Table 1: Predicted torsion-rotation parameters and nuclear quadrupole coupling tensor elements for different *o*-halotoluenes on the B3LYP/6-311++g(2d,2p) level of theory.

		^{19}F	^{35}Cl	^{37}Cl	^{79}Br	^{81}Br	^{127}I[a]
A_e	/ MHz	3249.75	2852.00	2843.44	2763.08	2762.06	2736.94
B_e	/ MHz	2184.32	1528.48	1490.24	977.63	967.37	725.34
C_e	/ MHz	1316.82	1001.24	983.67	725.33	719.60	575.40
τ'_{aaaa}	/ MHz	-2.87E-03	-2.239373E-03	-2.272676E-03	-1.759503E-03	-1.765403E-03	-
τ'_{bbaa}	/ MHz	-7.01E-04	-8.593302E-04	-8.539581E-04	-8.117399E-04	-8.043124E-04	-
τ'_{bbbb}	/ MHz	-5.34E-04	-3.051702E-04	-2.924190E-04	-1.600799E-04	-1.573219E-04	-
τ'_{ccaa}	/ MHz	-3.20E-04	-2.065958E-04	-2.049531E-04	-1.084426E-04	-1.076111E-04	-
τ'_{ccbb}	/ MHz	-1.55E-04	-1.094221E-04	-1.075298E-04	-8.578210E-05	-8.484871E-05	-
τ'_{cccc}	/ MHz	-9.28E-05	-6.973795E-05	-6.888219E-05	-5.393261E-05	-5.351865E-05	-
$3/2^*\chi_{aa}$	/ MHz	-	-9.868053E+01	-7.821515E+01	8.398216E+02	7.030273E+02	-2.731842E+03
$1/4^*(\chi_{bb}-\chi_{cc})$	/ MHz	-	1.619467E+00	1.350175E+00	-1.196950E+01	-1.002680E+01	3.328667E+01
χ_{ab}	/ MHz	-	1.455229E+01	1.037905E+01	1.082150E+01	7.749723E+00	2.595124E+01

[a]- For Iodine 6-311** was used.

Table 2: Predicted torsion-rotation parameters and nuclear quadrupole coupling tensor element for different *o*-halotoluenes on the MP2/6-311++g(2d,2p) level of theory.

		^{19}F	^{35}Cl	^{37}Cl	^{79}Br	^{81}Br	^{127}I[a]
A_e	/ MHz	3252.84	2865.52	2856.78	2773.80	2772.73	2750.39
B_e	/ MHz	2177.22	1529.46	1491.49	984.68	974.40	734.96
C_e	/ MHz	1314.73	1003.31	985.80	729.95	724.21	582.04
τ'_{aaaa}	/ MHz	-2.185001E-03	-2.275425E-03	-2.309020E-03	-1.789443E-03	-1.795451E-03	-
τ'_{bbaa}	/ MHz	-1.550701E-03	-8.379363E-04	-8.326919E-04	-7.925314E-04	-7.854262E-04	-
τ'_{bbbb}	/ MHz	-4.287970E-04	-2.965011E-04	-2.840024E-04	-1.524629E-04	-1.498322E-04	-
τ'_{ccaa}	/ MHz	-2.947381E-04	-2.054390E-04	-2.038181E-04	-1.076624E-04	-1.068412E-04	-
τ'_{ccbb}	/ MHz	-1.671904E-04	-1.054311E-04	-1.035583E-04	-8.112417E-05	-8.024697E-05	-
τ'_{cccc}	/ MHz	-9.343592E-05	-6.803187E-05	-6.715603E-05	-5.134336E-05	-5.094582E-05	-
$3/2^*\chi_{aa}$	/ MHz	-	-9.691183E+01	-7.680456E+01	8.089923E+02	6.772112E+02	-2.647378E+03
$1/4^*(\chi_{bb}-\chi_{cc})$	/ MHz	-	1.261928E+00	1.065615E+00	-1.006060E+01	-8.427282E+00	2.901938E+01
χ_{ab}	/ MHz	-	-1.399106E+01	-9.962374E+00	-8.466527E+00	-5.806944E+00	-4.507113E+01

[a]- For Iodine 6-311** was used.