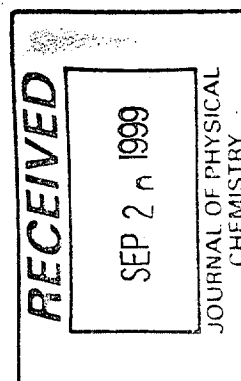


## SUPPLEMENTARY MATERIAL

Table S.1: Calculated *ab initio* MR-CI energies for the ground state ( $\tilde{X}^1A'$ ) of the  $N_2O$  molecule. The  $N_2$  distance is kept fixed at a value of  $2.13199 a_0$ . The Jacobi coordinate system used is defined in figure 1.

$R/a_0$	$\theta/deg$	$E_{MR-CI}/E_h$	$R/a_0$	$\theta/deg$	$E_{MR-CI}/E_h$
2.9	0.0	-184.12407142	3.0	0.0	-184.19856071
2.9	10.0	-184.14282305	3.0	10.0	-184.20348334
2.9	20.0	-184.16494424	3.0	20.0	-184.20978746
2.9	30.0	-184.17499046	3.0	30.0	-184.20485338
2.9	40.0	-184.16238803	3.0	40.0	-184.18045120
2.9	50.0	-184.13170284	3.0	50.0	-184.14464732
2.9	60.0	-184.14368242	3.0	60.0	-184.14687018
2.9	70.0	-184.15554791	3.0	70.0	-184.15540233
2.9	80.0	-184.16102346	3.0	80.0	-184.15920660
2.9	90.0	-184.16169270	3.0	90.0	-184.15967900
3.1	0.0	-184.23929900	3.2	0.0	-184.26152682
3.1	10.0	-184.23982930	3.2	10.0	-184.26016598
3.1	20.0	-184.23663336	3.2	20.0	-184.25111971
3.1	30.0	-184.22028182	3.2	30.0	-184.22734317
3.1	40.0	-184.19022902	3.2	40.0	-184.19362660
3.1	50.0	-184.15274193	3.2	50.0	-184.15936724
3.1	60.0	-184.14836594	3.2	60.0	-184.14892521
3.1	70.0	-184.15416562	3.2	70.0	-184.15266779
3.1	80.0	-184.15658647	3.2	80.0	-184.15435975
3.1	90.0	-184.15690968	3.2	90.0	-184.15402457
3.3	0.0	-184.27095496	3.4	0.0	-184.26994086
3.3	10.0	-184.26780884	3.4	10.0	-184.26633720
3.3	20.0	-184.25505565	3.4	20.0	-184.25170681
3.3	30.0	-184.22687753	3.4	30.0	-184.22474408
3.3	40.0	-184.19366812	3.4	40.0	-184.19162059
3.3	50.0	-184.16074647	3.4	50.0	-184.16043138
3.3	60.0	-184.14777850	3.4	60.0	-184.14653706
3.3	70.0	-184.15058551	3.4	70.0	-184.14732845
3.3	80.0	-184.15072254	3.4	80.0	-184.14824648
3.3	90.0	-184.15099201	3.4	90.0	-184.14765437
3.5	0.0	-184.26389950	3.7	0.0	-184.24628530
3.5	10.0	-184.26088577	3.7	10.0	-184.24135440
3.5	20.0	-184.24413538	3.7	20.0	-184.22616006
3.5	30.0	-184.21803520	3.7	30.0	-184.20344017
3.5	40.0	-184.18793189	3.7	40.0	-184.17644506
3.5	50.0	-184.15784541	3.7	50.0	-184.15367462
3.5	60.0	-184.14514470	3.7	60.0	-184.14285138
3.5	70.0	-184.14471324	3.7	70.0	-184.14045223
3.5	80.0	-184.14503531	3.7	80.0	-184.14098140
3.5	90.0	-184.14550865	3.7	90.0	-184.14135152



$R/a_0$	$\theta/\text{deg}$	$E_{MRCI}/E_h$	$R/a_0$	$\theta/\text{deg}$	$E_{MRCI}/E_h$
3.9	0.0	-184.22345155	4.0	0.0	-184.21238153
3.9	10.0	-184.21813955	4.0	10.0	-184.20867130
3.9	20.0	-184.20602745	4.0	20.0	-184.19602241
3.9	30.0	-184.18731368	4.0	30.0	-184.17959901
3.9	40.0	-184.16526384	4.0	40.0	-184.16091000
3.9	50.0	-184.14877047	4.0	50.0	-184.14664379
3.9	60.0	-184.14039298	4.0	60.0	-184.14043713
3.9	70.0	-184.13843418	4.0	70.0	-184.13938765
3.9	80.0	-184.13886151	4.0	80.0	-184.13994178
3.9	90.0	-184.13914785	4.0	90.0	-184.14026639
4.5	0.0	-184.16447727	5.0	0.0	-184.14881084
4.5	10.0	-184.16259777	5.0	10.0	-184.14826889
4.5	20.0	-184.15763686	5.0	20.0	-184.14692572
4.5	30.0	-184.15142226	5.0	30.0	-184.14539884
4.5	40.0	-184.14605549	5.0	40.0	-184.14418814
4.5	50.0	-184.14274402	5.0	50.0	-184.14344742
4.5	60.0	-184.14141198	5.0	60.0	-184.14308520
4.5	70.0	-184.14128632	5.0	70.0	-184.14296004
4.5	80.0	-184.14154212	5.0	80.0	-184.14294526
4.5	90.0	-184.14167550	5.0	90.0	-184.14295023
5.5	0.0	-184.14558939	6.0	0.0	-184.14487658
5.5	10.0	-184.14545203	6.0	10.0	-184.14481218
5.5	20.0	-184.14510103	6.0	20.0	-184.14463639
5.5	30.0	-184.14466581	6.0	30.0	-184.14438658
5.5	40.0	-184.14425146	6.0	40.0	-184.14411025
5.5	50.0	-184.14390473	6.0	50.0	-184.14384962
5.5	60.0	-184.14363721	6.0	60.0	-184.14363201
5.5	70.0	-184.14344936	6.0	70.0	-184.14346790
5.5	80.0	-184.14333874	6.0	80.0	-184.14336008
5.5	90.0	-184.14330110	6.0	90.0	-184.14321055

Table S.2: Calculated *ab initio* MR-CI energies for the  $2^1A'$  state of the  $N_2O$  molecule. The  $N_2$  distance is kept fixed at a value of  $2.13199 a_0$ . The Jacobi coordinate system used is defined in figure 1.

$R/a_0$	$\theta/deg$	$E_{MRCI}/E_h$	$R/a_0$	$\theta/deg$	$E_{MRCI}/E_h$
2.9	0.0	-183.82123799	3.0	0.0	-183.88568187
2.9	10.0	-183.86765740	3.0	10.0	-183.93109190
2.9	20.0	-183.94114170	3.0	20.0	-183.98920497
2.9	30.0	-184.01174804	3.0	30.0	-184.04459255
2.9	40.0	-184.06733607	3.0	40.0	-184.08702751
2.9	50.0	-184.10522137	3.0	50.0	-184.11572856
2.9	60.0	-184.08410304	3.0	60.0	-184.09509451
2.9	70.0	-184.04430246	3.0	70.0	-184.05928322
2.9	80.0	-184.01307066	3.0	80.0	-184.03342350
2.9	90.0	-184.00039083	3.0	90.0	-184.02376891
3.1	0.0	-183.94870081	3.2	0.0	-183.98100823
3.1	10.0	-183.97309594	3.2	10.0	-184.00225365
3.1	20.0	-184.02161151	3.2	20.0	-184.04386138
3.1	30.0	-184.06584724	3.2	30.0	-184.07939050
3.1	40.0	-184.1000927	3.2	40.0	-184.10864284
3.1	50.0	-184.12030284	3.2	50.0	-184.12239975
3.1	60.0	-184.10268257	3.2	60.0	-184.10823063
3.1	70.0	-184.07206137	3.2	70.0	-184.08201738
3.1	80.0	-184.05107953	3.2	80.0	-184.06609756
3.1	90.0	-184.04352941	3.2	90.0	-184.06053353
3.3	0.0	-184.00078302	3.4	0.0	-184.01211503
3.3	10.0	-184.01976125	3.4	10.0	-184.03075162
3.3	20.0	-184.05702407	3.4	20.0	-184.06501964
3.3	30.0	-184.08937226	3.4	30.0	-184.09604936
3.3	40.0	-184.11230341	3.4	40.0	-184.11554576
3.3	50.0	-184.12280751	3.4	50.0	-184.12329662
3.3	60.0	-184.11230976	3.4	60.0	-184.11368940
3.3	70.0	-184.09067826	3.4	70.0	-184.09799111
3.3	80.0	-184.07856846	3.4	80.0	-184.08918346
3.3	90.0	-184.07434148	3.4	90.0	-184.08636351
3.5	0.0	-184.01720547	3.7	0.0	-184.01083959
3.5	10.0	-184.03710068	3.7	10.0	-184.05148102
3.5	20.0	-184.07175335	3.7	20.0	-184.08116351
3.5	30.0	-184.09859584	3.7	30.0	-184.10429450
3.5	40.0	-184.11677051	3.7	40.0	-184.11700225
3.5	50.0	-184.12263231	3.7	50.0	-184.12124353
3.5	60.0	-184.11491485	3.7	60.0	-184.11833467
3.5	70.0	-184.10407493	3.7	70.0	-184.11454369
3.5	80.0	-184.09846495	3.7	80.0	-184.11220434
3.5	90.0	-184.09633472	3.7	90.0	-184.11165896

$R/a_0$	$\theta/deg$	$E_{MRCI}/E_h$	$R/a_0$	$\theta/deg$	$E_{MRCI}/E_h$
3.9	0.0	-184.04710057	4.0	0.0	-184.06277509
3.9	10.0	-184.06707188	4.0	10.0	-184.07551492
3.9	20.0	-184.09145596	4.0	20.0	-184.09544630
3.9	30.0	-184.10897460	4.0	30.0	-184.11196407
3.9	40.0	-184.11831674	4.0	40.0	-184.12040105
3.9	50.0	-184.12271704	4.0	50.0	-184.12449914
3.9	60.0	-184.12315848	4.0	60.0	-184.12584576
3.9	70.0	-184.12247218	4.0	70.0	-184.12590707
3.9	80.0	-184.12204962	4.0	80.0	-184.12603274
3.9	90.0	-184.12196674	4.0	90.0	-184.12603274
4.5	0.0	-184.11006228	5.0	0.0	-184.13074654
4.5	10.0	-184.11261597	5.0	10.0	-184.13152095
4.5	20.0	-184.11848472	5.0	20.0	-184.13345972
4.5	30.0	-184.12463502	5.0	30.0	-184.13578189
4.5	40.0	-184.12951329	5.0	40.0	-184.13787969
4.5	50.0	-184.13295041	5.0	50.0	-184.13947703
4.5	60.0	-184.13519448	5.0	60.0	-184.14052404
4.5	70.0	-184.13650798	5.0	70.0	-184.14111443
4.5	80.0	-184.13715848	5.0	80.0	-184.14139692
4.5	90.0	-184.13735066	5.0	90.0	-184.14147823
5.5	0.0	-184.13921956	6.0	0.0	-184.14230920
5.5	10.0	-184.13947324	6.0	10.0	-184.14238319
5.5	20.0	-184.14011998	6.0	20.0	-184.14257134
5.5	30.0	-184.14092284	6.0	30.0	-184.14279761
5.5	40.0	-184.14166544	6.0	40.0	-184.14299560
5.5	50.0	-184.14222641	6.0	50.0	-184.14313306
5.5	60.0	-184.14257214	6.0	60.0	-184.14320455
5.5	70.0	-184.14273988	6.0	70.0	-184.14322281
5.5	80.0	-184.14280563	6.0	80.0	-184.14321472
5.5	90.0	-184.14282314	6.0	90.0	-184.14321147

Table S.3: Calculated *ab initio* MR-CI energies for the  $1^1A''$  state of the  $N_2O$  molecule. The  $N_2$  distance is kept fixed at a value of  $2.13199 a_0$ . The Jacobi coordinate system used is defined in figure 1.

$R/a_0$	$\theta/deg$	$E_{MRCI}/E_h$	$R/a_0$	$\theta/deg$	$E_{MRCI}/E_h$
2.9	0.0	-183.81778605	3.0	0.0	-183.90025926
2.9	10.0	-183.88026693	3.0	10.0	-183.94386806
2.9	20.0	-183.95668413	3.0	20.0	-184.00489737
2.9	30.0	-184.02198606	3.0	30.0	-184.05422886
2.9	40.0	-184.05829132	3.0	40.0	-184.07968557
2.9	50.0	-184.07799046	3.0	50.0	-184.09209120
2.9	60.0	-184.08013928	3.0	60.0	-184.09028769
2.9	70.0	-184.06935066	3.0	70.0	-184.07917162
2.9	80.0	-184.04934843	3.0	80.0	-184.06282221
2.9	90.0	-184.03552535	3.0	90.0	-184.05275827
3.1	0.0	-183.95227375	3.2	0.0	-183.98418162
3.1	10.0	-183.98704519	3.2	10.0	-184.01396236
3.1	20.0	-184.03748538	3.2	20.0	-184.05685922
3.1	30.0	-184.07534814	3.2	30.0	-184.08660226
3.1	40.0	-184.09396911	3.2	40.0	-184.10266285
3.1	50.0	-184.10109521	3.2	50.0	-184.10674811
3.1	60.0	-184.09693001	3.2	60.0	-184.10144185
3.1	70.0	-184.08656512	3.2	70.0	-184.09251661
3.1	80.0	-184.07364923	3.2	80.0	-184.08274353
3.1	90.0	-184.06677211	3.2	90.0	-184.07867982
3.3	0.0	-184.00343385	3.4	0.0	-184.01388009
3.3	10.0	-184.02992528	3.4	10.0	-184.03976527
3.3	20.0	-184.06877723	3.4	20.0	-184.07654282
3.3	30.0	-184.09509693	3.4	30.0	-184.09980514
3.3	40.0	-184.10765242	3.4	40.0	-184.11169293
3.3	50.0	-184.11050204	3.4	50.0	-184.11234358
3.3	60.0	-184.10470205	3.4	60.0	-184.10744717
3.3	70.0	-184.09743824	3.4	70.0	-184.10199650
3.3	80.0	-184.09152160	3.4	80.0	-184.09893986
3.3	90.0	-184.08843188	3.4	90.0	-184.09688525
3.5	0.0	-184.01887117	3.7	0.0	-184.00782981
3.5	10.0	-184.04573166	3.7	10.0	-184.05550983
3.5	20.0	-184.08084977	3.7	20.0	-184.08715766
3.5	30.0	-184.10165557	3.7	30.0	-184.10634228
3.5	40.0	-184.11353769	3.7	40.0	-184.11466253
3.5	50.0	-184.11368891	3.7	50.0	-184.11511936
3.5	60.0	-184.10955754	3.7	60.0	-184.11417303
3.5	70.0	-184.10628677	3.7	70.0	-184.11490149
3.5	80.0	-184.10518719	3.7	80.0	-184.11549405
3.5	90.0	-184.10422900	3.7	90.0	-184.11583825

$R/a_0$	$\theta/\text{deg}$	$E_{MRCI}/E_h$	$R/a_0$	$\theta/\text{deg}$	$E_{MRCI}/E_h$
3.9	0.0	-184.04525421	4.0	0.0	-184.06216845
3.9	10.0	-184.06767708	4.0	10.0	-184.07635598
3.9	20.0	-184.09264364	4.0	20.0	-184.09774837
3.9	30.0	-184.10949739	4.0	30.0	-184.11300896
3.9	40.0	-184.11573018	4.0	40.0	-184.11910078
3.9	50.0	-184.11840837	4.0	50.0	-184.12165180
3.9	60.0	-184.11991718	4.0	60.0	-184.12360297
3.9	70.0	-184.12161347	4.0	70.0	-184.12557971
3.9	80.0	-184.12289490	4.0	80.0	-184.12704490
3.9	90.0	-184.12351399	4.0	90.0	-184.12764540
4.5	0.0	-184.11033903	5.0	0.0	-184.13080163
4.5	10.0	-184.11292008	5.0	10.0	-184.13156485
4.5	20.0	-184.11872081	5.0	20.0	-184.13346007
4.5	30.0	-184.12456541	5.0	30.0	-184.13569582
4.5	40.0	-184.12901710	5.0	40.0	-184.13767541
4.5	50.0	-184.13215451	5.0	50.0	-184.13917014
4.5	60.0	-184.13442622	5.0	60.0	-184.14019295
4.5	70.0	-184.13608366	5.0	70.0	-184.14085208
4.5	80.0	-184.13712448	5.0	80.0	-184.14123427
4.5	90.0	-184.13747698	5.0	90.0	-184.14136052
5.5	0.0	-184.13920556	6.0	0.0	-184.14228124
5.5	10.0	-184.13945605	6.0	10.0	-184.14235508
5.5	20.0	-184.14009657	6.0	20.0	-184.14254283
5.5	30.0	-184.14087625	6.0	30.0	-184.14276658
5.5	40.0	-184.14158051	6.0	40.0	-184.14295602
5.5	50.0	-184.14209745	6.0	50.0	-184.14307733
5.5	60.0	-184.14241557	6.0	60.0	-184.14312964
5.5	70.0	-184.14258280	6.0	70.0	-184.14313028
5.5	80.0	-184.14266412	6.0	80.0	-184.14310637
5.5	90.0	-184.14268962	6.0	90.0	-184.14309215

Table S.4: Calculated *ab initio* MR-CI electronic transition dipole moments for the  $2^1A' \leftarrow \tilde{X}^1A'$  of the  $N_2O$  molecule. The  $N_2$  distance is kept fixed at a value of  $2.13199 a_0$ . The Jacobi coordinate system used is defined in figure 1. The two non-zero parallel components are given.

$R/a_0$	$\theta/deg$	Trans. Moment Y /a.u.	Trans. Moment Z /a.u.
2.9	0.0	0.21063571	0.00000000
2.9	10.0	0.08284141	-0.36407771
2.9	20.0	-0.05889026	-0.45184803
2.9	30.0	-0.19020365	-0.46093927
2.9	40.0	-0.29131684	-0.39396986
2.9	50.0	-0.03857180	-0.09917666
2.9	60.0	0.36650959	0.24645177
2.9	70.0	0.33339020	0.20321075
2.9	80.0	0.21757403	0.18103811
2.9	90.0	0.00000000	-0.19075546
3.0	0.0	-0.15618245	0.00000000
3.0	10.0	-0.02047979	0.32343925
3.0	20.0	0.08050173	0.41048484
3.0	30.0	0.20938262	0.44481631
3.0	40.0	0.29646653	0.36970003
3.0	50.0	0.07655349	0.09981692
3.0	60.0	-0.36665051	-0.24808666
3.0	70.0	-0.31787982	-0.20420971
3.0	80.0	-0.19621522	-0.18155635
3.0	90.0	0.00000000	-0.17616206
3.1	0.0	0.00000000	-0.08429742
3.1	10.0	-0.00302044	-0.25557135
3.1	20.0	-0.10000990	-0.39023059
3.1	30.0	-0.22565828	-0.42842927
3.1	40.0	-0.29670732	-0.35242789
3.1	50.0	-0.08745012	-0.08600537
3.1	60.0	0.35582021	0.24532275
3.1	70.0	0.29050396	0.19401028
3.1	80.0	0.16911402	0.16821765
3.1	90.0	0.00000000	-0.16118953
3.2	0.0	0.00000000	-0.07071410
3.2	10.0	-0.01990493	-0.23480706
3.2	20.0	-0.12503962	-0.39436690
3.2	30.0	-0.22758759	-0.40866905
3.2	40.0	-0.29583066	-0.32128912
3.2	50.0	-0.10477753	-0.07975402
3.2	60.0	0.32903628	0.23615113
3.2	70.0	0.25715293	0.18348109
3.2	80.0	0.14441860	0.15737084
3.2	90.0	0.00000000	-0.14516387

$R/a_0$	$\theta/deg$	Trans. Moment Y /a.u.	Trans. Moment Z /a.u.
3.3	0.0	0.00000000	-0.08912964
3.3	10.0	-0.03416649	-0.22362216
3.3	20.0	-0.14483914	-0.39080282
3.3	30.0	-0.24774087	-0.41784283
3.3	40.0	-0.29047067	-0.31055423
3.3	50.0	-0.12180983	-0.07672804
3.3	60.0	0.29502424	0.21606299
3.3	70.0	0.22551199	0.16725832
3.3	80.0	0.11936679	0.14255854
3.3	90.0	0.00000000	-0.13189290
3.4	0.0	0.00000000	0.06323483
3.4	10.0	0.04903163	0.22904210
3.4	20.0	0.16172376	0.38479663
3.4	30.0	0.25908953	0.40467537
3.4	40.0	0.27702418	0.28110293
3.4	50.0	0.11664215	0.06263930
3.4	60.0	-0.22933545	-0.18463731
3.4	70.0	-0.19034217	-0.15202227
3.4	80.0	-0.10016395	-0.12629891
3.4	90.0	0.00000021	-0.11816904
3.5	0.0	0.00000000	-0.05407378
3.5	10.0	-0.06613748	-0.25360783
3.5	20.0	-0.17305637	-0.40048869
3.5	30.0	-0.26302507	-0.36900133
3.5	40.0	-0.26766823	-0.25629087
3.5	50.0	-0.11305264	-0.05185610
3.5	60.0	0.17108296	0.15222693
3.5	70.0	0.15163569	0.13397968
3.5	80.0	0.08701311	0.11527262
3.5	90.0	0.00000000	-0.10474421
3.7	0.0	-0.07441824	0.00000000
3.7	10.0	-0.11032382	-0.32447327
3.7	20.0	-0.20399009	-0.37598656
3.7	30.0	-0.24661789	-0.30952014
3.7	40.0	-0.22611248	-0.19155342
3.7	50.0	-0.10209227	-0.03106151
3.7	60.0	0.07517320	0.09731835
3.7	70.0	0.09998249	0.10812408
3.7	80.0	0.05188837	0.08906273
3.7	90.0	0.00000000	-0.08260382



$R/a_0$	$\theta/deg$	Trans. Moment Y /a.u.	Trans. Moment Z /a.u.
3.9	0.0	-0.07342178	0.00000000
3.9	10.0	-0.10663720	-0.21667116
3.9	20.0	-0.16297103	-0.26898623
3.9	30.0	-0.19526939	-0.21025137
3.9	40.0	-0.16233240	-0.11608271
3.9	50.0	-0.08060052	-0.01417316
3.9	60.0	0.01957168	0.06260086
3.9	70.0	0.05207445	0.07840919
3.9	80.0	0.03077072	0.06934711
3.9	90.0	-0.00000005	-0.06537529
4.0	0.0	-0.07726954	0.00000000
4.0	10.0	-0.10139534	-0.15246943
4.0	20.0	-0.14442024	-0.19064204
4.0	30.0	-0.15053551	-0.14289238
4.0	40.0	-0.11790414	-0.06801835
4.0	50.0	-0.05729847	0.00003311
4.0	60.0	0.00472801	0.04579369
4.0	70.0	0.02457882	0.05598705
4.0	80.0	0.01602744	0.05284929
4.0	90.0	0.00000000	-0.05104472
4.5	0.0	0.05328222	0.00000000
4.5	10.0	0.05631051	0.02543642
4.5	20.0	0.06061312	0.03527122
4.5	30.0	0.05798281	0.02738867
4.5	40.0	0.04574213	0.01025824
4.5	50.0	0.02780862	-0.00684531
4.5	60.0	0.01069148	-0.01877736
4.5	70.0	0.00023032	-0.02429693
4.5	80.0	-0.00216539	-0.02547534
4.5	90.0	0.00000000	-0.02544594
5.0	0.0	-0.02963507	0.00000000
5.0	10.0	-0.02963510	-0.00454535
5.0	20.0	-0.02888229	-0.00608654
5.0	30.0	-0.02617392	-0.00401710
5.0	40.0	-0.02132696	0.00028979
5.0	50.0	-0.01511552	0.00526583
5.0	60.0	-0.00866971	0.00975445
5.0	70.0	-0.00352785	0.01286140
5.0	80.0	-0.00082421	0.01438918
5.0	90.0	0.00000011	-0.01481572

$R/a_0$	$\theta/deg$	Trans. Moment Y /a.u.	Trans. Moment Z /a.u.
5.5	0.0	0.01615601	0.00000000
5.5	10.0	0.01594190	0.00071863
5.5	20.0	0.01523007	0.00089095
5.5	30.0	0.01391363	0.00026979
5.5	40.0	0.01199262	-0.00109855
5.5	50.0	0.00958114	-0.00300388
5.5	60.0	0.00680600	-0.00519506
5.5	70.0	0.00398295	-0.00720157
5.5	80.0	0.00169602	-0.00856537
5.5	90.0	0.00000000	-0.00910817
6.0	0.0	0.00945389	0.00000000
6.0	10.0	0.00934643	0.00013675
6.0	20.0	0.00902641	0.00019318
6.0	30.0	0.00850338	0.00004688
6.0	40.0	0.00779845	-0.00038924
6.0	50.0	0.00693720	-0.00113397
6.0	60.0	0.00587429	-0.00220674
6.0	70.0	0.00439718	-0.00360520
6.0	80.0	0.00230664	-0.00490541
6.0	90.0	0.00000002	-0.01066595

Table S.5: Calculated *ab initio* MR-CI electronic transition dipole moments for the  $1^1A'' \leftarrow \tilde{X}^1A'$  of the  $N_2O$  molecule. The  $N_2$  distance is kept fixed at a value of  $2.13199 a_0$ . The Jacobi coordinate system used is defined in figure 1. Only the non-zero perpendicular component is given.

$R/a_0$	$\theta/deg$	Trans. Moment /a.u.	$R/a_0$	$\theta/deg$	Trans. Moment /a.u.
2.9	0.0	0.00000000	3.0	0.0	0.00000000
2.9	10.0	0.13621323	3.0	10.0	0.08459552
2.9	20.0	0.08441557	3.0	20.0	0.05832867
2.9	30.0	0.03982808	3.0	30.0	0.01965571
2.9	40.0	-0.01115443	3.0	40.0	-0.02781472
2.9	50.0	-0.07827714	3.0	50.0	-0.08213484
2.9	60.0	-0.08142187	3.0	60.0	-0.08601238
2.9	70.0	-0.07462711	3.0	70.0	-0.07225951
2.9	80.0	-0.06722566	3.0	80.0	-0.06436547
2.9	90.0	-0.06518359	3.0	90.0	-0.06280756
3.1	0.0	0.00000000	3.2	0.0	0.00000000
3.1	10.0	-0.04698062	3.2	10.0	-0.02785672
3.1	20.0	-0.03514309	3.2	20.0	-0.01589033
3.1	30.0	-0.00033049	3.2	30.0	0.01580536
3.1	40.0	0.04162306	3.2	40.0	0.05311546
3.1	50.0	-0.08578251	3.2	50.0	0.09429098
3.1	60.0	0.08429792	3.2	60.0	0.08430493
3.1	70.0	0.07205229	3.2	70.0	0.06973365
3.1	80.0	0.06383949	3.2	80.0	0.06230654
3.1	90.0	-0.06164187	3.2	90.0	-0.06028346
3.3	0.0	0.00000000	3.4	0.0	0.00000000
3.3	10.0	0.01340328	3.4	10.0	0.00185308
3.3	20.0	-0.00113980	3.4	20.0	-0.01562930
3.3	30.0	-0.03062606	3.4	30.0	-0.04171279
3.3	40.0	-0.06035819	3.4	40.0	-0.07121391
3.3	50.0	-0.09393684	3.4	50.0	-0.09209406
3.3	60.0	-0.08266074	3.4	60.0	-0.08074488
3.3	70.0	-0.06802661	3.4	70.0	-0.06506690
3.3	80.0	-0.06005112	3.4	80.0	-0.05750757
3.3	90.0	-0.05819316	3.4	90.0	-0.05530846
3.5	0.0	0.00000000	3.7	0.0	0.07166278
3.5	10.0	-0.01128520	3.7	10.0	0.04677589
3.5	20.0	-0.02834357	3.7	20.0	0.05581726
3.5	30.0	-0.05182049	3.7	30.0	0.06823762
3.5	40.0	-0.07506110	3.7	40.0	0.07650592
3.5	50.0	-0.08642921	3.7	50.0	0.07249306
3.5	60.0	-0.07488427	3.7	60.0	0.06441675
3.5	70.0	-0.06148364	3.7	70.0	0.05211320
3.5	80.0	-0.05400351	3.7	80.0	0.04705495
3.5	90.0	-0.05235675	3.7	90.0	-0.04558094

$R/a_0$	$\theta/deg$	Trans. Moment /a.u.	$R/a_0$	$\theta/deg$	Trans. Moment /a.u.
3.9	0.0	-0.07056497	4.0	0.0	0.07481585
3.9	10.0	-0.06237533	4.0	10.0	0.06986718
3.9	20.0	-0.06519473	4.0	20.0	0.06623542
3.9	30.0	-0.06847572	4.0	30.0	0.06562656
3.9	40.0	-0.06683423	4.0	40.0	0.05995319
3.9	50.0	-0.06090999	4.0	50.0	0.05000385
3.9	60.0	-0.05180088	4.0	60.0	0.04163539
3.9	70.0	-0.04406563	4.0	70.0	0.03460640
3.9	80.0	-0.03991647	4.0	80.0	0.03066275
3.9	90.0	-0.03874321	4.0	90.0	-0.02945572
4.5	0.0	-0.04989768	5.0	0.0	0.02745262
4.5	10.0	-0.04845981	5.0	10.0	0.02658041
4.5	20.0	-0.04454667	5.0	20.0	0.02422064
4.5	30.0	-0.03899936	5.0	30.0	0.02104732
4.5	40.0	-0.03280996	5.0	40.0	0.01783496
4.5	50.0	-0.02709247	5.0	50.0	0.01512371
4.5	60.0	-0.02256245	5.0	60.0	0.01307949
4.5	70.0	-0.01931816	5.0	70.0	0.01160912
4.5	80.0	-0.01735693	5.0	80.0	0.01065899
4.5	90.0	-0.01670993	5.0	90.0	-0.01031850
5.5	0.0	-0.01481157	6.0	0.0	-0.00858359
5.5	10.0	-0.01440955	6.0	10.0	-0.00839802
5.5	20.0	-0.01331446	6.0	20.0	-0.00787823
5.5	30.0	-0.01182686	6.0	30.0	-0.00714739
5.5	40.0	-0.01031281	6.0	40.0	-0.00637343
5.5	50.0	-0.00903836	6.0	50.0	-0.00569427
5.5	60.0	-0.00810099	6.0	60.0	-0.00519609
5.5	70.0	-0.00745648	6.0	70.0	-0.00494929
5.5	80.0	-0.00703342	6.0	80.0	-0.00497421
5.5	90.0	-0.00687524	6.0	90.0	0.00000002