

Electronic Supplementary Material

1,2,4-Triazine-picolinamide functionalized, nonadentate chelates for the segregation of lanthanides(III) and actinides(III) in biphasic systems

*Gary L. Guillet, I.F. Dempsey Hyatt, Patrick C. Hillesheim, Michael J. Scott**

Department of Chemistry, P.O. Box 117200, University of Florida, Gainesville, Florida 32611

mjscott@chem.ufl.edu

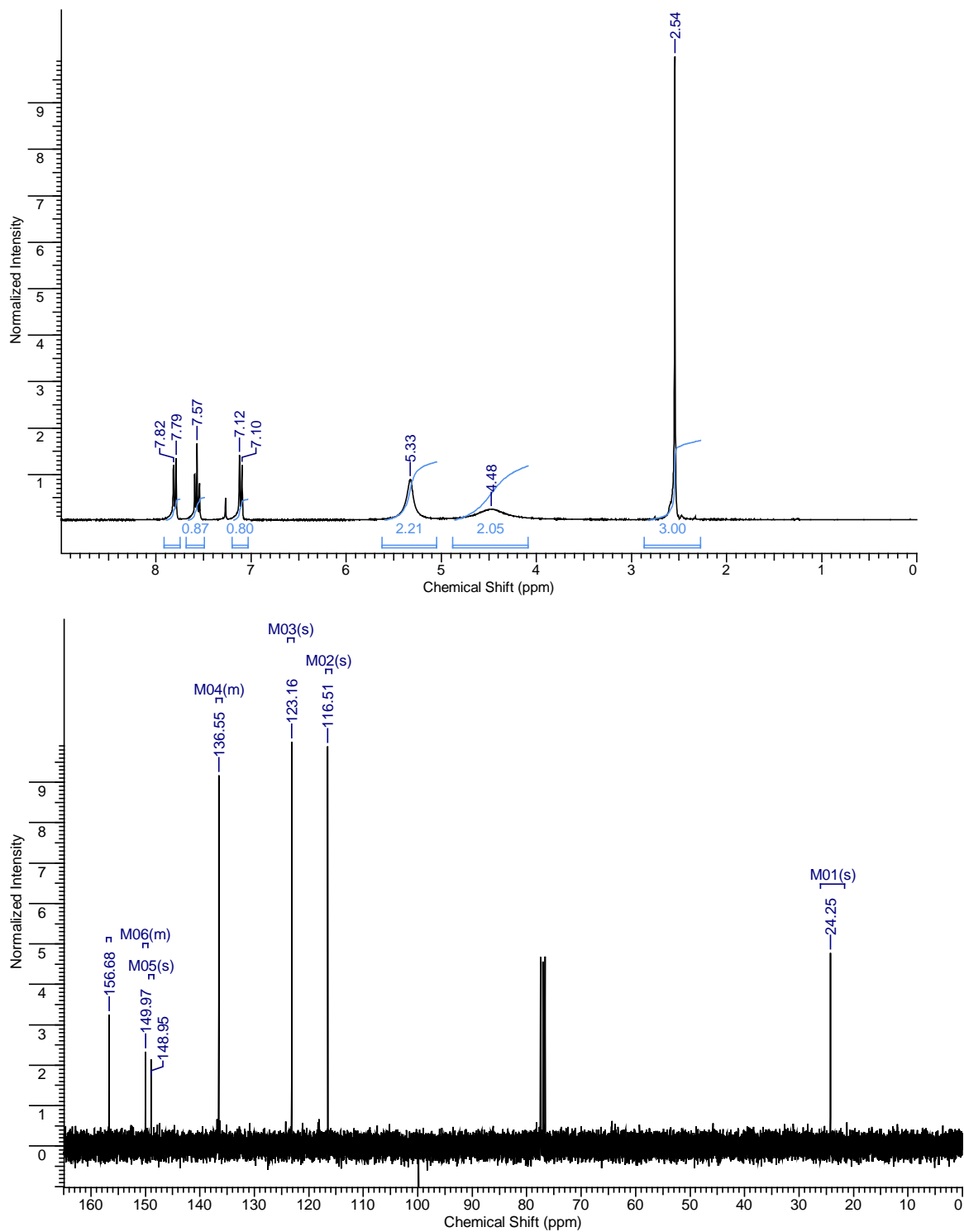


Figure S1 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **1** in CDCl₃

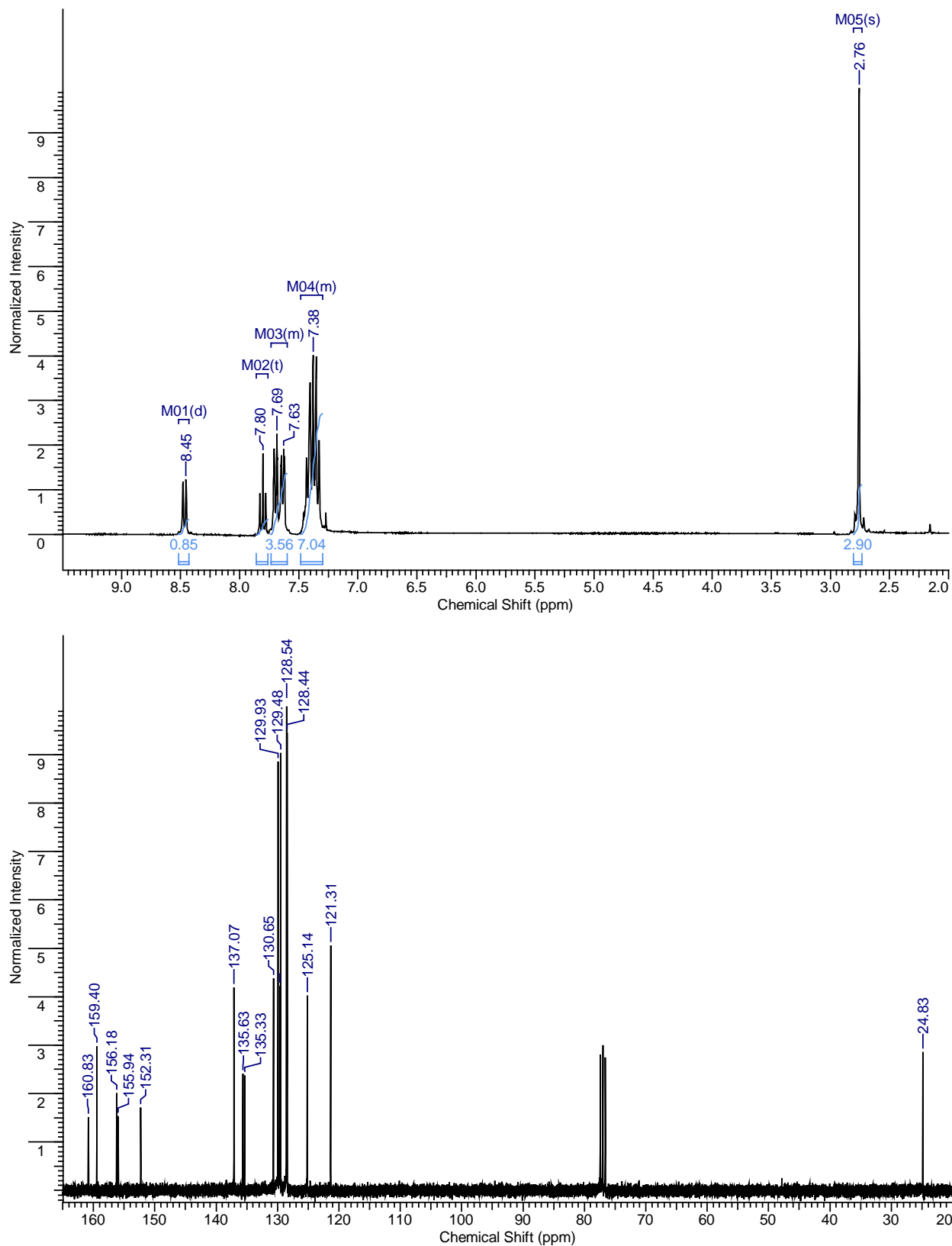


Figure S2 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **2** in CDCl₃

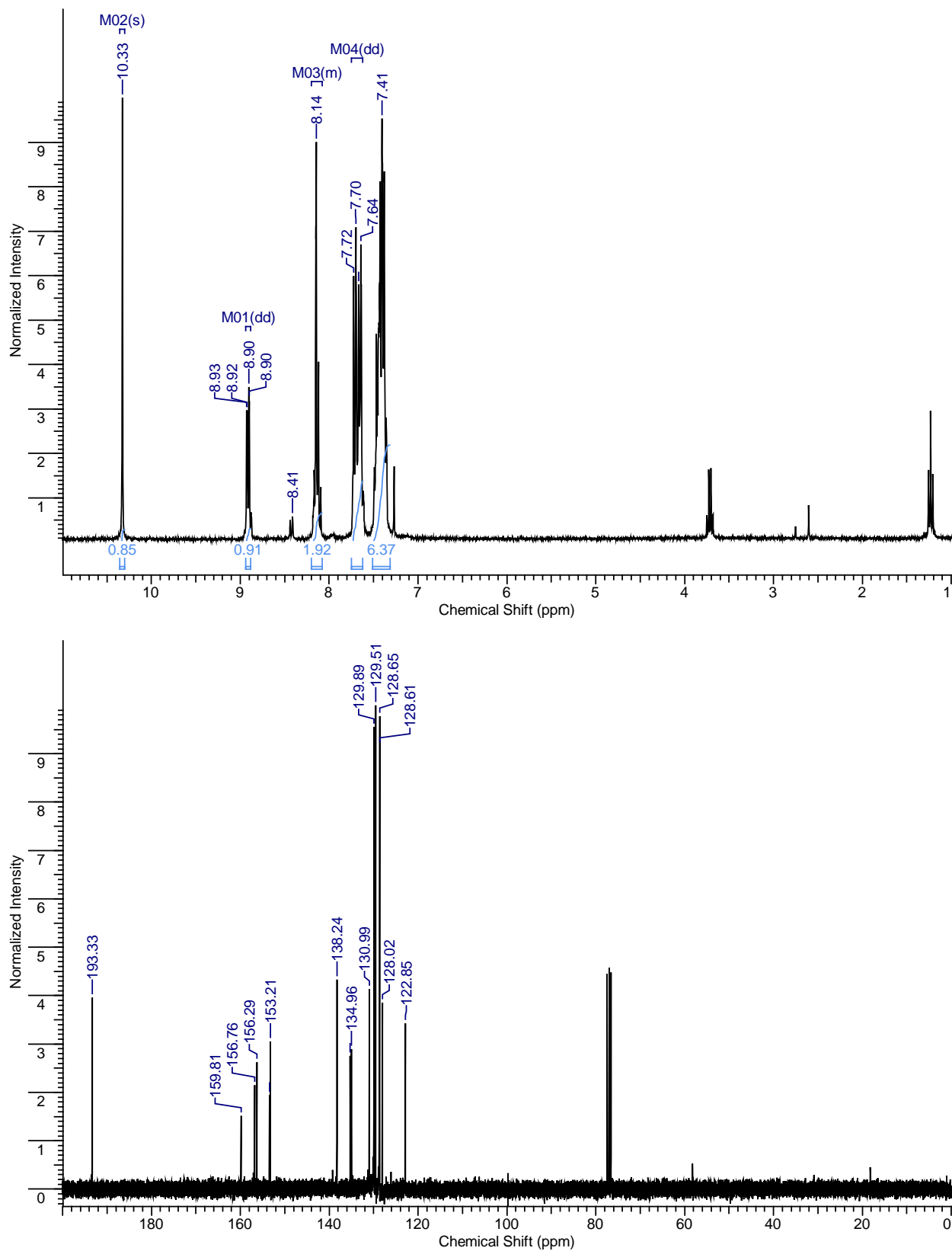


Figure S3 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **3** in CDCl₃

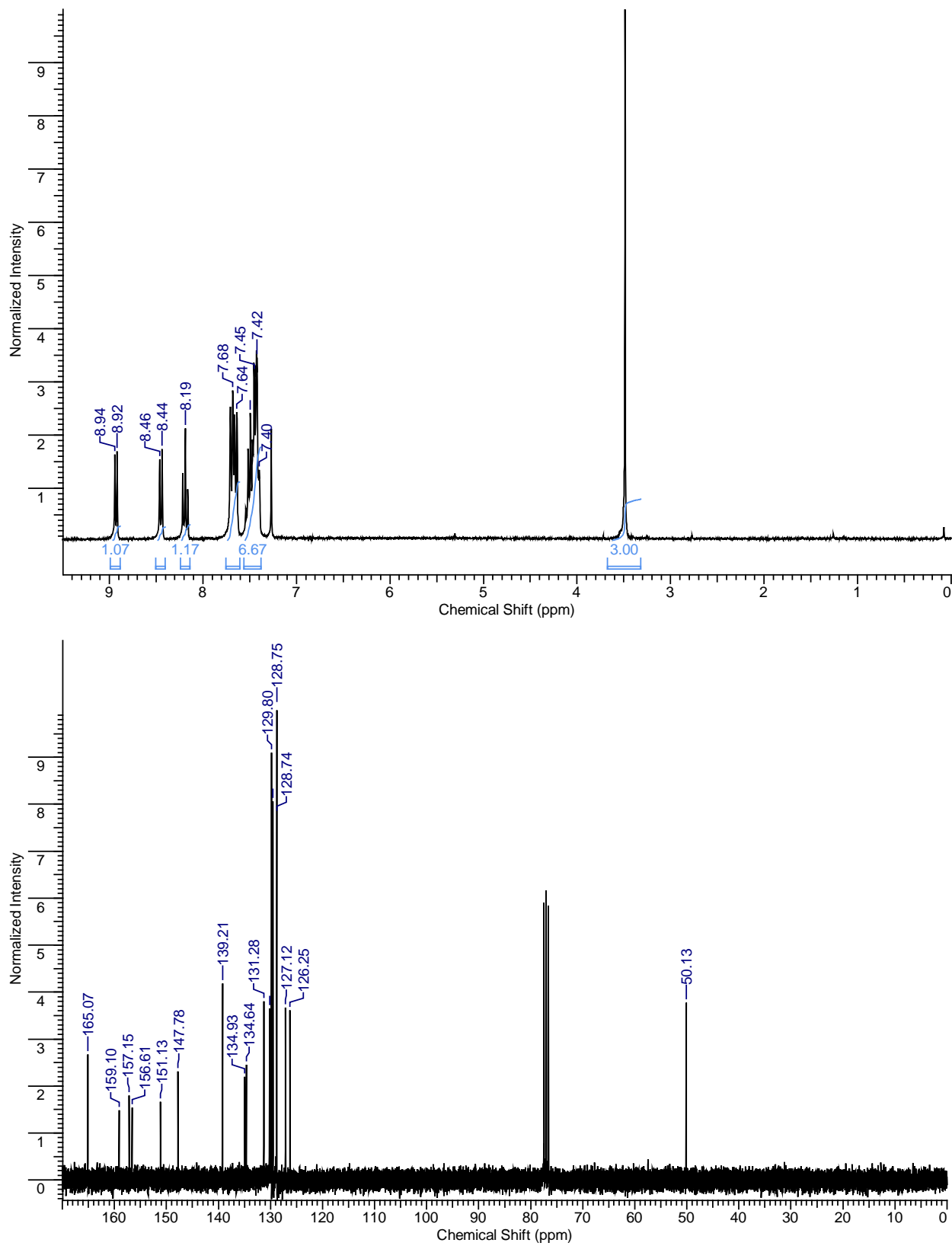


Figure S4 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **4** in CDCl₃

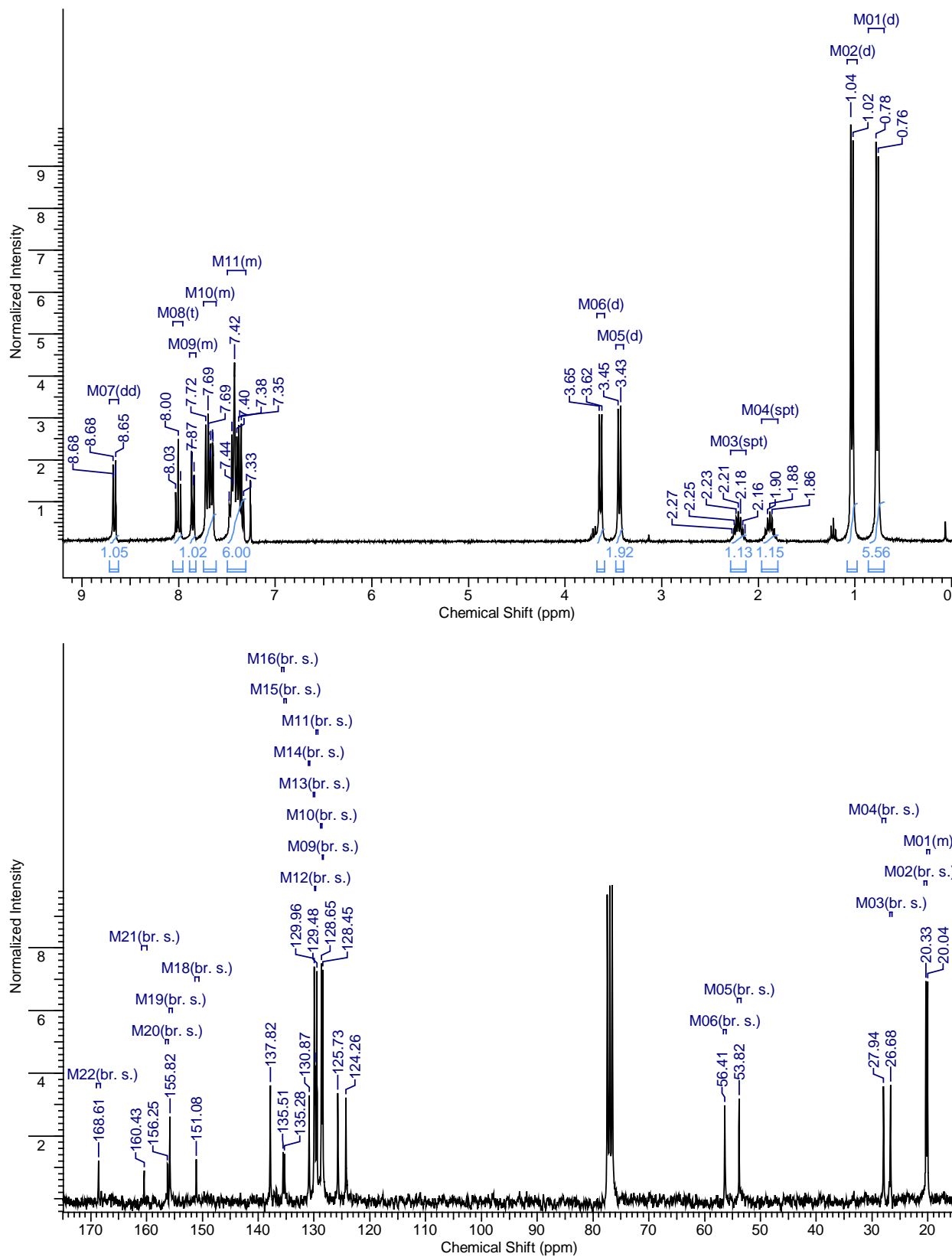


Figure S5 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **5** in CDCl₃

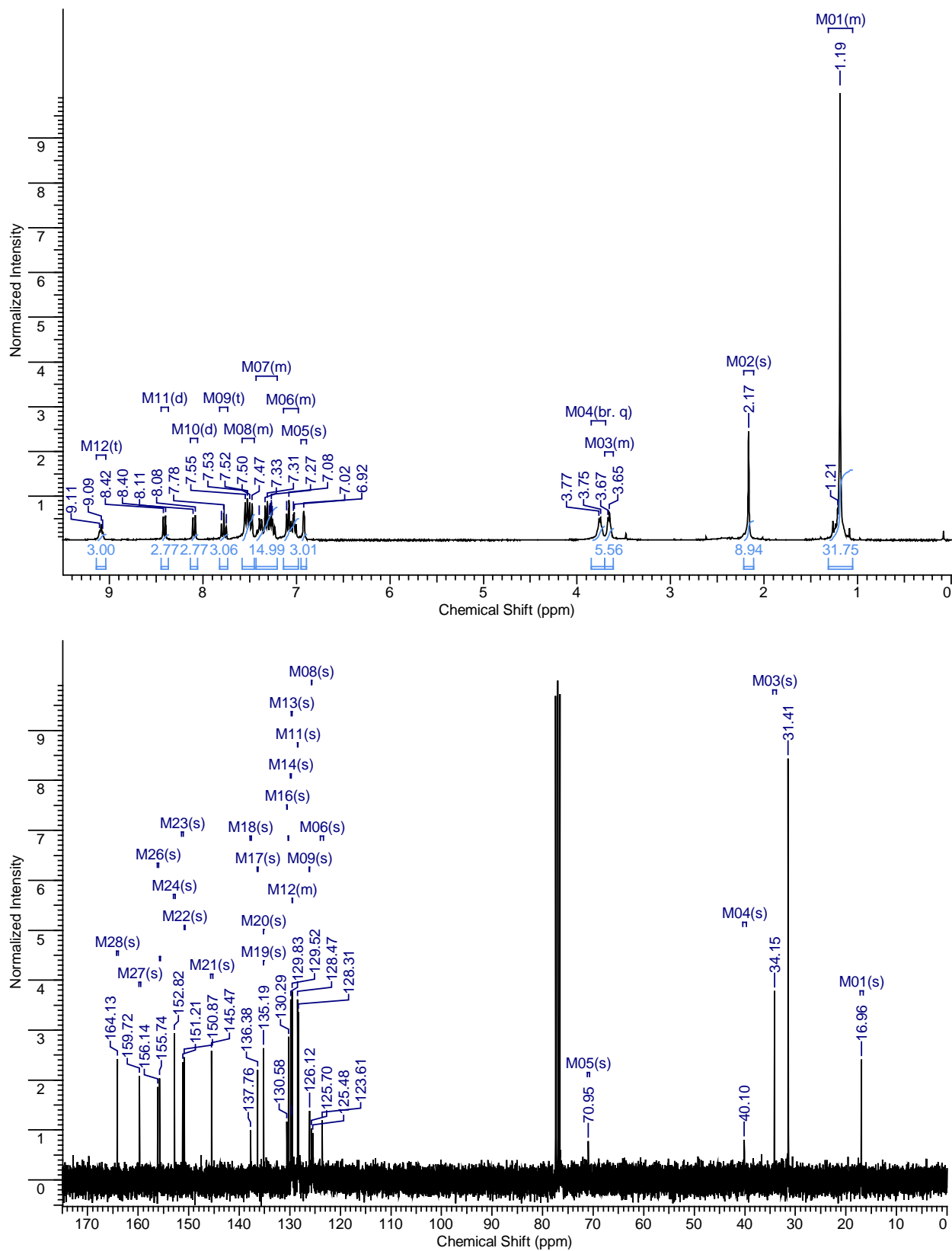


Figure S6 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **9** in CDCl₃

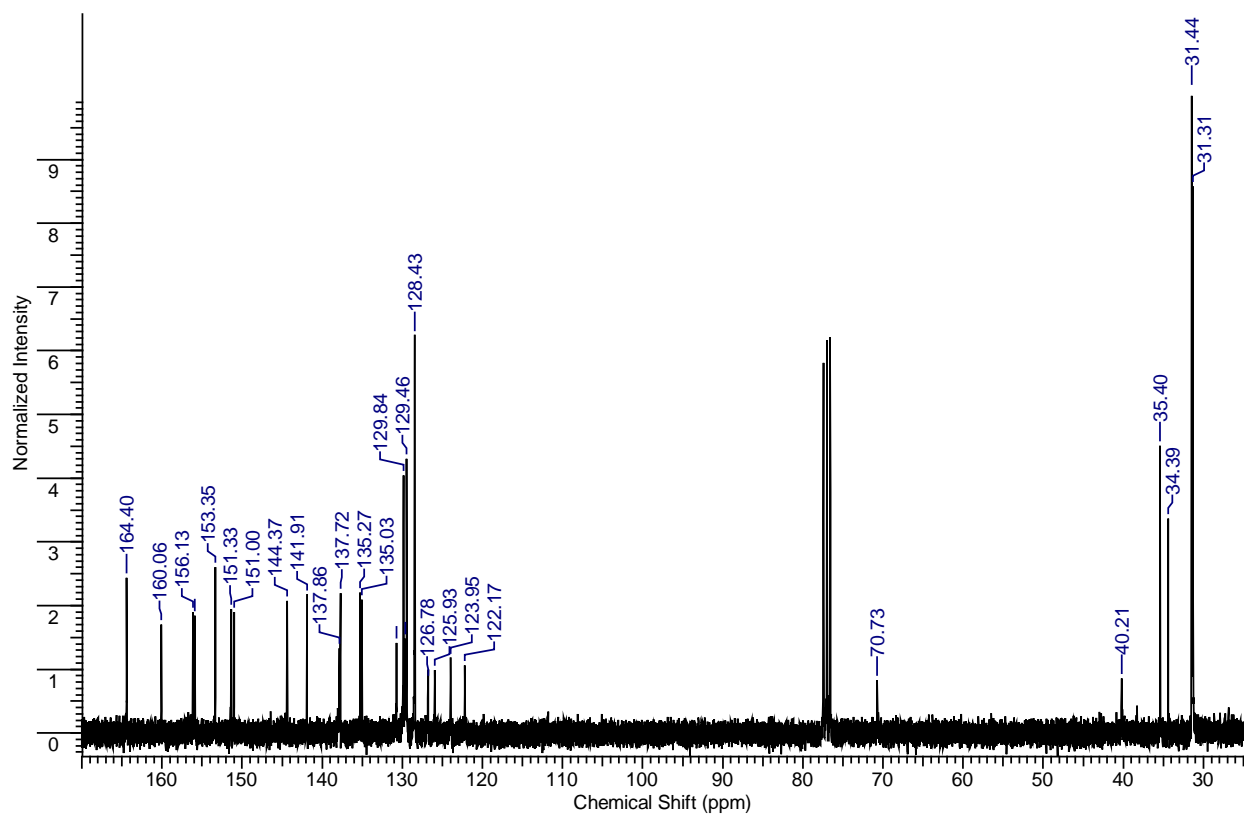
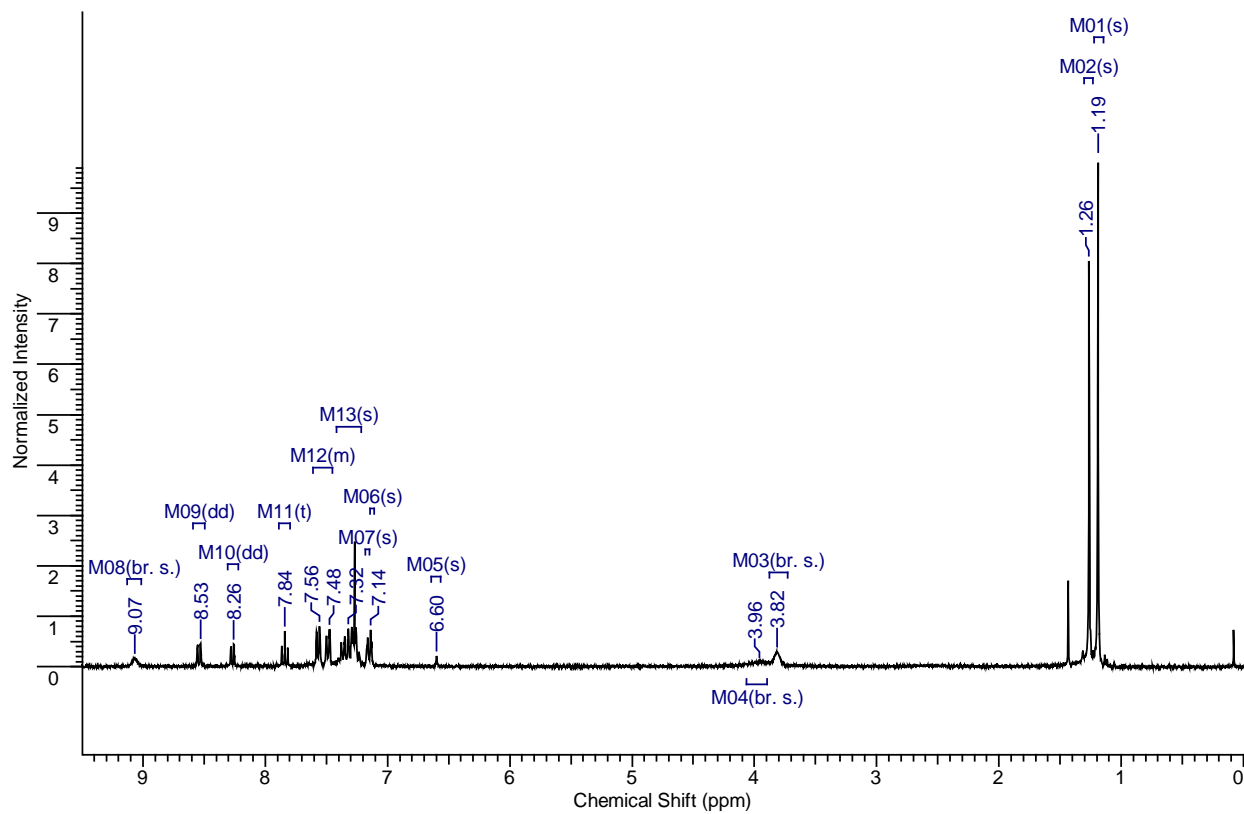


Figure S7 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **10** in CDCl₃

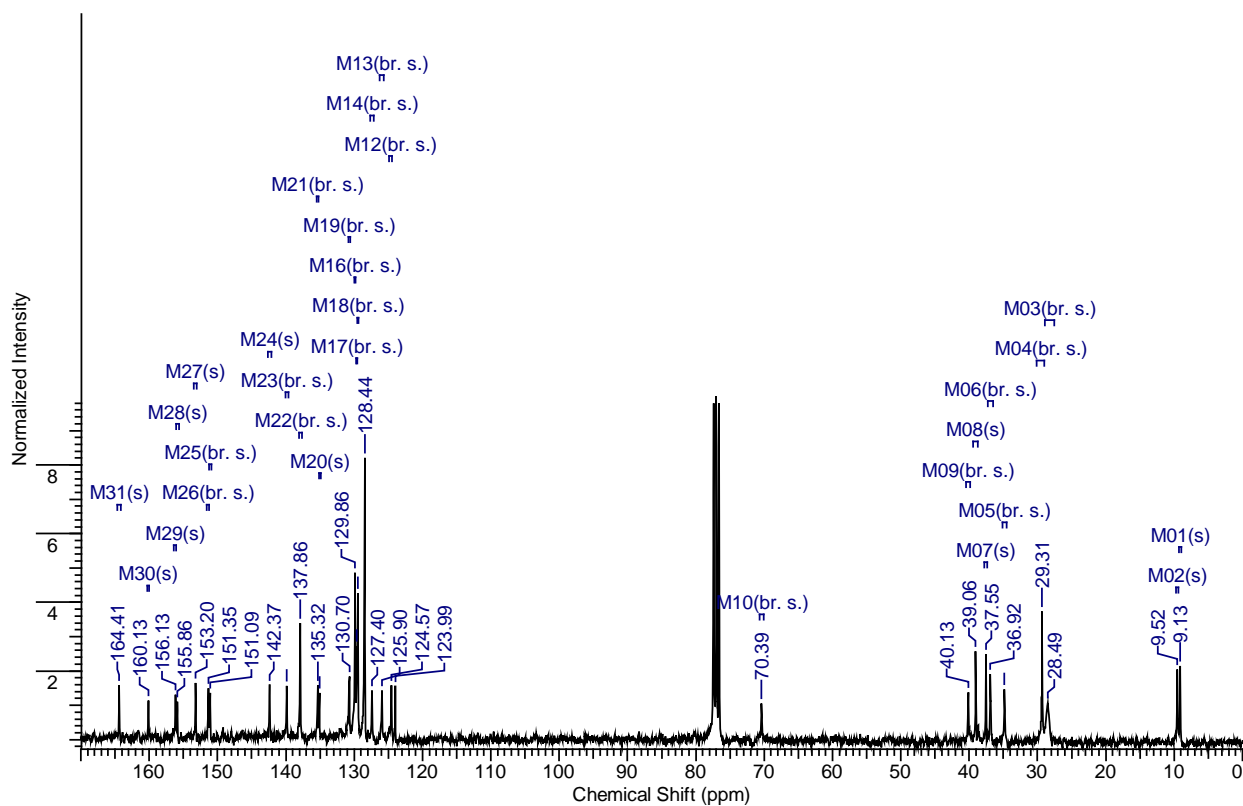
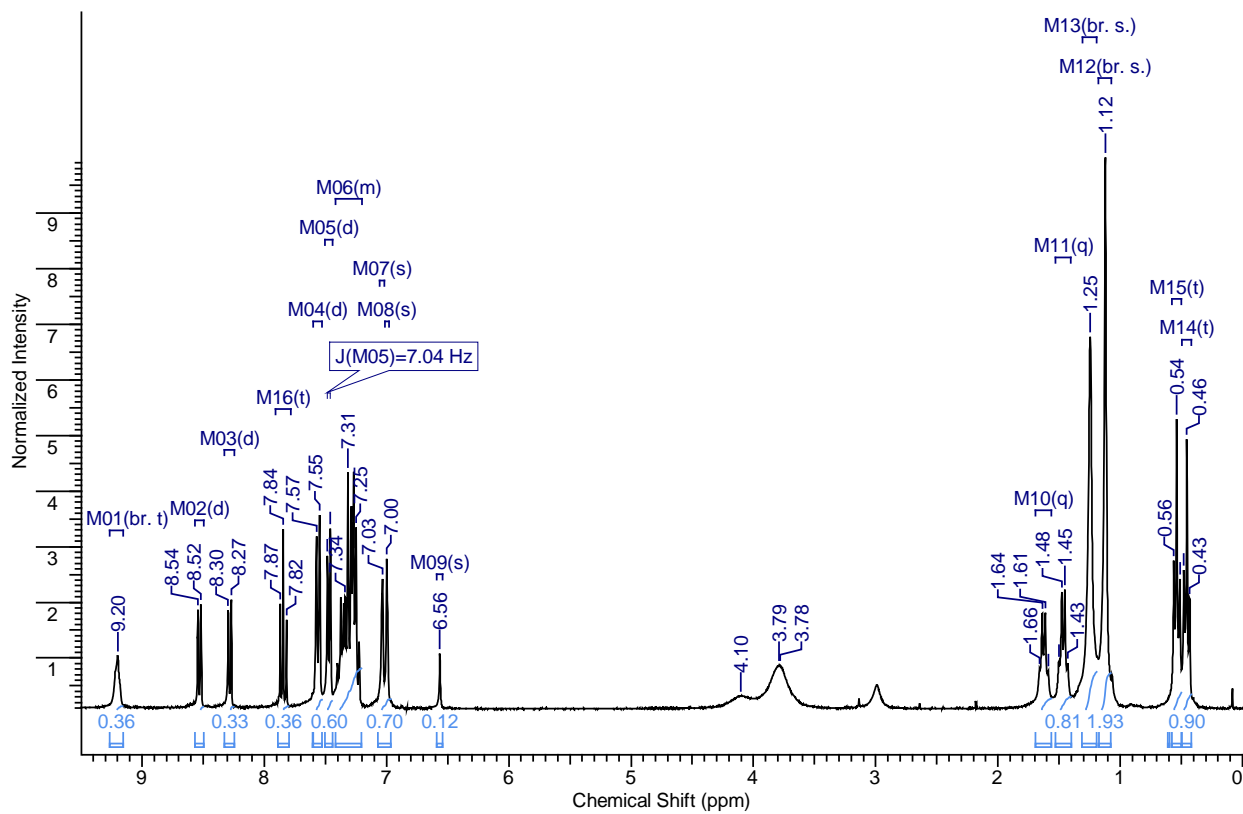


Figure S8 ¹H-NMR (top) and ¹³C-NMR (bottom) spectra of **11** in CDCl₃

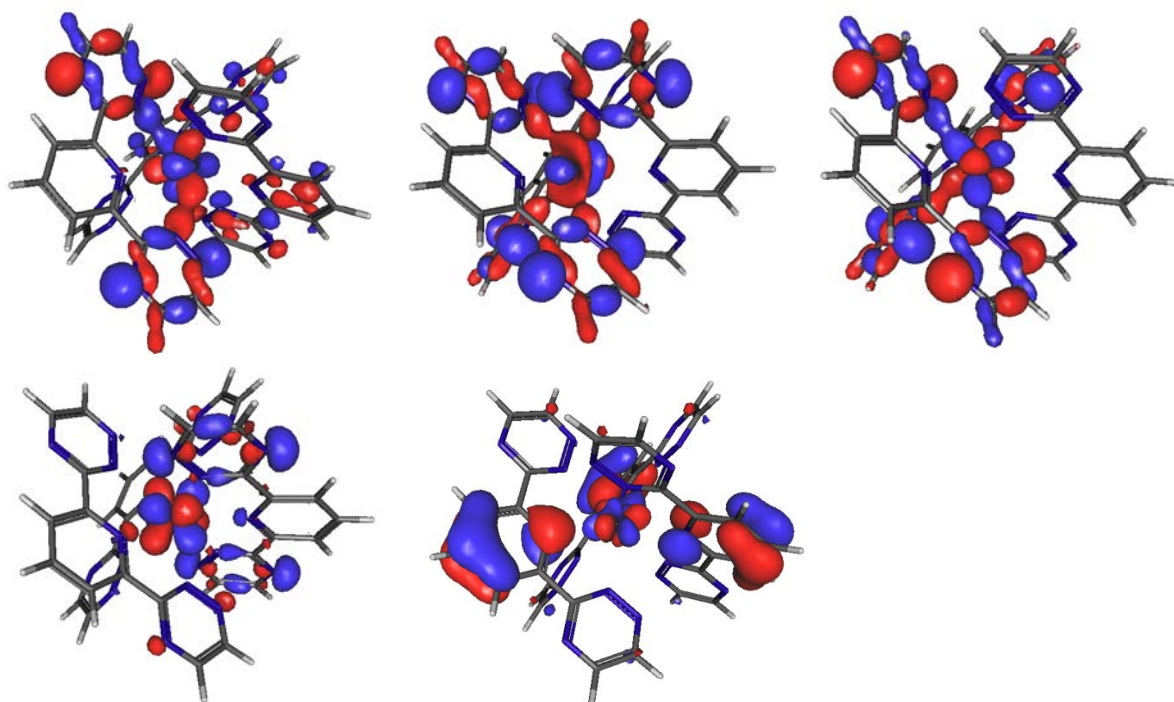


Figure S10 Am(BTP-H)₃³⁺ HOMO[-9], HOMO[-10], HOMO[-11], HOMO[-13], and HOMO[-17] bonding interactions from top left to bottom right, respectively.

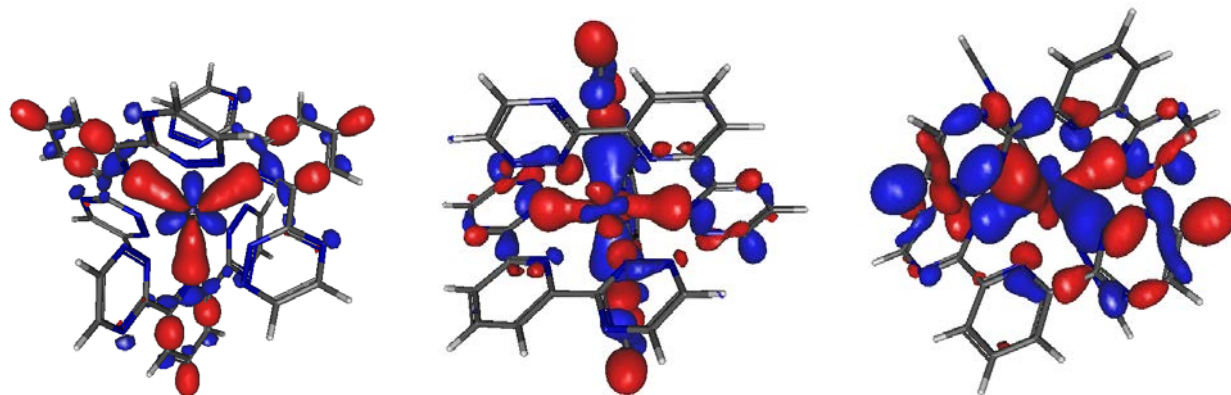


Figure S10 Cm(BTP-H)₃³⁺ HOMO[-33], HOMO[-43], and HOMO[-44] bonding interactions.

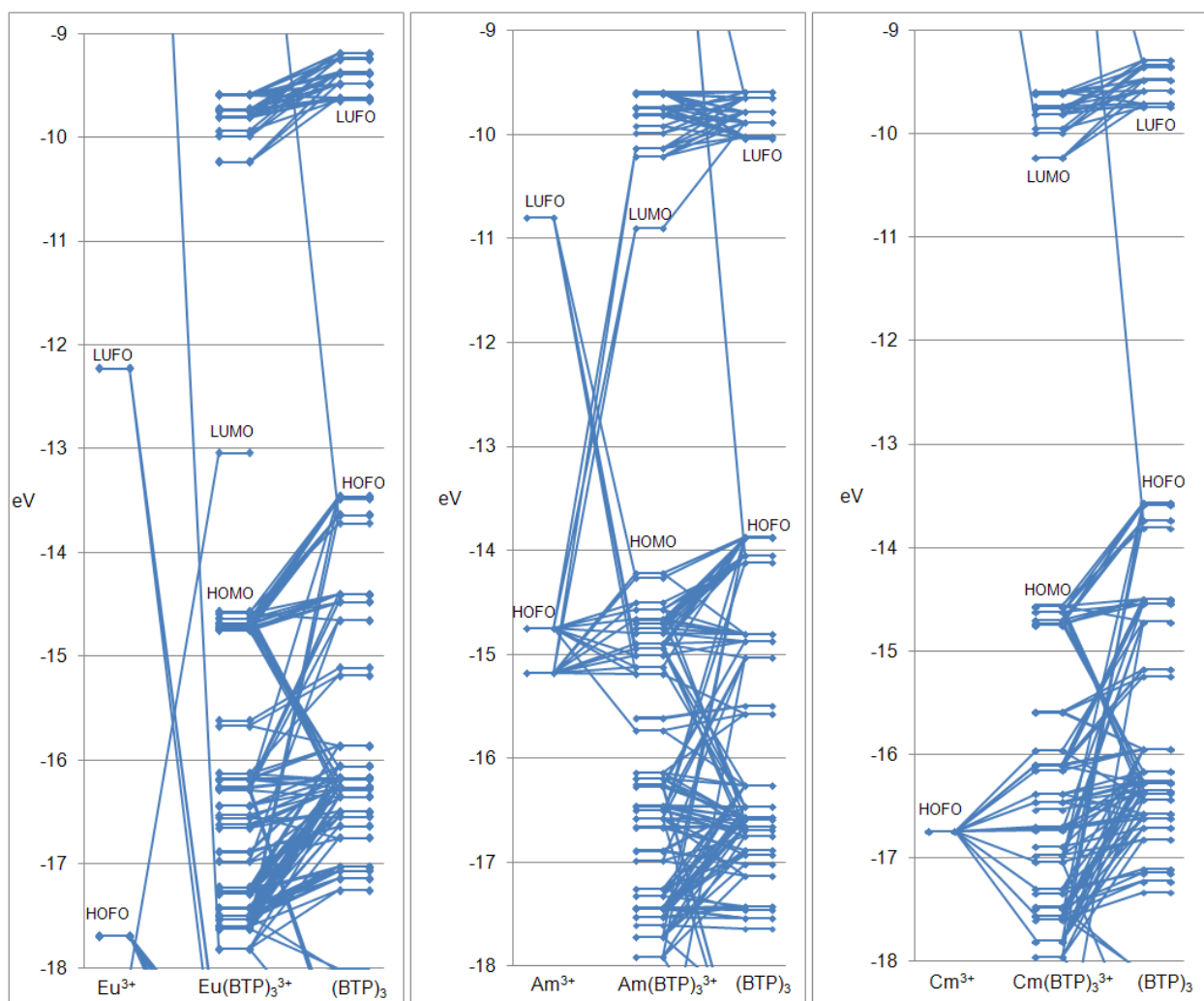


Figure S11 Orbital diagrams for $\text{Eu}(\text{BTP})_3^{3+}$, $\text{Am}(\text{BTP})_3^{3+}$, $\text{Cm}(\text{BTP})_3^{3+}$. Orbital contributions of less than 5% are not included. Diagrams made using AO Mix software.

Chart S1 Output file for $\text{Eu}(\text{BTP-H})_3^{3+}$

HOMO-35[#167, -17.598 eV]= Fr 1: 3.4%L+4

Fr 2: 47.1%H-19 S(0.123) OP(0.02)
13.7%H-31 S(0.039) OP(0.00)
6.3%H-24 S(-0.037) OP(0.00)
6.0%H-2 S(0.122) OP(0.01)
3.8%H-28 S(0.024) OP(0.00)
3.1%H-3 S(0.111) OP(0.00)
2.7%H-39 S(0.011) OP(0.00)

HOMO-36[#166, -17.602 eV]= Fr 2: 86.6%H-38 4.7%H-26 2.2%H-32 1.4%H-47 1.1%H-34

HOMO-37[#165, -17.627 eV]= Fr 1: 3.2%L+2

Fr 2: 50.1%H-18 S(0.120) OP(0.02)
13.9%H-30 S(0.038) OP(0.00)
5.0%H-40 S(0.011) OP(0.00)
4.9%H-1 S(-0.119) OP(0.01)
4.5%H-4 S(-0.108) OP(0.01)
3.7%H-23 S(0.036) OP(0.00)
2.1%H-21 S(-0.020) OP(0.00)

HOMO-38[#164, -17.822 eV]= Fr 1: 4.7%L+6

Fr 2: 34.7%H-26 S(0.301) OP(0.06)
28.8%H-32 S(-0.196) OP(0.03)
8.3%H-47 S(-0.218) OP(-0.02)
8.0%H-38 S(0.011) OP(0.00)
3.8%H-0 S(0.078) OP(0.00)
3.0%H-25 S(0.025) OP(0.00)
1.5%H-53 S(0.136) OP(-0.01)

HOMO-39[#163, -18.150 eV]= Fr 2: 84.7%H-39 6.3%H-48 1.9%H-19 1.5%H-41

HOMO-40[#162, -18.153 eV]= Fr 2: 85.7%H-40 6.3%H-49 2.6%H-18

HOMO-41[#161, -18.173 eV]= Fr 2: 86.1%H-41 5.9%H-50 2.1%H-29 1.5%H-39 1.1%H-22

HOMO-42[#160, -18.772 eV]= Fr 1: 53.0%L+0 16.0%H-1 7.6%H-0 1.8%H-5

Fr 2: 15.3%H-46 S(0.002 -0.001 -0.001 -0.002) OP(0.00 0.00 0.00 0.00)
2.5%H-44 S(0.005 -0.003 -0.002 0.002) OP(0.00 0.00 0.00 0.00)

HOMO-43[#159, -18.796 eV]= Fr 1: 24.1%H-0 18.5%H-4 11.4%H-1 8.4%H-5 2.0%H-3

Fr 2: 26.1%H-45 S(-0.002 0.000 0.002 0.000 0.002) OP(0.00 0.00 0.00 0.00 0.00)
1.5%H-47 S(-0.001 -0.005 0.001 -0.003 0.000) OP(0.00 0.00 0.00 0.00 0.00)
1.5%H-51 S(-0.002 0.001 0.001 0.000 -0.005) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-44[#158, -18.815 eV]= Fr 1: 32.0%L+0 19.9%H-5 19.8%H-1 9.4%H-0 9.0%H-4

Fr 2: 3.3%H-46 S(0.002 -0.002 -0.001 -0.001 0.001) OP(0.00 0.00 0.00 0.00 0.00)
1.1%H-52 S(-0.002 -0.003 -0.002 -0.002 0.002) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-45[#157, -18.933 eV]= Fr 1: 7.8%H-1 4.0%L+0 3.7%H-0 1.0%H-5

Fr 2: 69.3%H-46 S(-0.001 0.002 -0.001 -0.002) OP(0.00 0.00 0.00 0.00)
3.8%H-52 S(-0.002 -0.002 -0.002 -0.003) OP(0.00 0.00 0.00 0.00)
3.5%H-10 S(-0.003 0.004 -0.002 -0.003) OP(0.00 0.00 0.00 0.00)

HOMO-46[#156, -18.939 eV]= Fr 1: 10.7%H-4 5.3%H-0 4.8%H-5 2.5%H-1

Fr 2: 59.7%H-45 S(0.000 -0.002 0.000 0.002) OP(0.00 0.00 0.00 0.00)
4.3%H-47 S(-0.005 -0.001 -0.003 0.001) OP(0.00 0.00 0.00 0.00)

3.1%H-9 S(0.001 -0.005 0.000 0.004) OP(0.00 0.00 0.00 0.00)
1.4%H-51 S(0.001 -0.002 0.000 0.001) OP(0.00 0.00 0.00 0.00)

HOMO-47[#155, -18.990 eV]= Fr 2: 94.4%H-42 2.5%H-13
HOMO-48[#154, -18.992 eV]= Fr 2: 93.6%H-43 2.5%H-12
HOMO-49[#153, -19.073 eV]= Fr 1: 12.1%H-0 5.7%H-1 2.3%L+6
Fr 2: 55.7%H-47 S(-0.001 0.001 -0.218) OP(0.00 0.00 0.03)
9.9%H-53 S(-0.001 0.001 0.136) OP(0.00 0.00 -0.01)
2.9%H-11 S(-0.003 0.002 -0.319) OP(0.00 0.00 0.01)
2.4%H-26 S(-0.002 0.001 0.301) OP(0.00 0.00 0.01)
2.1%H-45 S(-0.002 0.002 0.000) OP(0.00 0.00 0.00)

HOMO-50[#152, -19.274 eV]= Fr 1: 4.8%L+0 1.3%H-5
Fr 2: 87.5%H-44 S(0.005 0.002) OP(0.00 0.00)
2.1%H-14 S(0.007 0.003) OP(0.00 0.00)
1.5%H-50 S(0.001 -0.001) OP(0.00 0.00)

HOMO-51[#151, -19.324 eV]= Fr 1: 37.0%H-3 16.7%H-4 14.6%H-0 7.6%H-5 6.9%H-1
Fr 2: 8.7%H-47 S(0.000 -0.005 -0.001 -0.003 0.001) OP(0.00 0.00 0.00 0.00 0.00)
3.4%H-51 S(-0.005 0.001 -0.002 0.000 0.001) OP(0.00 0.00 0.00 0.00 0.00)
1.2%H-11 S(0.000 -0.012 -0.003 -0.008 0.002) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-52[#150, -19.444 eV]= Fr 1: 7.0%H-5 3.8%H-1 3.2%H-4 1.8%H-0
Fr 2: 50.4%H-50 S(-0.001 0.002 0.001 0.001) OP(0.00 0.00 0.00 0.00)
25.1%H-48 S(-0.001 -0.001 0.001 0.000) OP(0.00 0.00 0.00 0.00)
2.3%H-41 S(0.000 0.000 0.000 0.000) OP(0.00 0.00 0.00 0.00)
1.1%H-39 S(-0.002 -0.001 0.001 -0.001) OP(0.00 0.00 0.00 0.00)

HOMO-53[#149, -19.468 eV]= Fr 1: 14.3%H-3 6.4%H-4 2.9%H-5
Fr 2: 66.3%H-49 S(0.001 0.001 0.000) OP(0.00 0.00 0.00)
2.9%H-40 S(-0.003 -0.001 0.000) OP(0.00 0.00 0.00)
1.6%H-73 S(0.001 0.001 0.000) OP(0.00 0.00 0.00)

HOMO-54[#148, -19.488 eV]= Fr 2: 59.6%H-48 26.4%H-50 3.0%H-39 1.7%H-72 1.2%H-41 1.0%H-44
HOMO-55[#147, -19.560 eV]= Fr 1: 35.9%H-3 12.5%H-4 8.8%H-0 5.7%H-5 4.2%H-1
Fr 2: 19.2%H-49 S(0.001 0.001 -0.003 0.000 0.002) OP(0.00 0.00 0.00 0.00 0.00)
6.1%H-51 S(-0.005 0.001 -0.002 0.000 0.001) OP(0.00 0.00 0.00 0.00 0.00)
1.7%H-47 S(0.000 -0.005 -0.001 -0.003 0.001) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-56[#146, -19.626 eV]= Fr 1: 32.3%H-5 17.3%H-1 14.6%H-4 8.2%H-0 3.1%L+0
Fr 2: 11.4%H-50 S(-0.001 0.002 0.001 0.001 0.001) OP(0.00 0.00 0.00 0.00 0.00)
4.2%H-44 S(0.002 -0.003 -0.001 -0.002 0.005) OP(0.00 0.00 0.00 0.00 0.00)
2.3%H-48 S(-0.001 -0.001 0.001 0.000 0.002) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-57[#145, -19.861 eV]= Fr 1: 1.0%H-5
Fr 2: 84.8%H-52 S(-0.003) OP(0.00)
3.0%H-55 S(0.000) OP(0.00)
2.6%H-46 S(-0.002) OP(0.00)
1.3%H-79 S(0.002) OP(0.00)
1.0%H-2 S(-0.010) OP(0.00)

HOMO-58[#144, -19.873 eV]= Fr 1: 7.3%H-3
Fr 2: 76.9%H-51 S(-0.005) OP(0.00)
3.0%H-54 S(0.000) OP(0.00)
2.6%H-45 S(0.002) OP(0.00)
1.3%H-78 S(-0.003) OP(0.00)

1.2%H-1 S(0.016) OP(0.00)
1.0%H-18 S(-0.011) OP(0.00)

HOMO-59[#143, -19.982 eV]= Fr 1: 2.2%L+6
Fr 2: 75.6%H-53 S(0.136) OP(0.02)
6.3%H-47 S(-0.218) OP(0.01)
5.3%H-56 S(0.055) OP(0.00)
3.0%H-80 S(0.164) OP(0.00)
2.2%H-26 S(0.301) OP(0.01)

Chart S2 Output for Am(BTP-H)₃³⁺

HOMO-0[#202, -14.217 eV]= Fr 1: 53.8%H-5 1.4%H-4 1.2%L+1 1.1%L+3
Fr 2: 23.1%H-0 S(0.037 -0.019 0.055 0.106) OP(-0.03 0.00 0.00 0.01)
9.1%H-1 S(-0.020 -0.019 -0.166 0.036) OP(-0.01 0.00 0.01 0.00)
4.1%H-6 S(0.006 -0.003 -0.015 -0.031) OP(0.00 0.00 0.00 0.00)
2.2%H-4 S(-0.008 -0.022 0.070 -0.138) OP(0.00 0.00 0.00 0.00)

HOMO-1[#201, -14.259 eV]= Fr 1: 33.3%H-3 18.8%L+0 2.6%H-2 2.5%L+2
Fr 2: 31.6%H-2 S(0.035 0.022 -0.006 -0.176) OP(-0.02 -0.01 0.00 0.02)
3.4%H-3 S(-0.006 -0.007 0.016 0.024) OP(0.00 0.00 0.00 0.00)
1.9%H-10 S(0.000 -0.002 0.010 0.093) OP(0.00 0.00 0.00 0.00)
1.1%H-24 S(-0.012 -0.008 0.001 0.078) OP(0.00 0.00 0.00 0.00)

HOMO-2[#200, -14.499 eV]= Fr 1: 6.4%H-2 4.9%H-3 3.2%L+0
Fr 2: 75.4%H-5 S(0.001 0.008 -0.015) OP(0.00 0.00 0.00)
5.7%H-22 S(0.001 -0.008 0.015) OP(0.00 0.00 0.00)
1.2%H-2 S(-0.006 0.035 0.022) OP(0.00 0.00 0.00)

HOMO-3[#199, -14.563 eV]= Fr 1: 39.6%H-2 11.2%H-0 1.8%H-3
Fr 2: 24.1%H-3 S(0.016 0.018 -0.006) OP(-0.01 -0.01 0.00)
6.8%H-5 S(0.001 -0.004 0.008) OP(0.00 0.00 0.00)
5.2%H-8 S(0.006 0.007 -0.001) OP(0.00 0.00 0.00)
2.3%H-19 S(0.004 0.010 0.015) OP(0.00 0.00 0.00)
2.1%H-2 S(-0.006 0.005 0.035) OP(0.00 0.00 0.00)

HOMO-4[#198, -14.659 eV]= Fr 1: 9.7%H-1 4.3%H-4
Fr 2: 44.4%H-4 S(0.009 -0.022) OP(0.00 -0.01)
11.5%H-6 S(-0.002 -0.003) OP(0.00 0.00)
6.2%H-7 S(0.005 -0.008) OP(0.00 0.00)
4.7%H-18 S(-0.019 0.000) OP(0.00 0.00)
3.8%H-9 S(-0.011 0.013) OP(0.00 0.00)
3.7%H-21 S(0.006 0.011) OP(0.00 0.00)

HOMO-5[#197, -14.675 eV]= Fr 1: 2.7%H-4
Fr 2: 30.3%H-6 S(-0.003) OP(0.00)
26.5%H-1 S(-0.019) OP(0.00)
15.0%H-7 S(-0.008) OP(0.00)
5.8%H-4 S(-0.022) OP(0.00)
5.4%H-18 S(0.000) OP(0.00)
3.5%H-0 S(-0.019) OP(0.00)
2.2%H-9 S(0.013) OP(0.00)

HOMO-6[#196, -14.704 eV]= Fr 2: 74.4%H-8 17.8%H-3 2.4%H-19 1.5%H-20
HOMO-7[#195, -14.704 eV]= Fr 1: 1.3%H-4
Fr 2: 68.6%H-7 S(-0.008) OP(0.00)

8.2%H-1 S(-0.019) OP(0.00)
8.0%H-6 S(-0.003) OP(0.00)
5.8%H-4 S(-0.022) OP(0.00)
1.6%H-23 S(0.006) OP(0.00)
1.2%H-21 S(0.011) OP(0.00)

HOMO-8[#194, -14.752 eV]= Fr 2: 43.0%H-6 15.4%H-1 15.3%H-0 5.1%H-4 4.0%H-9 2.2%H-21 2.1%H-25 1.6%H-23

HOMO-9[#193, -14.795 eV]= Fr 1: 39.2%H-2

Fr 2: 22.7%H-3 S(0.016) OP(0.01)
11.5%H-8 S(0.006) OP(0.00)
7.8%H-10 S(0.010) OP(0.00)
3.1%H-20 S(-0.006) OP(0.00)
2.7%H-14 S(-0.015) OP(0.00)
2.5%H-19 S(0.004) OP(0.00)
2.2%H-5 S(0.001) OP(0.00)

HOMO-10[#192, -14.894 eV]= Fr 1: 25.1%H-5 4.1%H-4 3.8%H-1

Fr 2: 23.6%H-0 S(0.037 -0.019 -0.013) OP(0.02 0.00 0.00)
10.4%H-11 S(0.026 -0.013 -0.009) OP(-0.01 0.00 0.00)
7.0%H-4 S(-0.008 -0.022 0.009) OP(0.00 0.00 0.00)
6.4%H-26 S(0.015 -0.008 -0.006) OP(0.00 0.00 0.00)
5.6%H-9 S(0.003 0.013 -0.011) OP(0.00 0.00 0.00)

HOMO-11[#191, -14.941 eV]= Fr 1: 21.8%L+0 14.5%H-3

Fr 2: 36.8%H-2 S(0.022 0.035) OP(0.01 0.02)
6.9%H-19 S(0.008 0.015) OP(0.00 0.00)
6.4%H-24 S(-0.008 -0.012) OP(0.00 0.00)
4.8%H-15 S(0.013 0.020) OP(0.00 0.00)
2.7%H-10 S(-0.002 0.000) OP(0.00 0.00)
2.2%H-20 S(0.008 0.011) OP(0.00 0.00)

HOMO-12[#190, -15.014 eV]= Fr 1: 53.9%H-0 15.6%H-3 9.9%L+0

Fr 2: 4.6%H-10 S(0.014 0.000 -0.002) OP(0.00 0.00 0.00)
3.8%H-3 S(0.018 -0.006 -0.007) OP(0.00 0.00 0.00)
2.8%H-5 S(-0.004 0.008 -0.015) OP(0.00 0.00 0.00)
2.5%H-14 S(0.011 -0.003 -0.002) OP(0.00 0.00 0.00)
1.8%H-22 S(0.002 -0.008 0.015) OP(0.00 0.00 0.00)

HOMO-13[#189, -15.125 eV]= Fr 1: 44.6%H-1 21.6%H-4 6.1%H-5

Fr 2: 8.2%H-1 S(-0.030 -0.019 -0.020) OP(0.01 0.00 0.00)
4.8%H-18 S(-0.019 0.000 -0.007) OP(-0.01 0.00 0.00)
2.6%H-23 S(0.012 0.006 0.007) OP(0.00 0.00 0.00)
2.4%H-4 S(0.009 -0.022 -0.008) OP(0.00 0.00 0.00)
2.0%H-9 S(-0.011 0.013 0.003) OP(0.00 0.00 0.00)

HOMO-14[#188, -15.188 eV]= Fr 1: 38.0%L+0 22.6%H-0 20.3%H-3 4.1%H-2

Fr 2: 6.0%H-14 S(-0.002 0.011 -0.003 -0.015) OP(0.00 0.00 0.00 0.00)
1.7%H-17 S(0.013 0.003 -0.006 -0.002) OP(0.00 0.00 0.00 0.00)
1.5%H-22 S(0.015 0.002 -0.008 0.001) OP(0.00 0.00 0.00 0.00)
1.3%H-2 S(0.022 0.005 0.035 -0.006) OP(0.00 0.00 0.00 0.00)

HOMO-15[#187, -15.610 eV]= Fr 2: 90.0%H-12 2.7%H-42 1.9%H-15 1.1%H-10

HOMO-16[#186, -15.619 eV]= Fr 2: 89.8%H-13 2.7%H-43 2.2%H-16 1.4%H-9

HOMO-17[#185, -15.736 eV]= Fr 1: 5.4%H-0 4.7%H-2 1.3%L+0

Fr 2: 76.8%H-14 S(0.011 -0.015 -0.002) OP(0.00 0.01 0.00)

5.7%H-17 S(0.003 -0.002 0.013) OP(0.00 0.00 0.00)
2.9%H-44 S(0.009 -0.012 -0.002) OP(0.00 0.00 0.00)

HOMO-18[#184, -16.141 eV]= Fr 2: 78.1%H-17 9.7%H-22 7.0%H-14
HOMO-19[#183, -16.197 eV]= Fr 2: 70.3%H-15 11.0%H-20 6.1%H-24 3.3%H-10 2.8%H-12 1.0%H-33
HOMO-20[#182, -16.198 eV]= Fr 2: 72.8%H-16 11.1%H-21 6.1%H-23 3.1%H-13 2.0%H-9
HOMO-21[#181, -16.253 eV]= Fr 1: 1.5%L+5
Fr 2: 61.5%H-9 S(0.135) OP(0.02)
12.3%H-21 S(0.061) OP(0.00)
4.8%H-45 S(0.050) OP(0.00)
3.8%H-7 S(-0.019) OP(0.00)
2.8%H-1 S(-0.069) OP(0.00)
2.0%H-13 S(0.024) OP(0.00)
1.6%H-4 S(-0.145) OP(0.00)

HOMO-22[#180, -16.269 eV]= Fr 1: 1.6%H-0 1.5%L+9
Fr 2: 59.2%H-10 S(0.014 -0.338) OP(0.00 0.03)
15.9%H-20 S(-0.003 -0.154) OP(0.00 0.01)
4.6%H-46 S(0.006 -0.160) OP(0.00 0.00)
3.6%H-8 S(0.007 -0.090) OP(0.00 0.00)
2.6%H-2 S(0.005 -0.037) OP(0.00 0.00)
1.9%H-12 S(0.003 -0.019) OP(0.00 0.00)

HOMO-23[#179, -16.455 eV]= Fr 2: 45.6%H-23 30.9%H-21 11.2%H-18 2.7%H-9 2.4%H-4 1.4%H-27
HOMO-24[#178, -16.461 eV]= Fr 2: 48.8%H-24 27.9%H-20 10.8%H-19 3.7%H-10 2.6%H-3 1.4%H-28
HOMO-25[#177, -16.496 eV]= Fr 1: 3.0%H-5
Fr 2: 45.4%H-25 S(0.002) OP(0.00)
33.2%H-11 S(0.026) OP(0.00)
6.3%H-26 S(0.015) OP(0.00)
2.9%H-47 S(0.010) OP(0.00)
2.2%H-0 S(0.037) OP(0.00)
2.1%H-23 S(0.007) OP(0.00)

HOMO-26[#176, -16.574 eV]= Fr 2: 49.6%H-29 42.6%H-22 4.0%H-5 1.8%H-17
HOMO-27[#175, -16.582 eV]= Fr 1: 3.4%H-5 1.0%L+6
Fr 2: 43.1%H-25 S(0.002 -0.039) OP(0.00 0.00)
23.3%H-11 S(0.026 0.328) OP(0.00 0.02)
12.2%H-0 S(0.037 -0.089) OP(0.00 0.00)
6.9%H-32 S(-0.002 0.176) OP(0.00 0.01)
4.5%H-47 S(0.010 0.200) OP(0.00 -0.01)

HOMO-28[#174, -16.661 eV]= Fr 2: 76.2%H-27 9.9%H-23 3.1%H-21 2.4%H-1 1.6%H-30
HOMO-29[#173, -16.663 eV]= Fr 2: 78.4%H-28 10.2%H-24 2.6%H-20 2.5%H-2 1.5%H-31
HOMO-30[#172, -16.888 eV]= Fr 2: 79.9%H-30 9.4%H-18 2.2%H-4 1.9%H-27 1.8%H-9 1.1%H-53
HOMO-31[#171, -16.893 eV]= Fr 2: 80.0%H-31 9.7%H-19 2.2%H-3 1.8%H-10 1.6%H-28 1.1%H-54
HOMO-32[#170, -16.985 eV]= Fr 2: 57.4%H-32 28.9%H-26 6.5%H-11 1.5%H-47 1.2%H-56
HOMO-33[#169, -17.251 eV]= Fr 1: 1.1%L+0
Fr 2: 42.0%H-35 S(0.004) OP(0.00)
24.6%H-29 S(0.007) OP(0.00)
18.0%H-22 S(0.015) OP(0.00)
7.0%H-17 S(0.013) OP(0.00)
2.0%H-5 S(-0.015) OP(0.00)
1.3%H-41 S(-0.001) OP(0.00)

HOMO-34[#168, -17.320 eV]= Fr 2: 77.4%H-34 5.7%H-16 4.5%H-21 2.6%H-23 2.0%H-27 1.1%H-59
1.0%H-4

HOMO-35[#167, -17.325 eV]= Fr 2: 81.7%H-33 4.7%H-15 3.1%H-20 1.7%H-24 1.2%H-58 1.2%H-28
HOMO-36[#166, -17.434 eV]= Fr 2: 46.0%H-36 12.9%H-21 12.8%H-34 5.4%H-27 5.1%H-23 4.1%H-16
3.0%H-4 2.5%H-18

HOMO-37[#165, -17.442 eV]= Fr 2: 47.7%H-37 13.0%H-20 9.4%H-33 4.8%H-28 4.6%H-24 4.2%H-15
3.4%H-19 3.3%H-3

HOMO-38[#164, -17.451 eV]= Fr 1: 1.1%L+8

Fr 2: 53.5%H-35 S(0.139) OP(0.01)

19.5%H-29 S(-0.239) OP(0.01)

15.2%H-22 S(-0.225) OP(0.01)

3.5%H-41 S(-0.107) OP(0.00)

1.8%H-5 S(0.262) OP(0.00)

1.3%H-17 S(-0.062) OP(0.00)

1.0%H-65 S(0.149) OP(0.00)

HOMO-39[#163, -17.525 eV]= Fr 2: 51.2%H-36 11.3%H-21 6.3%H-23 5.6%H-34 4.3%H-16 3.7%H-27
3.4%H-18 3.3%H-40

HOMO-40[#162, -17.529 eV]= Fr 2: 49.7%H-37 11.8%H-20 5.7%H-24 4.8%H-19 4.5%H-15 4.2%H-33
3.9%H-39 3.6%H-3

HOMO-41[#161, -17.603 eV]= Fr 2: 93.1%H-38 2.4%H-26

HOMO-42[#160, -17.717 eV]= Fr 1: 3.2%L+1

Fr 2: 45.0%H-18 S(-0.131) OP(0.02)

9.9%H-30 S(0.062) OP(0.00)

9.3%H-23 S(0.067) OP(0.00)

8.1%H-1 S(-0.166) OP(0.01)

4.4%H-40 S(0.010) OP(0.00)

4.0%H-27 S(-0.052) OP(0.00)

2.3%H-51 S(0.050) OP(0.00)

HOMO-43[#159, -17.723 eV]= Fr 2: 43.0%H-19 10.7%H-24 9.3%H-31 8.8%H-2 4.7%H-28 3.6%H-39
2.9%H-15 2.5%H-52

HOMO-44[#158, -17.915 eV]= Fr 1: 3.9%L+6

Fr 2: 39.0%H-26 S(-0.289) OP(0.05)

26.9%H-32 S(0.176) OP(0.02)

9.1%H-47 S(0.200) OP(-0.02)

5.4%H-0 S(-0.089) OP(0.01)

3.4%H-38 S(-0.010) OP(0.00)

3.2%H-25 S(-0.039) OP(0.00)

2.4%H-55 S(-0.119) OP(-0.01)

HOMO-45[#157, -18.161 eV]= Fr 2: 54.0%H-39 31.8%H-41 4.6%H-48 2.1%H-19 2.0%H-50

HOMO-46[#156, -18.174 eV]= Fr 2: 57.6%H-41 30.0%H-39 3.6%H-50 2.5%H-48 1.2%H-29 1.2%H-19

HOMO-47[#155, -18.175 eV]= Fr 2: 84.0%H-40 7.1%H-49 3.3%H-18

HOMO-48[#154, -18.877 eV]= Fr 2: 87.0%H-45 4.0%H-9 3.1%H-51 1.1%H-78

HOMO-49[#153, -18.888 eV]= Fr 2: 87.2%H-46 4.0%H-10 3.0%H-52 1.1%H-79

HOMO-50[#152, -18.977 eV]= Fr 2: 94.5%H-42 2.5%H-12

HOMO-51[#151, -18.988 eV]= Fr 2: 94.4%H-43 2.5%H-13

HOMO-52[#150, -19.130 eV]= Fr 1: 2.5%L+6

Fr 2: 69.1%H-47 S(0.200) OP(0.03)

13.9%H-55 S(-0.119) OP(-0.01)

5.3%H-11 S(0.328) OP(0.01)

2.8%H-26 S(-0.289) OP(0.01)

1.5%H-68 S(-0.082) OP(0.00)

1.4%H-80 S(-0.148) OP(0.00)

HOMO-53[#149, -19.254 eV]= Fr 1: 1.0%L+21

Fr 2: 91.8%H-44 S(-0.374) OP(0.02)

2.4%H-50 S(0.022) OP(0.00)
2.2%H-14 S(-0.159) OP(0.00)

HOMO-54[#148, -19.467 eV]= Fr 2: 70.6%H-50 17.2%H-48 3.0%H-41 2.2%H-44 1.6%H-74
HOMO-55[#147, -19.478 eV]= Fr 2: 70.2%H-48 17.4%H-50 3.5%H-39 1.9%H-72 1.1%H-3
HOMO-56[#146, -19.481 eV]= Fr 2: 87.3%H-49 4.3%H-40 2.3%H-73 1.3%H-4
HOMO-57[#145, -19.845 eV]= Fr 2: 89.1%H-51 2.5%H-45 1.3%H-78 1.0%H-1
HOMO-58[#144, -19.850 eV]= Fr 2: 89.2%H-52 2.4%H-46 1.3%H-79 1.0%H-2
HOMO-59[#143, -20.000 eV]= Fr 1: 1.8%L+6
Fr 2: 75.1%H-55 S(-0.119) OP(0.01)
7.8%H-47 S(0.200) OP(0.01)
3.9%H-56 S(-0.043) OP(0.00)
3.3%H-80 S(-0.148) OP(0.00)
2.7%H-26 S(-0.289) OP(0.01)
1.1%H-69 S(-0.058) OP(0.00)
1.1%H-11 S(0.328) OP(0.00)

Chart S3 Energy Output Diagram for Cm(BTP-H)₃³⁺

HOMO-0[#203, -14.557 eV]= Fr 2: 88.4%H-5 8.9%H-22
HOMO-1[#202, -14.569 eV]= Fr 1: 1.4%H-3
Fr 2: 53.6%H-0 S(0.029) OP(-0.01)
28.6%H-6 S(-0.005) OP(0.00)
6.0%H-11 S(0.020) OP(0.00)
4.0%H-26 S(-0.012) OP(0.00)
2.3%H-25 S(0.002) OP(0.00)

HOMO-2[#201, -14.616 eV]= Fr 1: 1.2%H-1
Fr 2: 57.9%H-1 S(-0.029) OP(-0.01)
11.0%H-4 S(-0.005) OP(0.00)
10.7%H-9 S(-0.006) OP(0.00)
4.5%H-21 S(-0.007) OP(0.00)
4.1%H-23 S(0.011) OP(0.00)
2.5%H-7 S(-0.003) OP(0.00)
1.5%H-18 S(-0.017) OP(0.00)

HOMO-3[#200, -14.616 eV]= Fr 1: 1.2%L+3
Fr 2: 57.8%H-2 S(-0.135) OP(0.02)
11.1%H-3 S(-0.052) OP(0.00)
10.7%H-10 S(0.095) OP(-0.01)
4.6%H-20 S(0.046) OP(0.00)
4.1%H-24 S(0.062) OP(0.00)
2.5%H-8 S(-0.024) OP(0.00)
1.5%H-19 S(0.035) OP(0.00)

HOMO-4[#199, -14.702 eV]= Fr 2: 43.8%H-7 40.6%H-4 5.8%H-18 3.3%H-21 1.6%H-1 1.0%H-9
HOMO-5[#198, -14.702 eV]= Fr 2: 43.9%H-8 40.6%H-3 5.8%H-19 3.3%H-20 1.7%H-2 1.0%H-10
HOMO-6[#197, -14.734 eV]= Fr 2: 38.4%H-7 22.2%H-4 16.4%H-1 10.2%H-9 5.7%H-18 2.4%H-23
1.2%H-21
HOMO-7[#196, -14.734 eV]= Fr 2: 38.7%H-8 22.1%H-3 16.4%H-2 10.1%H-10 5.7%H-19 2.4%H-24
1.1%H-20
HOMO-8[#195, -14.752 eV]= Fr 1: 1.2%L+0
Fr 2: 68.4%H-6 S(0.037) OP(0.00)
21.4%H-0 S(0.141) OP(0.01)
3.2%H-25 S(0.051) OP(0.00)

1.6%H-26 S(-0.002) OP(0.00)

HOMO-9[#194, -15.583 eV]= Fr 2: 88.7%H-13 4.1%H-16 2.6%H-43 1.2%H-9
HOMO-10[#193, -15.583 eV]= Fr 2: 88.7%H-12 4.1%H-15 2.6%H-42 1.2%H-10
HOMO-11[#192, -15.594 eV]= Fr 1: 2.2%H-5 1.2%H-6
Fr 2: 88.4%H-14 S(-0.012 0.011) OP(0.00 0.00)
4.0%H-17 S(0.004 0.008) OP(0.00 0.00)
2.1%H-44 S(-0.009 0.009) OP(0.00 0.00)

HOMO-12[#191, -15.957 eV]= Fr 1: 20.5%H-3 6.5%H-6 4.3%H-4 4.2%H-5 1.8%L+0
Fr 2: 43.9%H-11 S(0.020 0.011 0.009 0.009 0.110) OP(-0.01 0.00 0.00 0.00 0.01)
10.9%H-26 S(-0.012 -0.007 -0.005 -0.005 -0.002) OP(0.00 0.00 0.00 0.00 0.00)
2.7%H-0 S(0.029 0.016 0.013 0.013 0.141) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-13[#190, -16.099 eV]= Fr 1: 14.6%H-2 3.1%H-1 2.1%H-0
Fr 2: 30.1%H-16 S(-0.010 -0.015 -0.006) OP(0.00 0.00 0.00)
25.6%H-9 S(0.011 -0.006 0.002) OP(0.00 0.00 0.00)
5.5%H-13 S(0.002 -0.003 0.000) OP(0.00 0.00 0.00)
4.1%H-7 S(0.009 -0.003 0.002) OP(0.00 0.00 0.00)
2.4%H-18 S(0.001 -0.017 -0.003) OP(0.00 0.00 0.00)

HOMO-14[#189, -16.100 eV]= Fr 1: 15.4%H-4 2.4%H-3
Fr 2: 30.4%H-15 S(-0.012 0.006) OP(-0.01 0.00)
25.5%H-10 S(0.009 -0.003) OP(0.00 0.00)
5.5%H-12 S(0.001 0.000) OP(0.00 0.00)
4.1%H-8 S(-0.008 0.003) OP(0.00 0.00)
2.4%H-19 S(0.002 -0.002) OP(0.00 0.00)
1.5%H-20 S(0.011 -0.004) OP(0.00 0.00)

HOMO-15[#188, -16.106 eV]= Fr 1: 2.5%H-6
Fr 2: 85.8%H-17 S(0.008) OP(0.00)
4.2%H-14 S(0.011) OP(0.00)
4.1%H-22 S(-0.008) OP(0.00)

HOMO-16[#187, -16.154 eV]= Fr 1: 6.2%H-0 1.4%H-4
Fr 2: 44.4%H-15 S(0.012 -0.012) OP(0.00 0.00)
29.1%H-10 S(0.008 0.009) OP(0.00 0.00)
5.0%H-8 S(-0.005 -0.008) OP(0.00 0.00)
2.2%H-46 S(0.004 0.004) OP(0.00 0.00)
1.6%H-20 S(-0.005 0.011) OP(0.00 0.00)
1.3%H-2 S(0.024 -0.019) OP(0.00 0.00)

HOMO-17[#186, -16.154 eV]= Fr 1: 5.2%H-1 2.8%H-2
Fr 2: 44.7%H-16 S(-0.015 -0.010) OP(0.00 0.00)
28.9%H-9 S(-0.006 0.011) OP(0.00 0.00)
5.0%H-7 S(-0.003 0.009) OP(0.00 0.00)
2.2%H-45 S(-0.003 0.005) OP(0.00 0.00)
1.6%H-21 S(-0.007 -0.010) OP(0.00 0.00)
1.3%H-1 S(-0.029 -0.015) OP(0.00 0.00)

HOMO-18[#185, -16.377 eV]= Fr 1: 20.2%H-1 3.3%H-2 1.8%H-0
Fr 2: 49.6%H-21 S(-0.007 -0.010 -0.004) OP(0.00 0.00 0.00)
10.5%H-18 S(-0.017 0.001 -0.003) OP(-0.01 0.00 0.00)
4.9%H-27 S(-0.003 -0.005 -0.002) OP(0.00 0.00 0.00)
2.5%H-9 S(-0.006 0.011 0.002) OP(0.00 0.00 0.00)
1.6%H-16 S(-0.015 -0.010 -0.006) OP(0.00 0.00 0.00)

HOMO-19[#184, -16.377 eV]= Fr 1: 14.3%H-0 6.5%H-4 2.9%H-2 1.7%H-3
Fr 2: 49.6%H-20 S(-0.005 0.011 0.004 -0.004) OP(0.00 0.00 0.00 0.00)
10.5%H-19 S(-0.016 0.002 0.005 -0.002) OP(0.00 0.00 0.00 0.00)
4.9%H-28 S(0.001 -0.005 -0.002 0.002) OP(0.00 0.00 0.00 0.00)
2.4%H-10 S(0.008 0.009 -0.001 -0.003) OP(0.00 0.00 0.00 0.00)

HOMO-20[#183, -16.460 eV]= Fr 1: 17.5%H-0 1.6%H-2
Fr 2: 61.5%H-24 S(-0.009 0.004) OP(-0.01 0.00)
7.0%H-20 S(-0.005 0.004) OP(0.00 0.00)
2.1%H-28 S(0.001 -0.002) OP(0.00 0.00)
2.0%H-31 S(0.005 0.000) OP(0.00 0.00)
1.1%H-15 S(0.012 -0.006) OP(0.00 0.00)
1.0%H-22 S(-0.001 0.000) OP(0.00 0.00)

HOMO-21[#182, -16.461 eV]= Fr 1: 19.4%H-1
Fr 2: 63.0%H-23 S(0.011) OP(-0.01)
7.2%H-21 S(-0.007) OP(0.00)
2.2%H-27 S(-0.003) OP(0.00)
2.0%H-30 S(-0.004) OP(0.00)
1.1%H-16 S(-0.015) OP(0.00)

HOMO-22[#181, -16.462 eV]= Fr 1: 12.1%H-6 11.0%H-5 9.5%H-3
Fr 2: 44.9%H-22 S(-0.008 -0.007 0.007) OP(0.00 0.00 0.00)
15.6%H-29 S(0.005 0.002 -0.004) OP(0.00 0.00 0.00)
2.5%H-5 S(0.010 0.006 -0.008) OP(0.00 0.00 0.00)
1.4%H-24 S(0.000 -0.001 -0.003) OP(0.00 0.00 0.00)

HOMO-23[#180, -16.532 eV]= Fr 2: 88.0%H-25 3.6%H-32 3.2%H-26 1.3%H-0 1.2%H-11
HOMO-24[#179, -16.697 eV]= Fr 1: 48.7%H-5 10.3%H-3 1.1%H-4
Fr 2: 32.6%H-29 S(0.002 -0.004 -0.001) OP(0.00 0.00 0.00)
4.0%H-22 S(-0.007 0.007 0.001) OP(0.00 0.00 0.00)

HOMO-25[#178, -16.714 eV]= Fr 1: 29.2%H-2 2.9%H-0 1.3%H-4
Fr 2: 52.3%H-27 S(-0.005 -0.002 0.001) OP(0.00 0.00 0.00)
3.2%H-9 S(0.011 0.002 -0.001) OP(0.00 0.00 0.00)
1.7%H-18 S(0.001 -0.003 0.003) OP(0.00 0.00 0.00)
1.2%H-45 S(0.005 0.001 0.000) OP(0.00 0.00 0.00)
1.2%H-1 S(-0.015 -0.009 0.008) OP(0.00 0.00 0.00)

HOMO-26[#177, -16.714 eV]= Fr 1: 23.3%H-4 10.2%H-6 2.6%H-5 1.3%H-3
Fr 2: 46.1%H-28 S(-0.005 0.001 0.000 0.002) OP(0.00 0.00 0.00 0.00)
2.9%H-10 S(0.009 -0.002 0.000 -0.003) OP(0.00 0.00 0.00 0.00)
1.5%H-19 S(0.002 0.001 -0.001 -0.002) OP(0.00 0.00 0.00 0.00)
1.1%H-29 S(-0.001 0.005 0.002 -0.004) OP(0.00 0.00 0.00 0.00)

HOMO-27[#176, -16.716 eV]= Fr 1: 45.1%H-6 20.2%H-5 6.3%H-3 3.0%H-4
Fr 2: 9.3%H-29 S(0.005 0.002 -0.004 -0.001) OP(0.00 0.00 0.00 0.00)
7.7%H-28 S(0.001 0.000 0.002 -0.005) OP(0.00 0.00 0.00 0.00)
3.2%H-14 S(0.011 -0.012 -0.001 0.001) OP(0.00 0.00 0.00 0.00)
1.6%H-35 S(0.005 -0.001 -0.002 0.000) OP(0.00 0.00 0.00 0.00)

HOMO-28[#175, -16.731 eV]= Fr 1: 35.4%H-0 15.5%H-4 1.3%H-3
Fr 2: 24.3%H-28 S(0.001 -0.005 0.002) OP(0.00 0.00 0.00)
6.2%H-24 S(-0.009 0.006 -0.003) OP(0.00 0.00 0.00)
5.3%H-19 S(-0.016 0.002 -0.002) OP(0.00 0.00 0.00)

4.2%H-10 S(0.008 0.009 -0.003) OP(0.00 0.00 0.00)
1.5%H-8 S(-0.005 -0.008 0.003) OP(0.00 0.00 0.00)

HOMO-29[#174, -16.731 eV]= Fr 1: 26.3%H-1 25.4%H-2
Fr 2: 25.8%H-27 S(-0.003 -0.005) OP(0.00 0.00)
6.1%H-23 S(0.011 0.004) OP(0.00 0.00)
5.3%H-18 S(-0.017 0.001) OP(0.00 0.00)
4.1%H-9 S(-0.006 0.011) OP(0.00 0.00)
1.4%H-7 S(-0.003 0.009) OP(0.00 0.00)
1.2%H-21 S(-0.007 -0.010) OP(0.00 0.00)

HOMO-30[#173, -16.888 eV]= Fr 1: 1.9%H-1
Fr 2: 78.5%H-30 S(-0.004) OP(0.00)
8.9%H-18 S(-0.017) OP(0.00)
2.1%H-4 S(-0.005) OP(0.00)
1.5%H-9 S(-0.006) OP(0.00)
1.1%H-53 S(-0.001) OP(0.00)

HOMO-31[#172, -16.888 eV]= Fr 1: 1.3%H-0
Fr 2: 78.4%H-31 S(0.005) OP(0.00)
8.9%H-19 S(-0.016) OP(0.00)
2.1%H-3 S(-0.009) OP(0.00)
1.5%H-10 S(0.008) OP(0.00)
1.1%H-54 S(0.001) OP(0.00)

HOMO-32[#171, -16.974 eV]= Fr 1: 1.1%L+5 1.0%H-3
Fr 2: 49.0%H-32 S(0.170 -0.001) OP(-0.02 0.00)
26.8%H-26 S(0.282 -0.012) OP(0.02 0.00)
12.8%H-11 S(0.333 0.020) OP(0.02 0.00)
3.3%H-47 S(0.197 0.007) OP(-0.01 0.00)
1.0%H-56 S(0.048 0.003) OP(0.00 0.00)

HOMO-33[#170, -17.040 eV]= Fr 1: 29.8%H-3 9.4%H-6 6.3%H-4 6.0%H-5
Fr 2: 16.5%H-11 S(0.020 0.011 0.009 0.009) OP(0.01 0.00 0.00 0.00)
14.8%H-32 S(-0.001 -0.001 -0.001 -0.001) OP(0.00 0.00 0.00 0.00)
7.0%H-47 S(0.007 0.004 0.003 0.003) OP(0.00 0.00 0.00 0.00)
6.0%H-0 S(0.029 0.016 0.013 0.013) OP(0.01 0.00 0.00 0.00)

HOMO-34[#169, -17.302 eV]= Fr 1: 6.0%H-6 3.0%H-3
Fr 2: 57.2%H-35 S(0.005 -0.002) OP(0.00 0.00)
11.2%H-29 S(0.005 -0.004) OP(0.00 0.00)
11.1%H-22 S(-0.008 0.007) OP(0.00 0.00)
4.9%H-17 S(0.008 -0.006) OP(0.00 0.00)
1.4%H-5 S(0.010 -0.008) OP(0.00 0.00)
1.2%H-57 S(-0.005 0.003) OP(0.00 0.00)

HOMO-35[#168, -17.348 eV]= Fr 1: 1.3%H-4
Fr 2: 91.8%H-33 S(0.003) OP(0.00)
1.5%H-58 S(-0.004) OP(0.00)
1.2%H-15 S(-0.012) OP(0.00)

HOMO-36[#167, -17.348 eV]= Fr 1: 1.4%H-1
Fr 2: 91.8%H-34 S(0.004) OP(0.00)
1.5%H-59 S(-0.005) OP(0.00)
1.2%H-16 S(-0.015) OP(0.00)

HOMO-37[#166, -17.468 eV]= Fr 1: 2.3%H-2
Fr 2: 85.3%H-36 S(-0.002) OP(0.00)
3.1%H-21 S(-0.010) OP(0.00)
1.5%H-30 S(0.007) OP(0.00)
1.2%H-18 S(0.001) OP(0.00)
1.1%H-16 S(-0.010) OP(0.00)

HOMO-38[#165, -17.468 eV]= Fr 1: 2.3%H-4
Fr 2: 85.4%H-37 S(0.002) OP(0.00)
3.1%H-20 S(0.011) OP(0.00)
1.5%H-31 S(0.006) OP(0.00)
1.2%H-19 S(0.002) OP(0.00)
1.1%H-15 S(-0.012) OP(0.00)

HOMO-39[#164, -17.485 eV]= Fr 1: 1.7%H-5
Fr 2: 36.1%H-35 S(-0.001) OP(0.00)
23.8%H-29 S(0.002) OP(0.00)
21.9%H-22 S(-0.007) OP(0.00)
4.7%H-41 S(-0.001) OP(0.00)
2.9%H-5 S(0.006) OP(0.00)
2.1%H-17 S(0.004) OP(0.00)
1.3%H-65 S(-0.002) OP(0.00)

HOMO-40[#163, -17.558 eV]= Fr 1: 5.7%H-2
Fr 2: 20.0%H-21 S(-0.010) OP(0.00)
14.7%H-18 S(0.001) OP(0.00)
11.9%H-36 S(-0.002) OP(0.00)
8.4%H-40 S(0.006) OP(0.00)
7.5%H-4 S(0.020) OP(0.00)
6.5%H-16 S(-0.010) OP(0.00)
4.7%H-23 S(0.004) OP(0.00)

HOMO-41[#162, -17.558 eV]= Fr 1: 5.7%H-4
Fr 2: 20.0%H-20 S(0.011) OP(0.00)
14.7%H-19 S(0.002) OP(0.00)
11.8%H-37 S(0.002) OP(0.00)
8.4%H-39 S(0.005) OP(0.00)
7.5%H-3 S(-0.018) OP(0.00)
6.4%H-15 S(-0.012) OP(0.00)
4.6%H-24 S(0.006) OP(0.00)

HOMO-42[#161, -17.599 eV]= Fr 2: 94.0%H-38 1.9%H-26

HOMO-43[#160, -17.803 eV]= Fr 1: 12.1%H-1
Fr 2: 31.3%H-18 S(-0.017) OP(0.01)
12.8%H-23 S(0.011) OP(0.00)
9.9%H-1 S(-0.029) OP(0.01)
5.8%H-30 S(-0.004) OP(0.00)
4.9%H-27 S(-0.003) OP(0.00)
4.5%H-16 S(-0.015) OP(0.00)
3.1%H-51 S(-0.008) OP(0.00)

HOMO-44[#159, -17.803 eV]= Fr 1: 9.1%H-0
Fr 2: 31.2%H-19 S(-0.016) OP(0.01)
12.8%H-24 S(-0.009) OP(0.00)
9.9%H-2 S(0.024) OP(0.00)
5.8%H-31 S(0.005) OP(0.00)

4.9%H-28 S(0.001) OP(0.00)
4.5%H-15 S(0.012) OP(0.00)
3.1%H-52 S(0.007) OP(0.00)

HOMO-45[#158, -17.957 eV]= Fr 1: 3.8%L+5 3.3%H-3
Fr 2: 38.2%H-26 S(0.282 -0.012) OP(0.04 0.00)
23.7%H-32 S(0.170 -0.001) OP(0.02 0.00)
7.6%H-47 S(0.197 0.007) OP(-0.01 0.00)
6.3%H-0 S(-0.088 0.029) OP(0.01 0.00)
3.9%H-25 S(-0.050 0.002) OP(0.00 0.00)
2.6%H-55 S(0.113 -0.005) OP(-0.01 0.00)

Chart S4 Output file for $\text{Eu}(\text{TPAm})_3^{3+}$

HOMO-0[#187, -14.450 eV]= Fr 2: 64.5%H-2 8.4%H-11 8.3%H-23 6.6%H-5 2.0%H-1 1.4%H-8 1.2%H-3
1.2%H-4
HOMO-1[#186, -14.507 eV]= Fr 2: 35.5%H-0 26.5%H-3 11.6%H-4 4.6%H-21 3.7%H-18 3.6%H-10
3.5%H-19 2.4%H-22
HOMO-2[#185, -14.554 eV]= Fr 2: 31.9%H-1 25.3%H-4 8.8%H-3 4.8%H-22 4.2%H-19 3.7%H-9 3.4%H-
18 3.1%H-2
HOMO-3[#184, -14.743 eV]= Fr 2: 54.8%H-14 19.7%H-13 9.3%H-8 6.7%H-11 3.0%H-7 1.3%H-6
HOMO-4[#183, -14.764 eV]= Fr 2: 56.7%H-13 18.3%H-14 10.7%H-7 3.9%H-8 2.5%H-11 1.5%H-6
1.1%H-0
HOMO-5[#182, -14.772 eV]= Fr 2: 76.8%H-12 12.9%H-6 3.6%H-7 1.1%H-1 1.1%H-10 1.0%H-15
HOMO-6[#181, -15.129 eV]= Fr 2: 59.8%H-7 17.9%H-13 5.7%H-9 3.6%H-12 2.4%H-33 1.1%H-3
HOMO-7[#180, -15.148 eV]= Fr 2: 60.9%H-6 16.7%H-12 5.4%H-10 2.6%H-13 2.5%H-32 1.7%H-4
1.1%H-1
HOMO-8[#179, -15.265 eV]= Fr 2: 64.0%H-8 19.3%H-14 3.9%H-17 2.9%H-34 1.4%H-29 1.3%H-11
1.2%H-20 1.0%L+8
HOMO-9[#178, -15.605 eV]= Fr 2: 75.3%H-5 8.0%H-17 4.7%H-2 1.8%H-11 1.5%H-8
HOMO-10[#177, -15.693 eV]= Fr 2: 52.0%H-16 26.8%H-15 2.9%H-1 2.9%H-3 2.9%H-9 2.2%H-6
1.4%H-40 1.1%H-7
HOMO-11[#176, -15.709 eV]= Fr 2: 47.8%H-15 28.6%H-16 4.8%H-4 3.0%H-10 2.6%H-0 2.3%H-1
2.0%H-7 1.7%H-6
HOMO-12[#175, -15.758 eV]= Fr 2: 25.4%H-1 18.4%H-3 12.3%H-9 10.6%H-0 6.0%H-4 5.2%H-15
5.0%H-10 4.7%H-16
HOMO-13[#174, -15.787 eV]= Fr 2: 56.0%H-17 6.3%H-15 6.2%H-0 5.7%H-4 3.6%H-1 3.2%H-5 3.2%H-
10 1.8%H-20
HOMO-14[#173, -15.807 eV]= Fr 2: 21.1%H-17 17.3%H-0 11.6%H-10 10.7%H-4 6.5%H-5 5.4%H-1
4.9%H-16 4.5%H-15
HOMO-15[#172, -16.176 eV]= Fr 2: 44.1%H-11 9.9%H-2 8.4%H-10 4.8%H-9 3.4%H-3 3.0%H-8
3.0%H-20 2.6%H-14
HOMO-16[#171, -16.226 eV]= Fr 1: 2.8%L+1
Fr 2: 43.2%H-9 S(0.088) OP(0.01)
12.9%H-4 S(-0.117) OP(0.01)
6.8%H-10 S(0.046) OP(0.00)
6.0%H-21 S(-0.002) OP(0.00)
4.9%H-7 S(-0.044) OP(0.00)
3.6%H-1 S(0.007) OP(0.00)
2.2%H-0 S(-0.017) OP(0.00)
HOMO-17[#170, -16.256 eV]= Fr 2: 26.6%H-10 12.8%H-22 11.6%H-11 10.1%H-3 4.2%H-0 3.1%H-6
2.9%H-1 2.8%H-2
HOMO-18[#169, -16.298 eV]= Fr 2: 25.7%H-21 15.9%H-19 15.5%H-22 12.4%H-20 11.1%H-18 8.4%H-
23 3.8%H-9
HOMO-19[#168, -16.314 eV]= Fr 2: 27.0%H-21 21.8%H-20 20.5%H-18 13.6%H-23 6.2%H-22 2.7%H-
10 1.1%H-19

HOMO-20[#167, -16.324 eV]= Fr 2: 26.0%H-22 17.4%H-20 16.4%H-23 13.6%H-19 7.3%H-10 2.4%H-18 2.0%H-3 1.8%H-2
HOMO-21[#166, -16.553 eV]= Fr 2: 45.0%H-26 20.3%H-20 16.1%H-23 7.2%H-24 2.1%H-11 2.1%H-8 1.3%H-18 1.0%H-17
HOMO-22[#165, -16.575 eV]= Fr 2: 42.6%H-24 21.8%H-25 11.3%H-19 4.2%H-26 3.6%H-3 3.3%H-21 3.1%H-18 2.2%H-20
HOMO-23[#164, -16.594 eV]= Fr 2: 50.0%H-25 21.2%H-24 9.6%H-18 4.7%H-19 3.5%H-4 2.3%H-22 1.2%H-26
HOMO-24[#163, -17.202 eV]= Fr 2: 40.9%H-27 39.4%H-28 3.5%H-19 2.4%H-18 2.0%H-24 1.4%H-22 1.4%H-25 1.2%H-21
HOMO-25[#162, -17.225 eV]= Fr 2: 45.1%H-28 36.8%H-27 4.2%H-29 2.9%H-18 1.5%H-24 1.3%H-21
HOMO-26[#161, -17.268 eV]= Fr 2: 61.6%H-29 11.6%H-26 6.6%H-23 3.9%H-20 2.3%H-34 1.6%H-11 1.4%H-18 1.3%H-27
HOMO-27[#160, -17.323 eV]= Fr 2: 14.4%H-18 14.4%H-21 13.7%H-19 13.4%H-24 13.3%H-27 5.3%H-22 3.0%H-3 2.9%H-0
HOMO-28[#159, -17.362 eV]= Fr 2: 17.2%H-19 15.7%H-22 13.8%H-18 13.7%H-25 6.6%H-28 5.1%H-21 3.6%H-1 3.3%H-4
HOMO-29[#158, -17.478 eV]= Fr 1: 2.0%L+6 1.3%L+9
Fr 2: 28.0%H-26 S(-0.151 0.186) OP(0.02 0.01)
23.1%H-29 S(0.035 -0.052) OP(0.00 0.00)
21.8%H-23 S(0.136 -0.144) OP(0.01 0.01)
7.4%H-20 S(0.112 -0.066) OP(0.01 0.00)
2.9%H-2 S(0.045 -0.111) OP(0.00 0.00)
2.9%H-38 S(-0.116 -0.001) OP(0.00 0.00)

HOMO-30[#157, -17.991 eV]= Fr 2: 38.6%H-37 20.0%H-43 8.2%H-33 5.1%H-46 5.0%H-30 4.0%H-39 3.3%H-36 3.3%H-31
HOMO-31[#156, -17.996 eV]= Fr 2: 35.2%H-36 17.6%H-42 9.8%H-38 5.1%H-32 4.8%H-45 4.1%H-30 3.5%H-44 3.5%H-37
HOMO-32[#155, -18.019 eV]= Fr 2: 49.1%H-38 16.4%H-44 7.0%H-36 5.6%H-47 4.4%H-41 3.5%H-42 3.3%H-50 1.6%H-37
HOMO-33[#154, -18.057 eV]= Fr 2: 48.5%H-33 11.5%H-37 9.8%H-30 6.1%H-59 3.6%H-43 3.6%H-7 2.0%H-31 1.8%H-46
HOMO-34[#153, -18.082 eV]= Fr 1: 2.0%H-2
Fr 2: 52.1%H-32 S(-0.004) OP(0.00)
11.5%H-31 S(0.005) OP(0.00)
8.9%H-36 S(0.000) OP(0.00)
6.8%H-58 S(-0.001) OP(0.00)
4.1%H-6 S(0.005) OP(0.00)
2.2%H-42 S(0.000) OP(0.00)
1.9%H-30 S(0.007) OP(0.00)

HOMO-35[#152, -18.182 eV]= Fr 1: 9.2%H-0 8.6%H-5 3.3%H-3 3.2%L+0
Fr 2: 50.3%H-34 S(-0.005 -0.004 0.000 0.004) OP(0.00 0.00 0.00 0.00)
5.4%H-57 S(0.000 0.001 0.000 0.000) OP(0.00 0.00 0.00 0.00)
3.5%H-35 S(-0.002 -0.001 0.000 0.003) OP(0.00 0.00 0.00 0.00)
3.0%H-56 S(0.001 0.001 0.000 -0.001) OP(0.00 0.00 0.00 0.00)

HOMO-36[#151, -18.309 eV]= Fr 2: 15.5%H-37 9.0%H-35 8.4%H-43 7.8%H-46 6.6%H-31 5.0%H-38 4.8%H-34 4.8%H-42
HOMO-37[#150, -18.323 eV]= Fr 1: 14.7%H-2
Fr 2: 18.3%H-36 S(0.000) OP(0.00)
7.7%H-45 S(-0.003) OP(0.00)
7.6%H-43 S(0.001) OP(0.00)
7.6%H-30 S(0.007) OP(0.00)
6.9%H-42 S(0.000) OP(0.00)

6.6%H-37 S(0.000) OP(0.00)
5.6%H-46 S(-0.002) OP(0.00)

HOMO-38[#149, -18.348 eV]= Fr 2: 20.3%H-38 18.3%H-44 17.1%H-35 14.3%H-47 4.2%H-41 4.1%H-37 3.1%H-42 2.8%H-36

HOMO-39[#148, -18.405 eV]= Fr 1: 42.8%H-5 21.1%H-3 6.8%H-2
Fr 2: 4.6%H-34 S(-0.004 0.000 -0.003) OP(0.00 0.00 0.00)
3.6%H-37 S(-0.001 0.000 0.000) OP(0.00 0.00 0.00)
3.1%H-42 S(0.000 -0.002 0.000) OP(0.00 0.00 0.00)
2.1%H-46 S(-0.001 -0.002 -0.002) OP(0.00 0.00 0.00)
2.0%H-43 S(0.001 -0.001 0.001) OP(0.00 0.00 0.00)

HOMO-40[#147, -18.421 eV]= Fr 1: 42.8%H-2 9.2%H-3 7.7%H-1 2.7%L+0
Fr 2: 8.5%H-36 S(0.000 -0.002 0.000 0.000) OP(0.00 0.00 0.00 0.00)
6.5%H-42 S(0.000 -0.002 -0.001 0.000) OP(0.00 0.00 0.00 0.00)
5.8%H-45 S(-0.003 0.000 0.003 -0.002) OP(0.00 0.00 0.00 0.00)
3.4%H-43 S(0.001 -0.001 -0.001 0.001) OP(0.00 0.00 0.00 0.00)

HOMO-41[#146, -18.452 eV]= Fr 1: 29.5%H-0 22.0%L+0 8.6%H-1 7.5%H-3 3.5%H-4 3.5%H-2
Fr 2: 3.6%H-34 S(-0.005 0.004 -0.002 0.000 -0.002 -0.003) OP(0.00 0.00 0.00 0.00 0.00 0.00)
2.7%H-33 S(-0.001 0.003 -0.005 0.003 0.004 0.003) OP(0.00 0.00 0.00 0.00 0.00 0.00)

HOMO-42[#145, -18.849 eV]= Fr 1: 12.9%H-1 10.1%H-4 7.8%L+0 4.4%H-3
Fr 2: 17.4%H-43 S(-0.001 0.001 0.001 -0.001) OP(0.00 0.00 0.00 0.00)
13.2%H-46 S(-0.003 0.000 -0.002 -0.002) OP(0.00 0.00 0.00 0.00)
6.6%H-31 S(0.006 0.003 0.005 0.010) OP(0.00 0.00 0.00 0.00)
4.2%H-39 S(0.000 0.002 -0.002 -0.001) OP(0.00 0.00 0.00 0.00)

HOMO-43[#144, -18.876 eV]= Fr 1: 9.7%H-0 8.9%H-5 7.9%H-2 6.7%H-3 6.5%H-1
Fr 2: 15.2%H-42 S(0.001 0.000 0.000 -0.002 -0.001) OP(0.00 0.00 0.00 0.00 0.00)
13.3%H-45 S(0.000 0.000 -0.003 0.000 0.003) OP(0.00 0.00 0.00 0.00 0.00)
4.9%H-30 S(-0.002 0.003 0.007 -0.006 -0.009) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-44[#143, -18.987 eV]= Fr 1: 22.8%H-4 12.8%H-1 9.2%H-0 5.1%L+0 3.3%H-2
Fr 2: 20.9%H-40 S(0.001 -0.002 0.001 0.000 -0.001) OP(0.00 0.00 0.00 0.00 0.00)
3.9%H-46 S(0.000 -0.003 0.002 -0.002 -0.002) OP(0.00 0.00 0.00 0.00 0.00)
3.4%H-39 S(0.002 0.000 -0.002 -0.002 -0.002) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-45[#142, -19.003 eV]= Fr 1: 19.9%H-1 3.6%H-0
Fr 2: 15.4%H-45 S(0.003 0.000) OP(0.00 0.00)
11.4%H-40 S(-0.002 0.001) OP(0.00 0.00)
7.3%H-46 S(-0.003 0.002) OP(0.00 0.00)
7.0%H-42 S(-0.001 0.001) OP(0.00 0.00)
6.8%H-39 S(0.000 -0.002) OP(0.00 0.00)
5.6%H-47 S(-0.001 -0.002) OP(0.00 0.00)

HOMO-46[#141, -19.020 eV]= Fr 1: 10.9%H-3 10.0%H-4 8.3%H-5
Fr 2: 27.1%H-39 S(-0.001 0.002 0.001) OP(0.00 0.00 0.00)
8.7%H-43 S(-0.001 0.001 0.001) OP(0.00 0.00 0.00)
6.4%H-46 S(-0.002 0.000 -0.001) OP(0.00 0.00 0.00)
6.0%H-41 S(0.000 -0.001 0.004) OP(0.00 0.00 0.00)
3.7%H-58 S(0.000 0.001 0.001) OP(0.00 0.00 0.00)

HOMO-47[#140, -19.041 eV]= Fr 1: 5.3%H-4
Fr 2: 17.0%H-40 S(0.001) OP(0.00)
14.6%H-42 S(0.000) OP(0.00)

9.3%H-39 S(0.002) OP(0.00)
7.6%H-44 S(0.000) OP(0.00)
5.6%H-41 S(-0.001) OP(0.00)
4.8%H-58 S(0.001) OP(0.00)
4.6%H-46 S(0.000) OP(0.00)

HOMO-48[#139, -19.054 eV]= Fr 1: 13.8%H-4 6.2%H-3
Fr 2: 15.5%H-39 S(0.002 -0.001) OP(0.00 0.00)
13.3%H-40 S(0.001 0.003) OP(0.00 0.00)
10.5%H-43 S(0.001 -0.001) OP(0.00 0.00)
6.7%H-45 S(-0.002 0.000) OP(0.00 0.00)
5.9%H-59 S(0.000 -0.002) OP(0.00 0.00)
4.3%H-41 S(-0.001 0.000) OP(0.00 0.00)

HOMO-49[#138, -19.113 eV]= Fr 1: 8.1%L+0 5.5%H-3 4.9%H-5
Fr 2: 22.7%H-44 S(-0.004 0.000 -0.001) OP(0.00 0.00 0.00)
15.2%H-47 S(0.005 0.000 -0.001) OP(0.00 0.00 0.00)
12.0%H-41 S(0.002 0.000 0.004) OP(0.00 0.00 0.00)
6.9%H-46 S(-0.002 -0.002 -0.001) OP(0.00 0.00 0.00)
3.1%H-56 S(-0.001 0.000 0.001) OP(0.00 0.00 0.00)

HOMO-50[#137, -19.163 eV]= Fr 1: 6.6%H-4 4.7%H-5
Fr 2: 39.4%H-41 S(-0.001 0.004) OP(0.00 0.00)
14.5%H-47 S(0.000 -0.001) OP(0.00 0.00)
7.9%H-57 S(-0.001 0.001) OP(0.00 0.00)
5.7%H-35 S(-0.001 -0.001) OP(0.00 0.00)
5.6%H-50 S(-0.001 0.002) OP(0.00 0.00)
3.2%H-44 S(0.000 -0.001) OP(0.00 0.00)

HOMO-51[#136, -19.299 eV]= Fr 2: 43.5%H-50 16.2%H-47 15.4%H-35 3.8%H-49 3.7%H-34 1.7%H-41
1.5%H-71 1.5%H-46

HOMO-52[#135, -19.335 eV]= Fr 2: 25.6%H-49 19.4%H-48 10.9%H-46 8.4%H-31 5.7%H-45 3.5%H-30
3.2%H-50 2.9%H-32

HOMO-53[#134, -19.348 eV]= Fr 1: 2.7%H-4
Fr 2: 28.9%H-48 S(0.002) OP(0.00)
19.1%H-49 S(0.001) OP(0.00)
12.5%H-45 S(-0.002) OP(0.00)
9.0%H-30 S(0.007) OP(0.00)
4.8%H-46 S(0.000) OP(0.00)
3.0%H-31 S(0.003) OP(0.00)
2.8%H-33 S(0.004) OP(0.00)

HOMO-54[#133, -19.738 eV]= Fr 2: 48.1%H-55 9.8%H-54 8.6%H-31 8.3%H-49 3.4%H-58 2.7%H-48
1.6%H-62 1.5%H-52

HOMO-55[#132, -19.757 eV]= Fr 2: 48.6%H-54 9.9%H-55 8.3%H-30 7.8%H-48 3.1%H-59 2.9%H-49
1.5%H-61 1.4%H-51

HOMO-56[#131, -19.866 eV]= Fr 1: 2.1%L+6
Fr 2: 38.0%H-56 S(-0.053) OP(0.01)
15.5%H-53 S(0.020) OP(0.00)
15.3%H-57 S(-0.014) OP(0.00)
7.0%H-35 S(-0.249) OP(0.01)
5.4%H-50 S(0.100) OP(0.00)
5.1%H-44 S(-0.177) OP(0.01)
2.3%H-77 S(0.136) OP(0.00)

Chart S5 Output file for Am(TPAm)₃³⁺

HOMO-0[#187, -13.976 eV]= Fr 1: 27.4%H-3 19.8%H-1 11.5%H-0 5.3%H-5 4.5%H-4
Fr 2: 11.5%H-1 S(-0.013 -0.025 0.005 0.022 0.010) OP(0.00 -0.01 0.00 0.00 0.00)
7.6%H-0 S(-0.031 0.002 0.012 -0.011 0.000) OP(-0.01 0.00 0.00 0.00 0.00)
4.9%H-4 S(0.007 -0.017 0.014 0.007 0.008) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-1[#186, -13.982 eV]= Fr 1: 18.1%H-5 16.9%H-2 11.3%H-3 10.2%L+0 6.0%H-1 5.2%H-0
Fr 2: 11.6%H-0 S(-0.011 0.013 -0.031 0.012 0.002 0.012) OP(0.00 0.00 -0.01 0.00 0.00 0.00)
7.7%H-1 S(0.022 -0.007 -0.013 -0.006 -0.025 0.005) OP(-0.01 0.00 0.00 0.00 0.00 0.00)

HOMO-2[#185, -14.130 eV]= Fr 1: 20.0%H-1 15.5%H-5 14.8%L+0 7.1%H-2 6.7%H-0 1.3%H-4
Fr 2: 28.1%H-2 S(0.004 0.002 0.033 -0.023 0.003 -0.001) OP(0.00 0.00 -0.01 -0.01 0.00 0.00)
1.4%H-12 S(0.002 0.001 0.000 0.000 0.001 0.001) OP(0.00 0.00 0.00 0.00 0.00 0.00)

HOMO-3[#184, -14.449 eV]= Fr 1: 42.7%H-0 25.2%H-5 8.9%H-2 6.1%L+0 2.9%H-3
Fr 2: 5.6%H-5 S(-0.006 0.005 0.017 0.015 -0.012) OP(0.00 0.00 0.00 0.00 0.00)
1.5%H-9 S(0.010 -0.001 0.011 0.008 0.007) OP(0.00 0.00 0.00 0.00 0.00)
1.1%H-14 S(0.002 0.000 -0.002 -0.002 0.001) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-4[#183, -14.455 eV]= Fr 1: 40.8%H-3 18.2%H-2 11.8%L+0 7.9%H-1 6.0%H-0
Fr 2: 5.3%H-4 S(0.007 0.004 0.004 -0.017 0.014) OP(0.00 0.00 0.00 0.00 0.00)
1.6%H-10 S(-0.001 -0.010 -0.008 -0.003 0.014) OP(0.00 0.00 0.00 0.00 0.00)
1.1%H-13 S(0.000 -0.002 -0.002 0.002 -0.002) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-5[#182, -14.536 eV]= Fr 1: 10.0%H-1 7.7%H-4 5.2%L+0 3.2%H-2 3.2%H-5
Fr 2: 41.4%H-2 S(0.004 -0.001 0.033 -0.023 0.002) OP(0.00 0.00 0.01 0.01 0.00)
13.3%H-11 S(0.000 0.003 0.025 -0.017 0.000) OP(0.00 0.00 0.00 0.00 0.00)
7.9%H-23 S(0.004 -0.004 0.008 -0.005 0.004) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-6[#181, -14.642 eV]= Fr 1: 7.8%H-1 4.5%H-0
Fr 2: 24.6%H-0 S(0.002 0.012) OP(0.00 0.00)
15.7%H-4 S(-0.017 0.014) OP(0.00 0.00)
8.5%H-18 S(0.011 -0.009) OP(0.00 0.00)
7.2%H-21 S(-0.003 0.003) OP(0.00 0.00)
5.6%H-6 S(0.009 -0.013) OP(0.00 0.00)
5.2%H-1 S(-0.025 0.005) OP(0.00 0.00)

HOMO-7[#180, -14.645 eV]= Fr 1: 7.0%H-2 4.4%H-3
Fr 2: 24.3%H-1 S(-0.007 -0.013) OP(0.00 0.00)
15.4%H-5 S(0.017 -0.012) OP(0.00 0.00)
8.4%H-19 S(0.011 -0.009) OP(0.00 0.00)
7.1%H-22 S(-0.003 0.002) OP(0.00 0.00)
5.8%H-7 S(0.013 -0.002) OP(0.00 0.00)
5.1%H-0 S(0.013 -0.031) OP(0.00 0.00)

HOMO-8[#179, -14.768 eV]= Fr 2: 78.7%H-12 12.7%H-8 2.1%H-11

HOMO-9[#178, -14.796 eV]= Fr 2: 84.2%H-14 5.7%H-7 2.4%H-6 1.2%H-10

HOMO-10[#177, -14.796 eV]= Fr 1: 1.0%H-3
Fr 2: 84.3%H-13 S(0.000) OP(0.00)
5.7%H-6 S(0.008) OP(0.00)
2.3%H-7 S(-0.002) OP(0.00)
1.2%H-9 S(0.007) OP(0.00)

HOMO-11[#176, -14.941 eV]= Fr 1: 70.2%H-4 4.0%H-5 3.3%H-0 2.6%L+0
Fr 2: 3.7%H-17 S(-0.015 -0.001 0.000 0.000) OP(-0.01 0.00 0.00 0.00)
3.2%H-3 S(-0.016 0.019 0.015 -0.020) OP(0.00 0.00 0.00 0.00)

2.4%H-12 S(0.001 0.001 0.001 0.000) OP(0.00 0.00 0.00 0.00)
1.8%H-11 S(0.003 0.000 -0.001 0.025) OP(0.00 0.00 0.00 0.00)

HOMO-12[#175, -15.275 eV]= Fr 1: 3.8%H-0
Fr 2: 44.0%H-6 S(-0.013) OP(0.00)
11.7%H-7 S(-0.005) OP(0.00)
9.5%H-14 S(0.002) OP(0.00)
5.5%H-5 S(-0.006) OP(0.00)
5.2%H-10 S(0.014) OP(0.00)
3.0%H-9 S(0.010) OP(0.00)
2.1%H-30 S(-0.002) OP(0.00)

HOMO-13[#174, -15.276 eV]= Fr 1: 3.5%H-2
Fr 2: 43.9%H-7 S(0.013) OP(0.00)
11.7%H-6 S(-0.008) OP(0.00)
9.5%H-13 S(-0.002) OP(0.00)
5.5%H-4 S(0.004) OP(0.00)
5.2%H-9 S(0.011) OP(0.00)
2.9%H-10 S(-0.010) OP(0.00)
2.1%H-31 S(-0.003) OP(0.00)

HOMO-14[#173, -15.371 eV]= Fr 2: 62.0%H-8 13.3%H-12 6.0%H-17 4.4%H-34 2.5%H-3 2.3%H-11
2.0%H-29 1.1%H-20

HOMO-15[#172, -15.714 eV]= Fr 2: 74.0%H-15 9.9%H-16 2.4%H-4 1.9%H-6 1.9%H-40 1.7%H-1
1.2%H-9

HOMO-16[#171, -15.714 eV]= Fr 2: 74.0%H-16 9.9%H-15 2.4%H-5 1.9%H-7 1.9%H-39 1.7%H-0
1.2%H-10

HOMO-17[#170, -15.740 eV]= Fr 2: 54.9%H-17 34.6%H-3 1.7%H-34 1.2%H-41 1.0%H-23

HOMO-18[#169, -15.913 eV]= Fr 2: 23.3%H-9 18.8%H-5 17.0%H-1 5.9%H-6 5.1%H-16 5.1%H-0
3.5%H-7 3.0%H-10

HOMO-19[#168, -15.913 eV]= Fr 1: 3.6%H-3
Fr 2: 23.4%H-10 S(-0.001) OP(0.00)

18.7%H-4 S(0.007) OP(0.00)
16.9%H-0 S(-0.031) OP(0.00)
5.9%H-7 S(-0.002) OP(0.00)
5.1%H-15 S(0.007) OP(0.00)
5.1%H-1 S(-0.013) OP(0.00)
3.5%H-6 S(0.008) OP(0.00)

HOMO-20[#167, -15.968 eV]= Fr 1: 8.6%H-4

Fr 2: 48.2%H-3 S(-0.016) OP(0.01)
25.3%H-17 S(-0.015) OP(0.00)
4.1%H-8 S(0.007) OP(0.00)
3.6%H-20 S(-0.001) OP(0.00)
2.0%H-35 S(0.003) OP(0.00)

HOMO-21[#166, -16.268 eV]= Fr 2: 40.3%H-21 24.9%H-18 10.9%H-22 4.6%H-25 3.7%H-9 3.1%H-4
3.0%H-19 2.5%H-1

HOMO-22[#165, -16.268 eV]= Fr 2: 40.2%H-22 24.9%H-19 10.9%H-21 4.6%H-24 3.6%H-10 3.1%H-5
3.0%H-18 2.5%H-0

HOMO-23[#164, -16.271 eV]= Fr 2: 56.7%H-20 30.3%H-23 5.9%H-11 2.1%H-26 1.5%H-17

HOMO-24[#163, -16.363 eV]= Fr 2: 33.5%H-9 18.9%H-5 8.8%H-21 6.2%H-1 5.2%H-18 2.8%H-25
2.6%H-4 2.5%H-30

HOMO-25[#162, -16.363 eV]= Fr 2: 33.5%H-10 19.0%H-4 8.8%H-22 6.2%H-0 5.1%H-19 2.8%H-24
2.6%H-5 2.5%H-31

HOMO-26[#161, -16.394 eV]= Fr 1: 2.7%L+0 1.8%L+5

Fr 2: 50.8%H-11 S(0.025 0.124) OP(0.01 0.02)
15.7%H-2 S(0.033 0.108) OP(0.00 0.01)
7.4%H-23 S(0.008 0.026) OP(0.00 0.00)
4.0%H-8 S(-0.016 0.042) OP(0.00 0.00)
3.0%H-44 S(0.005 0.029) OP(0.00 0.00)
2.8%H-45 S(0.009 0.034) OP(0.00 0.00)

Chart S6 Output file for Cm(TPAm)₃³⁺

HOMO-0[#187, -14.450 eV]= Fr 2: 64.5%H-2 8.4%H-11 8.3%H-23 6.6%H-5 2.0%H-1 1.4%H-8 1.2%H-3
1.2%H-4

HOMO-1[#186, -14.507 eV]= Fr 2: 35.5%H-0 26.5%H-3 11.6%H-4 4.6%H-21 3.7%H-18 3.6%H-10
3.5%H-19 2.4%H-22

HOMO-2[#185, -14.554 eV]= Fr 2: 31.9%H-1 25.3%H-4 8.8%H-3 4.8%H-22 4.2%H-19 3.7%H-9 3.4%H-
18 3.1%H-2

HOMO-3[#184, -14.743 eV]= Fr 2: 54.8%H-14 19.7%H-13 9.3%H-8 6.7%H-11 3.0%H-7 1.3%H-6

HOMO-4[#183, -14.764 eV]= Fr 2: 56.7%H-13 18.3%H-14 10.7%H-7 3.9%H-8 2.5%H-11 1.5%H-6
1.1%H-0

HOMO-5[#182, -14.772 eV]= Fr 2: 76.8%H-12 12.9%H-6 3.6%H-7 1.1%H-1 1.1%H-10 1.0%H-15

HOMO-6[#181, -15.129 eV]= Fr 2: 59.8%H-7 17.9%H-13 5.7%H-9 3.6%H-12 2.4%H-33 1.1%H-3

HOMO-7[#180, -15.148 eV]= Fr 2: 60.9%H-6 16.7%H-12 5.4%H-10 2.6%H-13 2.5%H-32 1.7%H-4
1.1%H-1

HOMO-8[#179, -15.265 eV]= Fr 2: 64.0%H-8 19.3%H-14 3.9%H-17 2.9%H-34 1.4%H-29 1.3%H-11
1.2%H-20 1.0%L+8

HOMO-9[#178, -15.605 eV]= Fr 2: 75.3%H-5 8.0%H-17 4.7%H-2 1.8%H-11 1.5%H-8

HOMO-10[#177, -15.693 eV]= Fr 2: 52.0%H-16 26.8%H-15 2.9%H-1 2.9%H-3 2.9%H-9 2.2%H-6
1.4%H-40 1.1%H-7

HOMO-11[#176, -15.709 eV]= Fr 2: 47.8%H-15 28.6%H-16 4.8%H-4 3.0%H-10 2.6%H-0 2.3%H-1
2.0%H-7 1.7%H-6

HOMO-12[#175, -15.758 eV]= Fr 2: 25.4%H-1 18.4%H-3 12.3%H-9 10.6%H-0 6.0%H-4 5.2%H-15
5.0%H-10 4.7%H-16

HOMO-13[#174, -15.787 eV]= Fr 2: 56.0%H-17 6.3%H-15 6.2%H-0 5.7%H-4 3.6%H-1 3.2%H-5 3.2%H-
10 1.8%H-20

HOMO-14[#173, -15.807 eV]= Fr 2: 21.1%H-17 17.3%H-0 11.6%H-10 10.7%H-4 6.5%H-5 5.4%H-1
4.9%H-16 4.5%H-15

HOMO-15[#172, -16.176 eV]= Fr 2: 44.1%H-11 9.9%H-2 8.4%H-10 4.8%H-9 3.4%H-3 3.0%H-8
3.0%H-20 2.6%H-14

HOMO-16[#171, -16.226 eV]= Fr 1: 2.8%L+1

Fr 2: 43.2%H-9 S(0.088) OP(0.01)

12.9%H-4 S(-0.117) OP(0.01)

6.8%H-10 S(0.046) OP(0.00)

6.0%H-21 S(-0.002) OP(0.00)

4.9%H-7 S(-0.044) OP(0.00)

3.6%H-1 S(0.007) OP(0.00)

2.2%H-0 S(-0.017) OP(0.00)

HOMO-17[#170, -16.256 eV]= Fr 2: 26.6%H-10 12.8%H-22 11.6%H-11 10.1%H-3 4.2%H-0 3.1%H-6
2.9%H-1 2.8%H-2

HOMO-18[#169, -16.298 eV]= Fr 2: 25.7%H-21 15.9%H-19 15.5%H-22 12.4%H-20 11.1%H-18 8.4%H-
23 3.8%H-9

HOMO-19[#168, -16.314 eV]= Fr 2: 27.0%H-21 21.8%H-20 20.5%H-18 13.6%H-23 6.2%H-22 2.7%H-
10 1.1%H-19

HOMO-20[#167, -16.324 eV]= Fr 2: 26.0%H-22 17.4%H-20 16.4%H-23 13.6%H-19 7.3%H-10 2.4%H-
18 2.0%H-3 1.8%H-2

HOMO-21[#166, -16.553 eV]= Fr 2: 45.0%H-26 20.3%H-20 16.1%H-23 7.2%H-24 2.1%H-11 2.1%H-8
1.3%H-18 1.0%H-17

HOMO-22[#165, -16.575 eV]= Fr 2: 42.6%H-24 21.8%H-25 11.3%H-19 4.2%H-26 3.6%H-3 3.3%H-21
3.1%H-18 2.2%H-20

HOMO-23[#164, -16.594 eV]= Fr 2: 50.0%H-25 21.2%H-24 9.6%H-18 4.7%H-19 3.5%H-4 2.3%H-22
1.2%H-26

HOMO-24[#163, -17.202 eV]= Fr 2: 40.9%H-27 39.4%H-28 3.5%H-19 2.4%H-18 2.0%H-24 1.4%H-22
1.4%H-25 1.2%H-21

HOMO-25[#162, -17.225 eV]= Fr 2: 45.1%H-28 36.8%H-27 4.2%H-29 2.9%H-18 1.5%H-24 1.3%H-21

HOMO-26[#161, -17.268 eV]= Fr 2: 61.6%H-29 11.6%H-26 6.6%H-23 3.9%H-20 2.3%H-34 1.6%H-11
1.4%H-18 1.3%H-27

HOMO-27[#160, -17.323 eV]= Fr 2: 14.4%H-18 14.4%H-21 13.7%H-19 13.4%H-24 13.3%H-27 5.3%H-
22 3.0%H-3 2.9%H-0

HOMO-28[#159, -17.362 eV]= Fr 2: 17.2%H-19 15.7%H-22 13.8%H-18 13.7%H-25 6.6%H-28 5.1%H-
21 3.6%H-1 3.3%H-4

HOMO-29[#158, -17.478 eV]= Fr 1: 2.0%L+6 1.3%L+9

Fr 2: 28.0%H-26 S(-0.151 0.186) OP(0.02 0.01)

23.1%H-29 S(0.035 -0.052) OP(0.00 0.00)

21.8%H-23 S(0.136 -0.144) OP(0.01 0.01)

7.4%H-20 S(0.112 -0.066) OP(0.01 0.00)

2.9%H-2 S(0.045 -0.111) OP(0.00 0.00)

2.9%H-38 S(-0.116 -0.001) OP(0.00 0.00)

HOMO-30[#157, -17.991 eV]= Fr 2: 38.6%H-37 20.0%H-43 8.2%H-33 5.1%H-46 5.0%H-30 4.0%H-39
3.3%H-36 3.3%H-31

HOMO-31[#156, -17.996 eV]= Fr 2: 35.2%H-36 17.6%H-42 9.8%H-38 5.1%H-32 4.8%H-45 4.1%H-30
3.5%H-44 3.5%H-37

HOMO-32[#155, -18.019 eV]= Fr 2: 49.1%H-38 16.4%H-44 7.0%H-36 5.6%H-47 4.4%H-41 3.5%H-42
3.3%H-50 1.6%H-37

HOMO-33[#154, -18.057 eV]= Fr 2: 48.5%H-33 11.5%H-37 9.8%H-30 6.1%H-59 3.6%H-43 3.6%H-7
2.0%H-31 1.8%H-46

HOMO-34[#153, -18.082 eV]= Fr 1: 2.0%H-2

Fr 2: 52.1%H-32 S(-0.004) OP(0.00)

11.5%H-31 S(0.005) OP(0.00)

8.9%H-36 S(0.000) OP(0.00)

6.8%H-58 S(-0.001) OP(0.00)

4.1%H-6 S(0.005) OP(0.00)

2.2%H-42 S(0.000) OP(0.00)

1.9%H-30 S(0.007) OP(0.00)

HOMO-35[#152, -18.182 eV]= Fr 1: 9.2%H-0 8.6%H-5 3.3%H-3 3.2%L+0

Fr 2: 50.3%H-34 S(-0.005 -0.004 0.000 0.004) OP(0.00 0.00 0.00 0.00)

5.4%H-57 S(0.000 0.001 0.000 0.000) OP(0.00 0.00 0.00 0.00)

3.5%H-35 S(-0.002 -0.001 0.000 0.003) OP(0.00 0.00 0.00 0.00)

3.0%H-56 S(0.001 0.001 0.000 -0.001) OP(0.00 0.00 0.00 0.00)

HOMO-36[#151, -18.309 eV]= Fr 2: 15.5%H-37 9.0%H-35 8.4%H-43 7.8%H-46 6.6%H-31 5.0%H-38
4.8%H-34 4.8%H-42

HOMO-37[#150, -18.323 eV]= Fr 1: 14.7%H-2

Fr 2: 18.3%H-36 S(0.000) OP(0.00)

7.7%H-45 S(-0.003) OP(0.00)

7.6%H-43 S(0.001) OP(0.00)

7.6%H-30 S(0.007) OP(0.00)

6.9%H-42 S(0.000) OP(0.00)

6.6%H-37 S(0.000) OP(0.00)

5.6%H-46 S(-0.002) OP(0.00)

HOMO-38[#149, -18.348 eV]= Fr 2: 20.3%H-38 18.3%H-44 17.1%H-35 14.3%H-47 4.2%H-41 4.1%H-
37 3.1%H-42 2.8%H-36

HOMO-39[#148, -18.405 eV]= Fr 1: 42.8%H-5 21.1%H-3 6.8%H-2

Fr 2: 4.6%H-34 S(-0.004 0.000 -0.003) OP(0.00 0.00 0.00)
3.6%H-37 S(-0.001 0.000 0.000) OP(0.00 0.00 0.00)
3.1%H-42 S(0.000 -0.002 0.000) OP(0.00 0.00 0.00)
2.1%H-46 S(-0.001 -0.002 -0.002) OP(0.00 0.00 0.00)
2.0%H-43 S(0.001 -0.001 0.001) OP(0.00 0.00 0.00)

HOMO-40[#147, -18.421 eV]= Fr 1: 42.8%H-2 9.2%H-3 7.7%H-1 2.7%L+0
Fr 2: 8.5%H-36 S(0.000 -0.002 0.000 0.000) OP(0.00 0.00 0.00 0.00)
6.5%H-42 S(0.000 -0.002 -0.001 0.000) OP(0.00 0.00 0.00 0.00)
5.8%H-45 S(-0.003 0.000 0.003 -0.002) OP(0.00 0.00 0.00 0.00)
3.4%H-43 S(0.001 -0.001 -0.001 0.001) OP(0.00 0.00 0.00 0.00)

HOMO-41[#146, -18.452 eV]= Fr 1: 29.5%H-0 22.0%L+0 8.6%H-1 7.5%H-3 3.5%H-4 3.5%H-2
Fr 2: 3.6%H-34 S(-0.005 0.004 -0.002 0.000 -0.002 -0.003) OP(0.00 0.00 0.00 0.00 0.00 0.00)
2.7%H-33 S(-0.001 0.003 -0.005 0.003 0.004 0.003) OP(0.00 0.00 0.00 0.00 0.00 0.00)

HOMO-42[#145, -18.849 eV]= Fr 1: 12.9%H-1 10.1%H-4 7.8%L+0 4.4%H-3
Fr 2: 17.4%H-43 S(-0.001 0.001 0.001 -0.001) OP(0.00 0.00 0.00 0.00)
13.2%H-46 S(-0.003 0.000 -0.002 -0.002) OP(0.00 0.00 0.00 0.00)
6.6%H-31 S(0.006 0.003 0.005 0.010) OP(0.00 0.00 0.00 0.00)
4.2%H-39 S(0.000 0.002 -0.002 -0.001) OP(0.00 0.00 0.00 0.00)

HOMO-43[#144, -18.876 eV]= Fr 1: 9.7%H-0 8.9%H-5 7.9%H-2 6.7%H-3 6.5%H-1
Fr 2: 15.2%H-42 S(0.001 0.000 0.000 -0.002 -0.001) OP(0.00 0.00 0.00 0.00 0.00)
13.3%H-45 S(0.000 0.000 -0.003 0.000 0.003) OP(0.00 0.00 0.00 0.00 0.00)
4.9%H-30 S(-0.002 0.003 0.007 -0.006 -0.009) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-44[#143, -18.987 eV]= Fr 1: 22.8%H-4 12.8%H-1 9.2%H-0 5.1%L+0 3.3%H-2
Fr 2: 20.9%H-40 S(0.001 -0.002 0.001 0.000 -0.001) OP(0.00 0.00 0.00 0.00 0.00)
3.9%H-46 S(0.000 -0.003 0.002 -0.002 -0.002) OP(0.00 0.00 0.00 0.00 0.00)
3.4%H-39 S(0.002 0.000 -0.002 -0.002 -0.002) OP(0.00 0.00 0.00 0.00 0.00)

HOMO-45[#142, -19.003 eV]= Fr 1: 19.9%H-1 3.6%H-0
Fr 2: 15.4%H-45 S(0.003 0.000) OP(0.00 0.00)
11.4%H-40 S(-0.002 0.001) OP(0.00 0.00)
7.3%H-46 S(-0.003 0.002) OP(0.00 0.00)
7.0%H-42 S(-0.001 0.001) OP(0.00 0.00)
6.8%H-39 S(0.000 -0.002) OP(0.00 0.00)
5.6%H-47 S(-0.001 -0.002) OP(0.00 0.00)

HOMO-46[#141, -19.020 eV]= Fr 1: 10.9%H-3 10.0%H-4 8.3%H-5
Fr 2: 27.1%H-39 S(-0.001 0.002 0.001) OP(0.00 0.00 0.00)
8.7%H-43 S(-0.001 0.001 0.001) OP(0.00 0.00 0.00)
6.4%H-46 S(-0.002 0.000 -0.001) OP(0.00 0.00 0.00)
6.0%H-41 S(0.000 -0.001 0.004) OP(0.00 0.00 0.00)
3.7%H-58 S(0.000 0.001 0.001) OP(0.00 0.00 0.00)

HOMO-47[#140, -19.041 eV]= Fr 1: 5.3%H-4
Fr 2: 17.0%H-40 S(0.001) OP(0.00)
14.6%H-42 S(0.000) OP(0.00)
9.3%H-39 S(0.002) OP(0.00)
7.6%H-44 S(0.000) OP(0.00)
5.6%H-41 S(-0.001) OP(0.00)
4.8%H-58 S(0.001) OP(0.00)
4.6%H-46 S(0.000) OP(0.00)

HOMO-48[#139, -19.054 eV]= Fr 1: 13.8%H-4 6.2%H-3
Fr 2: 15.5%H-39 S(0.002 -0.001) OP(0.00 0.00)
13.3%H-40 S(0.001 0.003) OP(0.00 0.00)
10.5%H-43 S(0.001 -0.001) OP(0.00 0.00)
6.7%H-45 S(-0.002 0.000) OP(0.00 0.00)
5.9%H-59 S(0.000 -0.002) OP(0.00 0.00)
4.3%H-41 S(-0.001 0.000) OP(0.00 0.00)

HOMO-49[#138, -19.113 eV]= Fr 1: 8.1%L+0 5.5%H-3 4.9%H-5
Fr 2: 22.7%H-44 S(-0.004 0.000 -0.001) OP(0.00 0.00 0.00)
15.2%H-47 S(0.005 0.000 -0.001) OP(0.00 0.00 0.00)
12.0%H-41 S(0.002 0.000 0.004) OP(0.00 0.00 0.00)
6.9%H-46 S(-0.002 -0.002 -0.001) OP(0.00 0.00 0.00)
3.1%H-56 S(-0.001 0.000 0.001) OP(0.00 0.00 0.00)

HOMO-50[#137, -19.163 eV]= Fr 1: 6.6%H-4 4.7%H-5
Fr 2: 39.4%H-41 S(-0.001 0.004) OP(0.00 0.00)
14.5%H-47 S(0.000 -0.001) OP(0.00 0.00)
7.9%H-57 S(-0.001 0.001) OP(0.00 0.00)
5.7%H-35 S(-0.001 -0.001) OP(0.00 0.00)
5.6%H-50 S(-0.001 0.002) OP(0.00 0.00)
3.2%H-44 S(0.000 -0.001) OP(0.00 0.00)

HOMO-51[#136, -19.299 eV]= Fr 2: 43.5%H-50 16.2%H-47 15.4%H-35 3.8%H-49 3.7%H-34 1.7%H-41
1.5%H-71 1.5%H-46

HOMO-52[#135, -19.335 eV]= Fr 2: 25.6%H-49 19.4%H-48 10.9%H-46 8.4%H-31 5.7%H-45 3.5%H-30
3.2%H-50 2.9%H-32

HOMO-53[#134, -19.348 eV]= Fr 1: 2.7%H-4
Fr 2: 28.9%H-48 S(0.002) OP(0.00)
19.1%H-49 S(0.001) OP(0.00)
12.5%H-45 S(-0.002) OP(0.00)
9.0%H-30 S(0.007) OP(0.00)
4.8%H-46 S(0.000) OP(0.00)
3.0%H-31 S(0.003) OP(0.00)
2.8%H-33 S(0.004) OP(0.00)

HOMO-54[#133, -19.738 eV]= Fr 2: 48.1%H-55 9.8%H-54 8.6%H-31 8.3%H-49 3.4%H-58 2.7%H-48
1.6%H-62 1.5%H-52

HOMO-55[#132, -19.757 eV]= Fr 2: 48.6%H-54 9.9%H-55 8.3%H-30 7.8%H-48 3.1%H-59 2.9%H-49
1.5%H-61 1.4%H-51

HOMO-56[#131, -19.866 eV]= Fr 1: 2.1%L+6
Fr 2: 38.0%H-56 S(-0.053) OP(0.01)
15.5%H-53 S(0.020) OP(0.00)
15.3%H-57 S(-0.014) OP(0.00)
7.0%H-35 S(-0.249) OP(0.01)
5.4%H-50 S(0.100) OP(0.00)
5.1%H-44 S(-0.177) OP(0.01)
2.3%H-77 S(0.136) OP(0.00)