Supplementary information:

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Figure 1: The NO 4 σ orbital obtained from SA(3)-CASSCF(9,7)/aug-cc-pVTZ-333 calculation, using an iso-density values of (a) 0.025, (b) 0.030, and (c) 0.035 a.u. from left to right.

(a) (b) (c)

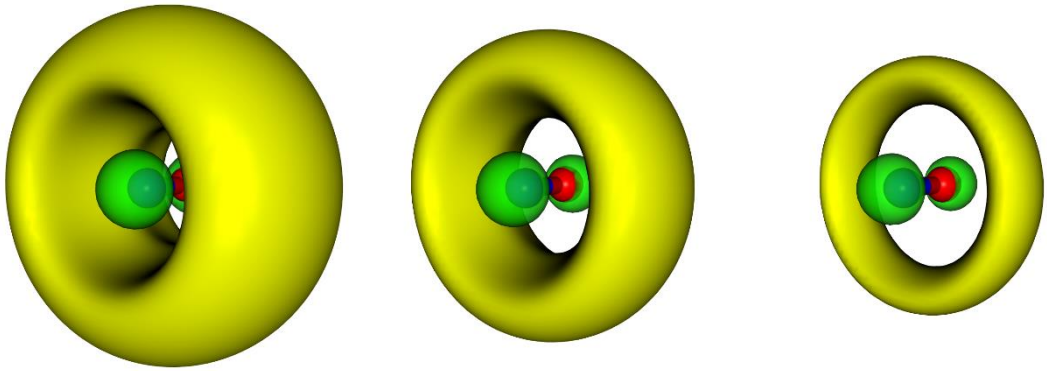

Table 1: Basis functions exponents computed in an even-tempered manner from the aug-cc-pVTZ basis set.

	Angular moment	1	2	3
nitrogen	*s*	0.01857	0.00598	0.00193
	p	0.01398	0.00398	0.00113
	d	0.04862	0.01565	0.00504
oxygen	*s*	0.02282	0.00706	0.00218
	p	0.01668	0.00466	0.00130
	d	0.07100	0.02356	0.00782

Table 2: Finite field RCCSD(T) dipole moment μ_z (Debye) and polarizability (a_0^3) of NO $X^2\Pi$ with ET-augmented basis sets. The nitric oxide molecule was aligned along the z-axis, and the isotropic polarizability $\langle\alpha\rangle$ was taken as the average of the trace values α_{xx} , α_{yy} and α_{zz}.

Method	Basis set	μ_z	α_{xx}	α_{yy}	α_{zz}	$\langle\alpha\rangle$
CCSD	aug-cc-pVTZ-100	-0.122	9.01	9.78	15.01	11.27
	aug-cc-pVTZ-200	-0.122	9.01	9.79	15.01	11.27
	aug-cc-pVTZ-300	-0.122	9.01	9.79	15.01	11.27
	aug-cc-pVTZ-110	-0.122	9.01	9.79	15.01	11.27
	aug-cc-pVTZ-210	-0.122	9.01	9.8	15.01	11.27
	aug-cc-pVTZ-310	-0.122	9.01	9.8	15.01	11.28
	aug-cc-pVTZ-220	-0.122	9.01	9.8	15.01	11.28
	aug-cc-pVTZ-320	-0.122	9.01	9.8	15.02	11.28
	aug-cc-pVTZ-330	-0.122	9.02	9.8	15.02	11.28
	aug-cc-pVTZ-111	-0.121	9.17	9.98	15.14	11.43
	aug-cc-pVTZ-211	-0.121	9.17	9.98	15.14	11.43
	aug-cc-pVTZ-311	-0.121	9.17	9.98	15.14	11.43
	aug-cc-pVTZ-221	-0.121	9.17	9.98	15.14	11.43
	aug-cc-pVTZ-222	-0.122	9.17	9.99	15.13	11.43
	aug-cc-pVTZ-321	-0.121	9.17	9.98	15.14	11.43
	aug-cc-pVTZ-331	-0.121	9.17	9.98	15.14	11.43
	aug-cc-pVTZ-322	-0.122	9.17	9.99	15.13	11.43
	aug-cc-pVTZ-332	-0.122	9.17	9.99	15.13	11.43
	aug-cc-pVTZ-333	-0.122	9.17	9.99	15.13	11.43
CCSD(T)	aug-cc-pVTZ-100	-0.167	9.12	9.97	15.04	11.38
	aug-cc-pVTZ-200	-0.167	9.12	9.97	15.04	11.38
	aug-cc-pVTZ-300	-0.167	9.12	9.97	15.04	11.38
	aug-cc-pVTZ-110	-0.166	9.13	9.97	15.05	11.38
	aug-cc-pVTZ-210	-0.166	9.13	9.98	15.05	11.38
	aug-cc-pVTZ-310	-0.166	9.13	9.98	15.05	11.39
	aug-cc-pVTZ-220	-0.166	9.13	9.98	15.05	11.39
	aug-cc-pVTZ-320	-0.166	9.13	9.99	15.05	11.39
	aug-cc-pVTZ-330	-0.166	9.13	9.99	15.05	11.39
	aug-cc-pVTZ-111	-0.165	9.29	10.18	15.19	11.55
	aug-cc-pVTZ-211	-0.165	9.29	10.18	15.19	11.55
	aug-cc-pVTZ-311	-0.165	9.29	10.18	15.19	11.56
	aug-cc-pVTZ-221	-0.165	9.29	10.18	15.19	11.55
	aug-cc-pVTZ-222	-0.166	9.30	10.18	15.17	11.55
	aug-cc-pVTZ-321	-0.165	9.29	10.18	15.19	11.55
	aug-cc-pVTZ-331	-0.165	9.29	10.18	15.19	11.55
	aug-cc-pVTZ-322	-0.166	9.30	10.19	15.17	11.55
	aug-cc-pVTZ-332	-0.166	9.30	10.19	15.17	11.55
	aug-cc-pVTZ-333	-0.166	9.30	10.19	15.17	11.55

Table 3: Finite field RCCSD(T) dipole moment μ_z (Debye) and polarizability (a_0^3) of NO $A^2\sum^+$ with ET-augmented basis sets. The nitric oxide molecule was aligned along the z-axis, and the isotropic polarizability <α> was taken as the average of the trace values α_{xx} , α_{yy} and α_{zz}.

Method	Basis set	μ_z	α_{xx}	α_{yy}	α_{zz}
CCSD	aug-cc-pVTZ-100	1.08	288.40	444.39	340.40
	aug-cc-pVTZ-200	1.12	286.96	467.67	347.19
	aug-cc-pVTZ-300	1.12	287.67	468.02	347.79
	aug-cc-pVTZ-110	1.09	609.29	444.93	554.50
	aug-cc-pVTZ-210	1.11	616.49	445.93	559.64
	aug-cc-pVTZ-310	1.11	616.67	445.36	559.57
	aug-cc-pVTZ-220	1.10	617.41	447.40	560.74
	aug-cc-pVTZ-320	1.10	616.06	447.43	559.85
	aug-cc-pVTZ-330	1.10	616.08	448.07	560.07
	aug-cc-pVTZ-111	1.08	616.85	448.81	560.84
	aug-cc-pVTZ-211	1.09	623.93	449.24	565.70
	aug-cc-pVTZ-311	1.09	623.93	448.92	565.59
	aug-cc-pVTZ-221	1.09	623.68	449.18	565.52
	aug-cc-pVTZ-222	1.09	626.29	449.87	567.48
	aug-cc-pVTZ-321	1.09	623.72	450.00	565.82
	aug-cc-pVTZ-331	1.09	623.54	449.55	565.54
	aug-cc-pVTZ-322	1.08	624.28	450.96	566.51
	aug-cc-pVTZ-332	1.09	626.20	449.93	567.44
	aug-cc-pVTZ-333	1.08	626.12	450.54	567.59
CCSD(T)	aug-cc-pVTZ-100	1.02	283.16	416.59	327.63
	aug-cc-pVTZ-200	1.05	283.13	435.34	333.87
	aug-cc-pVTZ-300	1.04	283.42	435.33	334.05
	aug-cc-pVTZ-110	1.02	575.72	416.99	522.81
	aug-cc-pVTZ-210	1.03	582.25	417.38	527.29
	aug-cc-pVTZ-310	1.03	581.55	418.66	527.25
	aug-cc-pVTZ-220	1.03	584.71	422.24	530.55
	aug-cc-pVTZ-320	1.03	580.63	418.51	526.59
	aug-cc-pVTZ-330	1.03	581.13	418.84	527.03
	aug-cc-pVTZ-111	1.00	583.46	419.12	528.68
	aug-cc-pVTZ-211	1.01	589.77	419.63	533.06
	aug-cc-pVTZ-311	1.01	590.33	419.18	533.28
	aug-cc-pVTZ-221	1.01	589.96	420.74	533.55
	aug-cc-pVTZ-222	1.01	591.59	420.56	534.58
	aug-cc-pVTZ-321	1.01	589.80	420.58	533.39
	aug-cc-pVTZ-331	1.01	589.35	420.14	532.94
	aug-cc-pVTZ-322	1.01	591.44	420.75	534.54
	aug-cc-pVTZ-332	1.01	590.76	421.17	534.23
	aug-cc-pVTZ-333	1.01	592.02	421.18	535.08