cis-3aA: AM1 geometry; DHf = -65.162427 kcal

C 0.000000 0 0.000000 0 0.000000 0 0 0 0 -0.0384

C 1.418070 1 0.000000 0 0.000000 0 1 0 0 -0.0316

C 1.421313 1 119.294139 1 0.000000 0 1 2 0 -0.1051

C 1.421642 1 118.947949 1 0.181003 1 2 1 3 -0.1185

C 1.376343 1 120.754366 1 0.219786 1 3 1 2 -0.1148

H 1.101569 1 118.010057 1 -179.856714 1 3 1 5 0.1334

C 1.371783 1 120.677142 1 -0.336335 1 4 2 1 -0.1114

H 1.100543 1 118.638601 1 179.973614 1 4 2 7 0.1351

C 1.517642 1 122.326388 1 179.305555 1 5 3 1 0.1178

H 1.102541 1 120.793659 1 178.930711 1 7 4 2 0.1481

N 1.455319 1 113.277844 1 -42.026668 1 9 5 3 -0.3279

C 1.510517 1 110.076915 1 126.363812 1 9 5 11 -0.2727

H 1.137620 1 107.759652 1 -116.080662 1 9 5 11 0.1335

C 1.400292 1 120.042511 1 94.350460 1 11 9 5 0.3762

C 1.375404 1 120.919550 1 -104.725484 1 12 9 5 0.1628

C 1.452152 1 118.006523 1 -178.752929 1 12 9 15 0.3752

H 0.998137 1 115.535131 1 -143.638604 1 11 9 14 0.2429

N 1.417264 1 118.702478 1 20.612183 1 14 11 9 -0.3219

O 1.250608 1 122.436939 1 176.426262 1 14 11 18 -0.3687

C 1.493784 1 121.730872 1 176.807967 1 15 12 9 -0.2105

O 1.240306 1 129.206025 1 189.550391 1 16 12 9 -0.3896

O 1.369631 1 115.569557 1 181.610988 1 16 12 21 -0.3200

H 0.995212 1 117.223755 1 170.179023 1 18 14 11 0.2621

H 1.120959 1 108.986970 1 -59.733601 1 20 15 12 0.1308

H 1.116660 1 113.163779 1 -121.545832 1 20 15 24 0.0894

H 1.121001 1 109.189724 1 116.842213 1 20 15 24 0.1278

H 0.971605 1 108.762431 1 179.570917 1 22 16 12 0.2451

C 1.421643 1 119.188665 1 -179.922397 1 2 1 3 -0.1191

H 1.100312 1 118.630913 1 179.989797 1 28 2 1 0.1337

C 1.422230 1 119.168320 1 -179.868978 1 1 2 4 -0.1152

H 1.100444 1 118.664018 1 -179.956176 1 30 1 2 0.1346

C 1.373186 1 120.456942 1 -0.035414 1 28 2 1 -0.1233

H 1.100110 1 120.639657 1 -179.983823 1 32 28 2 0.1333

C 1.416133 1 120.367702 1 0.020208 1 32 28 2 -0.1271

H 2.174000 1 146.635244 1 0.035979 1 32 28 2 0.1337

0 0.000000 0 0.000000 0 0.000000 0 0 0 0

cis-3aB: AM1 geometry; DHf = -65.114905 kcal

C 0.000000 0 0.000000 0 0.000000 0 0 0 0 -0.0407

C 1.418203 1 0.000000 0 0.000000 0 1 0 0 -0.0320

C 1.420812 1 119.265854 1 0.000000 0 1 2 0 -0.0904

C 1.421469 1 118.956898 1 -0.329313 1 2 1 3 -0.1149

C 1.377450 1 120.758621 1 0.079229 1 3 1 2 -0.1197

H 1.102810 1 118.808963 1 -181.192325 1 3 1 5 0.1467

C 1.372335 1 120.690635 1 0.232372 1 4 2 1 -0.1198

H 1.100611 1 118.647861 1 180.078359 1 4 2 7 0.1358

C 1.517287 1 119.371963 1 180.231376 1 5 3 1 0.1187

H 1.100967 1 120.034889 1 180.494116 1 7 4 2 0.1328

N 1.455109 1 113.217735 1 134.609124 1 9 5 3 -0.3268

C 1.510161 1 110.111877 1 126.589238 1 9 5 11 -0.2731

H 1.137920 1 107.799776 1 -116.022966 1 9 5 11 0.1314

C 1.400216 1 120.145694 1 95.557890 1 11 9 5 0.3767

C 1.375204 1 121.036567 1 -105.934160 1 12 9 5 0.1627

C 1.452260 1 117.824965 1 -178.786138 1 12 9 15 0.3751

H 0.998206 1 115.484377 1 -143.627307 1 11 9 14 0.2428

N 1.417276 1 118.754360 1 20.327443 1 14 11 9 -0.3223

O 1.250606 1 122.414380 1 176.349296 1 14 11 18 -0.3686

C 1.493836 1 121.717130 1 177.184426 1 15 12 9 -0.2104

O 1.240294 1 129.219310 1 -171.118216 1 16 12 9 -0.3898

O 1.369657 1 115.530819 1 -178.476138 1 16 12 21 -0.3198

H 0.995193 1 117.230292 1 170.026815 1 18 14 11 0.2621

H 1.120946 1 108.993254 1 -59.744896 1 20 15 12 0.1305

H 1.116657 1 113.163880 1 -121.552057 1 20 15 24 0.0894

H 1.120985 1 109.181105 1 116.837993 1 20 15 24 0.1278

H 0.971622 1 108.742171 1 179.789018 1 22 16 12 0.2450

C 1.421745 1 119.197848 1 -180.205852 1 2 1 3 -0.1187

H 1.100331 1 118.637252 1 180.037308 1 28 2 1 0.1340

C 1.422266 1 119.154323 1 -180.217320 1 1 2 4 -0.1146

H 1.100377 1 118.623862 1 -179.922556 1 30 1 2 0.1339

C 1.373127 1 120.448559 1 0.057216 1 28 2 1 -0.1232

H 1.100118 1 120.638710 1 -180.011954 1 32 28 2 0.1334

C 1.416237 1 120.372864 1 0.003259 1 32 28 2 -0.1274

H 2.173943 1 146.644337 1 0.033498 1 32 28 2 0.1335

0 0.000000 0 0.000000 0 0.000000 0 0 0 0

trans-3aA: AM1 geometry; DHf = -64.745057 kcal

C 0.000000 0 0.000000 0 0.000000 0 0 0 0 -0.0393

C 1.417987 1 0.000000 0 0.000000 0 1 0 0 -0.0319

C 1.421456 1 119.296271 1 0.000000 0 1 2 0 -0.1079

C 1.421733 1 118.949427 1 0.212734 1 2 1 3 -0.1179

C 1.376182 1 120.733829 1 0.260564 1 3 1 2 -0.1163

H 1.101562 1 117.897188 1 -179.853838 1 3 1 5 0.1325

C 1.371666 1 120.712423 1 -0.379276 1 4 2 1 -0.1129

H 1.100627 1 118.667679 1 179.935980 1 4 2 7 0.1366

C 1.517161 1 122.532470 1 179.209667 1 5 3 1 0.1129

H 1.103597 1 120.816889 1 178.763034 1 7 4 2 0.1590

N 1.453960 1 113.671263 1 -37.870691 1 9 5 3 -0.3260

C 1.515210 1 109.896242 1 126.601547 1 9 5 11 -0.2706

H 1.138317 1 107.573671 1 -116.332507 1 9 5 11 0.1370

C 1.400270 1 119.871430 1 93.938795 1 11 9 5 0.3762

C 1.372442 1 120.785007 1 -104.911950 1 12 9 5 0.1642

C 1.453341 1 114.383332 1 -178.217007 1 12 9 15 0.3769

H 0.998284 1 115.505207 1 -143.395530 1 11 9 14 0.2436

N 1.416739 1 118.630680 1 21.038937 1 14 11 9 -0.3232

O 1.250661 1 122.493687 1 176.377266 1 14 11 18 -0.3692

C 1.494565 1 122.469203 1 176.990912 1 15 12 9 -0.2051

O 1.239712 1 128.037633 1 11.983821 1 16 12 9 -0.3830

O 1.369218 1 116.897242 1 179.093679 1 16 12 21 -0.3269

H 0.995158 1 117.132073 1 170.217788 1 18 14 11 0.2613

H 1.120456 1 108.996435 1 -61.080817 1 20 15 12 0.1258

H 1.117146 1 113.155870 1 -121.209786 1 20 15 24 0.0895

H 1.120255 1 109.227269 1 117.510719 1 20 15 24 0.1222

H 0.971827 1 108.902929 1 179.469984 1 22 16 12 0.2455

C 1.421621 1 119.181338 1 -179.902043 1 2 1 3 -0.1188

H 1.100334 1 118.618702 1 179.966067 1 28 2 1 0.1339

C 1.422177 1 119.173285 1 -179.848976 1 1 2 4 -0.1157

H 1.100424 1 118.668651 1 180.061192 1 30 1 2 0.1340

C 1.373215 1 120.463944 1 -0.042253 1 28 2 1 -0.1243

H 1.100075 1 120.640597 1 180.000993 1 32 28 2 0.1329

C 1.416073 1 120.364616 1 0.022744 1 32 28 2 -0.1279

H 2.173952 1 146.630479 1 0.056057 1 32 28 2 0.1331

0 0.000000 0 0.000000 0 0.000000 0 0 0 0

trans3aB: AM1 geometry; DHf = -64.676093 kcal

C 0.000000 0 0.000000 0 0.000000 0 0 0 0 -0.0407

C 1.418174 1 0.000000 0 0.000000 0 1 0 0 -0.0324

C 1.420666 1 119.312331 1 0.000000 0 1 2 0 -0.0895

C 1.421269 1 118.934463 1 -0.462798 1 2 1 3 -0.1167

C 1.378066 1 120.714437 1 0.149707 1 3 1 2 -0.1251

H 1.103910 1 118.832062 1 178.656160 1 3 1 5 0.1574

C 1.372509 1 120.704357 1 0.280377 1 4 2 1 -0.1211

H 1.100563 1 118.652639 1 -179.911387 1 4 2 7 0.1349

C 1.516905 1 119.087142 1 -179.754205 1 5 3 1 0.1139

H 1.101017 1 119.866293 1 -179.392614 1 7 4 2 0.1316

N 1.453803 1 113.679295 1 139.766120 1 9 5 3 -0.3240

C 1.515023 1 109.906067 1 126.821465 1 9 5 11 -0.2708

H 1.138537 1 107.621398 1 -116.275860 1 9 5 11 0.1349

C 1.400453 1 119.910564 1 94.837956 1 11 9 5 0.3762

C 1.372215 1 120.876147 1 -105.876683 1 12 9 5 0.1640

C 1.453434 1 114.218366 1 -178.305407 1 12 9 15 0.3767

H 0.998417 1 115.410404 1 -143.131682 1 11 9 14 0.2431

N 1.416725 1 118.682687 1 20.907352 1 14 11 9 -0.3236

O 1.250666 1 122.474574 1 176.292802 1 14 11 18 -0.3691

C 1.494590 1 122.458145 1 177.263785 1 15 12 9 -0.2050

O 1.239763 1 127.989305 1 11.374484 1 16 12 9 -0.3835

O 1.369159 1 116.923291 1 -180.846744 1 16 12 21 -0.3270

H 0.995129 1 117.140355 1 170.047446 1 18 14 11 0.2613

H 1.120439 1 109.001215 1 -61.060078 1 20 15 12 0.1256

H 1.117136 1 113.157942 1 -121.219497 1 20 15 24 0.0894

H 1.120252 1 109.218930 1 117.501418 1 20 15 24 0.1221

H 0.971818 1 108.899812 1 179.401295 1 22 16 12 0.2454

C 1.421770 1 119.197393 1 179.698528 1 2 1 3 -0.1208

H 1.100255 1 118.642140 1 -179.890528 1 28 2 1 0.1331

C 1.422421 1 119.170997 1 179.623738 1 1 2 4 -0.1113

H 1.100637 1 118.579867 1 -179.741516 1 30 1 2 0.1377

C 1.373092 1 120.436563 1 0.115486 1 28 2 1 -0.1228

H 1.100093 1 120.631508 1 180.009800 1 32 28 2 0.1326

C 1.416290 1 120.384666 1 0.033110 1 32 28 2 -0.1298

H 2.173979 1 146.654487 1 0.111977 1 32 28 2 0.1333

0 0.000000 0 0.000000 0 0.000000 0 0 0 0

trans-4: HF/6-31G*; E = -604.4244317

C,0,-2.4229708374,0.0740845759,-1.0496855811

N,0,-2.3814249398,0.1815880695,0.2908557607

C,0,-1.243262527,-0.145383372,1.1312345807

C,0,0.0461891723,-0.0674127705,0.3433667348

C,0,0.0248705925,-0.0800914246,-0.9972581325

N,0,-1.1908205843,-0.1167740208,-1.6474362836

H,0,-1.2102475534,0.5557426843,1.9531343502

O,0,-3.4197391843,0.1595857028,-1.7118156763

H,0,-1.3591864783,-1.1351648975,1.5666768908

H,0,-3.2774709647,0.2627020031,0.7165805007

H,0,-1.2230524331,-0.1092075186,-2.6410700628

C,0,1.2074775053,-0.0619381277,-1.9367175713

H,0,1.7203930108,0.8890411191,-1.8822841077

H,0,1.9183405057,-0.8332276114,-1.6848579356

H,0,0.8757142366,-0.2158978852,-2.9561876337

C,0,1.2470016807,-0.0403529433,1.1903352633

O,0,1.1887887812,-0.1236903758,2.3811876682

O,0,2.4000742099,0.0880441785,0.5454703253

C,0,3.5730689605,0.1157439521,1.3405500666

H,0,3.5482830611,0.9522729778,2.0231252182

H,0,3.6720976933,-0.8006446043,1.9034898983

H,0,4.3936098555,0.2212700082,0.6477658409

cis-4: HF/6-31G*; E = -604.426433537

C	0	-2.2865	-0.04295	-1.25594
N	0	-2.25718	-0.15926	0.08484
C	0	-1.11177	0.07087	0.94787
C	0	0.17637	0.0227	0.15449
C	0	0.15854	0.05904	-1.18726

N	0	-1.04728	0.10718	-1.84806
H	0	-1.20945	1.02703	1.45628
O	0	-3.2848	-0.08617	-1.91955
H	0	-1.10543	-0.69222	1.71472
H	0	-3.15873	-0.22201	0.50173
H	0	-1.07169	0.12036	-2.84198
C	0	1.3609	0.05598	-2.09784
H	0	2.05018	0.84234	-1.83067
H	0	1.89735	-0.87906	-2.01179
H	0	1.05083	0.18963	-3.12732
C	0	1.44307	-0.01618	0.89592
O	0	2.54045	-0.08988	0.42683
O	0	1.23827	0.04561	2.2087
C	0	2.3841	0.01604	3.0397
H	0	2.9361	-0.90076	2.89248
H	0	3.02811	0.85705	2.82795
H	0	2.01428	0.07268	4.05172

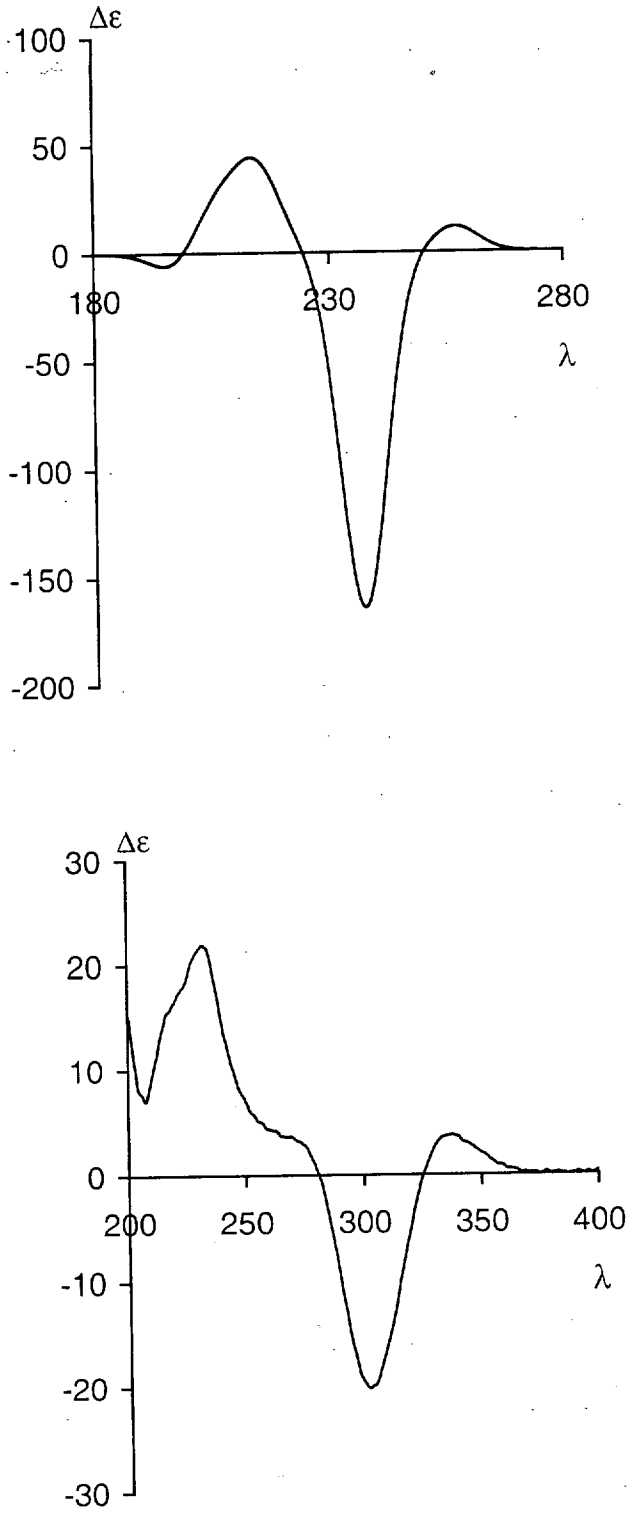

Figure S1. Comparison of the simulated[47] (RPA/6-31G*, AM1 geometry) CD spectrum for (*R*)-**5a** (**a**) with the experimental CD spectrum of (*R*)-**5b** (**b**).

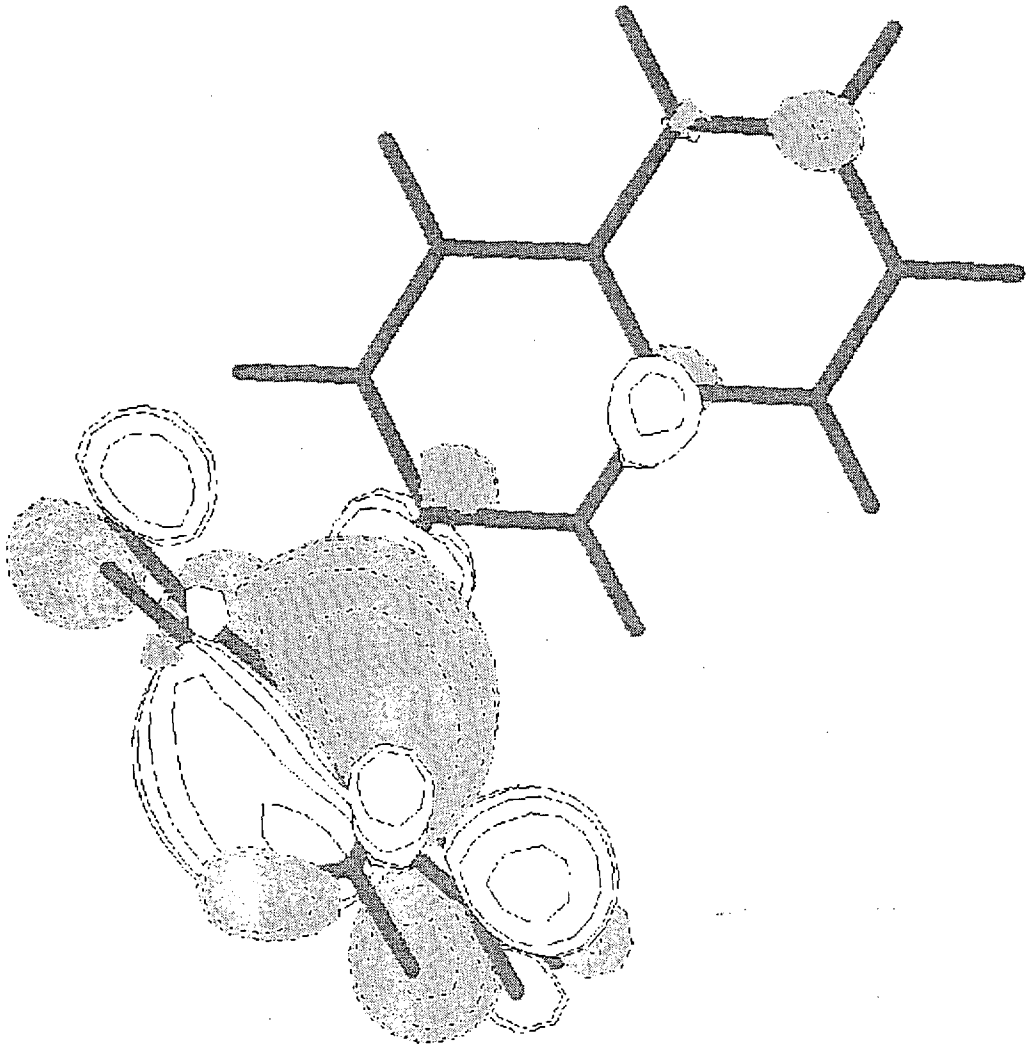

Figure S2a. Plot[49] of HOMO – 2 of s-cis-**(R)-3aA** (HF/6-31G*, AM1 geometry).

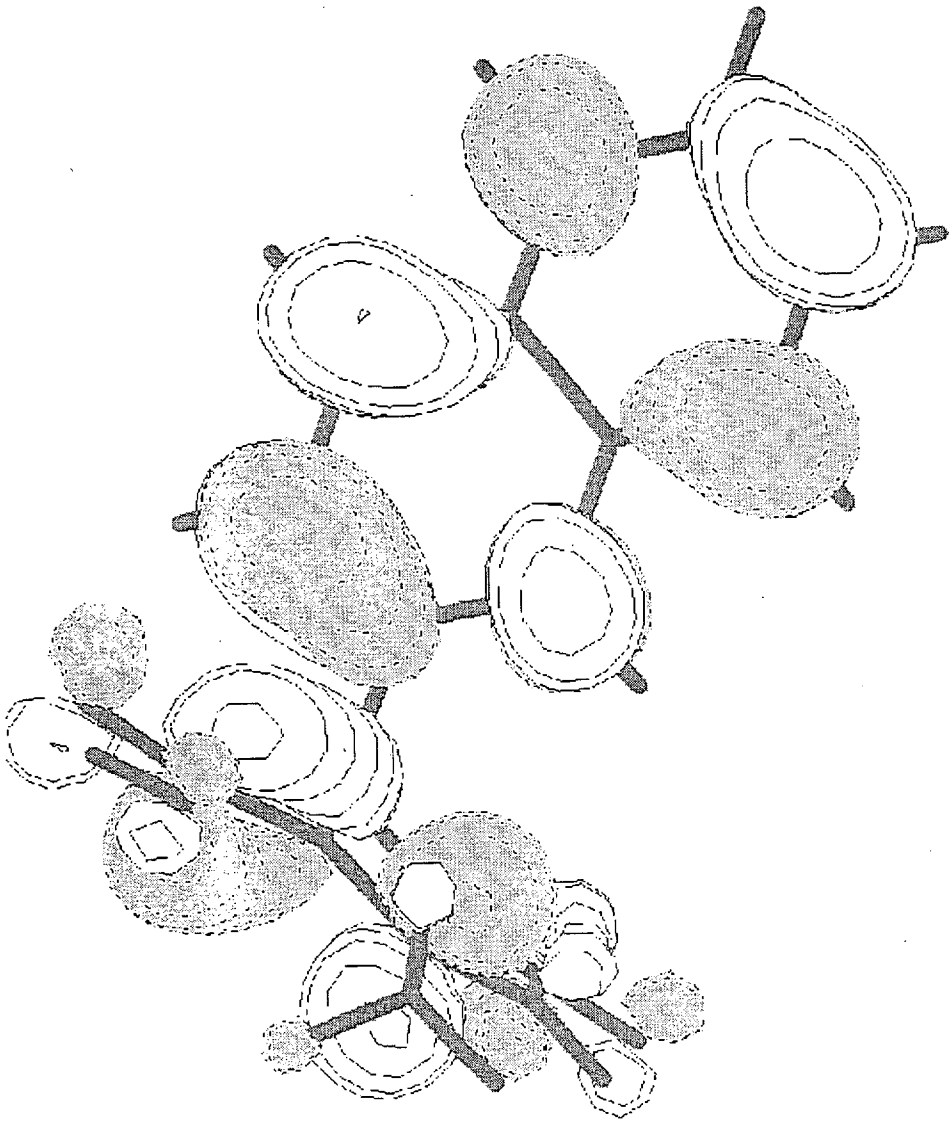

Figure S2b. Plots[49] of the LUMO of s-cis-**(R)-3aA** (HF/6-31G*, AM1 geometry).

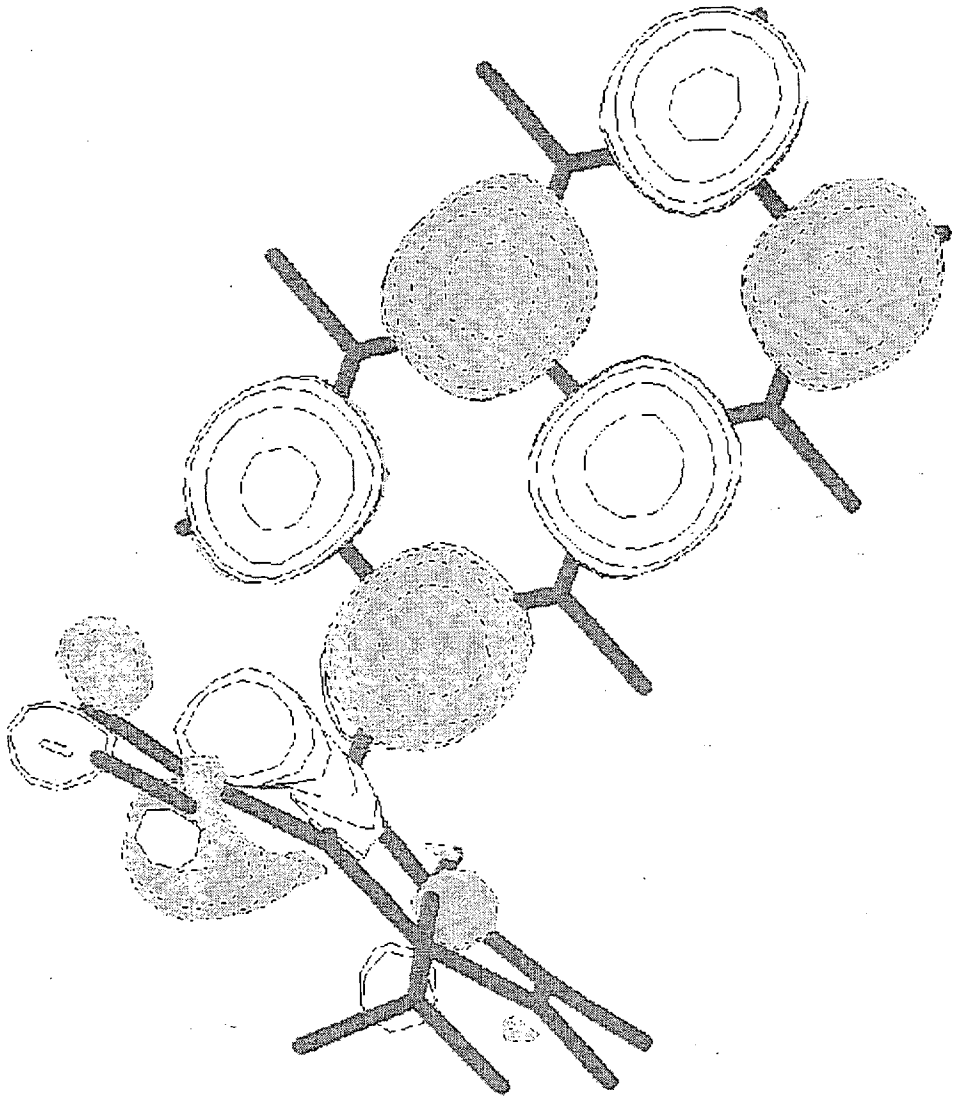

Figure S2. Plot[49] of the LUMO+1 of s-cis-**(R)-3aA** (HF/6-31G*, AM1 geometry).

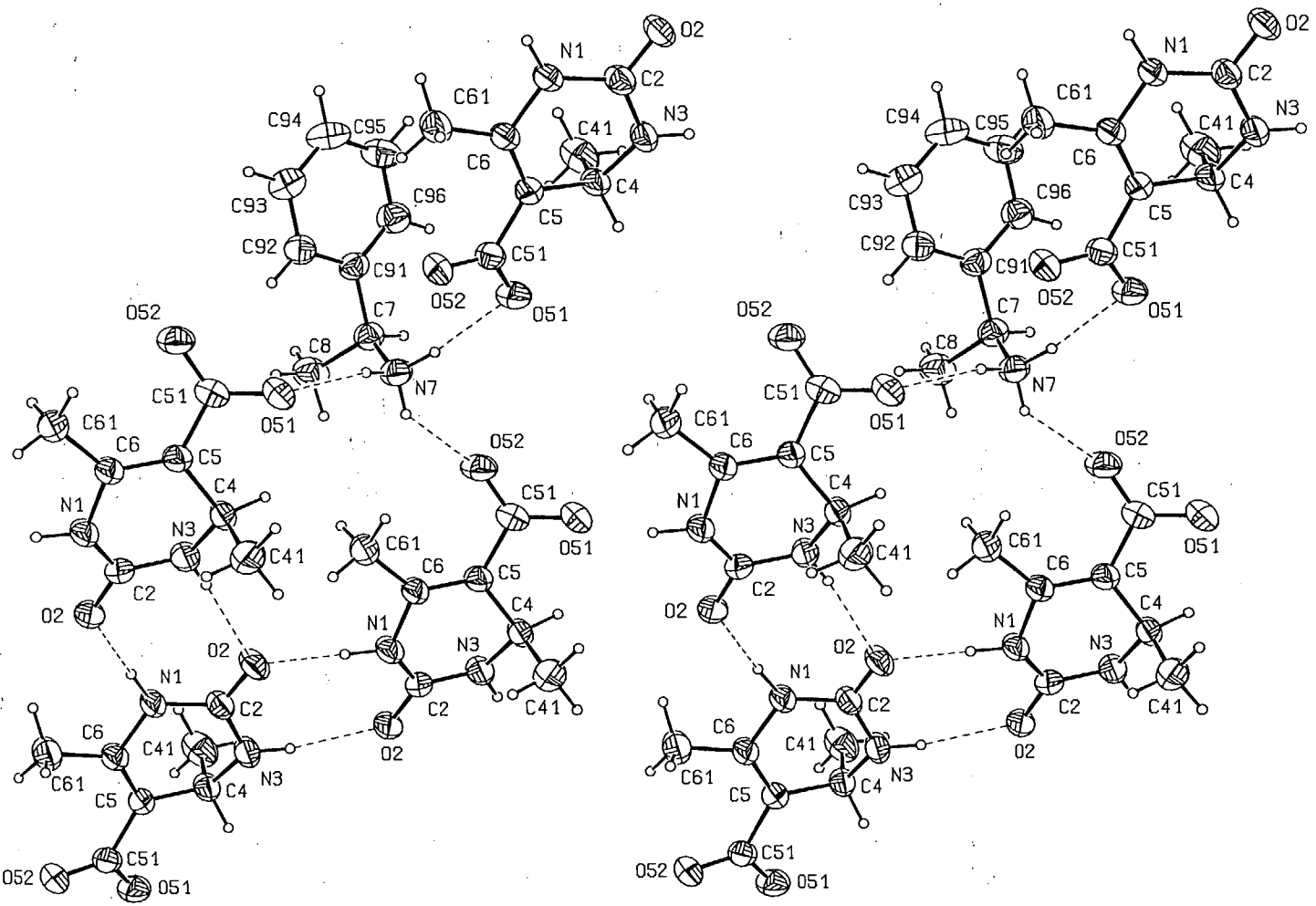

Figure S3. Stereoscopic ORTEP[37] plot of **6** showing the atomic numbering scheme and the hydrogen bonds drawn with dashed lines. One cation and four symmetry related anions are plotted in order to show all the five different hydrogen bonds. The probability ellipsoids are drawn at the 90% probability level.

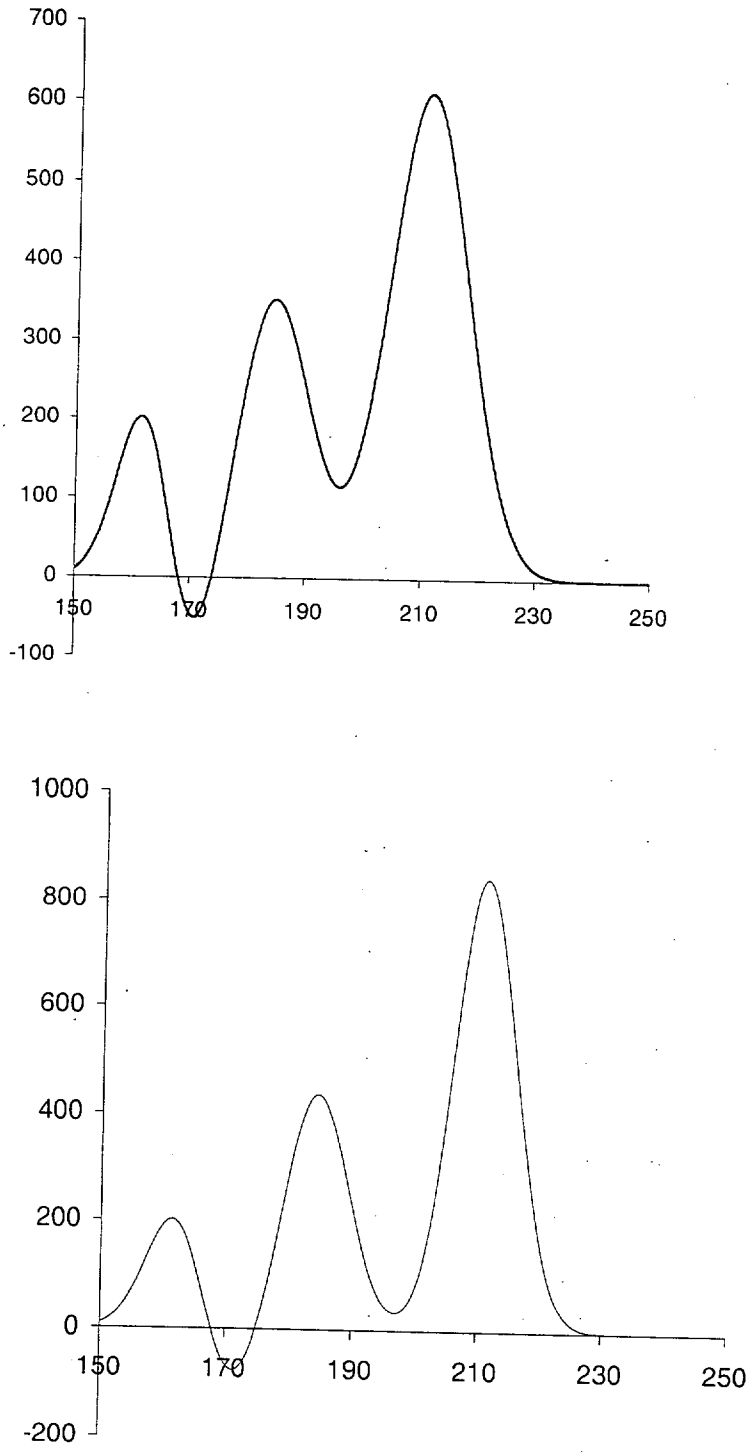

Figure S4 Influence of halfbandwidths on simulated CD spectra of **1a**: constant halfwidth (7 nm, upper part); variable halfwidts (< 170 nm: 7.02 nm, 171 – 200 nm: 8.72 nm; >200 nm: 9.68 nm, lower part)