

Table A. Geometries (in Å), rotational constants and dipole moments of uracil tautomers (RI-MP2/TZVPP level of theory).

| Atom number ^a Structure ^b | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | I ^c | μ ^d |
|--|--------|--------|--------|--------|-------|--------|--------|--------|--------|--------|--------|--------|----------------|----------------|
| u1 | | | | | | | | | | | | | | |
| x | 1.015 | -0.369 | -0.944 | -0.310 | 1.140 | 1.736 | 1.472 | -0.998 | -1.954 | -0.968 | 1.713 | 2.810 | 3.907 | 5.03 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.020 | |
| z | -1.174 | -1.218 | 0.035 | 1.287 | 1.199 | -0.010 | -2.071 | -2.257 | 0.048 | 2.312 | 2.110 | -0.127 | 1.332 | |
| u1o2l | | | | | | | | | | | | | | |
| x | 1.063 | -0.226 | -0.912 | -0.301 | 1.134 | 1.724 | -0.931 | -1.923 | -0.997 | 1.713 | 2.803 | -1.875 | 3.943 | 2.47 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.002 | |
| z | -1.223 | -1.138 | 0.034 | 1.310 | 1.204 | -0.022 | -2.284 | 0.060 | 2.312 | 2.113 | -0.104 | -2.101 | 1.328 | |
| u1o2r | | | | | | | | | | | | | | |
| x | 1.051 | -0.239 | -0.920 | -0.298 | 1.140 | 1.728 | -1.028 | -1.931 | -0.974 | 1.723 | 2.806 | -0.426 | 3.964 | 3.66 |
| y | 0.000 | 0.000 | -0.002 | 0.000 | 0.000 | 0.000 | 0.001 | -0.001 | 0.001 | 0.000 | 0.000 | 0.001 | 2.010 | |
| z | -1.234 | -1.125 | 0.038 | 1.305 | 1.187 | -0.039 | -2.209 | 0.024 | 2.320 | 2.094 | -0.128 | -2.966 | 1.334 | |
| u1o4l | | | | | | | | | | | | | | |
| x | 1.036 | -0.344 | -0.902 | -0.187 | 1.179 | 1.714 | -1.094 | -1.912 | -0.943 | 1.802 | 2.794 | -0.361 | 3.932 | 7.80 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.011 | |
| z | -1.305 | -1.243 | 0.061 | 1.197 | 1.123 | -0.184 | -2.198 | 0.113 | 2.308 | 2.002 | -0.291 | 3.076 | 1.330 | |
| u1o4r | | | | | | | | | | | | | | |
| x | 1.038 | -0.338 | -0.905 | -0.195 | 1.169 | 1.708 | -1.093 | -1.916 | -0.837 | 1.776 | 2.788 | -1.792 | 3.946 | 6.20 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 1.996 | |
| z | -1.308 | -1.253 | 0.054 | 1.200 | 1.124 | -0.179 | -2.205 | 0.058 | 2.379 | 2.012 | -0.280 | 2.247 | 1.326 | |
| u3o2l | | | | | | | | | | | | | | |
| x | 1.025 | -0.329 | -1.023 | -0.355 | 1.109 | 1.752 | 1.462 | -0.916 | -0.977 | 1.661 | 2.825 | -1.866 | 3.920 | 7.00 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.025 | |
| z | -1.146 | -1.061 | 0.021 | 1.262 | 1.207 | 0.022 | -2.051 | -2.270 | 2.312 | 2.133 | -0.086 | -2.086 | 1.335 | |
| u3o2r | | | | | | | | | | | | | | |
| x | 1.034 | -0.333 | -1.020 | -0.363 | 1.103 | 1.754 | 1.519 | -0.997 | -0.991 | 1.646 | 2.828 | -0.384 | 3.922 | 10.00 |
| y | 0.000 | 0.000 | 0.001 | 0.000 | 0.000 | 0.000 | -0.005 | -0.001 | 0.000 | 0.000 | 0.000 | 0.006 | 2.010 | |
| z | -1.132 | -1.065 | 0.015 | 1.258 | 1.223 | 0.045 | -2.012 | -2.240 | 2.304 | 2.154 | -0.058 | -2.981 | 1.329 | |
| u3o4l | | | | | | | | | | | | | | |
| x | 0.985 | -0.428 | -1.018 | -0.276 | 1.153 | 1.748 | 1.403 | -1.030 | -0.947 | 1.736 | 2.819 | -0.312 | 3.941 | 8.60 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.019 | |
| z | -1.194 | -1.181 | 0.065 | 1.129 | 1.139 | -0.082 | -2.112 | -2.237 | 2.299 | 2.045 | -0.221 | 3.022 | 1.335 | |
| u3o4r | | | | | | | | | | | | | | |
| x | 0.985 | -0.423 | -1.012 | -0.266 | 1.157 | 1.748 | 1.407 | -1.028 | -0.872 | 1.716 | 2.819 | -1.821 | 3.915 | 5.50 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.036 | |
| z | -1.187 | -1.183 | 0.066 | 1.131 | 1.151 | -0.071 | -2.103 | -2.238 | 2.329 | 2.070 | -0.213 | 2.137 | 1.340 | |
| uo2lo4l | | | | | | | | | | | | | | |
| x | 1.040 | -0.277 | -0.968 | -0.250 | 1.146 | 1.735 | -0.998 | -0.975 | 1.737 | 2.813 | -1.923 | -0.369 | 3.955 | 4.00 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.027 | |
| z | -1.257 | -1.097 | 0.045 | 1.156 | 1.138 | -0.114 | -2.230 | 2.293 | 2.041 | -0.213 | -1.951 | 3.042 | 1.340 | |
| uo2lo4r | | | | | | | | | | | | | | |
| x | 1.040 | -0.274 | -0.965 | -0.243 | 1.149 | 1.735 | -0.994 | -0.897 | 1.720 | 2.813 | -1.920 | -1.840 | 3.932 | 2.50 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.038 | |
| z | -1.250 | -1.099 | 0.046 | 1.158 | 1.148 | -0.104 | -2.234 | 2.333 | 2.062 | -0.204 | -1.961 | 2.117 | 1.342 | |
| uo2ro4l | | | | | | | | | | | | | | |
| x | 1.018 | -0.303 | -0.983 | -0.248 | 1.150 | 1.731 | -1.061 | -0.956 | 1.747 | 2.808 | -0.437 | -0.341 | 4.021 | 4.60 |

| | | | | | | | | | | | | | | |
|----------------|--------|--------|--------|--------|-------|--------|--------|--------|-------|--------|--------|--------|-------|------|
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.008 | |
| z | -1.254 | -1.083 | 0.059 | 1.157 | 1.131 | -0.122 | -2.191 | 2.305 | 2.031 | -0.233 | -2.929 | 3.045 | 1.339 | |
| uo2ro4r | | | | | | | | | | | | | | |
| x | 1.018 | -0.300 | -0.978 | -0.240 | 1.155 | 1.732 | -1.061 | -0.880 | 1.733 | 2.808 | -0.437 | -1.824 | 3.991 | 1.60 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.022 | |
| z | -1.249 | -1.085 | 0.061 | 1.159 | 1.141 | -0.113 | -2.192 | 2.340 | 2.050 | -0.227 | -2.930 | 2.130 | 1.342 | |

^a Atom 1 is placed at the origin. ^b For atom numbering and tautomer classification *cf.* Figure 1.

^c Rotational constants A, B, C in GHz. ^d Dipole moments in Debye.

Table B. Geometries (in Å), rotational constants and dipole moments of thymine tautomers (RI-MP2/TZVPP level of theory).

| Atom number ^a Structure ^b | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 | I ^c | μ ^d |
|--|--------|--------|--------|--------|-------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|----------------|----------------|
| t1 | | | | | | | | | | | | | | | | | |
| x | 1.241 | -0.039 | -1.005 | -0.835 | 0.558 | 1.520 | 1.979 | -0.282 | -1.960 | -1.808 | 0.855 | 2.571 | 1.421 | 1.421 | -0.084 | 3.200 | 4.00 |
| y | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | -0.882 | 0.882 | 0.000 | 1.405 | |
| z | -1.105 | -1.621 | -0.636 | 0.752 | 1.190 | 0.239 | -1.789 | -2.813 | -0.966 | 1.490 | 2.660 | 0.490 | 2.951 | 2.952 | 3.207 | 0.982 | |
| t1o2l | | | | | | | | | | | | | | | | | |
| x | 1.297 | 0.058 | -0.980 | -0.831 | 0.555 | 1.513 | -0.222 | -1.942 | -1.827 | 0.872 | 2.558 | -1.173 | -0.053 | 1.450 | 1.447 | 3.211 | 2.10 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.001 | 0.002 | -0.002 | -0.880 | 0.882 | 1.398 | |
| z | -1.139 | -1.498 | -0.622 | 0.780 | 1.188 | 0.215 | -2.815 | -0.934 | 1.491 | 2.654 | 0.500 | -2.959 | 3.224 | 2.929 | 2.930 | 0.980 | |
| t1o2r | | | | | | | | | | | | | | | | | |
| x | 1.292 | 0.045 | -0.988 | -0.832 | 0.561 | 1.520 | -0.332 | -1.936 | -1.817 | 0.879 | 2.566 | 0.491 | 1.465 | 1.446 | -0.046 | 3.215 | 3.20 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.001 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | -0.875 | 0.887 | -0.010 | 1.407 | |
| z | -1.147 | -1.488 | -0.621 | 0.775 | 1.178 | 0.208 | -2.776 | -0.973 | 1.501 | 2.644 | 0.490 | -3.284 | 2.918 | 2.922 | 3.214 | 0.985 | |
| t1o4l | | | | | | | | | | | | | | | | | |
| x | 1.321 | 0.016 | -0.977 | -0.735 | 0.557 | 1.527 | -0.321 | -1.935 | -1.861 | 0.955 | 2.572 | -1.617 | 1.531 | 1.561 | 0.100 | 3.091 | 8.10 |
| y | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.001 | 0.000 | -0.001 | 0.000 | 0.000 | -0.001 | 0.002 | 0.890 | -0.872 | -0.018 | 1.430 | |
| z | -1.167 | -1.624 | -0.620 | 0.705 | 1.159 | 0.127 | -2.792 | -0.942 | 1.442 | 2.605 | 0.430 | 2.374 | 2.855 | 2.845 | 3.282 | 0.984 | |
| t1o4r | | | | | | | | | | | | | | | | | |
| x | 1.313 | 0.011 | -0.981 | -0.729 | 0.568 | 1.527 | -0.349 | -1.927 | -1.765 | 0.951 | 2.573 | -2.602 | 1.543 | 1.543 | 0.069 | 3.119 | 6.40 |
| y | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | -0.880 | 0.881 | 0.000 | 1.423 | |
| z | -1.186 | -1.633 | -0.616 | 0.711 | 1.148 | 0.109 | -2.794 | -0.972 | 1.574 | 2.597 | 0.405 | 1.098 | 2.843 | 2.843 | 3.231 | 0.983 | |
| t3o2l | | | | | | | | | | | | | | | | | |
| x | 1.234 | -0.066 | -1.075 | -0.854 | 0.549 | 1.534 | 1.949 | -0.226 | -1.793 | 0.833 | 2.586 | -1.184 | 1.401 | 1.397 | -0.112 | 3.234 | 6.30 |
| y | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | -0.881 | 0.883 | -0.002 | 1.407 | |
| z | -1.090 | -1.459 | -0.663 | 0.724 | 1.180 | 0.254 | -1.797 | -2.795 | 1.507 | 2.652 | 0.502 | -2.930 | 2.947 | 2.948 | 3.187 | 0.986 | |
| t3o2r | | | | | | | | | | | | | | | | | |
| x | 1.234 | -0.075 | -1.073 | -0.851 | 0.549 | 1.533 | 1.980 | -0.329 | -1.790 | 0.829 | 2.586 | 0.490 | 1.395 | 1.393 | -0.118 | 3.246 | 9.60 |
| y | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | 0.001 | 0.000 | 0.000 | 0.000 | -0.001 | -0.001 | -0.882 | 0.882 | -0.001 | 1.396 | |
| z | -1.085 | -1.465 | -0.662 | 0.723 | 1.187 | 0.263 | -1.759 | -2.792 | 1.505 | 2.660 | 0.509 | -3.297 | 2.957 | 2.957 | 3.190 | 0.982 | |
| t3o4l | | | | | | | | | | | | | | | | | |
| x | 1.226 | -0.094 | -1.082 | -0.772 | 0.560 | 1.536 | 1.948 | -0.282 | -1.834 | 0.913 | 2.588 | -1.513 | 1.493 | 1.494 | 0.043 | 3.143 | 9.00 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | -0.882 | 0.882 | 0.000 | 1.424 | |
| z | -1.107 | -1.602 | -0.643 | 0.619 | 1.156 | 0.206 | -1.811 | -2.804 | 1.453 | 2.619 | 0.456 | 2.360 | 2.885 | 2.885 | 3.273 | 0.986 | |
| t3o4r | | | | | | | | | | | | | | | | | |
| x | 1.223 | -0.094 | -1.075 | -0.749 | 0.575 | 1.543 | 1.942 | -0.294 | -1.752 | 0.874 | 2.598 | -2.562 | 1.446 | 1.445 | -0.050 | 3.175 | 5.80 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | -0.881 | 0.882 | 0.000 | 1.425 | |
| z | -1.112 | -1.603 | -0.635 | 0.626 | 1.160 | 0.203 | -1.819 | -2.804 | 1.523 | 2.632 | 0.442 | 0.992 | 2.917 | 2.917 | 3.201 | 0.990 | |
| to2lo4l | | | | | | | | | | | | | | | | | |
| x | 1.302 | 0.015 | -1.030 | -0.760 | 0.549 | 1.534 | -0.253 | -1.858 | 0.916 | 2.578 | -1.217 | -1.575 | 1.507 | 1.503 | 0.053 | 3.148 | 4.40 |
| y | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | -0.001 | 0.002 | 0.001 | -0.879 | 0.882 | -0.003 | 1.431 | |
| z | -1.143 | -1.467 | -0.643 | 0.654 | 1.152 | 0.172 | -2.785 | 1.441 | 2.609 | 0.468 | -2.854 | 2.360 | 2.859 | 2.860 | 3.274 | 0.990 | |
| to2lo4r | | | | | | | | | | | | | | | | | |
| x | 1.297 | 0.015 | -1.028 | -0.742 | 0.563 | 1.540 | -0.261 | -1.779 | 0.885 | 2.586 | -1.223 | -2.577 | 1.482 | 1.452 | -0.024 | 3.175 | 2.70 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | -0.001 | -0.871 | 0.889 | -0.017 | 1.429 | |
| z | -1.147 | -1.471 | -0.637 | 0.658 | 1.156 | 0.170 | -2.788 | 1.521 | 2.620 | 0.456 | -2.858 | 0.975 | 2.885 | 2.891 | 3.214 | 0.992 | |
| to2ro4l | | | | | | | | | | | | | | | | | |
| x | 1.273 | -0.022 | -1.054 | -0.758 | 0.560 | 1.533 | -0.341 | -1.840 | 0.944 | 2.581 | 0.502 | -1.542 | 1.538 | 1.533 | 0.089 | 3.168 | 5.00 |

| | | | | | | | | | | | | | | | | | |
|----------------|--------|--------|--------|--------|-------|--------|--------|--------|-------|--------|--------|--------|--------|-------|--------|-------|------|
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.879 | 0.883 | -0.003 | 1.426 | |
| z | -1.154 | -1.463 | -0.630 | 0.660 | 1.146 | 0.157 | -2.769 | 1.467 | 2.599 | 0.435 | -3.238 | 2.381 | 2.842 | 2.843 | 3.273 | 0.990 | |
| to2ro4r | | | | | | | | | | | | | | | | | |
| x | 1.270 | -0.020 | -1.049 | -0.739 | 0.574 | 1.539 | -0.350 | -1.763 | 0.909 | 2.590 | 0.491 | -2.566 | 1.504 | 1.482 | 0.005 | 3.194 | 1.90 |
| y | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.001 | 0.000 | 0.000 | -0.001 | 0.000 | 0.000 | -0.873 | 0.887 | -0.013 | 1.426 | |
| z | -1.157 | -1.466 | -0.624 | 0.664 | 1.150 | 0.157 | -2.770 | 1.541 | 2.612 | 0.426 | -3.245 | 1.001 | 2.872 | 2.877 | 3.213 | 0.992 | |

^a Atom 1 is placed at the origin. ^b For atom numbering and tautomer classification *cf.* Figure 1.

^c Rotational constants A, B, C in GHz. ^d Dipole moments in Debye.

Part C





