= 2D_Z + 4D_+ \pm [4(D_Z - D_+)^2 + 12D_-^2]^{1/2} \\ a_0 &= (3/4)\sin^2 \theta_r \sin^2 \theta_{r'} \sin 2\varphi_r \sin 2\varphi_{r'} \\ a_1 &= (3/4)\sin 2\theta_r \sin 2\theta_{r'} \sin \varphi_r \sin \varphi_{r'} \\ a_{-1} &= (3/4)\sin 2\theta_r \sin 2\theta_{r'} \cos \varphi_r \cos \varphi_{r'} \\ a_{\pm 2} &= \frac{1}{12D_-^2 + d_{\pm}^2} \\ &\quad \cdot \left\{ (9D_-^2)\sin^2 \theta_r \cos 2\varphi_r \sin^2 \theta_{r'} \cos 2\varphi_{r'} - (3D_- d_{\pm}) [\sin^2 \theta_r \cos 2\varphi_r (1/2) \right. \\ &\quad \cdot (3\cos^2 \theta_{r'} - 1) + (1/2)(3\cos^2 \theta_r - 1) \\ &\quad \cdot \sin^2 \theta_{r'} \cos 2\varphi_{r'}] + (d_{\pm}^2)(1/2) \\ &\quad \left. (3\cos^2 \theta_r - 1)(1/2)(3\cos^2 \theta_{r'} - 1) \right\} \end{aligned}$$

with

$$d_{\pm} = 2(D_Z - D_+) \mp [4(D_Z - D_+)^2 + 12D_-^2]^{1/2}$$

Furthermore, we need three different diffusion coefficients:

$$D_X = \frac{1}{6\tau_X}; D_Y = \frac{1}{6\tau_Y}; D_Z = \frac{1}{6\tau_Z} \quad \text{and} \quad D_{\pm} = (D_X \pm D_Y)/2$$

Determination of Carbon-13 Chemical Shielding Tensor in the liquid state by combining NMR relaxation experiments and quantum chemical calculations

model	Carbon C3	liquid state				quantum chemistry			
		$\delta_{xx} \pm 5\text{ppm}$	$\delta_{yy} \pm 5\text{ppm}$	$\delta_{zz} \pm 5\text{ppm}$	$\delta$ iso (ppm)	$\delta_{xx}$	$\delta_{yy}$	$\delta_{zz}$	$\delta$ iso (ppm)
1	GIAO-HF/6-311+G	173.66	117.57	22.63	104.62	198.07	123.12	19.25	113.48
2	GIAO-HF/6-311+G**	174.55	115.68	23.63	104.62	188.41	114.94	19.82	107.72
3	GIAO-MP2/D95	173.67	118.93	21.27	104.62	148.94	102.20	28.47	93.20
4	GIAO-B3PW91/6-31G	174.79	115.16	23.92	104.62	157.57	112.01	21.63	97.07
5	GIAO-MP2/6-31G	176.40	113.51	23.95	104.62	149.10	102.89	25.06	92.35
6	GIAO-B3LYP/CC-PVDZ	173.39	118.13	22.35	104.62	162.16	108.81	22.18	97.72
7	CSGT-B3LYP/6-311+G**	174.18	116.79	23.19	104.62	166.38	113.50	22.75	100.88
8	GIAO-BPW91/6-311+G**	173.38	118.13	22.34	104.62	164.49	113.39	24.81	100.90
9	GIAO-BLYP/6-311+G**	172.79	119.31	21.75	104.62	165.26	111.89	25.53	100.89
10	IGAIM-B3LYP/6-311+G**	174.08	116.69	23.09	104.62	166.38	113.50	22.75	100.88
11	GIAO-B3LYP/TZVP	173.20	118.51	22.16	104.62	172.00	115.27	22.82	103.36
12	GIAO-B3PW91/6-311+G**	173.73	117.42	22.71	104.62	171.87	116.22	23.45	103.85
13	GIAO-B3LYP/6-311+G**	173.24	118.42	22.20	104.62	172.56	115.06	23.89	103.84
14	GIAO-B3PW91/CCPVTZ	173.90	117.04	22.90	104.62	170.37	115.71	23.65	103.24
15	GIAO-B3LYP/TZV	174.10	117.13	22.63	104.62	174.22	116.95	24.59	105.25
	min	172.79	113.51	21.27		148.94	102.20	19.25	92.35
	max	176.40	119.31	23.95		198.07	123.12	28.47	113.48

model	Carbon C5	liquid state				quantum chemistry			
		$\delta_{xx} \pm 5\text{ppm}$	$\delta_{yy} \pm 5\text{ppm}$	$\delta_{zz} \pm 5\text{ppm}$	$\delta$ iso (ppm)	$\delta_{xx}$	$\delta_{yy}$	$\delta_{zz}$	$\delta$ iso (ppm)
1	GIAO-HF/6-311+G	236.84	129.59	16.57	127.66	250.57	135.38	15.64	133.86
2	GIAO-HF/6-311+G**	236.58	130.39	16.02	127.66	245.37	130.35	16.57	130.76
3	GIAO-MP2/D95	235.51	133.67	13.83	127.66	208.44	113.49	20.79	114.24
4	GIAO-B3PW91/6-31G	236.37	131.12	15.50	127.66	209.04	125.30	19.12	117.82
5	GIAO-MP2/6-31G	235.93	132.44	14.631	127.66	202.20	113.90	21.58	112.56
6	GIAO-B3LYP/CC-PVDZ	235.85	132.63	14.50	127.66	217.04	124.91	21.51	121.16
7	CSGT-B3LYP/6-311+G**	235.73	132.92	14.34	127.66	226.32	129.03	19.96	125.10
8	GIAO-BPW91/6-311+G**	235.67	133.09	14.23	127.66	222.36	129.02	22.51	124.63
9	GIAO-BLYP/6-311+G**	235.54	133.46	13.99	127.66	224.66	126.89	23.91	125.15
10	IGAIM-B3LYP/6-311+G**	235.73	132.92	14.34	127.66	226.32	129.03	19.96	125.10
11	GIAO-B3LYP/TZVP	235.82	132.72	14.43	127.66	231.89	130.46	20.95	127.77
12	GIAO-B3PW91/6-311+G**	235.86	132.53	14.59	127.66	229.48	131.88	20.74	127.37
13	GIAO-B3LYP/6-311+G**	235.75	132.86	14.38	127.66	231.31	130.20	21.66	127.72
14	GIAO-B3PW91/CCPVTZ	235.90	132.51	14.58	127.66	228.36	132.23	20.88	127.16
15	GIAO-B3LYP/TZV	235.89	132.52	14.57	127.66	231.67	130.45	22.46	128.19
	min	235.54	129.59	13.83		202.20	113.49	15.64	112.56
	max	236.84	133.67	16.57		250.57	135.38	23.91	133.86

model	Carbon C6	liquid state				quantum chemistry			
		$\delta_{xx} \pm 5\text{ppm}$	$\delta_{yy} \pm 5\text{ppm}$	$\delta_{zz} \pm 5\text{ppm}$	$\delta$ iso (ppm)	$\delta_{xx}$	$\delta_{yy}$	$\delta_{zz}$	$\delta$ iso (ppm)
1	GIAO-HF/6-311+G	221.94	144.66	8.63	125.08	252.95	139.67	3.16	131.93
2	GIAO-HF/6-311+G**	221.77	144.82	8.64	125.08	245.85	134.38	3.88	128.04
3	GIAO-MP2/D95	220.46	145.96	8.82	125.08	207.98	116.92	11.44	112.12
4	GIAO-B3PW91/6-31G	221.05	145.37	8.82	125.08	210.44	129.33	4.83	114.87
5	GIAO-MP2/6-31G	220.64	145.86	8.74	125.08	202.54	118.84	6.47	109.28
6	GIAO-B3LYP/CC-PVDZ	220.71	145.62	8.91	125.08	218.05	128.28	7.76	118.03
7	CSGT-B3LYP/6-311+G**	221.15	145.65	8.93	125.08	230.21	131.54	5.20	122.32
8	GIAO-BPW91/6-311+G**	220.45	145.76	9.03	125.08	225.79	131.68	7.95	121.81
9	GIAO-BLYP/6-311+G**	220.38	145.79	8.79	125.08	228.29	129.49	9.00	122.26
10	IGAIM-B3LYP/6-311+G**	220.66	145.65	8.93	125.08	230.20	131.54	5.20	122.31
11	GIAO-B3LYP/TZVP	220.65	145.63	8.97	125.08	235.10	132.75	6.34	124.73
12	GIAO-B3PW91/6-311+G**	220.71	145.62	8.62	125.08	232.35	134.81	6.50	124.55
13	GIAO-B3LYP/6-311+G**	220.64	145.66	8.94	125.08	234.31	133.09	7.16	124.85
14	GIAO-B3PW91/CCPVTZ	221.21	145.59	8.95	125.08	231.25	135.19	6.24	124.23
15	GIAO-B3LYP/TZV	220.73	145.58	8.93	125.08	236.68	133.31	7.89	125.96
	min	220.38	144.66	8.62		202.54	116.92	3.16	109.28
	max	221.94	145.79	9.03		252.95	139.67	11.44	131.93

model	Carbon C1	quantum chemistry			
		$\delta_{xx}$	$\delta_{yy}$	$\delta_{zz}$	$\delta$ iso (ppm)
1	GIAO-HF/6-311+G	123.23	95.07	61.02	93.11
2	GIAO-HF/6-311+G**	116.49	90.67	60.96	89.37
3	GIAO-MP2/D95	145.91	103.50	63.98	104.46
4	GIAO-B3PW91/6-31G	133.56	93.15	60.95	95.89
5	GIAO-MP2/6-31G	136.50	93.11	58.61	96.07
6	GIAO-B3LYP/CC-PVDZ	132.61	95.83	62.01	96.81
7	CSGT-B3LYP/6-311+G**	130.73	103.47	67.60	100.60
8	GIAO-BPW91/6-311+G**	141.35	105.04	70.40	105.60
9	GIAO-BLYP/6-311+G**	141.79	106.25	71.17	106.40
10	IGAIM-B3LYP/6-311+G**	130.74	103.47	67.60	100.60
11	GIAO-B3LYP/TZVP	137.52	103.93	69.49	103.64
12	GIAO-B3PW91/6-311+G**	136.67	102.54	68.71	102.64
13	GIAO-B3LYP/6-311+G**	136.85	103.29	69.18	103.11
14	GIAO-B3PW91/CCPVTZ	137.73	104.17	68.36	103.42
15	GIAO-B3LYP/TZV	138.03	100.96	65.74	101.58

model	Carbon C2	quantum chemistry			
		$\delta_{xx}$	$\delta_{yy}$	$\delta_{zz}$	$\delta$ iso (ppm)
1	GIAO-HF/6-311+G	242.10	138.40	70.03	150.18
2	GIAO-HF/6-311++G**	238.10	136.75	71.45	148.77
3	GIAO-MP2/D95	186.77	134.42	83.00	134.73
4	GIAO-B3PW91/6-31G	197.37	133.68	76.03	135.69
5	GIAO-MP2/6-31G	190.33	132.40	77.23	133.32
6	GIAO-B3LYP/CC-PVDZ	205.22	139.88	79.80	141.64
7	CSGT-B3LYP/6-311++G**	215.86	144.17	80.53	146.85
8	GIAO-BPW91/6-311++G**	208.42	145.30	87.62	147.11
9	GIAO-BLYP/6-311++G**	210.79	146.89	86.96	148.21
10	IGAIM-B3LYP/6-311++G**	215.86	144.18	80.53	146.86
11	GIAO-B3LYP/TZVP	220.43	148.61	84.23	151.09
12	GIAO-B3PW91/6-311++G**	217.21	145.24	84.61	149.02
13	GIAO-B3LYP/6-311++G**	219.17	146.35	84.02	149.85
14	GIAO-B3PW91/CCPVTZ	217.20	146.48	84.38	149.35
15	GIAO-B3LYP/TZV	220.02	146.54	82.16	149.57

model	Carbon C4	quantum chemistry			
		$\delta_{xx}$	$\delta_{yy}$	$\delta_{zz}$	$\delta$ iso (ppm)
1	GIAO-HF/6-311+G	220.21	201.85	-6.68	138.46
2	GIAO-HF/6-311++G**	216.83	197.16	-8.40	135.20
3	GIAO-MP2/D95	183.62	170.98	-6.81	115.93
4	GIAO-B3PW91/6-31G	189.22	175.49	-4.10	120.20
5	GIAO-MP2/6-31G	183.18	171.00	-4.05	116.71
6	GIAO-B3LYP/CC-PVDZ	195.77	182.17	-1.59	125.45
7	CSGT-B3LYP/6-311++G**	204.04	189.57	-5.12	129.50
8	GIAO-BPW91/6-311++G**	200.63	186.75	-0.50	128.96
9	GIAO-BLYP/6-311++G**	203.33	189.09	-0.27	130.72
10	IGAIM-B3LYP/6-311++G**	204.04	189.57	-5.12	129.50
11	GIAO-B3LYP/TZVP	214.30	200.05	1.54	138.63
12	GIAO-B3PW91/6-311++G**	205.60	191.46	-2.56	131.50
13	GIAO-B3LYP/6-311++G**	207.77	193.28	-2.45	132.87
14	GIAO-B3PW91/CCPVTZ	205.92	191.87	-2.50	131.76
15	GIAO-B3LYP/TZV	208.31	193.96	-0.70	133.85

model	$\sigma$ TMS	
1	GIAO-HF/6-311+G	197.04
2	GIAO-HF/6-311++G**	190.58
3	GIAO-MP2/D95	203.54
4	GIAO-B3PW91/6-31G	192.37
5	GIAO-MP2/6-31G	205.55
6	GIAO-B3LYP/CC-PVDZ	188.40
7	CSGT-B3LYP/6-311++G**	177.47
8	GIAO-BPW91/6-311++G**	179.45
9	GIAO-BLYP/6-311++G**	175.28
10	IGAIM-B3LYP/6-311++G**	177.47
11	GIAO-B3LYP/TZVP	179.77
12	GIAO-B3PW91/6-311++G**	182.36
13	GIAO-B3LYP/6-311++G**	179.04
14	GIAO-B3PW91/CCPVTZ	182.14
15	GIAO-B3LYP/TZV	183.94