

Supplementary Information

Approaching the free rotor limit: Extremely low methyl torsional barrier observed in the microwave spectrum of 2,4-dimethylfluorobenzene

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Table S-1: Nuclear coordinates in the inertial principal axis system for 24DMFB calculated at the MP2/6-31G(d,p), MP2/6-311++G(d,p) and B3LYP-D3BJ/6-311++G(d,p) levels of theory. The atoms are numbered according to Figure 1.

	MP2/6-31G(d,p)			MP2/6-311++G(d,p)			B3LYP-D3BJ/6-311++G(d,p)		
	$a / \text{Å}$	$b / \text{Å}$	$c / \text{Å}$	$a / \text{Å}$	$b / \text{Å}$	$c / \text{Å}$	$a / \text{Å}$	$b / \text{Å}$	$c / \text{Å}$
C1	-1.091619	-0.628612	0.000027	-1.096500	-0.631568	-0.000035	-1.091250	-0.629009	0.000018
C2	-0.771234	0.729658	0.000016	-0.777654	0.729623	0.000096	-0.773706	0.727524	-0.000005
C3	0.586871	1.056668	-0.000063	0.584872	1.056407	-0.000217	0.583444	1.051135	-0.000051
C4	1.593234	0.080372	-0.000091	1.595743	0.080524	0.000068	1.589622	0.078124	-0.000066
C5	1.213607	-1.265283	-0.000065	1.215617	-1.268300	-0.000191	1.210594	-1.264464	-0.000061
C6	-0.134445	-1.630962	0.000009	-0.136321	-1.634647	0.000080	-0.136174	-1.626682	-0.000013
C7	-1.863904	1.760590	-0.000004	-1.865088	1.771408	0.000003	-1.855623	1.772807	-0.000009
C8	3.044420	0.484241	0.000086	3.049769	0.487213	0.000115	3.043051	0.482334	0.000075
F9	-2.408442	-0.974162	0.000038	-2.406196	-0.981016	0.000059	-2.406108	-0.983031	0.000055
H10	-2.501595	1.654757	0.877392	-2.505279	1.668577	0.881431	-2.499327	1.675536	0.878454
H11	-2.501481	1.654846	-0.877495	-2.504889	1.668866	-0.881743	-2.499321	1.675537	-0.878477
H12	-1.442291	2.764004	0.000073	-1.431384	2.774402	0.000267	-1.422714	2.774134	-0.000007
H13	3.287972	1.079436	0.880823	3.290835	1.083595	0.886104	3.287934	1.081304	0.882321
H14	3.287098	1.083247	-0.878295	3.289171	1.089412	-0.882360	3.287233	1.084557	-0.880131
H15	3.693098	-0.390488	-0.002119	3.701484	-0.390691	-0.003356	3.696356	-0.392022	-0.001769
H16	0.865638	2.105793	-0.000096	0.863316	2.109806	-0.000178	0.862004	2.100719	-0.000089
H17	1.973090	-2.038030	-0.000093	1.977625	-2.044121	-0.000146	1.969233	-2.038997	-0.000105
H18	-0.447126	-2.666130	-0.000018	-0.447741	-2.67466	-0.000059	-0.446170	-2.664100	-0.000021

Table S-2: Rotation constants of 24DMFB (in MHz) calculated at different levels of theory. The difference between the calculated and the experimental (in MHz) are given as ΔA , ΔB and ΔC .

Method/Basis set	<i>A</i>	ΔA	<i>B</i>	ΔB	<i>C</i>	ΔC
B3LYP-D3/6-31G(d,p)	3007.7	9.7	1276.9	-6.9	906.4	-2.7
B3LYP-D3/6-31+G(d,p)	2987.5	-10.5	1275.2	-8.6	903.7	-5.4
B3LYP-D3/6-31++G(d,p)	2987.4	-10.6	1275.2	-8.6	903.7	-5.4
B3LYP-D3/6-311G(d,p)	3011.8	13.8	1280.1	-3.7	908.4	-0.7
B3LYP-D3/6-311+G(d,p)	3001.9	3.9	1280.3	-3.5	907.6	-1.5
B3LYP-D3/6-311++G(d,p)	3001.9	3.9	1280.3	-3.5	907.6	-1.5
B3LYP-D3/6-311G(2d,2p)	3024.1	26.1	1284.5	0.7	911.7	2.6
B3LYP-D3/6-311+G(2d,2p)	3013.4	15.4	1284.4	0.6	910.6	1.5
B3LYP-D3/6-311++G(2d,2p)	3013.4	15.4	1284.4	0.6	910.6	1.5
B3LYP-D3/6-311G(df,pd)	3022.7	24.7	1284.7	0.9	911.7	2.6
B3LYP-D3/6-311+G(df,pd)	3012.6	14.6	1284.9	1.2	910.9	1.8
B3LYP-D3/6-311++G(df,pd)	3012.6	14.6	1284.9	1.2	910.9	1.8
B3LYP-D3/6-311G(2df,2pd)	3029.6	31.6	1286.4	2.6	913.1	4.0
B3LYP-D3/6-311+G(2df,2pd)	3018.4	20.4	1286.3	2.5	912.0	2.9
B3LYP-D3/6-311++G(2df,2pd)	3018.3	20.3	1286.3	2.5	912.0	2.9
B3LYP-D3/6-311G(3df,3pd)	3026.8	28.8	1287.3	3.5	913.3	4.2
B3LYP-D3/6-311+G(3df,3pd)	3017.7	19.7	1287.1	3.3	912.4	3.3
B3LYP-D3/6-311++G(3df,3pd)	3017.8	19.8	1287.0	3.3	912.4	3.3
B3LYP-D3/cc-pVDZ	3000.0	2.1	1273.1	-10.6	903.9	-5.2
B3LYP-D3/aug-cc-pVDZ	2984.5	-13.5	1274.1	-9.7	903.0	-6.1
B3LYP-D3/cc-pVTZ	3020.8	22.8	1286.3	2.5	912.3	3.2
B3LYP-D3/aug-cc-pVTZ	3016.5	18.5	1286.4	2.6	911.9	2.8
B3LYP-D3BJ/6-31G(d,p)	3007.7	9.7	1278.9	-4.9	907.4	-1.7
B3LYP-D3BJ/6-31+G(d,p)	2987.4	-10.5	1277.2	-6.6	904.7	-4.3
B3LYP-D3BJ/6-31++G(d,p)	2987.4	-10.6	1277.3	-6.5	904.7	-4.3
B3LYP-D3BJ/6-311G(d,p)	3011.6	13.7	1282.2	-1.6	909.4	0.3
B3LYP-D3BJ/6-311+G(d,p)	3001.7	3.8	1282.4	-1.4	908.6	-0.5
B3LYP-D3BJ/6-311++G(d,p)	3001.7	3.7	1282.4	-1.4	908.6	-0.5
B3LYP-D3BJ/6-311G(2d,2p)	3023.9	25.9	1286.6	2.8	912.7	3.6
B3LYP-D3BJ/6-311+G(2d,2p)	3013.2	15.2	1286.5	2.7	911.6	2.5
B3LYP-D3BJ/6-311++G(2d,2p)	3013.1	15.1	1286.5	2.7	911.6	2.5
B3LYP-D3BJ/6-311G(df,pd)	3022.5	24.5	1286.8	3.0	912.7	3.6
B3LYP-D3BJ/6-311+G(df,pd)	3012.3	14.3	1287.1	3.3	911.9	2.8
B3LYP-D3BJ/6-311++G(df,pd)	3012.3	14.3	1287.1	3.3	911.9	2.8
B3LYP-D3BJ/6-311G(2df,2pd)	3029.4	31.5	1288.5	4.7	914.1	5.1
B3LYP-D3BJ/6-311+G(2df,2pd)	3018.1	20.1	1288.4	4.6	913.1	4.0

B3LYP-D3BJ/6-311++G(2df,2pd)	3018.0	20.0	1288.4	4.6	913.1	4.0
B3LYP-D3BJ/6-311G(3df,3pd)	3026.6	28.6	1289.4	5.6	914.3	5.2
B3LYP-D3BJ/6-311+G(3df,3pd)	3017.8	19.8	1289.1	5.3	913.4	4.3
B3LYP-D3BJ/6-311++G(3df,3pd)	3017.5	19.5	1289.1	5.3	913.4	4.3
B3LYP-D3BJ/cc-pVDZ	3000.1	2.1	1275.2	-8.6	905.0	-4.1
B3LYP-D3BJ/aug-cc-pVDZ	2984.5	-13.5	1276.2	-7.6	904.0	-5.1
B3LYP-D3BJ/cc-pVTZ	3020.6	22.6	1288.4	4.6	913.3	4.2
B3LYP-D3BJ/aug-cc-pVTZ	3016.2	18.2	1288.4	4.6	912.9	3.8
CAM-B3LYP-D3BJ/6-311G(d,p)	3037.1	39.1	1291.1	7.3	916.2	7.1
CAM-B3LYP-D3BJ/6-311+G(d,p)	3027.2	29.2	1291.3	7.6	915.4	6.3
CAM-B3LYP-D3BJ/6-311++G(d,p)	3027.2	29.2	1291.3	7.5	915.4	6.3
CAM-B3LYP-D3BJ/cc-pVDZ	3025.5	27.5	1284.0	0.2	911.7	2.6
CAM-B3LYP-D3BJ/aug-cc-pVDZ	3008.9	11.0	1285.0	1.2	910.7	1.6
CAM-B3LYP-D3BJ/cc-pVTZ	3045.4	47.4	1297.4	13.6	920.1	11.0
CAM-B3LYP-D3BJ/aug-cc-pVTZ	3041.1	43.1	1297.4	13.6	919.7	10.6
CCSD/cc-pVDZ	2975.0	-22.9	1264.8	-19.0	897.6	-11.5
M06-2X/6-31G(d,p)	3029.7	31.7	1285.3	1.5	912.6	3.5
M06-2X/6-31+G(d,p)	3013.6	15.6	1283.9	0.1	910.5	1.4
M06-2X/6-31++G(d,p)	3013.5	15.5	1283.9	0.1	910.5	1.4
M06-2X/6-311G(d,p)	3033.3	35.3	1287.8	4.1	914.2	5.1
M06-2X/6-311+G(d,p)	3025.9	27.9	1288.1	4.3	913.7	4.6
M06-2X/6-311++G(d,p)	3025.9	27.9	1288.1	4.3	913.7	4.6
M06-2X/6-311G(2d,2p)	3045.3	47.4	1291.7	7.9	917.2	8.1
M06-2X/6-311+G(2d,2p)	3036.9	38.9	1291.6	7.8	916.4	7.3
M06-2X/6-311++G(2d,2p)	3036.8	38.8	1291.6	7.8	916.4	7.3
M06-2X/6-311G(df,pd)	3042.4	44.4	1291.4	7.6	916.8	7.7
M06-2X/6-311+G(df,pd)	3034.4	36.5	1291.8	8.0	916.3	7.2
M06-2X/6-311++G(df,pd)	3034.5	36.5	1291.8	8.0	916.3	7.2
M06-2X/6-311G(2df,2pd)	3050.2	52.2	1293.1	9.3	918.3	9.3
M06-2X/6-311+G(2df,2pd)	3040.9	42.9	1293.2	9.4	917.5	8.5
M06-2X/6-311++G(2df,2pd)	3040.8	42.8	1293.2	9.4	917.5	8.5
M06-2X/6-311G(3df,3pd)	3047.7	49.7	1293.7	9.9	918.4	9.3
M06-2X/6-311+G(3df,3pd)	3040.5	42.6	1293.5	9.8	917.7	8.6
M06-2X/6-311++G(3df,3pd)	3040.6	42.7	1293.5	9.7	917.7	8.6
M06-2X/cc-pVDZ	3024.6	26.6	1282.3	-1.5	910.8	1.7
M06-2X/aug-cc-pVDZ	3011.2	13.3	1283.3	-0.5	910.0	0.9
M06-2X/cc-pVTZ	3041.4	43.4	1293.2	9.5	917.6	8.5
M06-2X/aug-cc-pVTZ	3038.6	40.6	1293.3	9.5	917.4	8.3

MN15/6-31G(d,p)	3024.6	26.6	1281.4	-2.4	910.2	1.1
MN15/6-31+G(d,p)	3005.6	7.7	1279.9	-3.9	907.8	-1.3
MN15/6-31++G(d,p)	3005.5	7.5	1279.9	-3.9	907.8	-1.3
MN15/6-311G(d,p)	3032.0	34.0	1285.8	2.0	913.1	4.0
MN15/6-311+G(d,p)	3024.3	26.3	1286.0	2.2	912.5	3.4
MN15/6-311++G(d,p)	3024.3	26.3	1286.0	2.2	912.5	3.4
MN15/6-311G(2d,2p)	3043.5	45.6	1289.3	5.5	915.8	6.8
MN15/6-311+G(2d,2p)	3034.4	36.4	1289.3	5.5	915.0	5.9
MN15/6-311++G(2d,2p)	3034.4	36.4	1289.3	5.5	915.0	5.9
MN15/6-311G(df,pd)	3044.5	46.5	1290.8	7.0	916.7	7.6
MN15/6-311+G(df,pd)	3035.9	37.9	1291.1	7.3	916.1	7.0
MN15/6-311++G(df,pd)	3035.8	37.9	1291.1	7.3	916.0	7.0
MN15/6-311G(2df,2pd)	3049.7	51.7	1291.2	7.4	917.3	8.2
MN15/6-311+G(2df,2pd)	3039.6	41.7	1291.3	7.5	916.5	7.4
MN15/6-311++G(2df,2pd)	3039.6	41.6	1291.4	7.6	916.5	7.4
MN15/6-311G(3df,3pd)	3049.6	51.6	1292.9	9.1	918.2	9.1
MN15/6-311+G(3df,3pd)	3041.5	43.5	1292.7	8.9	917.3	8.3
MN15/6-311++G(3df,3pd)	3041.8	43.8	1292.7	8.9	917.4	8.3
MN15/cc-pVDZ	3021.1	23.1	1278.3	-5.5	908.4	-0.7
MN15/aug-cc-pVDZ	3006.0	8.0	1280.3	-3.5	908.0	-1.1
MN15/cc-pVTZ	3041.1	43.1	1292.3	8.5	917.1	8.0
MN15/aug-cc-pVTZ	3039.5	41.6	1292.7	8.9	917.2	8.1
PBE0/6-31G(d,p)	2978.6	-19.4	1266.9	-16.9	898.9	-10.2
PBE0/6-31+G(d,p)	2957.1	-40.9	1265.5	-18.3	896.2	-12.9
PBE0/6-31++G(d,p)	2957.0	-41.0	1265.5	-18.3	896.2	-12.9
PBE0/6-311G(d,p)	2983.0	-14.9	1270.8	-13.0	901.2	-7.9
PBE0/6-311+G(d,p)	2972.2	-25.8	1270.9	-12.9	900.2	-8.8
PBE0/6-311++G(d,p)	2972.2	-25.8	1270.8	-12.9	900.2	-8.9
PBE0/6-311G(2d,2p)	2993.9	-4.1	1274.4	-9.4	903.9	-5.2
PBE0/6-311+G(2d,2p)	2981.8	-16.2	1274.2	-9.5	902.8	-6.3
PBE0/6-311++G(2d,2p)	2981.7	-16.2	1274.3	-9.5	902.8	-6.3
PBE0/6-311G(df,pd)	2993.3	-4.6	1274.7	-9.0	904.1	-5.0
PBE0/6-311+G(df,pd)	2981.4	-16.5	1275.2	-8.6	903.3	-5.8
PBE0/6-311++G(df,pd)	2981.5	-16.5	1275.2	-8.6	903.2	-5.8
PBE0/6-311G(2df,2pd)	2999.7	1.7	1276.1	-7.7	905.3	-3.8
PBE0/6-311+G(2df,2pd)	2986.3	-11.7	1276.2	-7.6	904.2	-4.9
PBE0/6-311++G(2df,2pd)	2986.2	-11.8	1276.2	-7.6	904.2	-4.9
PBE0/6-311G(3df,3pd)	2996.6	-1.3	1277.3	-6.5	905.6	-3.5
PBE0/6-311+G(3df,3pd)	2986.3	-11.7	1277.0	-6.8	904.6	-4.5
PBE0/6-311++G(3df,3pd)	2986.4	-11.6	1277.0	-6.8	904.6	-4.5

PBE0/cc-pVDZ	2973.3	-24.6	1264.0	-19.8	897.0	-12.1
PBE0/aug-cc-pVDZ	2955.2	-42.8	1265.0	-18.8	895.8	-13.3
PBE0/cc-pVTZ	2990.0	-8.0	1276.3	-7.5	904.5	-4.6
PBE0/aug-cc-pVTZ	2984.8	-13.2	1276.2	-7.6	904.0	-5.1
PBE0-D3/6-31G(d,p)	2980.8	-17.1	1267.0	-16.8	899.1	-10.0
PBE0-D3/6-31+G(d,p)	2959.5	-38.4	1265.5	-18.3	896.4	-12.7
PBE0-D3/6-31++G(d,p)	2959.5	-38.5	1265.5	-18.2	896.4	-12.7
PBE0-D3/6-311G(d,p)	2985.3	-12.6	1270.8	-13.0	901.4	-7.7
PBE0-D3/6-311+G(d,p)	2974.6	-23.3	1270.9	-12.9	900.5	-8.6
PBE0-D3/6-311++G(d,p)	2974.6	-23.4	1270.9	-12.9	900.5	-8.6
PBE0-D3/6-311G(2d,2p)	2996.1	-1.9	1274.4	-9.4	904.2	-4.9
PBE0-D3/6-311+G(2d,2p)	2984.2	-13.7	1274.3	-9.5	903.0	-6.1
PBE0-D3/6-311++G(2d,2p)	2984.2	-13.8	1274.3	-9.5	903.0	-6.1
PBE0-D3/6-311G(df,pd)	2995.6	-2.4	1274.8	-9.0	904.3	-4.7
PBE0-D3/6-311+G(df,pd)	2983.9	-14.1	1275.2	-8.5	903.5	-5.6
PBE0-D3/6-311++G(df,pd)	2983.9	-14.1	1275.2	-8.6	903.5	-5.6
PBE0-D3/6-311G(2df,2pd)	3002.0	4.0	1276.1	-7.7	905.5	-3.6
PBE0-D3/6-311+G(2df,2pd)	2988.8	-9.2	1276.2	-7.5	904.4	-4.7
PBE0-D3/6-311++G(2df,2pd)	2988.6	-9.3	1276.3	-7.5	904.4	-4.7
PBE0-D3/6-311G(3df,3pd)	2998.9	0.9	1277.3	-6.5	905.9	-3.2
PBE0-D3/6-311+G(3df,3pd)	2988.7	-9.2	1277.0	-6.8	904.8	-4.3
PBE0-D3/6-311++G(3df,3pd)	2988.8	-9.1	1277.0	-6.8	904.8	-4.3
PBE0-D3/cc-pVDZ	2975.5	-22.5	1264.1	-19.7	897.2	-11.8
PBE0-D3/aug-cc-pVDZ	2957.7	-40.3	1265.0	-18.8	896.0	-13.0
PBE0-D3/cc-pVTZ	2992.3	-5.7	1276.3	-7.5	904.8	-4.3
PBE0-D3/aug-cc-pVTZ	2987.2	-10.8	1276.2	-7.6	904.2	-4.9
ω B97X-D/6-31G(d,p)	3015.9	17.9	1285.8	2.0	911.6	2.6
ω B97X-D/6-31+G(d,p)	3000.9	2.9	1284.4	0.7	909.6	0.5
ω B97X-D/6-31++G(d,p)	3000.9	2.9	1284.5	0.7	909.6	0.5
ω B97XD/6-311G(d,p)	3013.9	16.0	1289.3	5.5	913.2	4.1
ω B97XD/6-311+G(d,p)	3013.9	16.0	1289.3	5.5	913.2	4.1
ω B97XD/6-311++G(d,p)	3021.3	23.3	1289.1	5.3	913.8	4.7
ω B97XD/6-311G(2d,2p)	3032.6	34.7	1293.1	9.3	916.8	7.7
ω B97X-D/6-311+G(2d,2p)	3024.4	26.5	1293.0	9.2	916.0	6.9
ω B97X-D/6-311++G(2d,2p)	3024.4	26.4	1293.0	9.2	916.0	6.9
ω B97X-D/6-311G(df,pd)	3030.9	32.9	1293.3	9.5	916.7	7.6
ω B97X-D/6-311+G(df,pd)	3022.9	24.9	1293.6	9.8	916.2	7.1
ω B97X-D/6-311++G(df,pd)	3023.0	25.0	1293.6	9.8	916.2	7.1
ω B97X-D/6-311G(2df,2pd)	3037.8	39.8	1294.9	11.1	918.1	9.0

ω B97X-D/6-311+G(2df,2pd)	3028.6	30.6	1294.9	11.1	917.3	8.2
ω B97X-D/6-311++G(2df,2pd)	3028.5	30.6	1294.9	11.1	917.3	8.2
ω B97X-D/6-311G(3df,3pd)	3035.3	37.3	1295.7	12.0	918.3	9.3
ω B97X-D/6-311+G(3df,3pd)	3028.4	30.4	1295.6	11.8	917.6	8.5
ω B97X-D/6-311++G(3df,3pd)	3028.4	30.5	1295.5	11.8	917.6	8.5
ω B97X-D/cc-pVDZ	3008.8	10.8	1281.9	-1.9	909.2	0.1
ω B97X-D/aug-cc-pVDZ	2996.1	-1.9	1283.0	-0.8	908.5	-0.6
ω B97X-D/cc-pVTZ	3030.5	32.5	1295.2	11.4	917.6	8.5
ω B97X-D/aug-cc-pVTZ	3027.8	29.9	1295.3	11.6	917.5	8.4
MP2/6-31G(d,p)	3013.0	15.0	1279.2	-4.6	908.0	-1.1
MP2/6-31+G(d,p)	2991.5	-6.5	1276.6	-7.2	904.8	-4.3
MP2/6-31++G(d,p)	2991.8	-6.2	1276.2	-7.6	904.6	-4.5
MP2/6-311G(d,p)	2999.7	1.7	1277.3	-6.5	905.9	-3.2
MP2/6-311+G(d,p)	2989.8	-8.2	1276.8	-6.9	904.8	-4.3
MP2/6-311++G(d,p)	2989.7	-8.3	1276.7	-7.1	904.7	-4.4
MP2/6-311G(2d,2p)	3021.0	23.1	1282.2	-1.5	910.2	1.1
MP2/6-311+G(2d,2p)	3009.6	11.7	1282.1	-1.7	909.1	0.0
MP2/6-311++G(2d,2p)	3009.6	11.6	1282.1	-1.7	909.1	0.0
MP2/6-311G(df,pd)	3021.7	23.7	1286.8	3.0	912.7	3.6
MP2/6-311+G(df,pd)	3010.8	12.8	1286.3	2.5	911.4	2.3
MP2/6-311++G(df,pd)	3010.7	12.7	1286.2	2.5	911.4	2.3
MP2/6-311G(2df,2pd)	3037.3	39.3	1288.8	5.0	915.0	5.9
MP2/6-311+G(2df,2pd)	3025.0	27.0	1288.4	4.6	913.7	4.6
MP2/6-311++G(2df,2pd)	3024.9	26.9	1288.4	4.6	913.7	4.6
MP2/6-311G(3df,3pd)	3029.1	31.1	1288.0	4.2	913.9	4.8
MP2/6-311+G(3df,3pd)	3019.9	21.9	1287.4	3.6	912.7	3.6
MP2/6-311++G(3df,3pd)	3020.1	22.1	1287.4	3.6	912.7	3.7
MP2/cc-pVDZ	2980.0	-18.0	1265.8	-18.0	898.5	-10.6
MP2/aug-cc-pVDZ	2959.3	-38.7	1263.6	-20.2	895.5	-13.6
MP2/cc-pVTZ	3021.0	23.1	1287.0	3.3	912.7	3.6
MP2/aug-cc-pVTZ	3015.4	17.4	1286.5	2.7	911.9	2.8
Experimental	2998.0		1283.8		909.1	

Table S-3. Coefficients of Fourier expansion for the one-dimensional potential energy curves of 24DMFB given in Figures 2 and 3 calculated at the B3LYP-D3BJ/6-311++G(d,p), MP2/6-311++G(d,p) and MP2/6-311G(d,p) levels of theory. The potential is expanded as $V(\alpha) = \sum_{i=0}^n a_i f_i$.

B3LYP-D3BJ/6-311++G(d,p)				
	<i>o</i>-Me		<i>p</i>-Me	
f_i	a_i / Hartree	a_i /cm ⁻¹	a_i / Hartree	a_i /cm ⁻¹
1	-410.2653163		-410.2657503	
cos(3 α)	-0.000463388	-101.70	-0.000027632	-6.06
cos(6 α)	0.000008680	1.91	0.000010146	2.23

MP2/6-311++G(d,p)				
	<i>o</i>-Me		<i>p</i>-Me	
f_i	a_i / Hartree	a_i /cm ⁻¹	a_i / Hartree	a_i /cm ⁻¹
1	-409.0633023		-409.0637305	
cos(3 α)	-0.000536196	-117.68	-0.000017509	-3.84
cos(6 α)	0.000063160	13.86	0.000063209	13.87
cos(9 α)	–	–	0.000002456	0.54

MP2/6-31G(d,p)				
	<i>o</i>-Me		<i>p</i>-Me	
f_i	a_i / Hartree	a_i /cm ⁻¹	a_i / Hartree	a_i /cm ⁻¹
1	-408.8909866		-408.8913773	
cos(3 α)	-0.000461471	-101.28	-0.000019337	-4.24
cos(6 α)	0.000036636	8.04	0.000051809	11.37
cos(9 α)	–	–	-0.000003114	-0.68

Table S-4. Coefficients of Fourier expansion for the two-dimensional potential energy surface of 24DMFB given in Figure 4 calculated at the B3LYP-D3BJ/6-311++G(d,p), MP2/6-311++G(d,p) and MP2/6-311G(d,p) levels of theory. The potential is expanded as $V(\alpha) = \sum_{i=0}^n a_i f_i$.

i	f_i	B3LYP-D3BJ/ 6-311++G(d,p)		MP2/ 6-311++G(d,p)		MP2/ 6-31G(d,p)	
		a_i / Hartree	a_i /cm ⁻¹	a_i / Hartree	a_i /cm ⁻¹	a_i / Hartree	a_i /cm ⁻¹
0	1	-410.2652946		-409.0632178		-408.8909247	
1	cos(3 α_1)	0.000462578	101.52	0.000543278	119.24	0.000471359	103.45
2	cos(3 α_2)	0.000001395	0.31	0.000003791	0.83	0.000001457	0.32
3	cos(6 α_1)	0.000006685	1.47	0.000033305	7.31	0.000020280	4.45
4	cos(6 α_2)	0.000010325	2.27	0.000061306	13.46	0.000050444	11.07
5	cos(3 α_1)cos(3 α_2)	-0.000027476	-6.03	-0.000018683	-4.10	-0.000023375	-5.13
6	sin(3 α_1)sin(3 α_2)	-	-	0.000031097	6.83	-0.000001988	-0.44

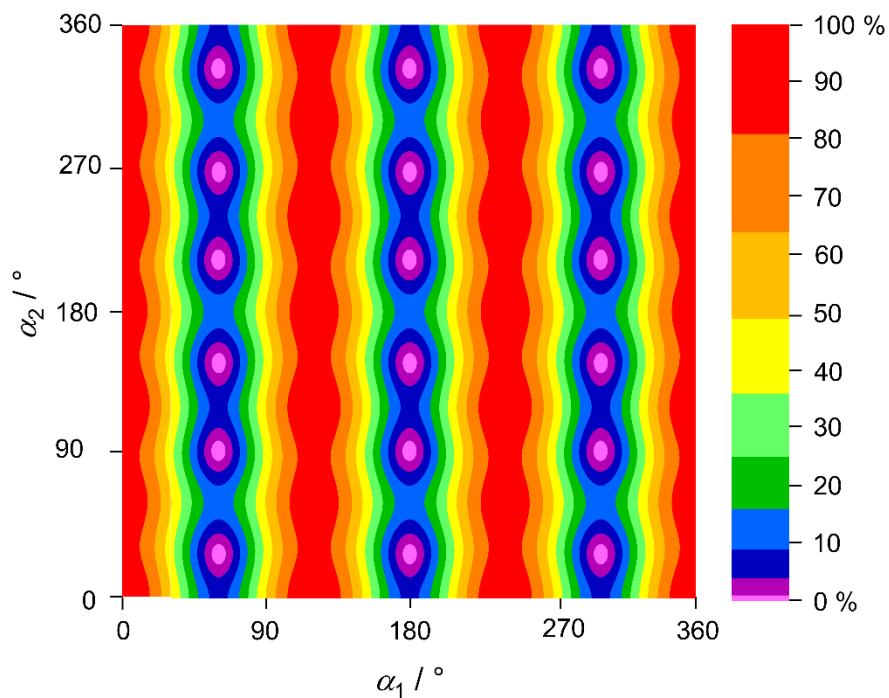


Figure S-1. A two-dimensional potential energy surface of 24DMFB in dependence on the dihedral angles $\alpha_1 = \angle(\text{C}_3, \text{C}_2, \text{C}_7, \text{H}_{12})$ and $\alpha_2 = \angle(\text{C}_5, \text{C}_4, \text{C}_8, \text{H}_{15})$ calculated at the MP2/6-31G(d,p) level of theory. The dihedral angles α_1 and α_2 were varied in steps of 10° while all other geometry parameters were optimized. The numbers in the color code indicate the energy (in percent) relative to the energetic minimum (0%) and maximum (100%) with $E_{\min} = -408.891426$, and $E_{\max} = -408.890357$ Hartree (100%).

Table S-5. Fitted frequencies (ν_{obs}) of 24DMFB. The $\nu_{\text{obs}} - \nu_{\text{calc}}$ residuals are obtained with the *XIAM_{mod}* and *BELGI-C_s-2Tops* programs.

J'	K_a'	K_c'	J	K_a	K_c	Species	ν_{obs} MHz	$\nu_{\text{obs}} - \nu_{\text{calc}}$ <i>XIAM</i> / kHz	$\nu_{\text{obs}} - \nu_{\text{calc}}$ <i>BELGI</i> / kHz
Upper level			Lower level						
2	0	2	1	0	1	(00)	4333.4249	24.9	1
2	1	1	1	1	0	(00)	4762.4717	34.1	0
2	1	2	1	0	1	(00)	5779.1966	108.4	1
2	1	2	1	1	1	(00)	4011.7336	9.3	-5
2	2	0	1	1	1	(00)	10493.8848	300.6	2
2	2	1	1	1	0	(00)	10064.8360	289.5	1
3	0	3	2	0	2	(00)	6372.8336	34.3	1
3	1	2	2	1	1	(00)	7107.3961	50.6	1
3	1	3	2	0	2	(00)	7431.9246	103	0
3	1	3	2	1	2	(00)	5986.1543	20.9	1
3	2	1	2	1	2	(00)	13270.6252	322.3	2
3	2	1	2	2	0	(00)	6788.4779	35.0	0
3	2	2	2	1	1	(00)	11883.0184	288.7	1
3	2	2	2	2	1	(00)	6580.6465	25.8	-8
3	3	0	3	2	1	(00)	9550.4607	355.6	1
3	3	1	3	2	2	(00)	9807.1074	356.8	1
4	0	4	3	0	3	(00)	8292.6417	41.1	1
4	1	3	3	1	2	(00)	9401.8697	64.1	2
4	1	4	3	0	3	(00)	8988.2414	93.0	0
4	1	4	3	1	3	(00)	7929.1510	24.8	1
4	2	2	3	1	3	(00)	16497.9156	350.6	4
4	2	2	3	2	1	(00)	9213.4437	48.2	3
4	2	3	3	1	2	(00)	13507.8615	279.3	-1
4	2	3	3	2	2	(00)	8732.2412	43.2	1
4	3	1	4	2	2	(00)	9234.2848	346.5	-3
4	3	2	3	3	1	(00)	8868.5307	48.9	8
4	3	2	4	2	3	(00)	9943.3883	353.9	-2
4	4	0	4	3	1	(00)	13568.7902	305.5	3
4	4	1	4	3	2	(00)	13602.0320	306.5	4
5	0	5	4	0	4	(00)	10119.3810	39.9	-2
5	0	5	4	1	4	(00)	9423.7839	-9.3	3
5	1	4	4	1	3	(00)	11618.4966	72.8	2
5	1	4	4	2	3	(00)	7512.5056	-141.6	6
5	1	5	4	0	4	(00)	10534.9414	79.6	0
5	1	5	4	1	4	(00)	9839.3384	24.5	-1
5	2	3	4	2	2	(00)	11694.1571	58.6	3

5	2	4	4	1	3	(00)	14954.6644	264.8	-1
5	2	4	4	2	3	(00)	10848.6692	46.2	-1
5	2	4	5	1	5	(00)	8835.3842	337.1	-2
5	3	2	4	3	1	(00)	11204.6107	47.2	1
5	3	2	5	2	3	(00)	8744.7368	333.5	-7
5	3	3	4	3	2	(00)	11106.9647	49.4	3
5	3	3	5	2	4	(00)	10201.6803	353.7	-1
5	4	1	4	4	0	(00)	11089.7140	53.9	10
5	4	1	5	3	2	(00)	13453.8826	301.3	0
5	4	2	4	4	1	(00)	11086.8190	44.4	0
5	4	2	5	3	3	(00)	13581.8839	299.1	-1
5	5	0	5	4	1	(00)	17482.4820	67.0	12
5	5	1	5	4	2	(00)	17485.7104	70.6	15
6	0	6	5	0	5	(00)	11904.7974	37.9	-1
6	0	6	5	1	5	(00)	11489.2413	2.4	2
6	1	5	5	1	4	(00)	13726.1036	83.7	6
6	1	5	5	2	4	(00)	10389.9349	-109.1	8
6	1	6	5	0	5	(00)	12135.6603	66.0	3
6	1	6	5	1	5	(00)	11720.0998	26.2	1
6	2	4	5	2	3	(00)	14166.8505	66.7	3
6	2	5	5	1	4	(00)	16259.2982	241.8	-2
6	2	5	5	2	4	(00)	12923.1338	53.3	4
6	2	5	6	0	6	(00)	10269.2722	383.4	-4
6	2	5	6	1	6	(00)	10038.4134	359.3	-4
6	3	3	5	3	2	(00)	13586.9565	50.6	4
6	3	3	6	2	4	(00)	8164.8418	316.4	-7
6	3	4	5	3	3	(00)	13341.0087	50.9	2
6	3	4	6	1	5	(00)	13152.7577	517.3	-4
6	3	4	6	2	5	(00)	10619.5555	351.6	-3
6	4	2	5	4	1	(00)	13352.6276	46.0	1
6	4	2	6	3	3	(00)	13219.5547	297.7	-2
6	4	3	5	4	2	(00)	13339.8509	47.2	2
6	4	3	6	3	4	(00)	13580.7267	296.0	0
6	5	1	5	5	0	(00)	13296.8992	42.8	1
6	5	1	6	4	2	(00)	17426.7496	59.9	8
6	5	2	5	5	1	(00)	13296.6531	41.3	0
6	5	2	6	4	3	(00)	17442.5101	62.2	10
7	0	7	6	0	6	(00)	13686.8064	34.2	2
7	0	7	6	1	6	(00)	13455.9484	11.0	3
7	1	6	6	1	5	(00)	15702.2764	80.7	1
7	1	6	6	2	5	(00)	13169.0780	-81.2	6

7	1	6	7	0	7	(00)	9751.5456	269.8	2
7	1	6	7	1	7	(00)	9629.8115	251.0	-1
7	1	7	6	0	6	(00)	13808.5363	48.8	1
7	1	7	6	1	6	(00)	13577.6772	24.4	1
7	2	5	6	2	4	(00)	16577.8623	70.8	2
7	2	6	6	1	5	(00)	17484.5005	211.3	-4
7	2	6	6	2	5	(00)	14951.3024	49.8	1
7	2	6	7	0	7	(00)	11533.7696	400.3	-3
7	2	6	7	1	7	(00)	11412.0393	385.4	-3
7	3	4	6	3	3	(00)	16059.4238	45.7	3
7	3	4	7	2	5	(00)	7646.4041	292.1	-5
7	3	5	6	3	4	(00)	15557.8054	48.8	2
7	3	5	7	1	6	(00)	13008.2828	481.5	-7
7	3	5	7	2	6	(00)	11226.0596	351.7	-1
7	4	3	6	4	2	(00)	15648.0812	42.8	1
7	4	3	7	3	4	(00)	12808.2115	294.1	-4
7	4	4	6	4	3	(00)	15606.6285	42.7	1
7	5	2	6	5	1	(00)	15548.0732	38.0	0
7	5	2	7	4	3	(00)	17326.7418	55.4	7
7	5	3	6	5	2	(00)	15546.7419	37.3	-1
7	5	3	7	4	4	(00)	17382.6218	55.1	7
8	0	8	7	0	7	(00)	15478.8442	26.2	2
8	0	8	7	1	7	(00)	15357.1143	11.6	2
8	1	7	7	1	6	(00)	17554.1598	76.4	1
8	1	7	7	2	6	(00)	15771.9354	-54.6	5
8	1	7	8	0	8	(00)	11826.8599	318.7	-1
8	1	7	8	1	8	(00)	11765.0277	310.6	0
8	1	8	7	0	7	(00)	15540.6774	35.3	2
8	1	8	7	1	7	(00)	15418.9468	20.0	2
8	2	6	7	2	5	(00)	18892.6349	70.4	3
8	2	6	7	3	5	(00)	12125.0505	-224.6	13
8	2	7	7	1	6	(00)	18714.5864	176.5	-3
8	2	7	7	2	6	(00)	16932.3613	44.8	1
8	2	7	8	0	8	(00)	12987.2872	419.5	-4
8	2	7	8	1	8	(00)	12925.4540	410.4	-3
8	3	5	7	3	4	(00)	18606.7429	38.9	4
8	3	5	8	2	6	(00)	7360.5128	261.3	-4
8	3	6	7	3	5	(00)	17744.2847	42.4	1
8	3	6	8	2	7	(00)	12037.9827	349.0	-1
8	4	4	7	4	3	(00)	17991.6367	33.4	1
8	4	4	8	3	5	(00)	12193.1062	289.5	-5

8	4	5	7	4	4	(00)	17882.5015	34.0	1
8	5	3	8	4	4	(00)	17152.1554	50.8	5
8	5	4	7	5	3	(00)	17811.8169	26.8	-2
8	5	4	8	4	5	(00)	17311.9388	49.4	6
8	6	2	7	6	1	(00)	17752.9054	20.5	-3
8	6	3	7	6	2	(00)	17752.7875	22.9	-1
8	7	1	7	7	0	(00)	17716.8778	18.7	1
8	7	2	7	7	1	(00)	17716.8661	8.3	-9
9	0	9	8	0	8	(00)	17281.0824	16.2	2
9	1	8	8	1	7	(00)	19325.0153	64.9	0
9	1	8	8	2	7	(00)	18164.5890	-34.9	4
9	1	9	8	0	8	(00)	17311.6355	21.2	1
9	1	9	8	1	8	(00)	17249.8034	13.2	3
9	2	7	8	2	6	(00)	21084.8908	63.7	2
9	2	8	8	1	7	(00)	20029.8104	136.5	-3
9	2	8	8	2	7	(00)	18869.3839	36.5	1
9	3	6	8	3	5	(00)	21175.7469	27.6	3
9	3	6	9	2	7	(00)	7451.3690	225.3	-2
9	3	7	8	3	6	(00)	19889.2882	31.6	1
9	3	7	9	2	8	(00)	13057.8861	343.3	-1
9	4	5	8	4	4	(00)	20403.6575	16.4	1
9	4	5	9	3	6	(00)	11421.0170	278.5	-7
9	4	6	8	4	5	(00)	20158.8991	19.6	0
9	5	4	8	5	3	(00)	20109.1479	12.9	-1
9	5	4	9	4	5	(00)	16857.6454	46.9	3
9	5	5	8	5	4	(00)	20092.5734	12.4	-1
9	6	3	8	6	2	(00)	20012.1348	4.9	-4
9	6	4	8	6	3	(00)	20011.5811	5.4	-3
9	7	2	8	7	1	(00)	19960.3545	-7.5	1
9	7	3	8	7	2	(00)	19960.3545	2.4	-9
9	8	1	8	8	0	(00)	19927.5564	-13.9	-7
9	8	2	8	8	1	(00)	19927.5564	-13.8	-7
10	0	10	9	0	9	(00)	19090.2981	4.3	2
10	0	10	9	1	9	(00)	19059.7449	-0.8	2
10	1	9	9	1	8	(00)	21068.9814	46.8	-1
10	1	10	9	0	9	(00)	19105.0796	7.5	2
10	1	10	9	1	9	(00)	19074.5265	2.6	2
10	2	8	9	2	7	(00)	23133.3035	50.2	0
10	2	8	10	1	9	(00)	11403.3653	176.9	2
10	2	8	10	2	9	(00)	10998.7898	129.5	5
10	2	9	9	1	8	(00)	21473.5573	94.7	-4

10	2	9	9	2	8	(00)	20768.7636	24.5	1
10	3	7	9	3	6	(00)	23699.3248	11.6	2
10	3	7	10	2	8	(00)	8017.3908	187.2	1
10	3	8	9	3	7	(00)	21984.9651	14.3	-1
10	3	8	10	2	9	(00)	14274.0875	333.0	-3
10	4	6	9	4	5	(00)	22902.5379	-7.4	2
10	4	6	10	3	7	(00)	10624.2288	258.3	-9
10	4	7	9	4	6	(00)	22423.9926	-1.8	-1
10	5	5	9	5	4	(00)	22432.6913	-9.4	-1
10	5	6	9	5	5	(00)	22387.9534	-10.2	-2
10	6	4	9	6	3	(00)	22286.4613	-17.8	-4
10	6	5	9	6	4	(00)	22284.4200	-17.6	-4
10	7	3	9	7	2	(00)	22214.2012	-29.4	-8
10	7	4	9	7	3	(00)	22214.1521	-26.4	-5
11	0	11	10	0	10	(00)	20903.6217	-9.6	2
11	0	11	10	1	10	(00)	20888.8398	-13.2	1
11	1	10	10	1	9	(00)	22820.9622	24.6	-1
11	1	11	10	0	10	(00)	20910.6536	-8.2	2
11	1	11	10	1	10	(00)	20895.8725	-11	2
11	2	9	10	2	8	(00)	25033.5778	31.6	-2
11	2	10	10	1	9	(00)	23043.3264	55.3	-3
11	2	10	10	2	9	(00)	22638.7500	7.0	-1
12	0	12	11	0	11	(00)	22719.1937	-26.7	2
12	0	12	11	1	11	(00)	22712.1618	-28.1	2
12	1	11	11	1	10	(00)	24591.8735	-2.0	-3
12	1	12	11	0	11	(00)	22722.4942	-25.6	2
12	1	12	11	1	11	(00)	22715.4618	-27.5	2
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2	1	2	1	0	1	(10)	5775.6664	-13.9	0
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2	2	0	1	1	0	(10)	10190.6585	612.3	6
2	2	0	1	1	1	(10)	10581.0515	802.6	-10
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3	1	2	2	1	1	(10)	7104.8527	-7.7	0
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4	0	4	3	0	3	(10)	8291.9296	23.4	1
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4	1	4	3	0	3	(10)	8987.1593	31.1	1
4	1	4	3	1	3	(10)	7929.1028	29.7	0
4	2	2	3	1	3	(10)	16508.3944	201.8	0
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4	2	3	3	1	2	(10)	13488.9417	-206	-3
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4	3	2	4	2	3	(10)	9808.1873	-1000.8	1
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5	1	4	4	2	3	(10)	7526.2626	198.9	1
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5	2	4	4	2	3	(10)	10855.2143	122.1	3
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6	0	6	5	0	5	(10)	11904.2636	33.5	0
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6	1	5	5	1	4	(10)	13723.5212	-3.1	-2
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7	0	7	6	1	6	(10)	13455.4538	29.6	2
7	1	6	6	1	5	(10)	15700.0313	0.4	0
7	1	6	6	2	5	(10)	13169.9782	25.2	1
7	1	6	7	0	7	(10)	9744.4152	-98.3	-2
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7	1	7	6	1	6	(10)	13577.1889	33.0	0
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7	2	6	7	1	7	(10)	11403.6925	-104.0	-4
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7	4	3	6	4	2	(10)	15625.3749	-29.4	0
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7	4	4	6	4	3	(10)	15625.2326	9.5	1
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8	1	8	7	0	7	(10)	15540.1804	37.7	0
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8	2	6	7	3	5	(10)	12150.5809	392.6	8
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8	7	1	7	7	0	(10)	17713.9930	-35.1	3
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9	5	4	9	4	5	(10)	17015.8521	-65.6	6
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10	0	10	9	0	9	(10)	19089.7986	26.1	1
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10	2	8	10	1	9	(10)	11394.9541	-214.8	-1
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11	0	11	10	0	10	(10)	20903.1257	17.8	0
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11	1	11	10	0	10	(10)	20910.1613	18.6	0
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12	0	12	11	0	11	(10)	22718.7034	7.7	1
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5	4	2	5	3	3	(01)	11740.6113	-459.7	-8
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6	0	6	5	0	5	(01)	11957.5582	57.0	-3
6	0	6	5	1	5	(01)	11468.5889	-87.2	-1
6	1	5	5	1	4	(01)	13388.4191	-212.1	-2
6	1	5	5	2	4	(01)	11435.8828	-145.3	-2
6	1	6	5	0	5	(01)	12237.9932	121.0	-3
6	1	6	5	1	5	(01)	11749.0228	-24.3	-2
6	2	4	5	2	3	(01)	13599.2245	-508.0	1
6	2	4	5	3	3	(01)	11717.6261	3623	-10
6	2	5	5	1	4	(01)	15366.5396	21.2	-3
6	2	5	5	2	4	(01)	13414.0047	89.5	-1
6	2	5	6	1	6	(01)	9176.0696	184.6	-1
6	3	3	5	3	2	(01)	13428.0360	-2.5	-2
6	3	3	6	2	4	(01)	10746.9053	1936.7	2
6	3	4	5	3	3	(01)	13720.0752	485.9	-4

6	3	4	6	2	5	(01)	7681.7609	-1110.2	-1
6	4	2	5	4	1	(01)	13331.9696	14.1	-2
6	4	2	6	3	3	(01)	14868.8207	1381	8
6	4	3	5	4	2	(01)	13400.8003	38.0	0
6	4	3	6	3	4	(01)	11421.3363	-907.7	-4
6	5	1	5	5	1	(01)	13290.3455	24.6	1
6	5	1	6	4	2	(01)	18827.9305	1634.2	-14
7	0	7	6	0	6	(01)	13726.1399	28.9	-2
7	0	7	6	1	6	(01)	13445.7054	-34.6	-1
7	1	6	6	1	5	(01)	15554.4749	-4.8	-1
7	1	6	6	2	5	(01)	13576.3559	-236.6	2
7	1	6	7	0	7	(01)	9306.7180	-19.5	0
7	1	7	6	0	6	(01)	13875.6188	46.7	-2
7	1	7	6	1	6	(01)	13595.1839	-17.3	-2
7	2	5	6	2	4	(01)	15846.1848	-774.0	3
7	2	6	6	1	5	(01)	17238.2364	262.3	-3
7	2	6	6	2	5	(01)	15260.1168	29.8	-1
7	2	6	7	0	7	(01)	10990.4793	247.4	-2
7	2	6	7	1	7	(01)	10841.0015	230.7	-1
7	3	4	6	3	3	(01)	15761.4873	-43.9	-1
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7	4	3	6	4	2	(01)	15602.2568	11.4	-1
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7	5	2	6	5	1	(01)	15535.6803	25.5	0
7	5	2	7	4	3	(01)	18761.3548	1649.1	-12
7	5	3	6	5	2	(01)	15584.9521	-3.5	1
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7	6	1	6	6	1	(01)	15500.2014	40.9	2
8	0	8	7	0	7	(01)	15504.9212	2.1	-2
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8	1	7	7	1	6	(01)	17552.5631	130.0	-2
8	1	7	7	2	6	(01)	15868.8011	-137.6	0
8	1	7	8	0	8	(01)	11354.3604	108.9	1
8	1	8	7	0	7	(01)	15580.9988	0.3	-2
8	1	8	7	1	7	(01)	15431.5202	-17.2	-2
8	2	6	7	2	5	(01)	18117.7315	-631.5	2
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8	4	4	7	4	3	(01)	17897.1329	1.9	0
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9	2	7	8	2	6	(01)	20446.7697	-331.6	0
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9	5	4	8	5	3	(01)	20072.1413	22.7	1
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2	0	2	1	0	1	(11)	4030.2113	-30.8	3
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3	0	3	2	0	2	(11)	6240.9617	-1.0	0
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4	0	4	3	0	3	(11)	8280.9653	14.3	-1
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5	2	4	4	1	3	(11)	13187.4573	0.3	1
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7	1	7	6	1	6	(11)	13591.5369	0.7	-2
7	2	5	6	2	4	(11)	15858.3828	-30.0	2
7	2	6	6	1	5	(11)	17316.7160	-162.8	5
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7	5	3	6	5	2	(11)	15583.3577	-86.1	2
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7	6	1	6	6	1	(11)	15499.5538	-26.7	0
8	0	8	7	0	7	(11)	15506.8921	-16.8	-2
8	0	8	7	1	7	(11)	15348.7633	42.6	-2
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11	1	11	10	0	10	(11)	20920.7650	-30.5	-1
11	1	11	10	1	10	(11)	20901.5626	-13	-1
11	2	9	10	2	8	(11)	24875.5027	176	-2
11	2	10	10	1	9	(11)	23162.2892	-180.7	6
11	2	10	10	2	9	(11)	22662.5033	72.1	1
12	0	12	11	0	11	(11)	22724.8226	-21.6	2
12	0	12	11	1	11	(11)	22715.7077	-19.5	-6
12	1	11	11	1	10	(11)	24633.9727	22.4	4
12	1	12	11	0	11	(11)	22729.0774	-31.7	-2
12	1	12	11	1	11	(11)	22719.9691	-23	-4
2	0	2	1	0	1	(12)	4045.2186	139	3
2	1	2	1	0	1	(12)	4954.8825	451.2	4
2	2	0	2	2	1	(12)	11239.5747	-1355.8	7
2	2	1	1	1	1	(12)	11820.9796	-1064.7	0
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3	0	3	2	0	2	(12)	6257.4435	22.1	0
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3	1	2	2	1	2	(12)	9824.9864	-982.2	-8
3	1	3	2	0	2	(12)	7187.4211	168.3	-1
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