Acta Chemica Scandinavica A 33 (1979) 289 – 296

X-Ray Structure and Normal Coordinate Analysis of *p*-Nitrosoanisole

HANS JØRGEN TALBERG

Department of Chemistry, University of Oslo, Oslo 3, Norway

The crystal structure of *p*-nitrosoanisole has been determined from X-ray diffraction data collected at −155 °C and refined by least-squares methods. The space group is $P2_1/c$ with cell dimensions $a=9.341(3)$ Å, $b=9.972(3)$ Å, $c=7.365(1)$ Å and $\beta=103.07(2)°$. The final R factor was 6.5 %. The crystal structure resembles closely that of *p*-nitroanisole. It is slightly disordered in the NO group. The N – O bond length obtained from a high angle data set is nearly identical to that estimated from a linear bond length – stretching frequency relationship. A theoretical analysis of the vibrational spectrum shows that *both* the NO and the CN stretch are fairly isolated. It also indicates that the shift in ν_{NO} and ν_{CN} following *para* substitution in nitrosobenzene is mainly due to mutual conjugation. Considering a linear R_{CN}/ν_{CN} relationship, the ν_{CN} frequency corresponds to an R_{CN} value close to that estimated from an R_{CN}/R_{NO} relationship. The molecular structure seems to be consistent with substantial mutual conjugation between the substituents.

Earlier the crystal and molecular structure of several monomeric nitrosobenzenes have been investigated in this laboratory. The present study of *p*-nitrosoanisole is part of this series of investigations.[1] Owing to the disorder in the structure some of the results are questionable. In an attempt to get more information about the molecular structure a theoretical interpretation of the vibrational spectrum has been performed.

EXPERIMENTAL

Using a standard method, *p*-nitrosoanisole was derived from *p*-anisidine in good yield.[2] Sublimation by use of a cold finger gave prismatic blue-green crystals melting sharply at 21 °C. This m.p. is the same as that reported in Refs. 2 and 3 but differs considerably from the m.p. found by other authors (see Ref. 11 in Refs. 3 and 4). Exposed to air the title compound is nearly completely oxidized to *p*-nitroanisole after one hour at 40 °C. All attempts to grow other crystal forms failed (Lüttke reports preparation of a sample containing traces of a dimer of the azo dioxide type).[5] Suitable crystals were transferred quickly from a cold trap into the low temperature gas stream at the diffractometer (SYNTEX $P\bar{1}$) (room temperature of 13 °C). During the measurements the temperature at the crystal site was −155 °C. The crystal had dimensions $0.3\times0.1\times0.1$ mm. A quadrant of reciprocal space was examined up to 70° in 2θ with Mo$K\alpha$ radiation. The scan limits were $2\theta(\alpha_1)-1.0°$ and $2\theta(\alpha_2)+1.0°$. Out of 1687 reflections measured 979 were above the $2.5\sigma(I)$ level and regarded as observed. The estimated standard deviation of the intensity, $\sigma(I)$, is based on counting statistics adding 2 % for uncertainty due to experimental fluctuations. Lattice points with $k=2n+1$ for the cell $a'=a$, $b'=2b$, $c'=c$ were also scanned in order to detect super reflections. None were found within a hemisphere of reciprocal space having $2\theta_{max}=15°$. For other details regarding the data collection, refer to earlier publications in this series. The atomic scattering factors for the heavy atoms were those of Doyle and Turner,[6] and for hydrogen those of Stewart *et al.*[7] Computer programs, except for ORTEP[8] and MULTAN,[9] applied during the X-ray investigation are described in Ref. 10.

The IR spectra were obtained using a Perkin Elmer 225 instrument. The solid state spectrum was recorded with the compound deposited from vapour on to a cryostate window at liquid nitrogen temperature. The vibrational analysis was performed applying a program by Gwinn.[11]

0302-4377/79/040289-08$02.50
© 1979 Acta Chemica Scandinavica

CRYSTAL DATA

p-Nitrosoanisole, $C_7H_7O_2N$, monoclinic, space group $P2_1/c$ (No. 14). Dimensions of the unit cell at −155 °C: $a=9.341(3)$ Å, $b=9.972(3)$ Å, $c=7.365(1)$ Å, $\beta=103.07(2)°$, $V=668.3$ Å^3, $M=137.14$, $D_x=1.363$ g cm^{-3}, $Z=4$, $F(000)=288$.

STRUCTURE DETERMINATION

The structure was determined applying the MULTAN program package. Full-matrix least-squares techniques were used to refine the structure. Including all atoms and all observed reflections the anisotropic refinement converged to a conventional R factor of 0.072, a weighted R_w of 0.063 and a goodness of fit S of 2.7. The starting positional parameters for the hydrogen atoms were obtained from ΔF maps. A common temperature factor was refined for the ring and for the methyl hydrogen atoms.

Using a data set with an inner cut-off value for $\sin\theta/\lambda$ of 0.4 Å^{-1}, comprising 645 reflections, the refinement converged to $R=0.073$, $R_w=0.059$ and $S=1.6$. All C−C bonds but one are 0.010 Å longer and the C−O bonds slightly shorter in the resulting high angle structure as compared to those of the structure based on the complete data set. These differences between the two structures are similar to those found for related compounds and contrast the larger differences occurring for the NO group parameters. The N−O bond is 0.027 Å longer, the C−N bond 0.037 Å shorter and the CNO angle 5.3° wider in the high angle structure. The final bond lengths and angles in the NO group are given in Fig. 1.

A difference Fourier synthesis using the high angle parameters was then calculated. The resulting ΔF map showed one residual peak of 0.5 e Å^{-3} at the nitrogen lone pair site and another of 0.7 e Å^{-3} between C4 and O4. Two types of disordered models were tried for interpretation of this situation. In the one examined first, each NO group atom occupies two sites related by rotation about the C3C4C5 bisector. This kind of disorder has previously been found for two *p*-nitrosoanilines.[1,2] In the present case it implies that both a *syn* and an *anti* rotamer are present in the crystal. Several proportions of the two rotamers were tried, refining with isotropic NO group atoms. However, in no case were significantly better R factors obtained along with acceptable structural results. Probably the structure is not conformationally disordered.

Also the second disordered model has two fractional isotropic nitrogen and nitroso oxygen atoms. The N−N and O−O vectors are near to the long

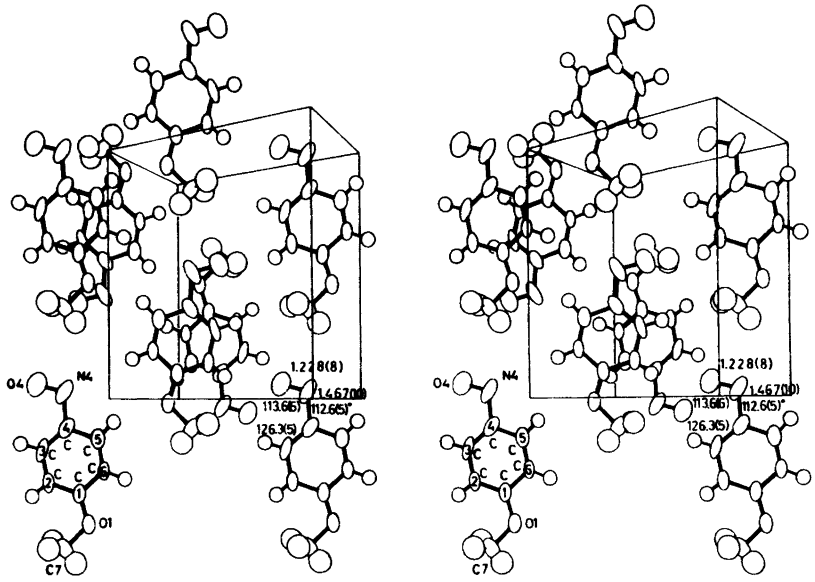

Fig. 1. A stereoscopic illustration of the structure including bond lengths (Å) and angles (°) in the nitroso group of the high angle ordered structure.

Table 1. Fractional atomic coordinates and thermal parameters from the refinement with the disordered model. For O4 and N4 also the parameters from the high angle refinement with the ordered model are given. The anisotropic temperature factors are expressed as: $\exp(-\pi^2(h^2a^{*2}U_{11}+\cdots+2klb^*c^*U_{23}))$. Estimated standard deviations in parentheses.

ATOM	x	y	z	U11	U22	U33	U12	U13	U23
O1	0.1562(2)	-0.0189(2)	0.4260(3)	0.033(1)	0.068(2)	0.040(1)	-0.002(1)	0.009(1)	-0.004(1)
C1	0.1948(3)	0.1091(4)	0.4602(4)	0.029(2)	0.067(3)	0.024(2)	-0.004(2)	0.009(1)	-0.002(2)
C2	0.3284(3)	0.1535(4)	0.5720(4)	0.020(2)	0.081(3)	0.025(2)	0.001(2)	0.005(1)	0.004(2)
C3	0.3548(4)	0.2907(4)	0.5940(4)	0.032(2)	0.097(3)	0.023(2)	-0.028(2)	0.009(1)	-0.012(2)
C4	0.2504(4)	0.3831(4)	0.5082(5)	0.044(2)	0.077(3)	0.027(2)	-0.011(2)	0.016(2)	-0.003(2)
C5	0.1161(3)	0.3368(4)	0.3972(4)	0.041(2)	0.071(3)	0.029(2)	-0.001(2)	0.012(2)	0.002(2)
C6	0.0903(3)	0.2047(4)	0.3730(4)	0.026(2)	0.065(3)	0.026(2)	0.000(2)	0.003(1)	-0.005(2)
C7	0.2578(5)	-0.1216(4)	0.5132(7)	0.048(2)	0.065(3)	0.053(2)	0.019(2)	0.017(2)	0.011(2)
O4	0.3777(5)	0.5685(6)	0.6205(6)	0.068(2)	0.103(3)	0.060(2)	-0.017(2)	0.024(2)	-0.018(2)
N4	0.2630(5)	0.5286(7)	0.5186(6)	0.052(2)	0.105(3)	0.038(2)	-0.037(2)	0.020(2)	-0.029(2)

ATOM	x	y	z	B	ATOM	x	y	z	B
H2	0.400(3)	0.103(3)	0.632(4)	3.3(3)	H3	0.443(3)	0.323(3)	0.674(4)	3.3
H5	0.049(3)	0.414(3)	0.335(4)	3.3	H6	-0.003(3)	0.168(3)	0.299(4)	3.3
H71	0.219(4)	-0.195(4)	0.476(6)	7.1(6)	H72	0.361(4)	-0.108(3)	0.471(5)	7.1
H73	0.280(4)	-0.104(4)	0.662(6)	7.1	O4A	0.3716(6)	0.5796(6)	0.6156(8)	2.8(1)
N4A	0.2710(6)	0.5137(7)	0.5244(8)	1.8(1)	O4B	0.3829(9)	0.5349(8)	0.6237(2)	7.1(3)
N4B	0.2454(8)	0.5602(7)	0.4990(11)	4.3(2)					

axis of, respectively, the N4 and O4 ellipsoid of the ordered model. Several sets of occupational factors were tried; that of equal weight (0.5) for all four fractional atoms yielding the best results. Unconstrained refinement with this set gave R factors significantly smaller than those presented above: $R=0.065$, $R_w=0.056$ and $S=2.4$. The final parameters are given in Table 1, the structural results in Fig. 2. A rigid body analysis for the methoxyphenyl part was performed using both the temperature factors from this refinement and those from the refinement with the ordered model. The analysis showed that the agreement between calculated and observed tensor elements ΔU_{rms}, is substantially better in the former than in the latter case: 0.0033 Å^2 as compared to 0.0047 Å^2, respectively.

A Fourier synthesis was calculated using the refined parameters for the ordered model. The resulting electron density map shows more egg-shaped than ellipsoidic O4 and N4 atoms, explaining the success of the second disordered model. Probably the egg-shape is due to slightly imperfect packing of the *syn* rotamers in the direction of the *b* axis.

VIBRATIONAL ANALYSIS

The IR spectrum of *p*-nitrosoanisole in the solid state is shown in Fig. 3; it is nearly identical to that for the CCl_4 solution. The main purpose of doing a normal coordinate analysis was to assign the mode which could be called the NO stretch. A tentative assignment of this mode to the band at 1508 cm^{-1} seemed somewhat dubious considering the electron donating power of the methoxy group and the extremely low-lying NO band in *p*-nitrosodimethylaniline, 1410 cm^{-1} in C_2Cl_4. The latter, which has been localized by ^{15}N substitution, is about 100 cm^{-1} from that of nitrosobenzene (1513 cm^{-1}) and *p*-fluoronitrosobenzene (1510 cm^{-1}).[13,14] In this connection it was of interest to investigate whether shifts in ν_{NO} due to *para* substitution reflect changes in the NO bonding or whether they mainly are of a mechanical nature.[15]

In order to get a reliable assignment for the *p*-nitrosoanisole modes, frequencies were calculated for the simpler parent compounds PhNO and $PhOCH_3$. A common ring force field and geometry were used.

Calculating the frequencies for *p*-nitrosoanisole, the force field and geometry were a composite of

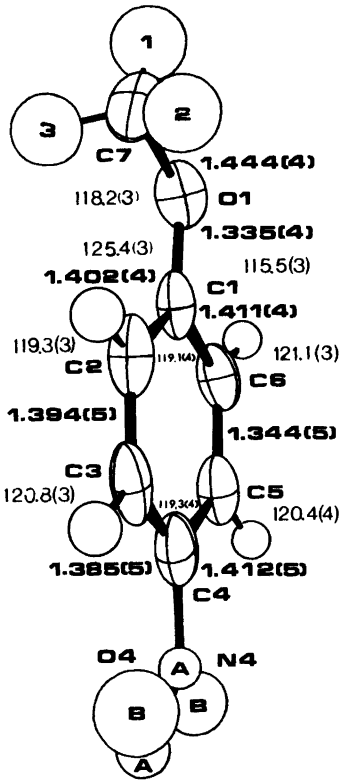

Fig. 2. 50 % probability ellipsoids, numbering of atoms and bond lengths (Å) and angles (°) in the methoxyphenyl fragment of the disordered model (using all data). The C – H bond lengths are in the range 0.83 – 1.08 Å with e.s.d.'s of 0.03 – 0.04 Å.

that of the two parent compounds. No change in the force field except for a necessary omittance of the *meta* and *para* CCH – CCH bend – bend interactions was introduced. The frequency shifts then calculated for the comparable modes upon *para* substitution are of a purely mechanical nature.

The ring geometry was that of a regular hexagon (R_{CC} = 1.397 Å, R_{CH} = 1.084 Å); other structural details being common to those found either for anisole, $PhOCH_3$, or the nitrosophenyl part of ethyl *m*-nitrosocinnamate.[16,1]

Non-planar vibrations of the ring and the CNO and COC groups, as well as torsional vibrations, were not considered.

The force field for the title compound is presented in Table 2. For the phenyl part, the 13 parameter model valence force field of Duinker and Mills for benzene was used.[17] The OCH_3 force field is symmetrical and generally used at this institute. Other stretching constants are estimated from bond length – force constant relationships.[18]

Remaining constants were guessed at and slightly adjusted in an attempt to improve the agreement between calculated and observed frequences. The calculated phenyl modes are comparable with those for benzene and the notation applied by Duinker and Mills for these has been adapted (interchanging 18 and 19, it is identical to that of Wilson).[14,17] Normal coordinate analyses have been performed previously for both the parent compounds (in combination with isotopic substitution for $PhOCH_3$). In the case of anisole all the presently calculated modes could be identified with those described by Bogatryeva *et al.*[19] Except for two

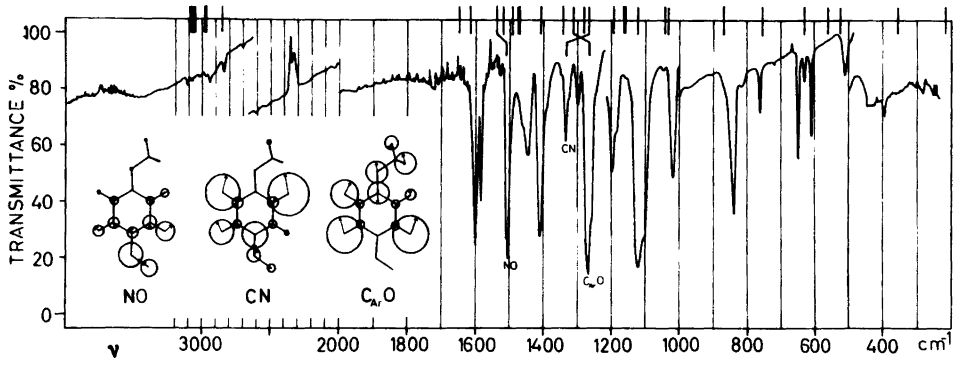

Fig. 3. The IR spectrum from 4000 to 200 cm^{-1} for *p*-nitrosoanisole in the solid state. The NO, CN and $C_{Ar}O$ normal coordinates enlarged six times are inserted. The bars crossing the upper frequency axis show the position of all (33) calculated frequencies but one (at 142 cm^{-1}).

Table 2. Valence force constants for *p*-nitrosoanisole in mdyn Å^{-1} for stretching, mdyn rad for bending-stretching and mdyn Å rad^{-2} for bending constants.

Stretch	
NO	10.5
CC	7.015
$C_{Ar}O$	5.7
CN	5.57
$C_{Ar}H$	5.125
$C_{Al}O$	4.7
$C_{Al}H$	4.7
Bend	
OCH	0.88
CCC	0.787
HCH	0.52
CCH	0.518
Other bends	0.9
Stretch-stretch interactions involving only heavy atoms	
$CC-CC_{ortho}$	0.531
$CC-CC_{meta}$	−0.531
$CC-CC_{para}$	0.531
CN−NO	0.52
CO−CO	0.50
Other neighbouring bonds	0.45
Other next-next bonds[a]	−0.2
Other "*para*" bonds[a]	0.2
Stretch-bend interactions with two atoms common	
CC−CCC	0.615
NO−CNO	0.51
$C_{Al}O-COC$	0.50
CN−CNO	0.37
CC−CCH	0.364
CO−OCH	0.36
$C_{Ar}O-COC$	0.35
Those involving exocyclic angles at C1 and C4	0.30
Stretch-bend interactions with central atom common	
CH−CCC	−0.014
Those involving the angles at C1 and C4	−0.1
Bend-bend interactions with two atoms common and terminal atoms *anti*	
CCC−CCH	0.043
Those involving the CN and the $C_{Ar}O$ bond	0.1

Table 2. Continued.

Bend-bend interactions with two atoms common and terminal atoms *syn*	
CCC−CCC	−0.098
Those involving the CN andthe $C_{Ar}O$ bond	−0.1
Bend-bend interactions with central atom in one being terminal atom in the other	
CCH−CCH	0.014

[a] Apart from those involving the OC7 bond.

modes (6a and b) their assignment has been used in the present analysis. With some modifications the previous assignment of Bradly and Straus has been applied in the case of PhNO.[20] Using the notation adapted by the authors; *18b* is shifted from the weak band at 1067 cm^{-1} to the strong band at 1112 cm^{-1}, while the combination $1+9b$ (=1066 cm^{-1}) is assigned to the former. The mode *13* (*CN* in the present notation) is assigned to a medium strong band at 1318 cm^{-1} in the C_2Cl_4 spectrum.[21] This band is missing in the gas phase spectrum.[20] Calculated and observed frequences are compared in Table 3.

The mean deviation is only slightly larger for *p*-nitrosoanisole than for PhNO and $PhOCH_3$; 29.6 cm^{-1} compared with 22.3 and 22.4 cm^{-1}. According to this analysis the aforehand assignment of the NO mode to the strong band at 1508 cm^{-1} seems to be correct.

In contrast to several of the ring modes the two interesting stretch modes *NO* and $C_{Ar}O$ are calculated to have very nearly the same frequency in *p*-nitrosoanisole and the respective parent compound. Then, probably mutual conjugation is mainly responsible for the shifts following *para* substitution. This is in close accordance with the results of Rao *et al.* in a theoretical (CNDO/2) investigation of PhCHO and some of its *para* substituted derivatives.[15]

Inspection of the normal coordinates for the NO and $C_{Ar}O$ stretching vibrations in Fig. 3 shows that the atoms of the other substituent are not involved in the vibration. An unexpected result of these calculations is the finding of a mode which could be called a *CN* stretch mode. Fig. 3 shows that the *CN* stretch is about as isolated as the two other stretches. Probably the *CN* stretch is at 1318 cm^{-1} in PhNO and at 1342 cm^{-1} in *p*-nitrosoanisole; the

Table 3. Observed and calculated frequencies (cm^{-1}) for *p*-nitrosoanisole and its two parent compounds. A tentative assignment for the most prominent bands in the spectrum of *p*-nitroanisole is included. The notation for the phenyl modes is that suggested by Wilson and used in the authoritative monography of Varsanyi.[14] Apart from those for the title compound the observed frequencies are taken from Refs. 19, 21 and 30 for $PhOCH_3$, PhNO and *p*-nitroanisole, respectively.

Normal mode type	Anisole		Nitrosobenzene		*p*-Nitrosoanisole		*p*-Nitroanisole
	Obs. (liq.)	Calc.	Obs. (C_2Cl_4)	Calc.	Obs. (CCl_4)	Calc.	Obs. (CCl_4)
NO			1513	1539	1508	1539	1505 (a.s.) 1322 (s.s.)
CN			1318	1270	1342	1267	1344
$C_{Ar}O$	1246	1310			1263	1313	1261
$C_{Al}O$	1039	1014			1030	1044	1036
CH_3 a.d.	1485	1490			1497	1493	
CH_3 a.d.	1466	1477			1460	1474	1462
CH_3 s.d.	1440	1474			1440	1473	1441
CH_3 rock	1151	1196				1161	
8a	1601	1627	1597	1625	1599	1647	
8b	1588	1593		1616	1585	1614	1586[a]
18a	1497	1511	1471	1484	1517	1520	1520
18b	1453	1441	1449	1438	1416	1409	
3	1335	1348	1312	1349	1330	1355	
14	1298	1296	1289	1299	1294	1288	1297
9a	1180	1191	1175	1183	1179	1193	1185
9b	1170	1163	1157	1160	1170	1166	1172
15	1152	1158					
19a	1076	1090	1112	1091	1113	1126	1109
19b	1022	1035	1017	1035		1035	1018
12	995	992	1000	1009	860	870	861
1	783	793	820	823	762[a]	758	754
6a	615	613		613	647	630	638
6b	553	580		599	610	562	

[a] Solid state.

increase upon substitution being due to mutual conjugation.

Bradly and Straus calculated a comparable mode at 1106 cm^{-1} in PhNO. However, six of their CNO group force constants are much smaller than the comparable constants for related molecules.[22-24]

DISCUSSION

The present crystal structure of *p*-nitrosoanisole, (I), is nearly isostructural with that found for *p*-nitroanisole, (II) ($\Delta x_{rms}=0.021$, $\Delta y_{rms}=0.019$, $\Delta z_{rms}=0.023$).[25] However, the planes containing edge-to-edge packed molecules are farther apart in (I).

The NO···ON distance is 0.385 Å longer than the ONO···ONO distance, and this difference may be of particular importance to the fact that *p*-nitrosoanisole is closer to being planar than *p*-nitroanisole. Apart from the two(three) terminal atoms both molecules have a planar heavy atom skeleton. The torsion about the C–OMe and the $C-NO(NO_2)$ bond is 1.0°(5.9°) and 2.0°(7.3°), respectively.

The structure of the methoxyphenyl fragment will be discussed with respect to the results given in Fig. 2 (the disordered model). The $C_{Ar}-O$ bond length is shorter in (I) than in (II): 1.335(4) Å as compared to 1.351(4) Å, respectively. Using e.s.d.'s being only slightly enlarged, however, one cannot say that the difference between these bond lengths is significant (note the nearly identical values for $\nu_{C_{Ar}O}$ for the two compounds; *cf.* Table 3). The

$O-CH_3$ bond length is negligibly longer in (I) than in (II). Comparing the methoxy group of (I) with those of 4,4-dimethoxy-α,β-diethylstilbene (III), one detects that this compound has $C_{Ar}-O$ and $O-CH_3$ bond lengths which are 0.039 Å longer and 0.023 Å shorter, respectively (the e.s.d.'s of the differences are 0.005 Å).[26] The two planar methoxyphenyl groups are rotated 57° out of the plane of the central fragment. Therefore, the difference between the methoxy group of (I) and those of (III) is probably due to mutual conjugation.

An INDO calculation for *p*-nitrosoanisole using an idealized planar geometry including a regular hexagonal ring has been performed. This calculation predicts the C6–C5 and the C3–C4 distance to be shorter than the C2–C3 and the C4–C5 distance, respectively. The differences are predicted to be of about the same magnitude and larger than that between C1–C2 and C1–C6. Actually all these trends are seen in the experimental results.

The N–O bond length (R_{NO}) of the high angle ordered structure, 1.228(8) Å, is nearly identical to the R_{NO} estimated from an R_{NO}/ν_{NO} relationship using $\nu_{NO}=1504$ cm^{-1} (solid *p*-nitrosoanisole), 1.231 Å. The R_{NO}/ν_{NO} relationship is linear and has a very good correlation coefficient (0.998 for 16 points) considering only nitroso compounds and oximes.[12] This correlation indicates that shifts in ν_{NO} reflect mainly valency shifts, in accordance with what has been found in the vibrational analysis.

According to an R_{NO}/R_{CN} relationship found previously for C–NO and C=NOH compounds, an R_{NO} of 1.231 Å corresponds to an R_{CN} of 1.431 Å.[27] The latter value is close to the R_{CN} estimated from a linear R_{CN}/ν_{CN} relationship (1.42 Å) based on the points (1365 cm^{-1}, 1.39 Å) and (1318 cm^{-1}, 1.45 Å) for *p*-nitrosodimethylaniline[13,28] and nitrosobenzene in C_2Cl_4 solution. The R_{CN} actually observed for the high angle ordered structure, 1.467(10), is 0.047 Å longer than the estimated R_{CN}. Although this is a comparatively large difference it is probably not significant if one doubles the e.s.d.; which seems reasonable in view of the disorder in the NO group. Therefore, R_{CN} is considered to be 1.42–1.43 Å in *p*-nitrosoanisole. It is gratifying that this is close to R_{CN} of 5-nitrososalicylic acid [1.426(2) Å] since this molecule has a hydroxyl group in *para* position to the nitroso group.[29] Further it implies that both the C–N and the $C_{Ar}-O$ bond length in (I) are shortened by about 0.03 Å due to mutual conjugation. This is in harmony with the fact that also the C–N and the $C_{Ar}-O$ bond lengths in (II) are equally shortened (about 0.02 Å).[25]

Because of the close resemblance of (I) and (II) as to the crystal structure, it was surprising to find a *syn* rotamer in the present structure. Owing to the efficient pairing of the individual molecular dipoles in the lattice and the higher dipole moment for the *anti* rotamer (3.06 D compared with 2.79 D according to INDO calculations) one would perhaps expect this form to be slightly favoured.

The author thanks ing. J. E. Gustavsen for recording the infrared spectra of the title compound.

REFERENCES

1. Talberg, H. J. *Acta Chem. Scand. A 32* (1978) 401.
2. Mijs, W. J., Hoekstra, S. E., Ulmann, R. M. and Havinga, E. *Recl. Trav. Chim. Pays-Bas 77* (1958) 746.
3. Calder, I. C. and Garrat, P. J. *Tetrahedron 25* (1969) 4023.
4. Hays, J. T. and Young, H. L. *J. Org. Chem. 32* (1967) 153.
5. Lüttke, W. *Ber. Bunsenges. Phys. Chem. 61* (1957) 340, and by *personal communication.*
6. Doyle, P. A. and Turner, P. S. *Acta Crystallogr. A 24* (1968) 390.
7. Stewart, R. F., Davidson, E. R. and Simpson, W. T. *J. Chem. Phys. 42* (1965) 3175.
8. Johnson, C. K. *ORTEP*, Report ORNL-3795, Oak Ridge National Laboratory, Oak Ridge 1965.
9. Germain, G., Main, P. and Woolfson, M. M. *Acta Crystallogr. A 27* (1971) 368.
10. Groth, P. *Acta Chem. Scand. 27* (1973) 1837.
11. Gwinn, W. D. *J. Chem. Phys. 55* (1971) 477.
12. Talberg, H. J. *Acta Chem. Scand. A 31* (1977) 743.
13. Knieriem, B. *Thesis*, Göttingen 1972.
14. Varsanyi, G. *Vibrational Spectra of Benzene Derivatives*, Academic, New York and London 1969.
15. Rao, C. N. R., Randhawa, H. S. and Reddy, N. V. R. *Spectrochim. Acta Part A 32* (1976) 685.
16. Seip, H. M. and Seip, R. *Acta Chem. Scand. 27* (1973) 4024.
17. Duinker, J. C. and Mills, I. M. *Spectrochim. Acta Part A 24* (1968) 417.
18. Stals, J. *Rev. Pure Appl. Chem. 20* (1970) 1.
19. Bogatryeva, I. K., Kotov, A. V. and Shishkina, M. V. *Acad. Sci. USSR, Bull. Div. Chem. Sci. 22* (1973) 2193.

20. Bradley, G. M. and Straus, H. L., *J. Phys. Chem. 79* (1975) 1953.
21. Lüttke, W. The spectrum is filed at the Collection of the Institute of Physical Chemistry, University of Freiburg/Breisgau.
22. Zwaric, R., Smolarek, J. and Goodman, L. *J. Mol. Spectrosc. 38* (1971) 336.
23. Heiberg, A. B. *Ab initio calc.* for CH_3NO. *Unpublished results.*
24. Dorko, E. A. and Buelov, L. *J. Chem. Phys. 62* (1975) 1869.
25. Talberg, H. J. *Acta Chem. Scand. A 32* (1978) 373.
26. Ruban, G. and Luger, P. *Acta Crystallogr. B 31* (1975) 2658.
27. Talberg, H. J. *Acta Chem. Scand. A 31* (1977) 37.
28. Rømming, C. and Talberg, H. J. *Acta Chem. Scand. 27* (1973) 2246.
29. Talberg, H. J. *Acta Chem. Scand. A 31* (1977) 485.
30. Srivastava, S. C., Srivastava, M. P. and Singh, I. S. *Indian J. Pure Appl. Phys. 8* (1970) 857.

Received November 2, 1978.