Table S-1: Coefficients of the one-dimensional Fourier expansion for the potential energy curves of 2A4MT given in Figure 2 calculated at the MP2/6-31G(d,p) level of theory. The potential is expanded as $V(\varphi) = \sum_{i=0}^{5} a_i f_i$.

i	$\mathbf{f}_i$	$\mathbf{a}_i$ **/ Hartree**	$\mathbf{a}_i$ **/ cm^{-1}**
0	1	−743.36102813	
1	cos 1α	−0.000721750	−158.4
2	cos 2α	−0.006602747	−1449.2
3	cos 3α	−0.000293569	−64.4
4	cos 4α	0.000439625	96.5
5	cos 5α	−0.000066190	−14.5

Table S-2: Nuclear coordinates in the principal axis system of *syn*-2A5MT and *anti*-2A5MT calculated at the MP2/6-311++G(d,p) level of theory (for atom numbering see Figure 1).

	***syn*-2A5MT**			***anti*-2A5MT**		
	***a* / Å**	***b* / Å**	***c* / Å**	***a* / Å**	***b* / Å**	***c* / Å**
S1	0.633959	−1.010491	−0.000088	−0.611102	−1.015933	0.000059
C2	−0.564937	0.232994	0.000042	0.561539	0.257459	0.000013
C3	0.017715	1.490700	−0.000031	−0.057695	1.494956	0.000063
C4	1.427528	1.433507	−0.000095	−1.464250	1.401443	0.000048
C5	1.917932	0.140589	−0.000129	−1.928946	0.099237	−0.000006
C6	−1.985912	−0.141843	0.000042	2.022440	0.038248	−0.000003
O7	−2.327220	−1.327833	0.000011	2.790051	1.001974	−0.000086
C8	−3.000748	0.979643	0.000083	2.532029	−1.385022	−0.000014
C9	3.352687	−0.286808	0.000227	−3.350664	−0.368558	−0.000118
H10	3.596419	−0.878304	0.882748	−3.579192	−0.967367	−0.881844
H11	3.992378	0.594172	−0.002335	−4.012277	0.496066	0.000136
H12	3.595536	−0.882770	−0.879514	−3.579165	−0.967860	0.881278
H13	2.073683	2.300829	−0.000153	−2.132887	2.251577	0.000055
H14	−0.554664	2.407808	−0.000048	0.515442	2.410700	0.000086
H15	−2.873377	1.608201	0.881088	2.173164	−1.918592	0.880557
H16	−3.996135	0.545438	0.000143	3.617934	−1.368476	−0.000193
H17	−2.873474	1.608169	−0.880959	2.172869	−1.918714	−0.880389

Table S-3a: Rotational constants and the dihedral angle *α* of *syn*-2A4MT optimized at various levels of theory. The deviations between the calculated and the experimental values of the rotational constants are given as ΔA, ΔB and ΔC. $V_{3,1}$ is the barrier of the acetyl methyl and $V_{3,2}$ that of the ring methyl group. All optimizations were performed under full-symmetry relaxation.

Method/Basis Set	*A* / MHz	*ΔA* / MHz	*B* / MHz	*ΔB* / MHz	*C* / MHz	*ΔC* / MHz	$V_{3,1}$ / cm^{-1}	$\Delta V_{3,1}$ / cm^{-1}	$V_{3,2}$ / cm^{-1}	$\Delta V_{3,2}$ / cm^{-1}	α / °	Planar
B3LYP-D3/6-31G(d,p)	3418.3619	–29.3	918.3812	–8.6	730.4766	–7.1	282.5	19.3	219.5	–62.2	0.03	Yes
B3LYP-D3/6-31+G(d,p)	3415.9261	–31.7	915.4731	–11.5	728.5392	–9.0	216.6	85.2	214.7	–57.4	0.00	Yes
B3LYP-D3/6-31++G(d,p)	3415.9593	–31.7	915.4634	–11.6	728.5349	–9.0	214.9	86.9	215.3	–58.0	0.00	Yes
B3LYP-D3/6-311G(d,p)	3431.1657	–16.5	919.7186	–7.3	731.8976	–5.6	272.9	28.9	196.3	–39.0	0.03	Yes
B3LYP-D3/6-311+G(d,p)	3430.6772	–16.9	918.1648	–8.9	730.8941	–6.6	227.2	74.6	197.2	–39.9	–0.04	Yes
B3LYP-D3/6-311++G(d,p)	3430.6089	–17.0	918.1501	–8.9	730.8818	–6.7	227.1	74.7	209.4	–52.1	–0.04	Yes
B3LYP-D3/6-311G(2d,2p)	3448.4943	0.9	922.4506	–4.6	734.3722	–3.2	292.3	9.5	168.3	–11.0	0.04	Yes
B3LYP-D3/6-311+G(2d,2p)	3447.3820	–0.2	921.5052	–5.5	733.7264	–3.8	237.2	64.6	170.4	–13.1	0.00	Yes
B3LYP-D3/6-311G(df,pd)	3443.4749	–4.1	921.9676	–5.1	733.8649	–3.7	262.7	39.1	194.9	–37.6	–0.04	Yes
B3LYP-D3/6-311+G(df,pd)	3443.1506	–4.5	920.5616	–6.5	732.9595	–4.6	217.8	84.0	192.9	–35.6	–0.04	Yes
B3LYP-D3/6-311++G(df,pd)	3443.2000	–4.4	920.5383	–6.5	732.9462	–4.6	212.2	89.6	198.1	–40.8	–0.04	Yes
B3LYP-D3/6-311++G(2d,2p)	3447.2525	–0.4	921.5282	–5.5	733.7355	–3.8	239.6	62.2	174.1	–16.8	0.00	Yes
B3LYP-D3/6-311G(2df,2pd)	3456.8451	9.2	924.3065	–2.7	735.9243	–1.6	295.9	5.9	155.5	1.8	0.02	Yes
B3LYP-D3/6-311+G(2df,2pd)	3455.7159	8.1	923.0726	–3.9	735.0944	–2.4	241.2	60.6	158.2	–0.9	0.00	Yes
B3LYP-D3/6-311++G(2df,2pd)	3455.6563	8.0	923.0699	–4.0	735.0905	–2.4	243.8	58.0	163.1	–5.8	0.00	Yes
B3LYP-D3/6-311G(3df,3pd)	3461.6822	14.1	924.7836	–2.2	736.4519	–1.1	279.8	22.0	149.2	8.1	0.00	Yes
B3LYP-D3/6-311+G(3df,3pd)	3459.5610	11.9	924.3036	–2.7	736.0521	–1.5	251.7	50.1	156.8	0.5	0.00	Yes
B3LYP-D3/6-311++G(3df,3pd)	3459.5068	11.9	924.3083	–2.7	736.0523	–1.5	251.2	50.6	157.0	0.3	0.00	Yes
B3LYP-D3/cc-pVDZ	3405.5805	–42.0	916.6983	–10.3	728.9029	–8.6	334.9	–33.1	195.7	–38.4	0.00	Yes
B3LYP-D3/aug-cc-pVDZ	3408.6021	–39.0	914.5675	–12.5	727.6775	–9.9	236.0	65.8	195.1	–37.8	0.00	Yes
B3LYP-D3/cc-pVTZ	3452.1936	4.6	922.9304	–4.1	734.8454	–2.7	254.4	47.4	172.1	–14.8	0.00	Yes
B3LYP-D3/aug-cc-pVTZ	3452.6106	5.0	922.5850	–4.4	734.6462	–2.9	238.0	63.8	169.2	–11.9	0.00	Yes
B3LYP-D3BJ/6-31G(d,p)	3418.6813	–28.9	921.8194	–5.2	732.6629	–4.9	249.0	52.8	200.4	–43.1	0.00	Yes

B3LYP-D3BJ/6-31+G(d,p)	3416.6542	–31.0	918.9596	–8.1	730.7763	–6.8	181.7	120.1	196.0	–38.7	0.00	Yes
B3LYP-D3BJ/6-31++G(d,p)	3416.6880	–30.9	918.9506	–8.1	730.7725	–6.8	179.8	122.0	196.5	–39.2	0.00	Yes
B3LYP-D3BJ/6-311G(d,p)	3432.3884	–15.2	923.1190	–3.9	734.1031	–3.4	239.4	62.4	176.5	–19.2	0.02	Yes
B3LYP-D3BJ/6-311+G(d,p)	3431.3484	–16.3	921.6454	–5.4	733.1266	–4.4	191.6	110.2	177.4	–20.1	0.00	Yes
B3LYP-D3BJ/6-311++G(d,p)	3431.3484	–16.3	921.6454	–5.4	733.1266	–4.4	191.5	110.3	189.6	–32.3	0.00	Yes
B3LYP-D3BJ/6-311G(2d,2p)	**3448.6950**	**1.1**	**926.0056**	**–1.0**	**736.6312**	**–0.9**	**259.3**	**42.5**	**147.8**	**9.5**	**–0.01**	**Yes**
B3LYP-D3BJ/6-311+G(2d,2p)	**3447.5436**	**–0.1**	**925.0572**	**–2.0**	**735.9810**	**–1.6**	**201.9**	**99.9**	**149.7**	**7.6**	**–0.01**	**Yes**
B3LYP-D3BJ/6-311++G(2d,2p)	3447.4083	–0.2	925.0767	–1.9	735.9877	–1.5	204.4	97.4	153.4	3.9	–0.01	Yes
B3LYP-D3BJ/6-311G(df,pd)	3443.5927	–4.0	925.3976	–1.6	736.0399	–1.5	228.3	73.5	174.9	–17.6	0.00	Yes
B3LYP-D3BJ/6-311+G(df,pd)	3443.3558	–4.3	924.0092	–3.0	735.1515	–2.4	181.6	120.2	173.0	–15.7	0.02	Yes
B3LYP-D3BJ/6-311++G(df,pd)	3443.3907	–4.2	923.9910	–3.0	735.1409	–2.4	176.0	125.8	178.0	–20.7	0.02	Yes
B3LYP-D3BJ/6-311G(2df,2pd)	3456.3326	8.7	927.7737	0.8	738.0961	0.6	262.2	39.6	134.7	22.6	0.00	Yes
B3LYP-D3BJ/6-311+G(2df,2pd)	3455.4266	7.8	926.6364	–0.4	737.3373	–0.2	205.7	96.1	137.3	20.0	0.00	Yes
B3LYP-D3BJ/6-311++G(2df,2pd)	3455.3680	7.7	926.6342	–0.4	737.3337	–0.2	208.2	93.6	142.2	15.1	0.00	Yes
B3LYP-D3BJ/6-311G(3df,3pd)	3461.3764	13.8	928.3088	1.3	738.6696	1.1	246.2	55.6	128.3	29.0	0.00	Yes
B3LYP-D3BJ/6-311+G(3df,3pd)	3459.0805	11.5	927.8741	0.9	738.2903	0.8	215.8	86.0	136.0	21.3	0.00	Yes
B3LYP-D3BJ/6-311++G(3df,3pd)	3459.0250	11.4	927.8789	0.9	738.2906	0.8	215.4	86.4	136.1	21.2	0.00	Yes
B3LYP-D3BJ/cc-pVDZ	3406.3694	–41.3	920.2583	–6.8	731.1857	–6.3	301.3	0.5	176.0	–18.7	0.00	Yes
B3LYP-D3BJ/aug-cc-pVDZ	3409.4634	–38.2	918.1520	–8.9	729.9816	–7.6	198.6	103.2	175.0	–17.7	0.01	Yes
B3LYP-D3BJ/cc-pVTZ	3452.0427	4.4	926.4325	–0.6	737.0548	–0.5	219.3	82.5	151.4	5.9	0.00	Yes
B3LYP-D3BJ/aug-cc-pVTZ	3452.2999	4.7	926.1371	–0.9	736.8804	–0.7	202.1	99.7	148.3	9.0	0.00	Yes
CAM-B3LYP-D3BJ/6-311G(d,p)	3469.6977	22.1	929.1899	2.2	739.6393	2.1	264.9	36.9	178.0	–20.7	0.00	Yes
CAM-B3LYP-D3BJ/6-311+G(d,p)	3469.4404	21.8	927.6204	0.6	738.6353	1.1	217.9	83.9	181.3	–24.0	0.00	Yes
CAM-B3LYP-D3BJ/6-311++G(d,p)	3469.3630	21.7	927.6105	0.6	738.6258	1.1	218.5	83.3	193.3	–36.0	0.00	Yes
CAM-B3LYP-D3BJ/cc-pVDZ	3444.2266	–3.4	926.1504	–0.9	736.6390	–0.9	318.5	–16.7	182.2	–24.9	0.00	Yes
CAM-B3LYP-D3BJ/aug-cc-pVDZ	3447.3200	–0.3	923.9464	–3.1	735.3687	–2.2	221.8	80.0	182.1	–24.8	0.01	Yes
CAM-B3LYP-D3BJ/cc-pVTZ	3489.6607	42.0	932.4014	5.4	742.5390	5.0	244.8	57.0	157.1	0.2	0.00	Yes
CAM-B3LYP-D3BJ/aug-cc-pVTZ	3489.8800	42.3	932.0109	5.0	742.3022	4.8	228.3	73.5	156.0	1.3	0.00	Yes
CCSD/cc-pVDZ	3391.1934	–56.4	912.0127	–15.0	725.3503	–12.2	403.7	–101.9	161.3	–4.0	0.01	Yes
M06-2X/6-31G(d,p)	**3448.3181**	**0.7**	**927.4369**	**0.4**	**737.5813**	**0.0**	**335.3**	**–33.5**	**205.7**	**–48.4**	**0.00**	**Yes**

M06-2X/6-31+G(d,p)	**3446.9602**	**–0.7**	**925.1120**	**–1.9**	**736.0613**	**–1.5**	**278.3**	**23.5**	**202.3**	**–45.0**	**0.00**	**Yes**
M06-2X/6-31++G(d,p)	**3446.9427**	**–0.7**	**925.1195**	**–1.9**	**736.0658**	**–1.5**	**277.3**	**24.5**	**204.0**	**–46.7**	**0.00**	**Yes**
M06-2X/6-311G(d,p)	3458.5736	11.0	928.3073	1.3	738.5914	1.1	340.2	–38.4	188.4	–31.1	0.01	Yes
M06-2X/6-311+G(d,p)	3458.7617	11.1	926.9316	–0.1	737.7305	0.2	306.3	–4.5	193.8	–36.5	0.00	Yes
M06-2X/6-311++G(d,p)	3458.6776	11.1	926.9272	–0.1	737.7245	0.2	306.6	–4.8	207.2	–49.9	0.00	Yes
M06-2X/6-311G(df,pd)	3468.7535	21.1	929.9338	2.9	740.0689	2.5	326.2	–24.4	188.4	–31.1	0.00	Yes
M06-2X/6-311+G(df,pd)	3469.1235	21.5	928.6382	1.6	739.2640	1.7	294.0	7.8	190.6	–33.3	0.00	Yes
M06-2X/6-311++G(df,pd)	3469.1104	21.5	928.6307	1.6	739.2586	1.7	288.0	13.8	196.3	–39.0	0.01	Yes
M06-2X/6-311G(3df,3pd)	3484.9651	37.3	932.2561	5.2	742.2514	4.7	363.3	–61.5	153.3	4.0	0.00	Yes
M06-2X/6-311+G(3df,3pd)	3483.0253	35.4	931.9722	4.9	741.9841	4.5	343.2	–41.4	160.2	–2.9	0.01	Yes
M06-2X/6-311++G(3df,3pd)	3482.9696	35.3	931.9736	5.0	741.9821	4.4	343.4	–41.6	160.5	–3.2	0.00	Yes
M06-2X/cc-pVDZ	3438.5948	–9.0	926.2620	–0.8	736.4632	–1.1	362.8	–61.0	186.8	–29.5	0.00	Yes
M06-2X/aug-cc-pVDZ	3441.3354	–6.3	924.1893	–2.8	735.2623	–2.3	297.3	4.5	188.9	–31.6	0.00	Yes
M06-2X/cc-pVTZ	3476.6829	29.1	931.3229	4.3	741.2847	3.8	324.6	–22.8	167.3	–10.0	0.00	Yes
M06-2X/aug-cc-pVTZ	3477.5311	29.9	931.0761	4.1	741.1666	3.6	316.5	–14.7	169.3	–12.0	0.00	Yes
MP2/6-31G(d,p)	3425.2312	–22.4	927.6229	0.6	736.5991	–0.9	359.0	–57.2	165.5	–8.2	–0.01	Yes
MP2/6-31+G(d,p)	3419.5932	–28.0	924.9485	–2.1	734.6713	–2.9	266.6	35.2	185.4	–28.1	0.01	Yes
MP2/6-31++G(d,p)	3419.4941	–28.1	924.9131	–2.1	734.6466	–2.9	262.2	39.6	194.2	–36.9	0.01	Yes
MP2/6-311G(d,p)	3422.6439	–25.0	926.4466	–0.6	735.8002	–1.7	347.3	–45.5	172.8	–15.5	–0.01	Yes
MP2/6-311+G(d,p)	3421.5236	–26.1	924.5314	–2.5	734.5427	–3.0	295.6	6.2	165.0	–7.7	–0.01	Yes
MP2/6-311++G(d,p)	3421.4884	–26.1	924.5102	–2.5	734.5289	–3.0	290.8	11.0	202.1	–44.8	–0.02	Yes
MP2/6-311G(2d,2p)	3433.9215	–13.7	928.9467	1.9	737.8014	0.3	340.8	–39.0	132.4	24.9	0.04	Yes
MP2/6-311+G(2d,2p)	3432.7451	–14.9	927.7451	0.7	736.9930	–0.5	283.3	18.5	137.0	20.3	0.03	Yes
MP2/6-311++G(2d,2p)	3432.5457	–15.1	927.7467	0.7	736.9867	–0.5	287.9	13.9	138.6	18.7	0.03	Yes
MP2/6-311G(df,pd)	3448.7375	1.1	931.6314	4.6	740.2461	2.7	326.7	–24.9	177.2	–19.9	–0.01	Yes
MP2/6-311+G(df,pd)	3445.5305	–2.1	929.8089	2.8	738.9522	1.4	272.0	29.8	168.3	–11.0	–0.02	Yes
MP2/6-311++G(df,pd)	3445.7387	–1.9	929.8698	2.8	739.0695	1.5	270.6	31.2	194.2	–36.9	–3.18	No
MP2/6-311G(2df,2pd)	3450.7935	3.2	934.6498	7.6	742.1877	4.7	335.8	–34.0	116.8	40.5	0.00	Yes
MP2/6-311+G(2df,2pd)	3448.8603	1.2	933.0737	6.1	741.1090	3.6	277.5	24.3	123.8	33.5	0.00	Yes
MP2/6-311++G(2df,2pd)	3448.7951	1.2	933.0566	6.0	741.0965	3.6	281.6	20.2	127.9	29.4	0.00	Yes

MP2/6-311G(3df,3pd)	3454.6254	7.0	934.2892	7.3	742.1542	4.6	339.2	−37.4	97.4	59.9	−0.03	Yes
MP2/6-311+G(3df,3pd)	3451.4295	3.8	933.6344	6.6	741.5942	4.1	295.2	6.6	108.1	49.2	−0.03	Yes
MP2/6-311++G(3df,3pd)	3451.3235	3.7	933.6862	6.7	741.6214	4.1	294.6	7.2	109.2	48.1	0.00	Yes
MP2/cc-pVDZ	3382.8111	−64.8	919.1623	−7.9	729.4494	−8.1	406.3	−104.5	166.8	−9.5	0.00	Yes
MP2/aug-cc-pVDZ	3373.8306	−73.8	916.4477	−10.6	727.3270	−10.2	271.2	30.6	152.0	5.3	0.00	Yes
MP2/cc-pVTZ	3441.6008	−6.0	931.9170	4.9	740.0479	2.5	293.6	8.2	137.9	19.4	0.00	Yes
ωB97X-D/6-311G(d,p)	3462.6795	15.1	925.5488	−1.5	737.0348	−0.5	262.7	39.1	183.4	−26.1	0.00	Yes
ωB97X-D/6-311+G(d,p)	3462.5874	15.0	925.5358	−1.5	737.0225	−0.5	263.8	38.0	195.7	−38.4	0.00	Yes
ωB97X-D/6-311++G(d,p)	3462.5550	14.9	926.9501	−0.1	737.9157	0.4	296.4	5.4	179.8	−22.5	0.00	Yes
ωB97X-D/aug-cc-pVDZ	3441.0349	−6.6	921.8369	−5.2	733.7647	−3.8	280.4	21.4	174.6	−17.3	0.02	Yes
ωB97X-D/aug-cc-pVTZ	3483.3105	35.7	929.6988	2.7	740.5541	3.0	283.1	18.7	154.5	2.8	0.00	Yes
ωB97X-D/cc-pVDZ	3438.4941	−9.1	923.6269	−3.4	734.7946	−2.7	334.4	−32.6	176.5	−19.2	0.00	Yes
ωB97X-D/cc-pVTZ	3482.9519	35.3	929.8956	2.9	740.6620	3.1	288.8	13.0	154.9	2.4	0.00	Yes
Experiment	3447.6227		927.0222		737.5335		301.8		157.3			

Table S-3b: Rotational constants, the dihedral angle *α* and the predicted torsional barriers of *anti*-2A3MT optimized at various levels of theory. The deviations between the calculated and the experimental values of the rotational constants are given as ΔA, ΔB, and ΔC. $V_{3,1}$ is the barrier of the acetyl methyl and $V_{3,2}$ that of the ring methyl group. All optimizations were performed under full geometry relaxation.

Method/Basis Set	*A* / MHz	*ΔA* / MHz	*B* / MHz	*ΔB* / MHz	*C* / MHz	*ΔC* / MHz	$V_{3.1}$ / cm^{-1}	$\Delta V_{3.1}$ / cm^{-1}	$V_{3.2}$ / cm^{-1}	$\Delta V_{3.2}$ / cm^{-1}	α / °	Planar
B3LYP-D3/6-31G(d,p)	1990.5429	–7.4	1398.6845	–13.3	829.8316	–6.5	306.1	0.1	253.6	68.2	179.99	Yes
B3LYP-D3/6-31+G(d,p)	1984.9254	–13.1	1397.2310	–14.7	828.3614	–7.9	243.8	62.4	334.6	–12.8	179.98	Yes
B3LYP-D3/6-31++G(d,p)	1985.0767	–12.9	1397.2922	–14.7	828.4097	–7.9	240.5	65.7	338.5	–16.7	179.97	Yes
B3LYP-D3/6-311G(d,p)	1995.3479	–2.6	1401.2186	–10.7	831.5498	–4.7	258.4	47.8	264.4	57.4	180.01	Yes
B3LYP-D3/6-311+G(d,p)	1992.0607	–5.9	1401.4298	–10.5	831.0581	–5.2	235.2	71.0	308.7	13.1	179.98	Yes
B3LYP-D3/6-311++G(d,p)	1992.0453	–5.9	1401.5310	–10.4	831.0911	–5.2	228.9	77.3	306.3	15.5	179.96	Yes
B3LYP-D3/6-311G(2d,2p)	2002.7562	4.8	1406.0075	–6.0	834.4760	–1.8	283.7	22.5	289.8	32.0	179.98	Yes
B3LYP-D3/6-311+G(2d,2p)	1999.4253	1.4	1406.8265	–5.1	834.1922	–2.1	251.9	54.3	330.0	–8.2	179.99	Yes
B3LYP-D3/6-311++G(2d,2p)	1999.3824	1.4	1406.9156	–5.1	834.2164	–2.1	250.8	55.4	328.2	–6.4	179.97	Yes
B3LYP-D3/6-311G(df,pd)	2001.6341	3.7	1405.3576	–6.6	834.0797	–2.2	248.1	58.1	289.9	31.9	179.96	Yes
B3LYP-D3/6-311+G(df,pd)	1998.5140	0.5	1405.5632	–6.4	833.6124	–2.7	223.2	83.0	325.2	–3.4	179.94	Yes
B3LYP-D3/6-311++G(df,pd)	1998.5126	0.5	1405.6433	–6.3	833.6390	–2.6	217.3	88.9	321.6	0.2	179.96	Yes
B3LYP-D3/6-311G(2df,2pd)	2007.5745	9.6	1408.9040	–3.1	836.3291	0.0	281.5	24.7	308.4	13.4	179.99	Yes
B3LYP-D3/6-311+G(2df,2pd)	2004.0825	6.1	1409.3829	–2.6	835.8958	–0.4	252.0	54.2	336.9	–15.1	179.99	Yes
B3LYP-D3/6-311++G(2df,2pd)	2004.0704	6.1	1409.4280	–2.5	835.9100	–0.4	255.8	50.4	336.1	–14.3	179.98	Yes
B3LYP-D3/6-311G(3df,3pd)	2009.2155	11.2	1410.7673	–1.2	837.2723	1.0	272.9	33.3	330.9	–9.1	180.00	Yes
B3LYP-D3/6-311+G(3df,3pd)	2006.4720	8.5	1411.4624	–0.5	837.0448	0.8	257.4	48.8	328.3	–6.5	179.97	Yes
B3LYP-D3/6-311++G(3df,3pd)	2006.4625	8.5	1411.4423	–0.5	837.0356	0.7	258.6	47.6	328.4	–6.6	179.97	Yes
B3LYP-D3/cc-pVDZ	1985.0693	–12.9	1395.8512	–16.1	827.9715	–8.3	328.2	–22.0	264.5	57.3	179.95	Yes
B3LYP-D3/aug-cc-pVDZ	1981.7207	–16.3	1396.3122	–15.7	827.5401	–8.7	260.4	45.8	336.6	–14.8	179.98	Yes
B3LYP-D3/cc-pVTZ	2003.6075	5.6	1408.3749	–3.6	835.4580	–0.8	267.5	38.7	309.7	12.1	179.98	Yes
B3LYP-D3/aug-cc-pVTZ	2002.7443	4.8	1408.8215	–3.1	835.4678	–0.8	255.8	50.4	331.3	–9.5	179.96	Yes
B3LYP-D3BJ/6-31G(d,p)	1989.3355	–8.6	1403.9340	–8.0	831.4701	–4.8	263.4	42.8	201.5	120.3	179.97	Yes

B3LYP-D3BJ/6-31+G(d,p)	1983.5530	–14.4	1402.5249	–9.4	829.9829	–6.3	200.3	105.9	291.5	30.3	179.99	Yes
B3LYP-D3BJ/6-31++G(d,p)	1983.5890	–14.4	1402.5097	–9.5	829.9838	–6.3	196.7	109.5	295.2	26.6	179.99	Yes
B3LYP-D3BJ/6-311G(d,p)	1993.9149	–4.1	1406.6604	–5.3	833.2169	–3.1	212.4	93.8	215.5	106.3	179.99	Yes
B3LYP-D3BJ/6-311+G(d,p)	1990.4953	–7.5	1406.7845	–5.2	832.6682	–3.6	188.8	117.4	265.9	55.9	179.97	Yes
B3LYP-D3BJ/6-311++G(d,p)	1990.4984	–7.5	1406.9541	–5.0	832.7281	–3.6	182.7	123.5	263.4	58.4	180.00	Yes
B3LYP-D3BJ/6-311G(2d,2p)	2001.1254	3.1	1411.5147	–0.5	836.1318	–0.2	237.3	68.9	241.3	80.5	180.00	Yes
B3LYP-D3BJ/6-311+G(2d,2p)	**1997.6762**	**–0.3**	**1412.2869**	**0.3**	**835.8065**	**–0.5**	**204.7**	101.5	287.5	34.3	**179.99**	**Yes**
B3LYP-D3BJ/6-311++G(2d,2p)	**1997.6437**	**–0.3**	**1412.3745**	**0.4**	**835.8317**	**–0.5**	**208.5**	97.7	285.7	36.1	**179.99**	**Yes**
B3LYP-D3BJ/6-311G(df,pd)	2000.0432	2.1	1410.8775	–1.1	835.7468	–0.5	201.0	105.2	240.8	81.0	179.99	Yes
B3LYP-D3BJ/6-311+G(df,pd)	1996.7614	–1.2	1411.0030	–1.0	835.2201	–1.1	175.8	130.4	282.1	39.7	179.98	Yes
B3LYP-D3BJ/6-311++G(df,pd)	1996.7909	–1.2	1411.0942	–0.9	835.2559	–1.0	170.1	136.1	278.3	43.5	179.99	Yes
B3LYP-D3BJ/6-311G(2df,2pd)	2005.7422	7.8	1414.4533	2.5	837.9635	1.7	234.8	71.4	260.0	61.8	180.00	Yes
B3LYP-D3BJ/6-311+G(2df,2pd)	2002.2228	4.2	1414.8468	2.9	837.4927	1.2	204.8	101.4	294.6	27.2	180.00	Yes
B3LYP-D3BJ/6-311++G(2df,2pd)	2002.2076	4.2	1414.8985	2.9	837.5085	1.2	208.6	97.6	293.7	28.1	180.00	Yes
B3LYP-D3BJ/6-311G(3df,3pd)	2007.3432	9.4	1416.3203	4.4	838.9013	2.6	226.4	79.8	283.9	37.9	180.00	Yes
B3LYP-D3BJ/6-311+G(3df,3pd)	2004.5105	6.5	1416.9671	5.0	838.6376	2.3	209.8	96.4	285.2	36.6	180.00	Yes
B3LYP-D3BJ/6-311++G(3df,3pd)	2004.5158	6.5	1416.9491	5.0	838.6317	2.3	210.9	95.3	285.3	36.5	180.00	Yes
B3LYP-D3BJ/cc-pVDZ	1983.7864	–14.2	1401.3538	–10.6	829.6832	–6.6	284.3	21.9	211.9	109.9	180.00	Yes
B3LYP-D3BJ/aug-cc-pVDZ	1980.2412	–17.7	1401.7828	–10.2	829.2029	–7.1	215.1	91.1	294.3	27.5	179.99	Yes
B3LYP-D3BJ/cc-pVTZ	2001.7832	3.8	1413.8759	1.9	837.0754	0.8	220.9	85.3	265.4	56.4	179.99	Yes
B3LYP-D3BJ/aug-cc-pVTZ	2000.8540	2.9	1414.3229	2.4	837.0719	0.8	209.0	97.2	289.1	32.7	179.99	Yes
CAM-B3LYP-D3BJ/6-311G(d,p)	2013.3968	15.4	1415.7122	3.7	839.7808	3.5	232.3	73.9	208.4	113.4	179.99	Yes
CAM-B3LYP-D3BJ/6-311+G(d,p)	2009.8536	11.9	1415.8869	3.9	839.2304	2.9	209.6	96.6	260.5	61.3	180.01	Yes
CAM-B3LYP-D3BJ/6-311++G(d,p)	2009.8346	11.9	1416.0094	4.0	839.2699	3.0	205.5	100.7	257.6	64.2	179.99	Yes
CAM-B3LYP-D3BJ/cc-pVDZ	2003.2950	5.3	1410.0786	–1.9	836.1384	–0.2	296.4	9.8	188.6	133.2	179.99	Yes
CAM-B3LYP-D3BJ/aug-cc-pVDZ	1999.5818	1.6	1410.6008	–1.4	835.6632	–0.6	230.6	75.6	277.5	44.3	179.99	Yes
CAM-B3LYP-D3BJ/aug-cc-pVTZ	2020.1182	22.1	1423.3993	11.4	843.6086	7.3	228.1	78.1	280.4	41.4	179.99	Yes
CAM-B3LYP-D3BJ/cc-pVTZ	2021.1858	23.2	1423.0308	11.1	843.6630	7.4	240.8	65.4	255.3	66.5	179.97	Yes
CCSD/cc-pVDZ	1970.5678	–27.4	1389.2197	–22.7	823.2340	–13.1	376.8	–70.6	335.8	–14.0	179.96	Yes
M06-2X/6-31G(d,p)	2006.5618	8.6	1413.6710	1.7	837.9107	1.6	332.9	–26.7	312.1	9.7	180.00	Yes

M06-2X/6-31+G(d,p)	2001.7245	3.7	1412.3014	0.3	836.6018	0.3	278.0	28.2	377.6	–55.8	179.99	Yes
M06-2X/6-31++G(d,p)	2001.6981	3.7	1412.3336	0.4	836.6083	0.3	274.5	31.7	381.4	–59.6	179.99	Yes
M06-2X/6-311G(d,p)	2009.6389	11.7	1415.9836	4.0	839.2501	3.0	305.1	1.1	340.8	–19.0	180.00	Yes
M06-2X/6-311+G(d,p)	2006.8048	8.8	1415.9801	4.0	838.7573	2.5	292.8	13.4	372.3	–50.5	179.98	Yes
M06-2X/6-311++G(d,p)	2006.7485	8.8	1416.1132	4.1	838.7948	2.5	288.4	17.8	369.8	–48.0	180.00	Yes
M06-2X/6-311G(df,pd)	2014.7269	16.8	1419.2845	7.3	841.2772	5.0	292.2	14.0	358.7	–36.9	179.99	Yes
M06-2X/6-311+G(df,pd)	2012.0969	14.1	1419.2922	7.3	840.8209	4.5	279.0	27.2	382.8	–61.0	179.99	Yes
M06-2X/6-311++G(df,pd)	2012.0133	14.0	1419.3965	7.4	840.8423	4.6	274.8	31.4	379.2	–57.4	179.98	Yes
M06-2X/6-311G(3df,3pd)	2021.5796	23.6	1424.1581	12.2	844.1515	7.9	344.0	–37.8	391.0	–69.2	179.96	Yes
M06-2X/6-311+G(3df,3pd)	2019.2558	21.3	1424.7666	12.8	843.9641	7.7	333.8	–27.6	386.4	–64.6	179.98	Yes
M06-2X/6-311++G(3df,3pd)	2019.2421	21.3	1424.7483	12.8	843.9546	7.7	335.4	–29.2	386.3	–64.5	179.98	Yes
M06-2X/cc-pVDZ	2002.7737	4.8	1411.5358	–0.4	836.5816	0.3	347.1	–40.9	312.1	9.7	179.98	Yes
M06-2X/aug-cc-pVDZ	1999.9174	1.9	1411.4927	–0.5	836.0539	–0.2	308.1	–1.9	367.0	–45.2	180.00	Yes
M06-2X/cc-pVTZ	2017.1889	19.2	1423.0174	11.1	842.9903	6.7	325.0	–18.8	364.4	–42.6	179.94	Yes
M06-2X/aug-cc-pVTZ	2016.5849	18.6	1423.3360	11.4	842.9959	6.7	318.5	–12.3	383.7	–61.9	179.98	Yes
MP2/6-31G(d,p)	**1998.0135**	**0.0**	**1411.1488**	**–0.8**	**835.4992**	**–0.8**	**365.0**	–58.8	343.4	–21.6	**180.01**	**Yes**
MP2/6-31+G(d,p)	1991.4849	–6.5	1406.9802	–5.0	834.6421	–1.6	288.2	18.0	412.8	–91.0	174.06	No
MP2/6-31++G(d,p)	1991.3537	–6.6	1406.0562	–5.9	835.5509	–0.7	303.5	2.7	394.7	–72.9	173.11	No
MP2/6-311G(d,p)	1994.1937	–3.8	1410.2776	–1.7	834.5987	–1.7	306.9	–0.7	410.8	–89.0	180.02	Yes
MP2/6-311+G(d,p)	1991.4344	–6.5	1407.1605	–4.8	836.0273	–0.3	305.0	1.2	439.8	–118.0	172.58	No
MP2/6-311++G(d,p)	1991.4877	–6.5	1406.2009	–5.8	836.8944	0.6	316.1	–9.9	424.8	–103.0	171.47	No
MP2/6-311G(2d,2p)	2000.1514	2.2	1416.5220	4.6	837.7125	1.4	301.0	5.2	358.8	–37.0	180.00	Yes
MP2/6-311+G(2d,2p)	1996.1020	–1.9	1417.2861	5.3	837.2749	1.0	279.7	26.5	423.5	–101.7	180.01	Yes
MP2/6-311++G(2d,2p)	1996.0517	–1.9	1417.4463	5.5	837.3241	1.0	244.3	61.9	417.2	–95.4	180.01	Yes
MP2/6-311G(df,pd)	2008.5029	10.5	1419.4563	7.5	840.5681	4.3	276.6	29.6	393.0	–71.2	177.91	No
MP2/6-311+G(df,pd)	2005.5448	7.6	1414.5150	2.5	842.3849	6.1	293.2	13.0	417.7	–95.9	171.16	No
MP2/6-311++G(df,pd)	2005.4402	7.5	1414.2446	2.3	843.0686	6.8	307.0	–0.8	404.8	–83.0	170.80	No
MP2/6-311G(2df,2pd)	2011.1884	13.2	1425.1979	13.2	842.6909	6.4	295.2	11.0	359.7	–37.9	179.93	Yes
MP2/6-311+G(2df,2pd)	2006.6522	8.7	1425.2066	13.2	841.9037	5.6	271.3	34.9	406.5	–84.7	180.00	Yes
MP2/6-311++G(2df,2pd)	2006.6585	8.7	1425.3283	13.4	841.9489	5.7	274.2	32.0	405.3	–83.5	180.00	Yes

MP2/6-311G(3df,3pd)	2010.2913	12.3	1425.6410	13.7	842.7080	6.4	303.1	3.1	382.2	–60.4	179.96	Yes
MP2/6-311+G(3df,3pd)	2006.8342	8.9	1425.8502	13.9	842.1781	5.9	266.5	39.7	389.4	–67.6	179.97	Yes
MP2/6-311++G(3df,3pd)	2006.8395	8.9	1425.8420	13.9	842.1758	5.9	273.7	32.5	389.2	–67.4	179.96	Yes
MP2/cc-pVDZ	1975.7803	–22.2	1398.1949	–13.8	827.2509	–9.0	378.2	–72.0	332.7	–10.9	179.99	Yes
MP2/aug-cc-pVDZ	1966.2899	–31.7	1398.5114	–13.5	825.7057	–10.6	271.7	34.5	400.0	–78.2	179.97	Yes
MP2/cc-pVTZ	2003.7456	5.8	1421.8441	9.9	840.2247	3.9	281.5	24.7	361.4	–39.6	179.96	Yes
MP2/aug-cc-pVTZ	2001.9133	3.9	1422.6234	10.7	840.1936	3.9	[a]		[a]		180.00	Yes
ωB97X-D/6-311G(d,p)	2005.5701	7.6	1411.6054	–0.4	837.0132	0.7	232.4	73.8	285.7	36.1	180.00	Yes
ωB97X-D/6-311+G(d,p)	2005.5318	7.6	1411.7392	–0.2	837.0534	0.8	227.1	79.1	282.9	38.9	180.00	Yes
ωB97X-D/6-311++G(d,p)	2008.5172	10.5	1411.5709	–0.4	837.5148	1.2	248.9	57.3	243.9	77.9	179.69	Yes
ωB97X-D/aug-cc-pVDZ	1995.3205	–2.7	1406.4217	–5.5	833.4901	–2.8	268.1	38.1	290.9	30.9	179.68	Yes
ωB97X-D/aug-cc-pVTZ	2015.7562	17.8	1418.8771	6.9	841.2891	5.0	264.4	41.8	307.6	14.2	179.99	Yes
ωB97X-D/cc-pVDZ	1997.9542	0.0	1405.5094	–6.5	833.6362	–2.7	301.8	4.4	207.2	114.6	179.97	Yes
ωB97X-D/cc-pVTZ	2016.2760	18.3	1418.3646	6.4	841.1981	4.9	265.9	40.3	283.0	38.8	179.99	Yes
Experiment	1997.9755		1411.9663		836.2886		306.2		321.8			Yes

[a] Not done due to high quantum chemical cost.

Table S-3c: Rotational constants and the dihedral angle α of *syn*-2A4MT optimized at various levels of theory. The deviations between the calculated and the experimental values of the rotational constants are given as ΔA, ΔB and ΔC. $V_{3,1}$ is the barrier of the acetyl methyl and $V_{3,2}$ that of the ring methyl group. All optimizations were performed under full-symmetry relaxation.

Method/Basis Set	*A* / MHz	*ΔA* / MHz	*B* / MHz	*ΔB* / MHz	*C* / MHz	*ΔC* / MHz	$V_{3,1}$ / cm^{-1}	$\Delta V_{3.1}$ / cm^{-1}	$V_{3,2}$ / cm^{-1}	$\Delta V_{3.2}$ / cm^{-1}	*α* / °	Planar
B3LYP-D3/6-31G(d,p)	2601.4782	–43.4	1002.7369	–3.5	730.3342	–5.5	307.4	17.5	214.1	–3.4	0.01	Yes
B3LYP-D3/6-31+G(d,p)	2591.2296	–53.7	1001.5151	–4.7	728.8929	–6.9	244.8	80.1	198.2	12.5	0.01	Yes
B3LYP-D3/6-31++G(d,p)	2591.3320	–53.6	1001.4868	–4.7	728.8861	–6.9	243.4	81.5	200.3	10.4	0.01	Yes
B3LYP-D3/6-311G(d,p)	2607.2534	–37.7	1005.0833	–1.2	732.0281	–3.8	298.1	26.8	206.3	4.4	0.02	Yes
B3LYP-D3/6-311+G(d,p)	2599.5519	–45.4	1004.9105	–1.3	731.3305	–4.5	255.6	69.3	206.5	4.2	0.02	Yes
B3LYP-D3/6-311++G(d,p)	2599.4792	–45.4	1004.9117	–1.3	731.3251	–4.5	257.1	67.8	203.1	7.6	0.02	Yes
B3LYP-D3/6-311G(2d,2p)	2619.2060	–25.7	1008.0091	1.8	734.4795	–1.3	316.0	8.9	194.5	16.2	0.00	Yes
B3LYP-D3/6-311+G(2d,2p)	2612.8810	–32.0	1008.3118	2.1	734.1448	–1.6	264.9	60.0	193.0	17.7	0.01	Yes
B3LYP-D3/6-311++G(2d,2p)	2612.9388	–32.0	1008.3026	2.1	734.1445	–1.6	267.7	57.2	191.6	19.1	0.01	Yes
B3LYP-D3/6-311G(df,pd)	2613.8997	–31.0	1008.0801	1.8	734.1254	–1.7	287.5	37.4	203.0	7.7	0.00	Yes
B3LYP-D3/6-311+G(df,pd)	2606.6722	–38.3	1008.0817	1.8	733.5557	–2.2	246.4	78.5	204.7	6.0	0.01	Yes
B3LYP-D3/6-311++G(df,pd)	2606.6226	–38.3	1008.0911	1.9	733.5556	–2.2	251.3	73.6	201.7	9.0	0.01	Yes
B3LYP-D3/6-311G(2df,2pd)	2626.7024	–18.2	1009.6178	3.4	735.9201	0.1	320.1	4.8	189.9	20.8	0.00	Yes
B3LYP-D3/6-311+G(2df,2pd)	2620.3221	–24.6	1009.7213	3.5	735.4757	–0.3	268.5	56.4	189.6	21.1	0.01	Yes
B3LYP-D3/6-311++G(2df,2pd)	2620.3391	–24.6	1009.7076	3.5	735.4699	–0.3	271.2	53.7	187.2	23.5	0.01	Yes
B3LYP-D3/6-311G(3df,3pd)	2631.2874	–13.6	1009.9583	3.7	736.4635	0.7	304.5	20.4	186.1	24.6	0.01	Yes
B3LYP-D3/6-311+G(3df,3pd)	2626.1748	–18.8	1010.3115	4.1	736.2511	0.5	279.0	45.9	187.6	23.1	0.01	Yes
B3LYP-D3/6-311++G(3df,3pd)	2626.1571	–18.8	1010.3074	4.1	736.2472	0.5	278.3	46.6	186.5	24.2	0.01	Yes
B3LYP-D3/cc-pVDZ	2595.6788	–49.2	1000.2675	–6.0	728.6418	–7.1	361.8	–36.9	221.2	–10.5	0.03	Yes
B3LYP-D3/aug-cc-pVDZ	2585.1179	–59.8	1000.7469	–5.5	728.0478	–7.7	266.7	58.2	206.8	3.9	0.01	Yes
B3LYP-D3/cc-pVTZ	2619.1081	–25.8	1009.3731	3.1	735.1963	–0.6	279.6	45.3	196.6	14.1	0.01	Yes
B3LYP-D3/aug-cc-pVTZ	2617.2079	–27.7	1009.5244	3.3	735.1275	–0.7	265.1	59.8	194.5	16.2	0.01	Yes
B3LYP-D3BJ/6-31G(d,p)	2612.8340	–32.1	1003.6541	–2.6	731.7123	–4.1	273.8	51.1	216.0	–5.3	0.01	Yes

B3LYP-D3BJ/6-31+G(d,p)	2602.3153	–42.6	1002.5002	–3.7	730.2883	–5.5	209.8	115.1	200.2	10.5	0.01	Yes
B3LYP-D3BJ/6-31++G(d,p)	2602.4267	–42.5	1002.4709	–3.8	730.2817	–5.5	208.4	116.5	202.4	8.3	0.01	Yes
B3LYP-D3BJ/6-311G(d,p)	2618.3454	–26.6	1006.0078	–0.2	733.3895	–2.4	264.4	60.5	208.3	2.4	0.02	Yes
B3LYP-D3BJ/6-311+G(d,p)	2610.5933	–34.3	1006.0068	–0.2	732.7816	–3.0	219.9	105.0	208.4	2.3	0.01	Yes
B3LYP-D3BJ/6-311++G(d,p)	2610.5255	–34.4	1006.0075	–0.2	732.7764	–3.0	221.4	103.5	205.0	5.7	0.01	Yes
B3LYP-D3BJ/6-311G(2d,2p)	2630.3615	–14.6	1008.9671	2.7	735.8633	0.1	282.9	42.0	196.5	14.2	0.02	Yes
B3LYP-D3BJ/6-311+G(2d,2p)	2623.9337	–21.0	1009.3067	3.1	735.5415	–0.2	229.5	95.4	194.9	15.8	0.00	Yes
B3LYP-D3BJ/6-311++G(2d,2p)	2623.9948	–20.9	1009.2975	3.1	735.5415	–0.2	232.3	92.6	193.6	17.1	0.00	Yes
B3LYP-D3BJ/6-311G(df,pd)	2624.7854	–20.1	1009.0603	2.8	735.5007	–0.3	253.0	71.9	204.9	5.8	0.00	Yes
B3LYP-D3BJ/6-311+G(df,pd)	2617.4033	–27.5	1009.1352	2.9	734.9602	–0.8	210.0	114.9	206.6	4.1	0.00	Yes
B3LYP-D3BJ/6-311++G(df,pd)	2617.3557	–27.6	1009.1443	2.9	734.9601	–0.8	214.9	110.0	203.6	7.1	0.00	Yes
B3LYP-D3BJ/6-311G(2df,2pd)	2637.7344	–7.2	1010.5188	4.3	737.2615	1.5	286.2	38.7	191.9	18.8	0.00	Yes
B3LYP-D3BJ/6-311+G(2df,2pd)	2631.0756	–13.9	1010.7070	4.5	736.8427	1.1	232.8	92.1	191.4	19.3	0.00	Yes
B3LYP-D3BJ/6-311++G(2df,2pd)	2631.0962	–13.8	1010.6930	4.5	736.8370	1.0	235.5	89.4	189.1	21.6	0.00	Yes
B3LYP-D3BJ/6-311G(3df,3pd)	2642.2993	–2.6	1010.8504	4.6	737.7972	2.0	270.7	54.2	188.1	22.6	0.00	Yes
B3LYP-D3BJ/6-311+G(3df,3pd)	2636.8637	–8.1	1011.2921	5.1	737.6087	1.8	243.1	81.8	189.5	21.2	0.00	Yes
B3LYP-D3BJ/6-311++G(3df,3pd)	2636.8489	–8.1	1011.2878	5.1	737.6050	1.8	242.4	82.5	188.4	22.3	0.00	Yes
B3LYP-D3BJ/cc-pVDZ	2607.2897	–37.6	1001.2629	–5.0	730.0814	–5.7	327.9	–3.0	223.1	–12.4	0.02	Yes
B3LYP-D3BJ/aug-cc-pVDZ	2596.2485	–48.7	1001.5802	–4.7	729.3670	–6.4	229.1	95.8	208.8	1.9	0.02	Yes
B3LYP-D3BJ/cc-pVTZ	2630.0196	–14.9	1010.3374	4.1	736.5644	0.8	244.4	80.5	198.5	12.2	0.00	Yes
B3LYP-D3BJ/aug-cc-pVTZ	2628.0400	–16.9	1010.5210	4.3	736.5073	0.7	229.1	95.8	196.4	14.3	0.00	Yes
CAM-B3LYP-D3BJ/6-311G(d,p)	2645.0327	0.1	1012.3437	6.1	738.8379	3.0	289.0	35.9	220.1	–9.4	0.00	Yes
CAM-B3LYP-D3BJ/6-311+G(d,p)	2636.7089	–8.2	1012.4586	6.2	738.2510	2.5	245.9	79.0	223.0	–12.3	0.01	Yes
CAM-B3LYP-D3BJ/6-311++G(d,p)	2636.6305	–8.3	1012.4620	6.2	738.2466	2.5	248.3	76.6	220.0	–9.3	0.01	Yes
CAM-B3LYP-D3BJ/cc-pVDZ	2634.6978	–10.2	1007.3015	1.1	735.4288	–0.4	343.6	–18.7	237.4	–26.7	0.01	Yes
CAM-B3LYP-D3BJ/aug-cc-pVDZ	2622.2020	–22.7	1007.9473	1.7	734.7802	–1.0	251.8	73.1	229.9	–19.2	0.01	Yes
CAM-B3LYP-D3BJ/cc-pVTZ	2655.7364	10.8	1016.8809	10.6	742.0484	6.3	269.5	55.4	215.6	–4.9	0.01	Yes
CAM-B3LYP-D3BJ/aug-cc-pVTZ	2653.3824	8.5	1017.0339	10.8	741.9462	6.2	254.9	70.0	215.6	–4.9	0.02	Yes
CCSD-cc-pVDZ	2589.7646	–55.2	992.8722	–13.4	724.3161	–11.5	416.8	–91.9	181.7	29.0	0.01	Yes
M06-2X/6-31G(d,p)	2635.9870	–8.9	1009.0625	2.8	736.4066	0.6	355.9	–31.0	226.3	–15.6	0.01	Yes

M06-2X/6-31+G(d,p)	2627.7528	–17.2	1007.8931	1.7	735.1554	–0.6	303.6	21.3	215.9	–5.2	0.01	Yes
M06-2X/6-31++G(d,p)	2627.8255	–17.1	1007.8828	1.6	735.1559	–0.6	302.4	22.5	218.0	–7.3	0.01	Yes
M06-2X/6-311G(d,p)	2637.7788	–7.1	1011.3133	5.1	737.7395	1.9	362.7	–37.8	216.9	–6.2	0.02	Yes
M06-2X/6-311+G(d,p)	2631.4442	–13.5	1011.1984	5.0	737.1835	1.4	333.8	-8.9	221.6	–10.9	0.00	Yes
M06-2X/6-311++G(d,p)	2631.3433	–13.6	1011.2132	5.0	737.1836	1.4	335.3	–10.4	217.8	–7.1	0.00	Yes
M06-2X/6-311G(df,pd)	2642.4006	–2.5	1013.7400	7.5	739.3770	3.6	348.6	–23.7	211.7	–1.0	0.01	Yes
M06-2X/6-311+G(df,pd)	2636.3481	–8.6	1013.6732	7.4	738.8661	3.1	321.3	3.6	217.6	–6.9	0.01	Yes
M06-2X/6-311++G(df,pd)	2636.2790	–8.6	1013.6931	7.5	738.8705	3.1	327.1	–2.2	214.3	–3.6	0.01	Yes
M06-2X/6-311G(3df,3pd)	2657.3824	12.5	1015.3633	9.1	741.3826	5.6	385.4	–60.5	208.7	2.0	0.02	Yes
M06-2X/6-311+G(3df,3pd)	2653.2325	8.3	1015.7277	9.5	741.2544	5.5	367.6	–42.7	210.6	0.1	0.02	Yes
M06-2X/6-311++G(3df,3pd)	2653.2271	8.3	1015.7186	9.5	741.2488	5.5	367.2	–42.3	209.7	1.0	0.02	Yes
M06-2X/cc-pVDZ	2632.8818	–12.0	1007.0152	0.8	735.1449	–0.6	386.6	–61.7	231.0	–20.3	0.01	Yes
M06-2X/aug-cc-pVDZ	2621.4279	–23.5	1007.5902	1.4	734.5411	–1.2	325.5	–0.6	232.2	–21.5	0.00	Yes
M06-2X/cc-pVTZ	2647.5473	2.6	1015.6045	9.4	740.7453	5.0	348.4	–23.5	213.6	–2.9	0.01	Yes
M06-2X/aug-cc-pVTZ	2646.0467	1.1	1015.7707	9.5	740.7156	4.9	341.2	–16.3	215.3	–4.6	0.02	Yes
MP2/6-31G(d,p)	**2638.4658**	**–6.5**	**1006.2736**	**0.0**	**735.0768**	**–0.7**	**370.2**	**–45.3**	**184.2**	**26.5**	**0.00**	**Yes**
MP2/6-31+G(d,p)	2627.0088	–17.9	1004.5288	–1.7	733.2786	–2.5	285.2	39.7	176.2	34.5	0.01	Yes
MP2/6-31++G(d,p)	2626.8646	–18.1	1004.4746	–1.8	733.2406	–2.6			182.3	28.4	-0.06	Yes
MP2/6-311G(d,p)	2635.1569	–9.8	1004.6614	–1.6	734.0230	–1.8	352.6	–27.7	164.8	45.9	0.04	Yes
MP2/6-311+G(d,p)	2625.8529	–19.1	1004.4667	–1.8	733.2009	–2.6	311.4	13.5	172.3	38.4	0.00	Yes
MP2/6-311++G(d,p)	2625.8529	–19.1	1004.4667	–1.8	733.2009	–2.6	314.3	10.6	165.7	45.0	0.00	Yes
MP2/6-311G(2d,2p)	2635.6490	–9.3	1009.4822	3.2	736.5367	0.7	355.2	–30.3	156.4	54.3	0.02	Yes
MP2/6-311+G(2d,2p)	2628.5531	–16.4	1009.7794	3.5	736.1437	0.4	303.7	21.2	153.6	57.1	0.01	Yes
MP2/6-311++G(2d,2p)	2628.6870	–16.2	1009.7233	3.5	736.1260	0.3	307.2	17.7	151.7	59.0	0.01	Yes
MP2/6-311G(df,pd)	2654.3153	9.4	1010.4405	4.2	738.5643	2.8	334.9	–10.0	161.8	48.9	0.00	Yes
MP2/6-311+G(df,pd)	**2644.8388**	**–0.1**	**1009.9215**	**3.7**	**737.5579**	**1.8**	**286.0**	**38.9**	**167.1**	**43.6**	**–0.03**	**Yes**
MP2/6-311++G(df,pd)	**2644.5603**	**–0.4**	**1009.8853**	**3.7**	**737.5160**	**1.7**			**162.5**	**48.2**	**–0.12**	**Yes**
MP2/6-311G(2df,2pd)	2660.6414	15.7	1013.3557	7.1	740.5544	4.8	350.8	–25.9	143.5	67.2	0.01	Yes
MP2/6-311+G(2df,2pd)	2652.4601	7.5	1013.2104	7.0	739.8459	4.1	295.4	29.5	144.4	66.3	0.01	Yes
MP2/6-311++G(2df,2pd)	2652.3683	7.4	1013.2005	7.0	739.8344	4.0	301.0	23.9	141.2	69.5	0.02	Yes

MP2/6-311G(3df,3pd)	2666.0479	21.1	1011.9974	5.8	740.2604	4.5	353.9	–29.0	137.5	73.2	0.01	Yes
MP2/6-311+G(3df,3pd)	2659.9035	15.0	1012.1462	5.9	739.8668	4.1	311.3	13.6	141.1	69.6	0.01	Yes
MP2/6-311++G(3df,3pd)	2659.9087	15.0	1012.1573	5.9	739.8733	4.1	311.0	13.9	140.6	70.1	0.01	Yes
MP2/cc-pVDZ	2613.4345	–31.5	995.3465	–10.9	727.4509	–8.3	418.0	–93.1	184.3	26.4	0.00	Yes
MP2/aug-cc-pVDZ	2597.4151	–47.5	994.5343	–11.7	725.7778	–10.0	290.1	34.8	166.0	44.7	0.00	Yes
MP2/cc-pVTZ	2650.9636	6.0	1011.1248	4.9	738.6209	2.8	308.6	16.3	153.5	57.2	0.00	Yes
MP2/aug-cc-pVTZ	2648.3007	3.4	1011.2585	5.0	738.4997	2.7					0.01	Yes
ωB97X-D/6-311G(d,p)	2629.8921	–15.0	1010.8145	4.6	736.8607	1.1	290.7	34.2	210.2	0.5	0.00	Yes
ωB97X-D/6-311+G(d,p)	2629.7781	–15.2	1010.8188	4.6	736.8538	1.1	293.9	31.0	206.8	3.9	0.00	Yes
ωB97X-D/6-311++G(d,p)	2636.7931	–8.1	1010.8193	4.6	737.4018	1.6	320.4	4.5	206.1	4.6	0.00	Yes
ωB97X-D/aug-cc-pVDZ	2616.6259	–28.3	1005.9957	–0.2	733.3220	–2.5	310.2	14.7	221.8	–11.1	0.01	Yes
ωB97X-D/aug-cc-pVTZ	2646.6516	1.7	1015.1397	8.9	740.4266	4.6	308.3	16.6	202.9	7.8	0.01	Yes
ωB97X-D/cc-pVDZ	2628.3348	–16.6	1005.1752	–1.1	733.8127	–2.0	358.4	–33.5	222.0	–11.3	0.02	Yes
ωB97X-D/cc-pVTZ	2648.4300	3.5	1014.8613	8.6	740.4173	4.6	312.4	12.5	202.0	8.7	0.01	Yes
Experiment	2644.9282		1006.2336		735.7908		324.9		210.7			

Table S-3d: Rotational constants of *anti*-2A4MT optimized at various levels of theory. The deviations between the calculated and the experimental values of the rotational constants are given as ΔA, ΔB and ΔC. $V_{3,1}$ is the barrier of the acetyl methyl and $V_{3,2}$ that of the ring methyl group. All optimizations were performed under full-symmetry relaxation.

Method/Basis Set	*A* / MHz	*ΔA* / MHz	*B* / MHz	*ΔB* / MHz	*C* / MHz	*ΔC* / MHz	$V_{3,1}$ / cm^{-1}	$\Delta V_{3.1}$ / cm^{-1}	$V_{3,2}$ / cm^{-1}	$\Delta V_{3.2}$ / cm^{-1}	α / °	Planar
B3LYP-D3/6-31G(d,p)	2673.5389	–57.0	988.7175	–2.7	728.3155	–6.0	293.4	-12.2	234.3	–21.3	179.98	Yes
B3LYP-D3/6-31+G(d,p)	2677.1494	–53.4	986.1828	–5.3	727.2193	–7.1	221.0	60.2	215.6	–2.6	–179.97	Yes
B3LYP-D3/6-31++G(d,p)	2677.3617	–53.2	986.1525	–5.3	727.2187	–7.1	215.3	65.9	216.4	–3.4	–179.99	Yes
B3LYP-D3/6-311G(d,p)	2682.3126	–48.3	990.6725	–0.8	730.0208	–4.3	251.3	29.9	224.3	–11.3	–179.99	Yes
B3LYP-D3/6-311+G(d,p)	2686.9337	–43.6	989.4651	–2.0	729.7072	–4.6	220.2	61.0	220.6	–7.6	–179.99	Yes
B3LYP-D3/6-311++G(d,p)	2686.9523	–43.6	989.4849	–2.0	729.7195	–4.6	223.6	57.6	216.2	–3.2	180.01	Yes
B3LYP-D3/6-311G(2d,2p)	2698.0544	–32.5	993.1695	1.7	732.5025	–1.8	276.4	4.8	200.9	12.1	–179.99	Yes
B3LYP-D3/6-311+G(2d,2p)	2702.5715	–28.0	992.4287	1.0	732.4325	–1.8	234.5	46.7	200.4	12.6	180.01	Yes
B3LYP-D3/6-311++G(2d,2p)	2702.6846	–27.9	992.4343	1.0	732.4441	–1.8	239.2	42.0	198.6	14.4	180.01	Yes
B3LYP-D3/6-311G(df,pd)	2690.6927	–39.9	993.7649	2.3	732.3055	–2.0	239.7	41.5	220.5	–7.5	–179.96	Yes
B3LYP-D3/6-311+G(df,pd)	2695.4562	–35.1	992.5417	1.1	731.9909	–2.3	207.9	73.3	219.3	–6.3	–179.99	Yes
B3LYP-D3/6-311++G(df,pd)	2695.4669	–35.1	992.5676	1.1	732.0049	–2.3	213.4	67.8	214.8	–1.8	180.00	Yes
B3LYP-D3/6-311G(2df,2pd)	2706.7202	–23.9	994.8561	3.4	734.0552	–0.2	271.4	9.8	196.8	16.2	180.01	Yes
B3LYP-D3/6-311+G(2df,2pd)	2711.6058	–19.0	993.8990	2.4	733.8926	–0.4	234.5	46.7	196.7	16.3	180.01	Yes
B3LYP-D3/6-311++G(2df,2pd)	2711.6608	–18.9	993.8964	2.4	733.8956	–0.4	238.6	42.6	194.1	18.9	180.01	Yes
B3LYP-D3/6-311G(3df,3pd)	2715.7476	–14.8	995.1724	3.7	734.8911	0.6	254.6	26.6	188.5	24.5	–179.98	Yes
B3LYP-D3/6-311+G(3df,3pd)	2718.0482	–12.5	994.6161	3.2	734.7565	0.5	240.9	40.3	193.7	19.3	180.01	Yes
B3LYP-D3/6-311++G(3df,3pd)	2718.0005	–12.6	994.5847	3.1	734.7356	0.5	239.8	41.4	192.5	20.5	180.00	Yes
B3LYP-D3/cc-pVDZ	2664.4117	–66.2	986.8474	–4.6	726.6988	–7.6	323.5	–42.3	237.8	–24.8	179.99	Yes
B3LYP-D3/aug-cc-pVDZ	2674.4695	–56.1	984.9770	–6.5	726.4110	–7.9	236.1	45.1	209.5	3.5	180.01	Yes
B3LYP-D3/cc-pVTZ	2705.5037	–25.1	994.1679	2.7	733.5924	–0.7	252.8	28.4	205.5	7.5	–179.97	Yes
B3LYP-D3/aug-cc-pVTZ	2707.5402	–23.0	993.9172	2.5	733.6058	–0.7	237.3	43.9	200.6	12.4	180.01	Yes
B3LYP-D3BJ/6-31G(d,p)	2683.6800	–46.9	989.8207	–1.6	729.6631	–4.6	257.1	24.1	235.8	–22.8	179.99	Yes

B3LYP-D3BJ/6-31+G(d,p)	2687.9700	–42.6	987.2505	–4.2	728.5947	–5.7	182.9	98.3	217.1	–4.1	–179.96	Yes
B3LYP-D3BJ/6-31++G(d,p)	2688.1636	–42.4	987.1791	–4.3	728.5702	–5.7	176.9	104.3	217.9	–4.9	180.02	Yes
B3LYP-D3BJ/6-311G(d,p)	2692.7856	–37.8	991.7359	0.3	731.3705	–2.9	211.0	70.2	226.0	–13.0	180.01	Yes
B3LYP-D3BJ/6-311+G(d,p)	2697.7977	–32.8	990.5326	–0.9	731.0855	–3.2	178.8	102.4	222.0	–9.0	179.99	Yes
B3LYP-D3BJ/6-311++G(d,p)	2697.8456	–32.7	990.5528	–0.9	731.1000	–3.2	182.3	98.9	217.7	–4.7	179.99	Yes
B3LYP-D3BJ/6-311G(2d,2p)	2708.6540	–21.9	994.2569	2.8	733.8718	–0.4	235.3	45.9	202.3	10.7	180.03	Yes
B3LYP-D3BJ/6-311+G(2d,2p)	2713.5010	–17.1	993.4740	2.0	733.8009	–0.5	191.8	89.4	201.7	11.3	–179.98	Yes
B3LYP-D3BJ/6-311++G(2d,2p)	2713.5976	–17.0	993.4957	2.0	733.8201	–0.5	196.7	84.5	199.9	13.1	–179.98	Yes
B3LYP-D3BJ/6-311G(df,pd)	2701.0908	–29.5	994.8229	3.4	733.6465	–0.6	197.9	83.3	222.2	–9.2	180.00	Yes
B3LYP-D3BJ/6-311+G(df,pd)	2706.2617	–24.3	993.6018	2.1	733.3608	–0.9	165.2	116.0	220.8	–7.8	179.96	Yes
B3LYP-D3BJ/6-311++G(df,pd)	2706.3329	–24.2	993.6205	2.2	733.3752	–0.9	170.6	110.6	216.2	–3.2	179.99	Yes
B3LYP-D3BJ/6-311G(2df,2pd)	2717.2634	–13.3	995.9366	4.5	735.4154	1.1	229.6	51.6	198.3	14.7	180.03	Yes
B3LYP-D3BJ/6-311+G(2df,2pd)	2722.4213	–8.2	994.9458	3.5	735.2518	1.0	191.5	89.7	197.9	15.1	–179.99	Yes
B3LYP-D3BJ/6-311++G(2df,2pd)	2722.4917	–8.1	994.9462	3.5	735.2575	1.0	195.6	85.6	195.4	17.6	–179.99	Yes
B3LYP-D3BJ/6-311G(3df,3pd)	2726.3224	–4.3	996.2316	4.8	736.2396	2.0	212.7	68.5	190.0	23.0	–179.97	Yes
B3LYP-D3BJ/6-311+G(3df,3pd)	**2728.8828**	**–1.7**	**995.6649**	**4.2**	**736.1168**	**1.8**	**197.1**	**84.1**	**195.0**	**18.0**	**–179.99**	**Yes**
B3LYP-D3BJ/6-311++G(3df,3pd)	**2728.8540**	**–1.7**	**995.6462**	**4.2**	**736.1042**	**1.8**	**196.0**	**85.2**	**193.9**	**19.1**	**–179.99**	**Yes**
B3LYP-D3BJ/cc-pVDZ	2675.1523	–55.4	987.9894	–3.5	728.1135	–6.2	285.3	–4.1	239.2	–26.2	179.99	Yes
B3LYP-D3BJ/aug-cc-pVDZ	2685.6086	–45.0	986.1277	–5.3	727.8551	–6.4	195.6	85.6	210.9	2.1	–179.97	Yes
B3LYP-D3BJ/cc-pVTZ	2716.2177	–14.4	995.2439	3.8	734.9621	0.7	210.5	70.7	206.8	6.2	–179.97	Yes
B3LYP-D3BJ/aug-cc-pVTZ	2718.4714	–12.1	994.9671	3.5	734.9763	0.7	194.7	86.5	201.9	11.1	–179.99	Yes
CAM-B3LYP-D3BJ/6-311G(d,p)	2716.7748	–13.8	998.6185	7.2	736.8736	2.6	230.6	50.6	240.0	–27.0	180.00	Yes
CAM-B3LYP-D3BJ/6-311+G(d,p)	2722.1944	–8.4	997.3387	5.9	736.5744	2.3	199.7	81.5	239.9	–26.9	179.98	Yes
CAM-B3LYP-D3BJ/6-311++G(d,p)	2722.2625	–8.3	997.3623	5.9	736.5923	2.3	204.3	76.9	235.3	–22.3	180.00	Yes
CAM-B3LYP-D3BJ/cc-pVDZ	2698.9992	–31.6	994.6806	3.2	733.5039	–0.8	299.3	–18.1	256.7	–43.7	180.02	Yes
CAM-B3LYP-D3BJ/aug-cc-pVDZ	2709.9589	–20.6	992.7073	1.3	733.2164	–1.1	213.6	67.6	234.4	–21.4	–179.98	Yes
CAM-B3LYP-D3BJ/cc-pVTZ	2740.8239	10.2	1002.0625	10.6	740.4722	6.2	231.5	49.7	226.6	–13.6	–179.97	Yes
CAM-B3LYP-D3BJ/aug-cc-pVTZ	2743.2407	12.7	1001.7193	10.3	740.4609	6.2	215.6	65.6	224.1	–11.1	–179.98	Yes
CCSD-cc-pVDZ	2659.9505	–70.6	979.3844	–12.1	722.3826	–11.9	366.6	–85.4	193.0	20.0	–179.95	Yes
M06-2X/6-31G(d,p)	2713.0282	–17.6	995.0118	3.6	734.6556	0.4	320.2	–39.0	247.0	–34.0	–180.00	Yes

M06-2X/6-31+G(d,p)	2716.6462	–13.9	992.6814	1.2	733.6610	–0.6	256.7	24.5	233.7	–20.7	180.01	Yes
M06-2X/6-31++G(d,p)	2716.7567	–13.8	992.6909	1.2	733.6747	–0.6	250.9	30.3	233.7	–20.7	180.00	Yes
M06-2X/6-311G(d,p)	2720.5492	–10.0	996.5336	5.1	736.0299	1.8	292.3	–11.1	235.2	–22.2	–179.98	Yes
M06-2X/6-311+G(d,p)	2725.3481	–5.2	995.3236	3.9	735.7194	1.4	274.4	6.8	236.2	–23.2	–179.98	Yes
M06-2X/6-311++G(d,p)	2725.4583	–5.1	995.3539	3.9	735.7445	1.5	279.0	2.2	231.0	–18.0	–179.98	Yes
M06-2X/6-311G(df,pd)	2727.5835	–3.0	998.9027	7.5	737.8219	3.5	276.6	4.6	229.6	–16.6	–179.96	Yes
M06-2X/6-311+G(df,pd)	2732.3101	1.7	997.7455	6.3	737.5321	3.3	259.1	22.1	232.6	–19.6	180.00	Yes
M06-2X/6-311++G(df,pd)	2732.4539	1.9	997.7724	6.3	737.5570	3.3	266.1	15.1	227.4	–14.4	180.00	Yes
M06-2X/6-311G(3df,3pd)	2751.5364	21.0	999.9629	8.5	740.1171	5.8	320.9	–39.7	211.3	1.7	180.01	Yes
M06-2X/6-311+G(3df,3pd)	2753.6205	23.0	999.5225	8.1	740.0272	5.8	314.2	–33.0	216.3	–3.3	–179.98	Yes
M06-2X/6-311++G(3df,3pd)	2753.5907	23.0	999.5047	8.1	740.0150	5.7	313.0	–31.8	215.3	–2.3	–179.98	Yes
M06-2X/cc-pVDZ	2706.2011	–24.4	993.5869	2.1	733.4492	–0.8	340.9	–59.7	248.6	–35.6	–179.99	Yes
M06-2X/aug-cc-pVDZ	2716.0771	–14.5	991.6555	0.2	733.1005	–1.2	286.3	–5.1	233.5	–20.5	–179.99	Yes
M06-2X/cc-pVTZ	2742.9568	12.4	999.7467	8.3	739.3785	5.1	305.5	–24.3	222.6	–9.6	180.01	Yes
M06-2X/aug-cc-pVTZ	2745.6248	15.0	999.4736	8.0	739.4220	5.1	297.2	–16.0	221.4	–8.4	–179.97	Yes
MP2/6-31G(d,p)	2703.1755	–27.4	993.2393	1.8	732.9337	–1.3	341.4	–60.2	197.7	15.3	180.01	Yes
MP2/6-31+G(d,p)	2706.1712	–24.4	989.2247	–2.2	731.8120	–2.5	253.9	27.3	188.2	24.8	175.23	No
MP2/6-31++G(d,p)	2703.0127	–27.6	989.3621	–2.1	732.3712	–1.9	252.6	28.6	191.0	22.0	174.93	No
MP2/6-311G(d,p)	2711.9214	–18.7	990.3318	–1.1	732.0507	–2.2	295.8	–14.6	177.3	35.7	180.00	Yes
MP2/6-311+G(d,p)	2714.3525	–16.2	988.2229	–3.2	732.2135	–2.1	269.9	11.3	176.8	36.2	175.10	No
MP2/6-311++G(d,p)	2712.0149	–18.6	988.1640	–3.3	732.7234	–1.6	266.4	14.8	172.3	40.7	174.08	No
MP2/6-311G(2d,2p)	2717.9579	–12.6	994.3983	2.9	734.6183	0.3	290.0	-8.8	159.1	53.9	–179.98	Yes
MP2/6-311+G(2d,2p)	2722.9976	–7.6	993.4229	2.0	734.4538	0.2	256.5	24.7	156.2	56.8	180.01	Yes
MP2/6-311++G(2d,2p)	2723.4295	–7.2	993.3735	1.9	734.4604	0.2	257.9	23.3	155.4	57.6	180.01	Yes
MP2/6-311G(df,pd)	2734.1347	3.6	996.3579	4.9	736.9312	2.7	264.6	16.6	173.3	39.7	–179.98	Yes
MP2/6-311+G(df,pd)	**2732.7276**	**2.1**	**993.8389**	**2.4**	**737.4365**	**3.2**	**253.2**	**28.0**	**173.7**	**39.3**	**173.67**	**No**
MP2/6-311++G(df,pd)	**2730.7259**	**0.1**	**993.8610**	**2.4**	**737.8011**	**3.5**	**254.3**	**26.9**	**170.1**	**42.9**	**173.28**	**No**
MP2/6-311G(2df,2pd)	2742.1858	11.6	998.8556	7.4	738.8272	4.6	280.6	0.6	148.0	65.0	–179.99	Yes
MP2/6-311+G(2df,2pd)	2747.4517	16.9	997.3091	5.9	738.3627	4.1	246.9	34.3	147.9	65.1	180.00	Yes
MP2/6-311++G(2df,2pd)	2747.5992	17.0	997.3011	5.8	738.3705	4.1	249.4	31.8	145.9	67.1	180.01	Yes

MP2/6-311G(3df,3pd)	2752.9610	22.4	997.3915	5.9	738.8151	4.5	274.8	6.4	138.5	74.5	–179.99	Yes
MP2/6-311+G(3df,3pd)	2755.1990	24.6	996.4282	5.0	738.4483	4.2	247.7	33.5	144.5	68.5	180.02	Yes
MP2/6-311++G(3df,3pd)	2755.2768	24.7	996.3739	4.9	738.4242	4.1	243.8	37.4	142.6	70.4	180.02	Yes
MP2/cc-pVDZ	2678.4668	–52.1	982.6398	–8.8	725.4915	–8.8	260.1	21.1	159.1	53.9	180.01	Yes
MP2/aug-cc-pVDZ	2690.2845	–40.3	978.5267	–12.9	724.1102	–10.2	248.6	32.6	163.0	50.0	180.00	Yes
MP2/cc-pVTZ	2738.3196	7.7	996.0755	4.6	737.0309	2.8					–179.99	Yes
MP2/aug-cc-pVTZ	2740.5904	10.0	995.6018	4.1	736.9496	2.7	366.4	–85.2	193.7	19.3	180.02	Yes
ωB97X-D/6-311G(d,p)	2719.1353	–11.4	994.6788	3.2	734.9196	0.6	221.5	59.7	226.6	–13.6	180.00	Yes
ωB97X-D/6-311+G(d,p)	2719.1954	–11.4	994.6938	3.2	734.9322	0.7	226.4	54.8	221.8	–8.8	179.99	Yes
ωB97X-D/6-311++G(d,p)	2714.7557	–15.8	995.7710	4.3	735.1950	0.9	244.8	36.4	225.5	–12.5	–179.99	Yes
ωB97X-D/aug-cc-pVDZ	2707.7899	–22.8	989.9923	–1.5	731.5950	–2.7	250.8	30.4	226.2	–13.2	–179.99	Yes
ωB97X-D/aug-cc-pVTZ	2741.1694	10.6	998.7434	7.3	738.7002	4.4	249.5	31.7	210.2	2.8	–179.97	Yes
ωB97X-D/cc-pVDZ	2697.0119	–33.6	991.5958	0.1	731.6937	–2.6	300.3	–19.1	240.2	–27.2	180.00	Yes
ωB97X-D/cc-pVTZ	2738.9395	8.4	998.8403	7.4	738.5911	4.3	253.9	27.3	211.6	1.4	–179.97	Yes
Experiment	2730.5803		991.4523		734.2752		281.2		213.0			

Table S-4: Coefficients of the one-dimensional Fourier expansion for the potential energy curves of *syn*- and *anti*-2A5MT given in Figure 3 and Figure S-1 calculated at the MP2/6-31G(d,p), level of theory. The potential is expanded as $V(\varphi) = \sum_{i=0}^{2} a_i f_i$.

		syn				***anti***			
		Acetyl Methyl		**Ring Methyl**		**Acetyl Methyl**		**Ring Methyl**	
i	**f_i**	**a_i / Hartree**	**a_i / cm^{-1}**	**a_i / Hartree**	**a_i / cm^{-1}**	**a_i / Hartree**	**a_i / cm^{-1}**	**a_i / Hartree**	**a_i / cm^{-1}**
0	1	−743.3675393		−743.3679628		−743.3654078		−743.3658014	
1	cos 3α	−0.0008114	−178.1	0.0003787	83.1	0.0007944	174.3	0.0004014	88.1
2	cos 6α	−0.0000496	−10.9	0.0000325	7.1	0.0000360	7.9	0.0000329	7.2

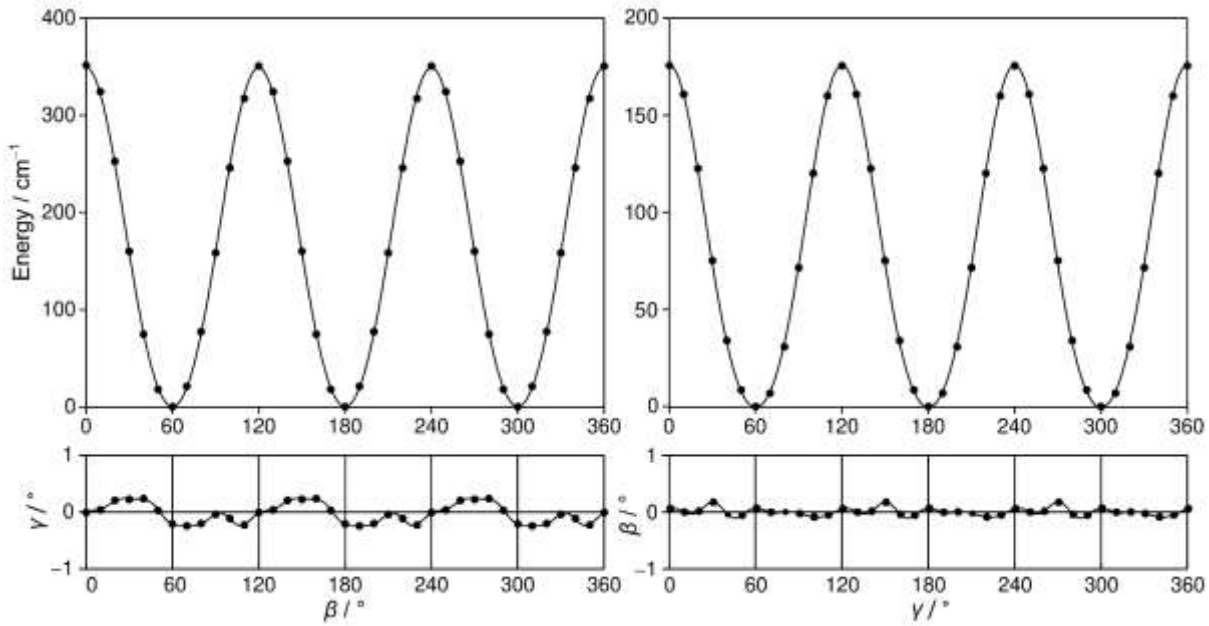

Figure S-1: Left hand side: The potential energy curve of *anti*-2A5MT obtained at the MP2/6-31G(d,p) level of theory by rotating the acetyl methyl group about the C6–C8 bond by varying the dihedral angle *β* in steps of 10°. The energies are given relative to the lowest energy with $E = -743.3661677$ Hartree. The calculated barrier height is 351.6 cm^{-1}. The lower trace depicts the oscillation of the ring methyl group upon the rotation of the acetyl methyl group. The deviations of the dihedral angle *γ* are given relative to the value of 179.98° of the fully optimized geometry. **Right hand side:** The potential energy curve of *syn*-2A5MT obtained by rotating the ring methyl group about the C3–C9 bond by varying the dihedral angle *γ* in steps of 10°. The energies are given relative to the lowest energy with $E = -743.3661678$ Hartree. The calculated barrier height is 175.5 cm^{-1}. The lower trace depicts the oscillation of the acetyl methyl group upon the ring methyl torsion. The deviations of the dihedral angle *β* are given relative to the values of 0.00° of the fully optimized geometry.

Table S-5: Coefficients of the two-dimensional Fourier expansion for the potential energy surface calculated at the MP2/6-31G(d,p) level of theory shown in Figure 4. Due to symmetry, only data points in the range from 0° to 120° were calculated for both *β* and *γ*. The potential is expanded as $V(\beta,\gamma) = \sum_i f_i V_i$. Max. dev. stands for maximum deviations between the fitted and the calculated values.

i	$\mathbf{f}_i$	$\mathbf{V}_i$ **/ Hartree**
1	1	−743.3673255
2	cos(3β)	0.0007590
3	cos(3γ)	0.0003762
4	cos(6β)	0.0001384
5	cos(6γ)	0.0000323
	Max. dev. (%)	8.0

Table S-6: Observed frequencies ν_{obs} of 427 rotational transitions of *syn*-2A5MT with $\nu_{obs}-\nu_{calc}$ values obtained from a fit with the program *XIAM*.

No.	J	K_a	K_c	J	K_a	K_c	Species	ν_{obs}	$\nu_{obs}-\nu_{calc}$
	upper level			lower level				MHz	kHz
1	4	0	4	3	0	3	(00)	6557.6150	0.1
2	4	0	4	3	0	3	(01)	6557.4625	–0.5
3	4	0	4	3	0	3	(10)	6556.4996	0.3
4	4	0	4	3	0	3	(11)	6556.1676	2.1
5	4	0	4	3	0	3	(12)	6556.5281	–0.3
6	5	0	5	4	0	4	(00)	8126.3405	–0.8
7	5	0	5	4	0	4	(01)	8126.1754	–0.3
8	5	0	5	4	0	4	(10)	8124.7950	1.1
9	5	0	5	4	0	4	(11)	8124.4691	–0.7
10	5	0	5	4	0	4	(12)	8124.7950	7.8
11	6	0	6	5	0	5	(00)	9657.5524	–1.3
12	6	0	6	5	0	5	(01)	9657.3814	–1.2
13	6	0	6	5	0	5	(10)	9655.5293	–1.2
14	6	0	6	5	0	5	(11)	9655.2258	–0.7
15	6	0	6	5	0	5	(12)	9655.4940	0.6
16	7	0	7	6	0	6	(00)	11155.6346	–1.5
17	7	0	7	6	0	6	(01)	11155.4632	–2.0
18	7	0	7	6	0	6	(10)	11153.1946	–1.6
19	7	0	7	6	0	6	(11)	11152.9114	–0.7
20	7	0	7	6	0	6	(12)	11153.1397	–0.3
21	8	0	8	7	0	7	(00)	12629.7351	–2.4
22	8	0	8	7	0	7	(01)	12629.5681	–1.9
23	8	0	8	7	0	7	(10)	12627.0447	0.3
24	8	0	8	7	0	7	(11)	12626.7765	–2.5
25	9	0	9	8	0	8	(00)	14090.3852	–0.1
26	9	0	9	8	0	8	(01)	14090.2212	–1.8
27	9	0	9	8	0	8	(10)	14087.6369	–2.0
28	9	0	9	8	0	8	(11)	14087.3919	–1.5
29	9	0	9	8	0	8	(12)	14087.5598	–2.1
30	10	0	10	9	0	9	(00)	15546.2084	–1.0
31	10	0	10	9	0	9	(01)	15546.0510	–1.8
32	10	0	10	9	0	9	(10)	15543.5683	–0.5
33	10	0	10	9	0	9	(11)	15543.3444	–0.5
34	11	0	11	10	0	10	(00)	17002.5530	1.6
35	11	0	11	10	0	10	(01)	17002.3983	–2.1
36	11	0	11	10	0	10	(10)	17000.1034	2.0
37	11	0	11	10	0	10	(11)	16999.8980	–1.2
38	12	0	12	11	0	11	(00)	18461.8418	0.4
39	12	0	12	11	0	11	(01)	18461.6951	–0.2
40	12	0	12	11	0	11	(10)	18459.6003	–1.8
41	12	0	12	11	0	11	(11)	18459.4195	0.2

42	12	0	12	11	0	11	(12)	18459.4962	1.7
43	13	0	13	12	0	12	(00)	19924.6580	−1.2
44	13	0	13	12	0	12	(01)	19924.5179	0.4
45	13	0	13	12	0	12	(10)	19922.6094	−2.4
46	13	0	13	12	0	12	(11)	19922.4457	0.5
47	13	0	13	12	0	12	(12)	19922.4959	−0.5
48	15	0	15	14	0	14	(00)	22859.2859	1.9
49	15	0	15	14	0	14	(01)	22859.1452	−3.6
50	6	0	6	5	1	5	(00)	8175.0415	−1.0
51	6	0	6	5	1	5	(01)	8175.3098	−0.1
52	6	0	6	5	1	5	(10)	8206.9829	2.5
53	6	0	6	5	1	5	(11)	8210.5275	3.1
54	6	0	6	5	1	5	(12)	8203.9260	0.1
55	7	0	7	6	1	6	(00)	9984.4456	−0.2
56	7	0	7	6	1	6	(01)	9984.5986	−2.0
57	7	0	7	6	1	6	(10)	10007.2031	2.6
58	7	0	7	6	1	6	(11)	10009.5094	3.7
59	7	0	7	6	1	6	(12)	10005.1763	−0.9
60	8	0	8	7	1	7	(00)	11739.5480	−0.8
61	8	0	8	7	1	7	(01)	11739.6208	0.2
62	8	0	8	7	1	7	(10)	11755.8016	0.1
63	8	0	8	7	1	7	(11)	11757.3152	3.1
64	8	0	8	7	1	7	(12)	11754.4157	−0.4
65	9	0	9	8	1	8	(00)	13436.6391	−6.9
66	9	0	9	8	1	8	(01)	13436.6569	0.1
67	9	0	9	8	1	8	(10)	13448.0691	−0.1
68	9	0	9	8	1	8	(11)	13449.0485	−1.2
69	9	0	9	8	1	8	(12)	13447.0967	−0.8
70	10	0	10	9	1	9	(00)	15079.9323	−7.2
71	10	0	10	9	1	9	(01)	15079.9066	0.7
72	10	0	10	9	1	9	(10)	15087.7347	−0.5
73	10	0	10	9	1	9	(11)	15088.3555	0.5
74	10	0	10	9	1	9	(12)	15087.0368	−2.6
75	11	0	11	10	1	10	(00)	16677.9040	4.0
76	11	0	11	10	1	10	(01)	16677.8320	−2.9
77	11	0	11	10	1	10	(10)	16682.9896	−5.0
78	11	0	11	10	1	10	(11)	16683.3675	−0.3
79	11	0	11	10	1	10	(12)	16682.4849	−0.3
80	12	0	12	11	1	11	(00)	18240.1688	−2.1
81	12	0	12	11	1	11	(01)	18240.0824	−1.5
82	12	0	12	11	1	11	(10)	18243.2871	0.0
83	12	0	12	11	1	11	(11)	18243.4949	2.5
84	13	0	13	12	1	12	(00)	19775.6876	−0.2
85	13	0	13	12	1	12	(01)	19775.5860	0.0
86	13	0	13	12	1	12	(10)	19777.3793	−0.7
87	13	0	13	12	1	12	(11)	19777.4690	−3.3
88	14	0	14	13	1	13	(00)	21291.8609	−2.0

89	14	0	14	13	1	13	(01)	21291.7539	2.3
90	14	0	14	13	1	13	(10)	21292.5444	−2.9
91	3	1	3	2	0	2	(00)	7051.7346	−0.8
92	3	1	3	2	0	2	(01)	7050.7847	−1.6
93	3	1	3	2	0	2	(10)	6978.4485	−4.5
94	3	1	3	2	0	2	(11)	6968.7094	1.2
95	3	1	3	2	0	2	(12)	6986.5604	0.0
96	4	1	4	3	0	3	(00)	8358.6082	−0.1
97	4	1	4	3	0	3	(01)	8357.8640	−1.6
98	4	1	4	3	0	3	(10)	8309.7424	−3.6
99	4	1	4	3	0	3	(11)	8303.5045	−0.3
100	4	1	4	3	0	3	(12)	8314.5922	−0.7
101	5	1	5	4	0	4	(00)	9608.8518	−0.7
102	5	1	5	4	0	4	(01)	9608.2478	−0.7
103	5	1	5	4	0	4	(10)	9573.3398	−4.2
104	5	1	5	4	0	4	(11)	9569.1702	−1.6
105	5	1	5	4	0	4	(12)	9576.3520	−2.7
106	6	1	6	5	0	5	(00)	10828.7436	−0.5
107	6	1	6	5	0	5	(01)	10828.2453	−1.9
108	6	1	6	5	0	5	(10)	10801.5221	−4.0
109	6	1	6	5	0	5	(11)	10798.6281	−4.7
110	6	1	6	5	0	5	(12)	10803.4546	−1.6
111	7	1	7	6	0	6	(00)	12045.8262	1.3
112	7	1	7	6	0	6	(01)	12045.4120	−2.5
113	7	1	7	6	0	6	(10)	12024.4350	−4.0
114	7	1	7	6	0	6	(11)	12022.3755	−3.5
115	7	1	7	6	0	6	(12)	12025.6992	−1.4
116	8	1	8	7	0	7	(00)	13283.4759	−1.0
117	8	1	8	7	0	7	(01)	13283.1345	−1.7
118	8	1	8	7	0	7	(10)	13266.6123	−1.7
119	8	1	8	7	0	7	(11)	13265.1194	−3.3
120	8	1	8	7	0	7	(12)	13267.4391	−2.1
121	9	1	9	8	0	8	(00)	14556.6558	0.6
122	9	1	9	8	0	8	(01)	14556.3703	0.4
123	9	1	9	8	0	8	(10)	14543.4708	−1.7
124	9	1	9	8	0	8	(11)	14542.3823	−1.1
125	9	1	9	8	0	8	(12)	14544.0031	−1.0
126	10	1	10	9	0	9	(00)	15870.8658	5.0
127	10	1	10	9	0	9	(01)	15870.6179	−0.4
128	10	1	10	9	0	9	(10)	15860.6720	−3.6
129	10	1	10	9	0	9	(11)	15859.8720	−4.3
130	10	1	10	9	0	9	(12)	15860.9997	−0.2
131	11	1	11	10	0	10	(00)	17224.2229	1.1
132	11	1	11	10	0	10	(01)	17224.0117	−0.1
133	11	1	11	10	0	10	(10)	17216.4154	−1.0
134	11	1	11	10	0	10	(11)	17215.8247	−1.4
135	12	1	12	11	0	11	(00)	18610.8160	3.2

136	12	1	12	11	0	11	(01)	18610.6271	0.3
137	12	1	12	11	0	11	(10)	18604.8322	–1.6
138	12	1	12	11	0	11	(11)	18604.3937	1.5
139	12	1	12	11	0	11	(12)	18604.9140	5.1
140	13	1	13	12	0	12	(00)	20023.4811	1.4
141	13	1	13	12	0	12	(01)	20023.3128	1.7
142	13	1	13	12	0	12	(10)	20018.8587	1.1
143	13	1	13	12	0	12	(11)	20018.5205	0.4
144	14	1	14	13	0	13	(00)	21455.5323	1.0
145	14	1	14	13	0	13	(01)	21455.3761	0.8
146	8	1	7	8	0	8	(00)	7321.6212	–3.4
147	8	1	7	8	0	8	(01)	7320.1696	–0.8
148	5	1	5	4	1	4	(00)	7807.8578	–1.3
149	6	1	6	5	1	5	(00)	9346.2336	0.8
150	6	1	6	5	1	5	(01)	9346.1733	–1.2
151	6	1	6	5	1	5	(10)	9352.9761	0.1
152	6	1	6	5	1	5	(11)	9353.9304	–0.3
153	6	1	6	5	1	5	(12)	9351.8885	–0.2
154	7	1	7	6	1	6	(00)	10874.6336	–0.9
155	7	1	7	6	1	6	(01)	10874.5433	–6.6
156	7	1	7	6	1	6	(10)	10878.4439	0.6
157	7	1	7	6	1	6	(11)	10878.9722	–0.5
158	7	1	7	6	1	6	(12)	10877.7370	–0.9
159	8	1	8	7	1	7	(00)	12393.2883	0.2
160	8	1	8	7	1	7	(01)	12393.1861	–0.8
161	8	1	8	7	1	7	(10)	12395.3720	0.8
162	8	1	8	7	1	7	(11)	12395.6557	–0.1
163	8	1	8	7	1	7	(12)	12394.8807	0.2
164	9	1	9	8	1	8	(00)	13902.9158	–0.1
165	9	1	9	8	1	8	(01)	13902.8020	–1.7
166	9	1	9	8	1	8	(10)	13903.9021	–0.7
167	9	1	9	8	1	8	(11)	13904.0393	–0.4
168	9	1	9	8	1	8	(12)	13903.5395	–0.2
169	10	1	10	9	1	9	(00)	15404.5896	–1.3
170	10	1	10	9	1	9	(01)	15404.4698	–1.6
171	10	1	10	9	1	9	(10)	15404.8413	–0.7
172	10	1	10	9	1	9	(11)	15404.8841	–2.2
173	11	1	11	10	1	10	(00)	16899.5698	–0.6
174	11	1	11	10	1	10	(01)	16899.4470	0.8
175	11	1	11	10	1	10	(10)	16899.3091	–0.5
176	12	1	12	11	1	11	(00)	18389.1410	–1.4
177	12	1	12	11	1	11	(01)	18389.0137	–1.7
178	12	1	12	11	1	11	(10)	18388.5181	–0.7
179	12	1	12	11	1	11	(11)	18388.4652	–0.1
180	12	1	12	11	1	11	(12)	18388.3168	–1.5
181	5	1	4	4	1	3	(00)	8748.2981	–0.4
182	5	1	4	4	1	3	(01)	8747.9334	–0.8

183	5	1	4	4	1	3	(10)	8734.8292	–0.6
184	5	1	4	4	1	3	(11)	8732.5953	0.5
185	5	1	4	4	1	3	(12)	8736.3795	–0.9
186	6	1	5	5	1	4	(00)	10464.2402	–1.8
187	6	1	5	5	1	4	(01)	10463.8449	–2.0
188	6	1	5	5	1	4	(10)	10455.6184	0.3
189	6	1	5	5	1	4	(11)	10453.9683	1.8
190	6	1	5	5	1	4	(12)	10456.4924	–1.0
191	7	1	6	6	1	5	(00)	12158.6604	–0.5
192	7	1	6	6	1	5	(01)	12158.2245	–1.0
193	7	1	6	6	1	5	(10)	12152.2409	0.5
194	7	1	6	6	1	5	(11)	12150.8515	2.1
195	7	1	6	6	1	5	(12)	12152.7654	0.0
196	8	1	7	7	1	6	(00)	13825.7137	–1.2
197	8	1	7	7	1	6	(01)	13825.2410	–2.1
198	8	1	7	7	1	6	(10)	13820.2302	0.9
199	8	1	7	7	1	6	(11)	13818.9794	1.4
200	8	1	7	7	1	6	(12)	13820.5395	1.0
201	9	1	8	8	1	7	(00)	15459.3203	–0.7
202	9	1	8	8	1	7	(01)	15458.8198	–2.9
203	9	1	8	8	1	7	(10)	15454.0766	–1.5
204	9	1	8	8	1	7	(11)	15452.9399	1.2
205	9	1	8	8	1	7	(12)	15454.2201	–1.8
206	10	1	9	9	1	8	(00)	17054.1776	–1.3
207	10	1	9	9	1	8	(01)	17053.6655	0.0
208	10	1	9	9	1	8	(10)	17048.7473	–0.2
209	11	1	10	10	1	9	(00)	18607.3727	–2.0
210	11	1	10	10	1	9	(01)	18606.8577	1.3
211	11	1	10	10	1	9	(10)	18601.4806	–1.5
212	11	1	10	10	1	9	(11)	18600.5700	0.2
213	11	1	10	10	1	9	(12)	18601.3596	–1.2
214	4	2	2	3	1	3	(00)	15244.7807	2.7
215	4	2	2	3	1	3	(01)	15253.7595	–3.7
216	4	2	2	3	1	3	(12)	15703.8532	3.4
217	5	2	3	4	1	4	(00)	17481.8957	4.5
218	5	2	3	4	1	4	(01)	17484.7595	–3.0
219	5	2	3	4	1	4	(12)	17841.7554	2.2
220	4	2	3	3	1	3	(01)	15080.5163	–3.2
221	2	2	1	1	1	0	(00)	11101.8884	0.8
222	2	2	1	1	1	0	(01)	11060.7186	–5.8
223	3	2	2	2	1	1	(00)	12576.9674	–0.4
224	3	2	2	2	1	1	(01)	12550.9348	–5.1
225	3	2	2	2	1	1	(12)	11963.1193	–4.1
226	4	2	3	3	1	2	(00)	13955.8834	–1.3
227	4	2	3	3	1	2	(01)	13943.0319	–2.8
228	4	2	3	3	1	2	(10)	13384.6547	–4.9
229	4	2	3	3	1	2	(12)	13432.9934	0.2

230	5	2	4	4	1	3	(00)	15240.2237	–0.6
231	5	2	4	4	1	3	(01)	15233.3890	–1.6
232	5	2	4	4	1	3	(10)	14774.2798	–3.1
233	5	2	4	4	1	3	(12)	14817.3896	2.6
234	6	2	5	5	1	4	(00)	16433.5593	1.0
235	6	2	5	5	1	4	(01)	16429.2475	–1.3
236	6	2	5	5	1	4	(10)	16082.0018	–1.0
237	6	2	5	5	1	4	(12)	16118.1409	2.3
238	7	2	6	6	1	5	(00)	17542.2279	1.2
239	7	2	6	6	1	5	(01)	17539.1338	–0.6
240	7	2	6	6	1	5	(10)	17292.6300	–1.5
241	7	2	6	6	1	5	(12)	17319.7174	3.9
242	8	2	7	7	1	6	(00)	18576.2726	1.5
243	8	2	7	7	1	6	(01)	18573.8688	–0.3
244	8	2	7	7	1	6	(10)	18400.8544	–0.8
245	8	2	7	7	1	6	(12)	18419.4213	3.2
246	9	2	8	8	1	7	(00)	19550.3480	4.2
247	9	2	8	8	1	7	(01)	19548.3978	2.8
248	9	2	8	8	1	7	(11)	19407.6975	7.4
249	9	2	8	8	1	7	(12)	19435.8104	3.7
250	10	2	9	9	1	8	(00)	20484.2381	2.8
251	10	2	9	9	1	8	(01)	20482.6209	2.9
252	10	2	9	9	1	8	(10)	20388.9186	2.8
253	10	2	9	9	1	8	(11)	20377.8167	4.2
254	10	2	9	9	1	8	(12)	20396.9345	4.0
255	11	2	10	10	1	9	(01)	21401.0981	4.8
256	11	2	10	10	1	9	(10)	21328.2621	1.8
257	11	2	10	10	1	9	(11)	21320.4017	9.8
258	11	2	10	10	1	9	(12)	21333.5073	4.6
259	2	2	1	2	1	2	(00)	8151.7294	6.2
260	2	2	1	2	1	2	(10)	7687.1121	–1.0
261	2	2	1	2	1	2	(12)	7712.8350	–1.8
262	4	2	3	4	1	4	(00)	8832.9001	–1.5
263	4	2	3	4	1	4	(01)	8820.5540	–1.7
264	4	2	3	4	1	4	(10)	8365.6140	0.1
265	4	2	3	4	1	4	(12)	8398.9048	1.7
266	5	2	4	5	1	5	(00)	9325.3494	0.6
267	5	2	4	5	1	5	(01)	9318.6528	–3.5
268	5	2	4	5	1	5	(10)	8926.7637	5.2
269	5	2	4	5	1	5	(12)	8959.5405	–0.8
270	6	2	5	6	1	6	(00)	9920.7550	6.6
271	6	2	5	6	1	6	(01)	9916.2765	2.2
272	6	2	5	6	1	6	(10)	9616.3320	–0.3
273	6	2	5	6	1	6	(12)	9644.7861	1.5
274	7	2	6	7	1	7	(00)	10619.0277	3.4
275	7	2	6	7	1	7	(01)	10615.4562	–0.6
276	7	2	6	7	1	7	(10)	10404.1299	–5.9

277	7	2	6	7	1	7	(11)	10377.7589	10.0
278	7	2	6	7	1	7	(12)	10425.1169	1.8
279	8	2	7	8	1	8	(00)	11418.4458	4.4
280	8	2	7	8	1	8	(01)	11415.2297	–0.5
281	8	2	7	8	1	8	(10)	11269.2239	–4.8
282	8	2	7	8	1	8	(12)	11282.7084	3.9
283	5	2	3	4	2	2	(00)	8496.9987	0.1
284	5	2	3	4	2	2	(01)	8490.9627	–0.4
285	5	2	3	4	2	2	(10)	8416.4825	–1.4
286	5	2	3	4	2	2	(11)	8414.5496	–0.4
287	5	2	3	4	2	2	(12)	8418.2337	–1.5
288	6	2	4	5	2	3	(00)	10271.4562	0.0
289	6	2	4	5	2	3	(01)	10268.8126	0.5
290	6	2	4	5	2	3	(10)	10171.2451	–0.4
291	7	2	5	6	2	4	(00)	12068.4516	1.4
292	7	2	5	6	2	4	(01)	12067.0519	2.3
293	7	2	5	6	2	4	(10)	11975.9092	–3.2
294	6	2	5	5	2	4	(00)	9941.6318	–0.7
295	6	2	5	5	2	4	(01)	9943.7931	0.7
296	6	2	5	5	2	4	(10)	10042.5508	1.0
297	6	2	5	5	2	4	(11)	10047.0351	–2.0
298	6	2	5	5	2	4	(12)	10037.1327	0.6
299	7	2	6	6	2	5	(00)	11572.9107	0.3
300	7	2	6	6	2	5	(01)	11573.7327	0.2
301	7	2	6	6	2	5	(10)	11666.2480	1.2
302	7	2	6	6	2	5	(11)	11673.5155	–4.3
303	7	2	6	6	2	5	(12)	11658.0685	0.2
304	8	2	7	7	2	6	(00)	13192.7059	0.7
305	8	2	7	7	2	6	(01)	13192.9604	0.1
306	8	2	7	7	2	6	(10)	13260.4653	1.2
307	8	2	7	7	2	6	(11)	13267.8829	–3.9
308	8	2	7	7	2	6	(12)	13252.4694	–0.5
309	9	2	8	8	2	7	(00)	14799.7883	0.6
310	9	2	8	8	2	7	(01)	14799.7642	–4.8
311	9	2	8	8	2	7	(10)	14842.9312	1.5
312	9	2	8	8	2	7	(11)	14848.5052	–2.1
313	9	2	8	8	2	7	(12)	14836.9263	–0.9
314	10	2	9	9	2	8	(00)	16393.2112	–1.2
315	10	2	9	9	2	8	(01)	16393.0461	0.4
316	10	2	9	9	2	8	(10)	16419.4386	0.2
317	10	2	9	9	2	8	(11)	16423.0586	–2.5
318	10	2	9	9	2	8	(12)	16415.3452	–0.4
319	11	2	10	10	2	9	(00)	17972.3952	–0.5
320	11	2	10	10	2	9	(01)	17972.1412	0.4
321	11	2	10	10	2	9	(10)	17988.0908	–1.2
322	11	2	10	10	2	9	(11)	17990.2991	–2.4
323	11	2	10	10	2	9	(12)	17985.3208	0.8

324	3	3	1	2	2	0	(00)	18102.1494	−2.2
325	4	3	2	3	2	1	(00)	19745.4348	−1.4
326	5	3	3	4	2	2	(00)	21348.9383	−0.3
327	3	3	0	2	2	1	(00)	18112.7617	−2.4
328	4	3	1	3	2	2	(00)	19798.9903	−1.2
329	5	3	2	4	2	3	(00)	21510.6267	−0.4
330	6	3	3	5	2	4	(00)	23264.4563	1.2
331	3	3	0	3	2	1	(00)	13067.7714	0.6
332	3	3	0	3	2	1	(01)	13104.0048	5.7
333	3	3	0	3	2	1	(10)	13318.1201	−9.3
334	4	3	1	4	2	2	(00)	12996.4916	−7.0
335	4	3	1	4	2	2	(01)	13044.4298	−7.0
336	4	3	1	4	2	2	(10)	13293.2651	−5.2
337	4	3	1	4	2	2	(12)	13268.8990	−8.3
338	5	3	2	5	2	3	(00)	12861.6355	−2.0
339	5	3	2	5	2	3	(01)	12911.8650	−2.6
340	5	3	2	5	2	3	(10)	13234.0452	−1.9
341	5	3	2	5	2	3	(12)	13207.8057	−6.7
342	6	3	3	6	2	4	(00)	12644.3159	0.2
343	6	3	3	6	2	4	(01)	12688.6471	−1.2
344	6	3	3	6	2	4	(10)	13105.4014	1.0
345	6	3	3	6	2	4	(12)	13074.3549	−4.6
346	7	3	4	7	2	5	(00)	12335.8003	4.5
347	7	3	4	7	2	5	(01)	12367.5601	−3.0
348	7	3	4	7	2	5	(10)	12865.5908	6.2
349	7	3	4	7	2	5	(12)	12827.1842	0.2
350	8	3	5	8	2	6	(00)	11943.1300	5.1
351	8	3	5	8	2	6	(01)	11961.1336	−0.8
352	8	3	5	8	2	6	(10)	12496.5742	10.8
353	8	3	5	8	2	6	(12)	12451.3147	1.7
354	6	3	4	6	2	4	(01)	12560.4294	−1.2
355	3	3	1	3	2	2	(00)	13118.5457	−0.3
356	3	3	1	3	2	2	(01)	13076.4492	4.0
357	4	3	2	4	2	3	(00)	13146.0467	−10.7
358	4	3	2	4	2	3	(01)	13092.2471	−2.3
359	5	3	3	5	2	4	(00)	13200.6245	1.2
360	5	3	3	5	2	4	(01)	13144.5275	−2.5
361	6	3	4	6	2	5	(00)	13293.8937	0.4
362	6	3	4	6	2	5	(01)	13243.7099	−0.7
363	7	3	5	7	2	6	(00)	13438.1755	2.8
364	7	3	5	7	2	6	(01)	13400.5763	−1.3
365	8	3	6	8	2	7	(00)	13645.6944	1.9
366	8	3	6	8	2	7	(01)	13621.8919	2.1
367	5	3	3	4	2	2	(00)	21348.9381	−0.5
368	5	3	3	4	2	2	(01)	21277.2348	1.9
369	6	3	4	5	2	3	(00)	22886.8430	0.6
370	6	3	4	5	2	3	(01)	22829.2407	−2.0

371	5	3	3	4	3	2	(00)	8354.8742	2.0
372	6	3	4	5	3	3	(00)	10034.9051	2.7
373	6	3	4	5	3	3	(01)	10042.9756	2.7
374	6	3	4	5	3	3	(10)	10046.2123	–0.5
375	6	3	4	5	3	3	(11)	10046.1744	2.1
376	6	3	4	5	3	3	(12)	10045.7794	–1.0
377	7	3	5	6	3	4	(00)	11717.1916	1.8
378	6	3	3	5	3	2	(00)	10054.1375	3.2
379	6	3	3	5	3	2	(01)	10045.5930	0.3
380	6	3	3	5	3	2	(10)	10042.5993	0.6
381	4	4	0	4	3	1	(00)	18336.8924	1.2
382	4	4	0	4	3	1	(01)	18354.8502	3.6
383	4	4	1	4	3	2	(00)	18339.3122	–1.4
384	4	4	1	4	3	2	(01)	18313.1564	6.7
385	5	4	2	5	3	3	(00)	18330.3416	–8.1
386	5	4	2	5	3	3	(01)	18300.7197	–3.2
387	6	4	3	6	3	4	(00)	18317.9510	–4.5
388	6	4	3	6	3	4	(01)	18280.2716	–0.5
389	7	4	4	7	3	5	(00)	18303.7482	–0.7
390	7	4	4	7	3	5	(01)	18253.0651	–2.4
391	8	4	5	8	3	6	(00)	18291.2675	7.0
392	8	4	5	8	3	6	(01)	18227.9332	1.9
393	9	4	6	9	3	7	(00)	18286.2245	6.2
394	9	4	6	9	3	7	(01)	18216.9769	3.3
395	6	4	2	5	4	1	(00)	10022.8666	7.2
396	6	4	2	5	4	1	(01)	10022.3802	–1.3
397	6	4	2	5	4	1	(10)	10021.6967	1.2
398	6	4	2	5	4	1	(11)	10021.4084	4.0
399	6	4	2	5	4	1	(12)	10021.5244	2.1
400	7	4	3	6	4	2	(00)	11704.1500	0.8
401	7	4	3	6	4	2	(01)	11703.1996	7.3
402	7	4	3	6	4	2	(10)	11702.1573	2.6
403	7	4	3	6	4	2	(11)	11701.7957	3.6
404	7	4	3	6	4	2	(12)	11701.9728	0.9
405	8	4	4	7	4	3	(00)	13390.9302	5.0
406	8	4	4	7	4	3	(01)	13388.8996	2.2
407	8	4	4	7	4	3	(10)	13387.3739	0.9
408	8	4	4	7	4	3	(11)	13386.9303	2.4
409	8	4	4	7	4	3	(12)	13387.1931	3.2
410	9	4	5	8	4	4	(00)	15084.5591	5.0
411	9	4	5	8	4	4	(10)	15078.1214	5.9
412	6	4	3	5	4	2	(01)	10022.5244	2.3
413	6	4	3	5	4	2	(10)	10023.1795	0.6
414	6	4	3	5	4	2	(11)	10023.0299	–0.1
415	7	4	4	6	4	3	(00)	11702.9854	2.2
416	7	4	4	6	4	3	(01)	11703.3984	3.5
417	7	4	4	6	4	3	(10)	11704.4884	3.4

418	7	4	4	6	4	3	(11)	11704.3353	1.7
419	7	4	4	6	4	3	(12)	11704.0945	4.1
420	8	4	5	7	4	4	(00)	13387.7405	3.9
421	8	4	5	7	4	4	(01)	13389.1385	2.3
422	8	4	5	7	4	4	(10)	13390.8384	1.0
423	8	4	5	7	4	4	(11)	13390.6908	–4.0
424	8	4	5	7	4	4	(12)	13390.3521	1.1
425	9	4	6	8	4	5	(00)	15076.9689	4.3
426	9	4	6	8	4	5	(10)	15083.0464	3.1
427	9	4	6	8	4	5	(11)	15082.9237	0.1