

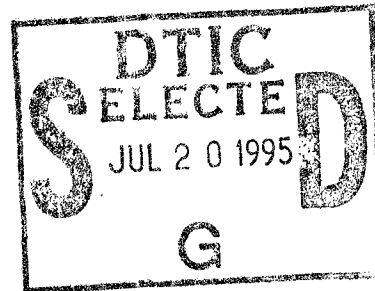
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Technical Report ARAED-TR-95008

**AB INITIO STUDY OF THE STRUCTURE, VIBRATIONAL
FREQUENCIES, AND FORCE FIELD FOR THE SYMMETRIC
FORM OF N_2O_3**

T. Vladimiroff



June 1995



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13. ABSTRACT (Maximum 200 words) In this work, the structure of symmetric N ₂ O ₃ was investigated using ab initio methods. Bond lengths and bond angles were determined using the 6-31G* and the 6-311G* basis sets using both SCF and MP2 theory. At the highest level of theory considered in this work, the N-O bond was determined to be 1.492A, the N=O bond was 1.168A, the N-O-N angle was 103.5 deg, and the O=N-O bond angle was found to be 109.9 deg. The molecule was determined to be planar at all levels of theory considered. Vibrational frequencies were computed for the symmetric structure of N ₂ O ₃ at the 6-31G*/MP2 level of theory. The calculated vibrational frequencies are in better agreement with the alternate assignments than the preferred assignments of Nour, Chen, and Laane. A new force field was derived for this molecule by starting with the theoretical values and making small adjustments in such a way as to reproduce the experimentally observed vibrational frequencies. An attempt was made to determine the relative stability of the symmetric and asymmetric forms of this molecule. Results are also compared with recent density functional calculations on these molecules.				
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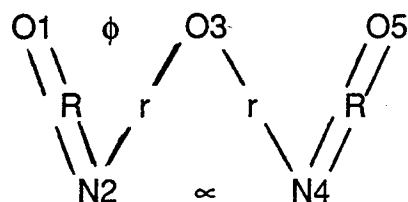
INTRODUCTION

Oxygen combines with nitrogen in order to form a variety of molecules. Some of these molecules are relatively unstable so that they can only be studied at cold temperatures using spectroscopic techniques. However, recent improvements in computational methods and digital computers have made theoretical investigations increasingly popular. Theoretical calculations can be used to supplement and elucidate some of the experimental findings. There have not been many theoretical studies of $s\text{-N}_2\text{O}_3$ reported in the literature. Some time ago Jubert, Varetti, Villar, and Castro (ref 1) performed an ab initio SCF study of this molecule using GAUSSIAN 70 in conjunction with STO-3G, 4-31G, and 6-31G basis sets. More recently, extensive calculations on the nitrogen oxides were conducted employing the density functional method (ref 2) in conjunction with a triple zeta plus polarization orbital basis set.

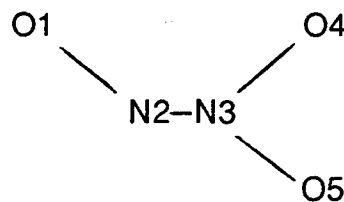
The symmetric form of N_2O_3 was first postulated by Fateley, Bent, and Crawford (ref 3) in order to explain some of their spectroscopic observation in an argon matrix. Hisatsune, Devlin and Wada (ref 4) also detected bands that could be assigned to $s\text{-N}_2\text{O}_3$. Varetti and Pimentel (ref 5) discovered that symmetric N_2O_3 could be obtained from the asymmetric form using 720 nm irradiation. Nour, Chen and Laane (ref 6) performed normal coordinate analysis on symmetric N_2O_3 , however, since all the vibrational bands could not be uniquely assigned, two different analysis were performed and two different sets of force constants were published. Even though there does not seem to be overwhelming support for the preferred assignments, the alternate assignment was completely ignored in a recent compilation (ref 7) of vibrational bands of $\text{H}_x\text{N}_y\text{O}_z$ molecules. In this work, ab initio theoretical calculations were performed which indicate that the alternate assignment may be correct.

COMPUTATIONAL DETAILS

All the computations in this study were performed with the GAUSSIAN 92 (ref 8) system of quantum chemistry programs. Standard 6-31G, 6-31G*, and 6-311G* (ref 9) basis sets were used as indicated. Electron correlation effects were introduced at the MP2 (ref 10) level of theory. Molecular geometries were determined by minimizing the total energy using gradient techniques. The force on each atom was reduced to less than 0.00045 au. The second derivative matrix was computed analytically. The two structures of N_2O_3 considered in this work are depicted in fig. 1 with the atoms numbered as in ref 1. For the force constants of symmetric N_2O_3 the notation in ref 6 was employed: the $\text{N}=\text{O}$ bond is R , the $\text{N}-\text{O}$ bond is r , the $\text{O}-\text{N}-\text{O}$ angle is ϕ , and the $\text{N}-\text{O}-\text{N}$ angle is α .



symmetric N_2O_3



asymmetric N_2O_3

Figure 1
 N_2O_3 molecules studied.

The only previous work on N_2O_3 which includes electron correlation effects was a density functional study (ref 2). In this work, three different exchange-correlation potentials were employed. At the local spin density approximation, the Dirac-Slater (ref 11) exchange term was used along with Vosko-Wilk-Nusair's (VWN) parameterization (ref 12) of the correlation energy of the homogeneous electron gas. This functional relationship was further augmented by introducing Perdew's correction (ref 13) for the correlation energy and either Becke's (B/P) (ref 14) or Perdew-Wang's (PW/P) (ref 15) corrections for the exchange term. By comparing known experimental properties with calculated values it was found (ref 2) that the VWN functional gave the best structures for the nitrogen oxides and that the B/P approach gave the best frequencies.

RESULTS AND DISCUSSIONS

The geometric parameters calculated in this work are listed in table 1, along with the results of the density functional study (ref 2). To our knowledge pure symmetric N_2O_3 has never been isolated so that an experimental geometry is not available. There is very good agreement between the VWN calculations in ref 2 and the MP2/6-311G* results. In the absence of experimental values, these have to be considered the best estimates currently available for the structure of symmetric N_2O_3 . From table 1 it is obvious that at the SCF level of theory, the computed bond lengths are too short and that going to a larger basis set only makes the situation worse. The 6-31G* basis set is not flexible enough to give a good geometry for N_2O_3 at the MP2 level of theory.

The vibrational frequencies are reported in table 2 along with the assignments of Nour, Chen, and Laane (ref 6) and the B/P calculations in ref 2. The vibrational frequencies were calculated at the MP2/6-31G* level of theory. Comparing our calculations with the work of Liu and Zhou (ref 16) on N_2O_4 and the study on HONO by Murto, Rasanen, Aspiala, and Lotta (ref 17) suggests that our calculations should not be in error by more than 200 cm^{-1} . This observation effectively rules out the preferred

assignment of Nour, et. al. (ref 6). At this level of theory it is unlikely that our calculated frequency for the antisymmetric O=N-O bend would be in error by more than 400 cm⁻¹. All of the computed frequencies are within 100 cm⁻¹ of the alternate assignments of Nour, et. al. (ref 6). Stirling, et. al. (ref 2) came to the same conclusion. It can also be pointed out that as reported in ref 5 the infrared (IR) intensity of the band at 704 cm⁻¹ is very weak while the band at 366 cm⁻¹ is strong. As can be seen from table 2, the calculations predict a weak IR band at 703 cm⁻¹ and strong fundamental at 273 cm⁻¹, which also support the alternate assignment of Nour, et. al. (ref 6).

The quadratic force field calculated at the MP2/6-31G* level of theory is reported in table 3 along with the normal coordinate analysis performed by Nour, et. al. (ref 6) and by Varetti (ref 18). The overall agreement was not very good. The calculations were biased by the truncation of the one and two electron basis set. The experimental analysis was biased by incorrect assignments, the assumption that certain cross terms are zero, the inclusion of anharmonic contributions in the experimental frequencies and the fact that the forces due to the matrix were not included in the analysis. Since the assignments were the same, the agreement should be best for the Nour et. al. (ref 6) calculation 2. Nevertheless, there was substantial disagreement. While the N=O force constant was about the same, the N-O force constant was almost a factor of two weaker and was more like the value in N₂O₅ (ref 6). The bond angle force constants are almost reversed with O=N-O bond angle being stronger than the N-O-N bond angle force constant for the theoretical analysis. The ab initio calculation also produces an N=O,O-N interaction term which was almost 10 times larger than the value obtained from experiment.

In view of these discrepancies, it was decided to perform a normal coordinate analysis starting with the theoretically computed force constants. In reference 6, a total of 28 frequencies were reported which should be sufficient to determine all of the 18 required parameters. The General Vibrational Analysis System (ref 19) of computer programs was employed as described by McIntosh and Michaelian (ref 20). The MP2/6-311G* geometry from table 1 was used. After several iterations of the SIMPLEX optimization algorithm employed by this code, the parameters in the last column of table 3 were derived. The rms value of 2.2 cm⁻¹ for 28 frequencies was obtained, which was slightly better than the value of 2.7 cm⁻¹ computed for calculation 2 in ref 6. It can be seen that there is generally good agreement between the the last two columns in table 3. Good agreement for the torsions can not be expected since they are highly anharmonic.

It can be argued that a better fit of the experimental data was received because more parameters were employed. On the other hand, ignoring some of the interaction force constants can only be justified if they are indeed small. In fact, it may be possible to obtain reasonably good fits of the experimental frequencies by ignoring some of the interaction constants and adjusting other constants to compensate for this omission. It

is believed that the force field obtained in this work is more reliable than previously obtained values (refs 6 and 18) because it is in good agreement with both the experimental frequencies and the calculated force constants using ab initio methods.

Finally, a comment on the relative stability of the symmetric and asymmetric forms of dinitrogen trioxide. The computations were performed with the 6-31G, 6-31G*, and 6-311G* basis sets using MP2 theory. The optimized geometric parameters of ONNO₂ are summarized in table 4 and compared to the experimental values (ref 21). At this level of theory, it is particularly difficult to obtain an accurate representation of the N-N bond. The literature on N₂O₄ indicated that with MP2 theory, f functions would be required on the nitrogen atoms (ref 16) in order to get an N-N distance of 1.794Å [experimental 1.782Å(22)]. Bauschlicher, Komornicki, and Roos (ref 23) obtained a value of 1.8Å using a CAS-SCF calculation. The length of the N-N bond seems to be much more accurately computed using density functional theory (ref 2). This inability to produce an accurate geometry for the asymmetric structure seriously jeopardizes the calculation of the energy differences between the two isomers of N₂O₃. Nevertheless, the energy differences were determined and are depicted in table 4. The asymmetric form was found to be more stable using MP2 theory. The MP2 results are in reasonable agreement with the values of 5.3, 5.5, and 7.7 kcal/mole computed in reference 2 using different density functionals and somewhat higher than the experimental value of 1.8 +/-0.2 kcal/mole observed by Holland and Maier (ref 24) in liquid xenon.

CONCLUSIONS

The structure of s-N₂O₃ was investigated using ab initio theoretical methods. At the highest level of theory considered (6-311G*/MP2), a reasonable geometry for this molecule was obtained. The vibrational frequencies computed in this work are within 100 cm⁻¹ of the alternate assignment proposed by Nour, Chen and Laane (ref 6), but do not support their preferred assignment. For the structure of s-N₂O₃, good agreement was obtained with the density functional calculations (ref 2); however, for the asymmetric form of this molecule the density functional method was noticeably better in predicting the length of the N-N bond than the more conventional approach. A new force field for s-N₂O₃ was obtained which is in good agreement with both the experimental and theoretical results.

Table 1
Optimized bond angles and bond lengths for symmetric N₂O₃

	<u>SCF</u>		<u>MP2</u>		
	<u>6-31G*</u>	<u>6-311G*</u>	<u>6-31G*</u>	<u>6-311G*</u>	<u>VWN^a</u>
O ₁ N ₂	1.145	1.136	1.184	1.168	1.167
N ₂ O ₃	1.379	1.377	1.502	1.492	1.487
O ₁ N ₂ O ₃	110.1	110.4	109.5	109.9	109.8
N ₂ O ₃ N ₄	107.6	108.0	102.4	103.5	103.2
O ₁ N ₂ N ₃ O ₄	180.0	180.0	180.0	180.0	180.0
energy	-333.25410	-333.34266	-334.13094	-334.28161	

NOTE: The energy is in Hartrees, the bond lengths are in Angstroms, and the bond angles are in degrees.

^aReference 2.

Table 2
Calculated and experimental frequencies for s-N₂O₃ in cm⁻¹

<u>Symbol</u>	<u>Vibrational mode</u>	<u>Experimental^a</u>		<u>Theoretical^b</u>	
		<u>Preferred</u>	<u>Alternate</u>	<u>This work^c</u>	<u>B/P^d</u>
A1	1 sym N=O str	1740	1740	1712(0.08)	1786(1)
	2 sym N-O st	973	973	980(51)	945(39)
	3 N-O-N bend	395	395	398(3.8)	362(10)
	4 sym O=N-O bend	366	275w	232(0.02)	199(0.2)
A2	5 torsion	140	140	241(0.0)	217(0.0)
B2	6 antisym N=O str	1687	1687	1664(354)	1731(744)
	7 antisym N-O str	877	704w	703(2.0)	667(16)
	8 antisym O=N-O bend	704	366s	273(1085)	326(896)
B1	9 torsion	105	105	136(0.02)	119(0.3)

^aReference 6.

^bIR intensities are given in parentheses in km/mole.

^cComputed at the MP2, 6-31G* level of theory.

^dReference 2.

Table 3
Calculated and derived force constants for s-N₂O₃^a

Force constant	Calculation 1 ^b	Calculation 2 ^c	Varetti ^d	This work	
				MP2	exp
diagonal force constants					
R	12.11	12.47	12.10	12.87	13.02
r	3.61	2.89	4.14	1.56	1.62
alpha	1.87	1.29	1.74	0.99	1.27
phi	1.61	0.80	1.51	2.03	2.03
tau	0.05	0.05		0.12	0.056
interaction force constants					
R,r	0.19	0.28	0.26	2.13	2.16
R,alpha	-0.39	-0.20	0.28	0.13	0.14
r,r'	0.36	0.79	0.95	0.72	0.55
r,alpha	0.22	0.21	0.16	0.28	0.28
r,phi	0.16	0.10	0.26	0.25	0.21
phi,phi'	-0.16	0.34	-0.29	0.16	0.14
alpha,phi	-0.33	0.02	-0.42	0.33	0.31
tau,tau	-0.01	-0.01		0.014	-0.004
R,R'			-0.01	0.30	0.27
R,r'			0.16	-0.44	-0.43
R,phi			0.12	0.61	0.49
R,phi'			0.01	0.18	0.12
r,phi'			0.19	-0.023	-0.02
RMS (28 frequencies)		2.7cm ⁻¹			2.2cm ⁻¹

^aThe N=O bond is R, the N-O bond is r, the O-N-O angle is phi, the N-O-N angle is alpha and tau is the torsion. Primes indicate the more distant interactions. Stretching constants are in mdyn/Å and bending constants and torsions are in mdyn Å/rad².

^bNormal coordinate analysis based on preferred assignment (ref 6).

^cNormal coordinate analysis based on alternate assignment (ref 6).

^dReference 18.

Table 4
Optimized bond angles and bond lengths for asymmetric N₂O₃

	<u>Experimental</u> ^a	<u>MP2</u>		
		<u>6-31G</u>	<u>6-31G*</u>	<u>6-311G*</u>
N ₂ N ₃	1.864	2.130	1.938	1.907
O ₁ N ₂	1.142	1.220	1.174	1.157
N ₃ O ₄	1.202	1.264	1.223	1.209
N ₃ O ₅	1.217	1.259	1.221	1.208
O ₁ N ₂ N ₃	105.05	95.6	100.8	102.7
N ₂ N ₃ O ₄	112.72	101.3	106.0	107.9
N ₂ N ₃ O ₅	117.47	126.9	121.8	119.8
energy (a.u.)		-333.72662	-334.14166	-334.29657
E(sym-asym) (kcal/mole)	1.8 +/- .2 ^b	7.7	6.7	9.4

NOTE: The energy is in Hartrees, the bond lengths are in Angstroms, and the bond angles are in degrees.

^aReference 21.

^bReference 24.

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