Table S3: Calculated Frequencies of $(CH_3)_3CCN–BF_3$.[a]

Mode	Symmetry[b]	Frequency (^{11}B)[c]	Frequency (^{10}B)[c]	Intensity[d]	Approximate Description[e]
ν_1	A1	147	147	32.5	B-N Stretch
ν_2	A1	399	399	48.6	$(CH_3)_3CCN$ Deformation
ν_3	A1	585	601	442	BF_3 Symmetric Deformation
ν_4	A1	699	699	9.20	$(CH_3)_3CCN$ Breathing
ν_5	A1	834	840	167	BF_3 Symmetric Stretch
ν_6	A1	883	883	23.2	C-C Stretch
ν_7	A1	1260	1304	17.1	$(CH_3)_3CCN$ Deformation
ν_8	A1	1426	1426	0.470	CH_3 Deformaiton
ν_9	A1	1503	1503	13.4	CH_3 Deformaiton
ν_{10}	A1	2313	2313	107	CN Stretch
ν_{11}	A1	2928	2928	8.14	CH Stretch
ν_{12}	A1	3004	3004	27.1	CH Stretch
ν_{13}	A2	3	3	0	Intermolecular Torsion
ν_{14}	A2	198	198	0	CH_3 Torsion
ν_{15}	A2	971	971	0	CH_3 Torsion
ν_{16}	A2	1461	1461	0	CH_3 Torsion
ν_{17}	A2	3003	3003	0	CH Stretch
ν_{18}	E	47	47	0.0354	Intermolecular Bend
ν_{19}	E	165	165	0.631	C-N Bend
ν_{20}	E	266	266	0.0658	Methyl Torsion
ν_{21}	E	331	331	0.0455	$(CH_3)_3CCN$ Deformation
ν_{21}	E	371	371	0.0094	$(CH_3)_3CCN$ Deformation
ν_{22}	E	486	488	5.98	BF_3 Asymmetric Deformation
ν_{23}	E	605	605	0.628	C-C-N Bend
ν_{24}	E	942	942	1.48	CH_3 Rock
ν_{25}	E	1048	1048	0.0001	CH_3 Rock
ν_{26}	E	1225	1225	0.798	$(CH_3)_3CCN$ Def./CH_3 Rock
ν_{27}	E	1258	1258	357	BF_3 Asymmetric Stretch
ν_{28}	E	1393	1393	5.83	CH_3 Deformation
ν_{29}	E	1469	1469	0.461	CH_3 Deformation
ν_{30}	E	1485	1485	9.78	CH_3 Deformation
ν_{31}	E	2923	2923	10.7	CH Stretch
ν_{32}	E	2999	2999	1.28	CH Stretch
ν_{33}	E	3006	3006	20.5	CH Stretch

a) Frequencies were calculated using the B3LYP/aug-cc-pV(T|D)Z method, and frequencies above 2000 cm^{-1} were scaled by a factor of 0.96. See text for discussion. b) The point group is C_{3v}. c) Units of cm^{-1}. d) Units of km/mol. e) Given the size of the system, most of the normal vibrational modes are mixtures of motions about several internal coordinates, and as such, the brief descriptions listed are very much approximate.