Henn, Powell & Venanzi, 1963) and bis(triphenylmethylarsonium)tetrachloronickel(II) (Pauling, 1966).

The phase diagram of the CsCl–$NiCl_2$ system has been determined (Iberson, Gut & Gruen, 1962) and it is found that below 50 mole% $NiCl_2$ there is a solid-to-solid color change from yellow to blue at 417°C caused by the formation of the compound Cs_3NiCl_5, which was shown to be isomorphous with Cs_3CoCl_5 by powder methods. Cs_3CoCl_5 is known to contain $[CoCl_4]^{2-}$ tetrahedra; therefore, Cs_3NiCl_5 presumably contains $[NiCl_4]^{2-}$ tetrahedra. We have prepared this compound in powder form and have found it to be stable at room temperature for long periods of time when stored in a desiccator. If single crystals of it can be prepared, it would probably provide a compound for an accurate structure determination of the tetrahedral $[NiCl_4]^{2-}$ ion.

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A Neutron Structure Analysis of $S_4N_4H_4$

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The hydrogen atoms in $S_4N_4H_4$ have been found to be covalently bonded to the nitrogen atoms in agreement with the inference of Sass & Donohue (*Acta Cryst.* (1958) **11**, 497] from an X-ray study. There is trigonal bonding about the nitrogen atom with coplanar SNHS groupings. The $S_4N_4H_4$ molecules are connected by N–H···N and N–H···S hydrogen bonds.

Introduction

A study of the structure of $S_4N_4H_4$ by X-ray methods was made by Sass & Donohue (1958) in an attempt to resolve conflicting chemical evidence regarding the molecular structure. Arnold (1938) and Goehring (1947) favoured a structure containing N–H bonds while Meuwsen (1929) and Sidgwick (1950) preferred a structure with S–H bonds. Measurements of the infrared and Raman spectra (Lippinscott & Tobin, 1953) showed strong absorption bands at 3220, 3285, and 3320 cm^{-1} which are characteristic of N–H bonds. Sass & Donohue located the sulphur and nitrogen atoms, and from considerations of ring angles and molecular packing, inferred that the hydrogen atoms were bound to nitrogen.

The present neutron structure analysis was carried out to obtain direct evidence for the location of the hydrogen atoms in the molecule and to study the nature of the intermolecular hydrogen bonding.

Experimental

The crystal, in the form of a rough cube of volume 0·92 mm^3, was supplied by Dr B. Dawson of the C.S.I.R.O. Division of Chemical Physics, Melbourne. The crystal data (taken from Sass & Donohue) are:

$$a=8{\cdot}010,\ b=12{\cdot}20,\ c=6{\cdot}727\ \text{Å}.$$

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The space group is *Pnma* D_{2h}^{16} (No. 62) and there are four molecules per unit cell.

The crystal was mounted on a four circle diffractometer installed on the A.A.E.C.'s reactor HIFAR at Lucas Heights, N.S.W., and the intensity data were collected by hand setting the φ and χ angles of the instrument and step scanning each reflexion in the $\theta:2\theta$ mode under monitor control. The neutron wavelength was 1·09 Å and the monochromatic beam intensity at the specimen was 4×10^6 neutrons. cm^{-2} sec^{-1}. The step size was 2′ of 2θ and the scanning rate was about one third of a degree of 2θ per minute. 328 reflexions with $\sin\theta/\lambda<0{\cdot}7$ were examined of which 146 were below the minimum observable intensity.

Analysis of data

The measured intensities were not corrected for extinction or absorption. There is no evidence in the data of extinction effects and a rough calculation shows that for a crystal of this size the maximum reduction in intensity due to absorption can only be a few per cent.

Standard deviations were assigned to the measured data through the expression $\sigma^2=\sigma_1^2+\sigma_2^2$ where σ_1 is the error due to counting statistics and $\sigma_2=\alpha+\beta P$ where P is the observed integrated intensity and α and β are constants chosen as 50·0 and 0·05 by a series of repeated measurements on several reflexions. The unobserved reflexions were allocated an intensity and a standard deviation equal to one half the intensity of the weakest observed reflection.

The data were refined by full-matrix least-squares based on F, using the programs ORFLS and ORFFE (Busing, Martin & Levy 1962*a*,*b*) on an IBM 360 computer. The variable parameters were one scale factor and the positional and anisotropic thermal parameters of each atom. The scattering lengths were taken as constants with the values given by Bacon (1962). The initial parameters were the sulphur and nitrogen parameters given by Sass & Donohue with hydrogen atoms placed at the positions they postulate.

After the first cycle it was obvious that Sass & Donohue were substantially correct, and the least-squares refinement converged to an R value of 0·137 for all reflexions (0·061 for observed reflexions only). The weighted R for all reflexions was 0·087 defined as:

$$R=[\Sigma w(|F_o|-|F_c|)^2/w|F_o'^2]^{\frac{1}{2}}.$$

The parameter shift in the final cycle was less than one third the standard deviation for all parameters.

The calculated and observed structure amplitudes are listed in Table 1 and the atomic parameters in Table 2. Of the 13 variable non-hydrogen atom positional parameters, 7 are within one standard deviation of the X-ray measurements, 10 within 2 and all within 3. The standard deviation was taken as the sum of that found by Sass & Donohue and that deduced in the present refinement.

Discussion

In the discussion the nomenclature adopted by Sass & Donohue is used. The basic molecule is designated M

Table 1. *Observed and calculated structure factors with the standard deviation of F_o* ($\times$ 100)

The unobserved reflexions are marked with an asterisk.

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 (H= 0,K=  0)    .  (H= 0,K= 12)    . 4   187  -200 29 . 2*   84  -122 42 | 5*  121    61 61 . 2   340   328 18 . 2   313   263 18 . 2   174  -170 24 .
2   503  -485 16 . 0   416   397 22 . 5*  117   -19 58 . 3   133  -129 25 |  (H= 2,K=  9)    . 3   358   350 18 . 3   353  -359 20 . 3   110   166 39 .
4  1200  1229 32 . 2   150   264 32 . 6*  121  -116 61 . 4   228   231 21 | 0*  110  -105 55 . 4   449  -444 20 . 4   375   360 19 . 4   269   301 22 .
6*  116   107 58 .  (H= 1,K=  0)    .  (H= 1,K=  7)    . 5   193   219 26 | 1   423  -423 20 . 5   465   441 21 . 6*  121    63 61 .  (H= 5,K=  1)    .
 (H= 0,K=  1)    . 1   497   486  7 . 1   605   554 20 . 6   152  -130 35 | 2   883   898 27 . 6   321   390 23 .  (H= 4,K=  2)    . 1   264   293 19 .
1*   54    15 27 . 2*   74   -56 37 . 2*  103    91 52 .  (H= 2,K=  3)    | 3   291   316 22 .  (H= 3,K=  5)    . 0   413  -375 17 . 2   184  -171 31 .
3*   87   162 44 . 3   316   318 16 . 3   548   531 20 . 0   386  -422 15 | 4*  120   -51 60 . 2   143   189 29 . 1   348   369 17 . 3*  110   140 55 .
5*  109   180 54 . 4   334   342 18 . 4*  114   -95 57 . 1   144   250 25 |  (H= 2,K= 10)    . 3   206  -212 22 . 2   128  -203 31 . 4   203   216 25 .
7*  122  -203 61 . 5   109   260 39 . 5*  119   151 59 . 2   972  -936 27 | 1*  115    15 58 . 4   209  -232 24 . 3   194  -225 24 .  (H= 5,K=  2)    .
 (H= 0,K=  2)    . 6   117   153 42 .  (H= 1,K=  8)    . 3   337  -326 17 | 2   403  -422 21 . 5*  118  -109 59 . 4   216  -204 23 . 1*  103    78 52 .
0   413  -414  6 . 7   339   366 23 . 1*  105   -51 52 . 4   269  -272 20 | 3*  119   -22 60 . 6*  122    70 61 . 5*  117    37 59 . 2   111   131 37 .
2   663   641 19 .  (H= 1,K=  1)    . 2   181  -194 26 . 5*  113    16 56 | 4*  122   -71 61 .  (H= 3,K=  6)    . 6   211  -160 28 . 3   157   171 30 .
6   446   433 21 . 1   314  -301 12 . 4*  116    35 58 . 6   482  -496 21 |  (H= 2,K= 11)    . 1   588   583 20 .  (H= 4,K=  3)    .  (H= 5,K=  3)    .
 (H= 0,K=  3)    . 2   106  -154 20 . 5*  120   -67 60 .  (H= 2,K=  4)    | 0   168   190 30 . 2   182  -177 24 . 0   531   542 19 . 1   333   393 19 .
1   157   120 14 . 4*  100   175 50 .  (H= 1,K=  9)    . 0*   83    51 42 | 1   168   219 32 . 3   615   599 22 . 1   620  -708 20 . 2   151   163 27 .
3*   92   -94 46 . 5*  109     8 55 . 1   389   413 20 . 1   177  -161 19 | 2*  120  -111 60 . 4   379   351 15 . 2   505   550 19 . 3   614   619 22 .
5   533   556 21 . 6*  117   -16 58 . 2*  112  -117 56 . 2   583   532 19 | 3   122  -130 44 . 5   238   218 26 . 3   628   602 21 . 4*  116  -160 58 .
 (H= 0,K=  4)    . 7   440  -448 22 . 3   349   300 21 . 3   317   295 18 |  (H= 2,K= 12)    .  (H= 3,K=  7)    . 4   367   400 21 .  (H= 5,K=  4)    .
0   580  -578  9 .  (H= 1,K=  2)    . 4*  119    96 59 . 4*  107    88 54 | 0   371  -346 22 . 1*  106  -108 53 .  (H= 4,K=  4)    . 1    99  -121 42 .
2   430  -645 16 . 1   227   224  9 .  (H= 1,K= 10)    . 5   433  -424 20 | 1*  121  -110 61 . 2*  109   -65 54 . 0   257   275 18 . 2*  110     8 55 .
4*  105  -136 52 . 3*   91    29 45 . 1*  114   198 57 . 6*  120    74 60 |  (H= 3,K=  0)    . 3   244  -214 24 . 1   171  -146 26 . 3   165  -167 31 .
6   608  -565 23 . 4*  101    43 51 . 2*  116    81 58 .  (H= 2,K=  5)    | 1   745  -688 22 . 4*  117    91 41 . 2*  104   -83 52 .  (H= 5,K=  5)    .
 (H= 0,K=  5)    . 5   329   349 20 . 4*  121   -82 61 . 0   388  -417 16 | 2*   89    77 45 . 5*  121   106 60 . 3   147    76 34 . 1   288   304 20 .
1*   86    14 43 . 6*  117   107 59 .  (H= 1,K= 11)    . 1   300  -292 17 | 3   907  -884 12 .  (H= 3,K=  8)    . 4   316   282 21 . 2*  112  -140 56 .
3   390   362 18 .  (H= 1,K=  3)    . 1*  117  -106 59 . 2*   96     5 48 | 4   255  -316 21 . 2   148   144 33 .  (H= 4,K=  5)    . 3*  115   -39 58 .
5   161  -224 31 . 1   404  -382 14 . 2*  119   -31 59 . 3*  103    69 51 | 5*  113  -168 57 . 3*  116   -69 58 . 0   114   201 35 .  (H= 5,K=  6)    .
 (H= 0,K=  6)    . 2*   83   -42 41 . 3*  121    68 60 . 4   378  -356 20 | 6*  119   141 60 . 4*  119  -105 60 . 1   745   729 23 . 1   231  -232 23 .
0   902  -878 15 . 3   266  -263 17 .  (H= 1,K= 12)    . 5   231  -237 26 |  (H= 3,K=  1)    .  (H= 3,K=  9)    . 2*  106   116 53 . 2*  114    97 57 .
2*   97  -119 49 . 4   183   178 24 . 1*  121   107 60 . 6*  121   -54 60 | 1*   83  -113 42 . 1*  114   -40 57 . 3   490  -492 14 . 3   169  -129 36 .
4   878  -862 26 . 5   694  -690 23 . 2*  122   -22 61 .  (H= 2,K=  6)    | 2*   90    89 45 . 2*  116    51 58 . 4*  116   122 58 .  (H= 5,K=  7)    .
6*  121  -153 61 . 6*  118   -69 59 .  (H= 2,K=  0)    . 0   368   367 17 | 3*   98    83 49 . 3*  118   125 59 .  (H= 4,K=  6)    . 1   152  -182 33 .
 (H= 0,K=  7)    .  (H= 1,K=  4)    . 0*   66   -62 33 . 1*   97    56 48 | 4   106   -92 40 .  (H= 3,K= 10)    . 0   139   195 31 . 2*  117   113 58 .
1*   98   -36 49 . 1   392  -364 15 . 1*   70    78 35 . 3   199  -186 23 | 5   210  -239 27 . 1   380  -332 21 . 1   250   278 21 .  (H= 6,K=  0)    .
3*  107    -5 54 . 2*   88  -119 44 . 2*   81    52 40 . 4   394   377 20 | 6*  119   -39 60 . 2*  119  -102 59 . 2   341   339 20 . 0   187   159 24 .
5*  118  -122 59 . 3   130   123 26 . 3*   92    70 46 . 5*  118   104 59 |  (H= 3,K=  2)    . 3*  121  -164 60 . 3*  113  -125 57 . 1*  110   114 55 .
 (H= 0,K=  8)    . 4*  105   -30 53 . 4   440  -437 18 . 6*  122    89 61 | 1   319  -335 16 .  (H= 3,K= 11)    .  (H= 4,K=  7)    . 2   689   664 23 .
0   847   849 14 . 5   566  -564 21 . 5*  111   135 55 .  (H= 2,K=  7)    | 2   315  -307 17 . 1*  120    65 60 . 0   223  -273 22 . 3*  115   -89 58 .
2   618  -654 21 . 6*  119   -30 60 . 6*  118   132 59 . 0   914   911 26 | 3    99  -152 36 . 2*  121  -102 61 . 1   359  -370 20 .  (H= 6,K=  1)    .
 (H= 0,K=  9)    .  (H= 1,K=  5)    .  (H= 2,K=  1)    . 1   432   448 19 | 4   302   265 19 .  (H= 4,K=  0)    . 2*  113  -219 56 . 2*  112  -148 56 .
1*  109    38 54 . 1*   87   110 44 . 0  1090-1127 28 . 2*  105  -169 53 | 5   457  -472 20 . 0   565  -526 19 . 3   355   354 21 . 3*  115    -9 58 .
3*  115  -144 57 . 2*   93    94 47 . 1   657  -646 19 . 3   294  -274 21 | 6   358  -351 22 . 1   664  -606 21 .  (H= 4,K=  8)    .  (H= 6,K=  2)    .
5*  122    57 61 . 3*  100   -97 50 . 2   372   371 15 . 4   696   675 23 |  (H= 3,K=  3)    . 2   430  -398 18 . 0   161   134 30 . 0   555   571 21 .
 (H= 0,K= 10)    . 4*  108  -142 54 . 3   267   274 17 . 5*  119   106 60 | 1   109  -238 29 . 3   251   265 21 . 1   391  -418 20 .  (H= 6,K=  5)    .
0*  113    95 56 . 5   326   283 21 . 4   854  -854 25 .  (H= 2,K=  8)    | 2   269  -256 17 . 4   491  -483  9 . 2   216   161 26 . 1*  115  -110 57 .
2   998   928 29 . 6*  120   168 60 . 5*  111  -169 56 . 0*  106  -221 53 | 3*  101  -103 51 . 5*  116  -167 58 .  (H= 4,K=  9)    . 2*  117   -82 58 .
4*  121    29 60 .  (H= 1,K=  6)    . 6*  118    22 59 . 1*  107    56 53 | 4*  109   130 54 . 6*  121   -20 61 . 0   190  -292 26 .  (H= 6,K=  6)    .
 (H= 0,K= 11)    . 1   318  -306 16 .  (H= 2,K=  2)    . 2*  109    26 55 | 5*  115   -47 58 .  (H= 4,K=  1)    . 1   271   277 24 . 0*  116    44 58 .
1*  117   -30 59 . 2*   98    23 49 . 0   116   227 23 . 3*  113   -47 57 |  (H= 3,K=  4)    . 0   276   244 17 .  (H= 5,K=  0)    . 1   266  -234 23 .
3*  121  -126 60 . 3   449  -459 19 . 1*   75    57 38 . 4   268  -290 27 | 1   379   390 17 . 1   557   495 19 . 1   338   353 18 .                  .
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with atom coordinates in Table 2 for atoms S(1), S(1′), N(2), H(2), H(3), H(3′) and these coordinates with a translation of **c** for atoms S(2), S(2′), N(1), N(3), N(3′), H(1). Three other molecules related to M by an a-glide perpendicular to **c**, an n-glide perpendicular to **a** and a centre of symmetry are designated A, N, I respectively. All other molecules are related to these by lattice translations such that M_{qrs} is related to M by $q\mathbf{a}+r\mathbf{b}+s\mathbf{c}$.

The $S_4N_4H_4$ *molecule*

The molecule is shown in Fig. 1 with intramolecular distances and bond angles given in Table 3. The average of the four crystallographically independent S–N bonds is 1·653 Å with all deviations within three standard deviations. The sulphur atoms lie in the plane $0{\cdot}966\,X+0{\cdot}259\,Z=0{\cdot}687$ while the best least-squares plane through the nitrogen atoms is $0{\cdot}967\,X+0{\cdot}254\,Z=1{\cdot}26$ with no atom further than 0·024 Å from it. The two planes are parallel to within 1° and are 0·573 Å apart. Only one mirror plane is required by the space group; however, the molecule has the symmetry $4mm$ in the crystal to the accuracy of the present results.

Sass & Donohue conclude from the nitrogen bond angle that the bonding about the nitrogen is trigonal and that the groupings are coplanar S–NH–S. The best least-squares planes through the three such groups are:

$0{\cdot}175\,X+0{\cdot}985\,Z=-1{\cdot}468$ through N(1) H(1) S(2) S(2′),

$-0{\cdot}724\,X+0{\cdot}691\,Z=0{\cdot}5931$ through S(1) S(1′) N(2) H(2), and

$0{\cdot}526\,X+0{\cdot}840\,Y+0{\cdot}139\,Z=1{\cdot}680$ through S(1) S(2) N(3) H(3).

In each case the nitrogen atom is furthest from the plane at 0·07, 0·04, 0·06 Å respectively. These deviations from planarity are not considered significant.

Hydrogen bonding

Sass & Donohue suggest that H(2) of molecule M is hydrogen-bonded to N(1) of molecule M_{001}, H(1) is roughly equidistant from the sulphur square of $A_{10\bar{1}}$ and H(3) is hydrogen bonded to one of S(2′) of $N_{0\bar{1}0}$, N(3) of $I_{10\bar{1}}$, S(2) of $I_{10\bar{1}}$. The hydrogen bonds found in this investigation are

(1) $N(1)M_{000}$–$H(1)M_{000}\cdots S(2)A_{10\bar{1}}$ with
N(1)–H(1) 1·08, H(1)···S(2) 2·71, N(1)···S(2) 3·65 Å
The NHS angle is 145·4°

(2) $N(2)M_{000}$–$H(2)M_{000}\cdots N(1)M_{001}$ with
N(2)–H(2) 0·96, H(2)···N(1) 2·39, N(2)···N(1) 3·18 Å.
The NHN angle is 139·1°.

(3) $N(3)M_{000}$–$H(3)M_{000}\cdots S(2')N_{0\bar{1}0}$ with
N(3)–H(3) 1·08, H(3)···S(2′) 2·73, N(3)···S(2′) 3·57 Å.
The NHS angle is 133·8°.

While the angle at the hydrogen atom is far from the ideal value of 180° the N···H and S···H contacts are significantly shorter than the van der Waals contacts of 2·7 and 3·05 Å respectively and show the existence of a weak covalent bond. Baur (1965) has discussed hydrogen bonds in crystalline hydrates and has shown that bond angles of 140° are consistent with hydrogen bonding.

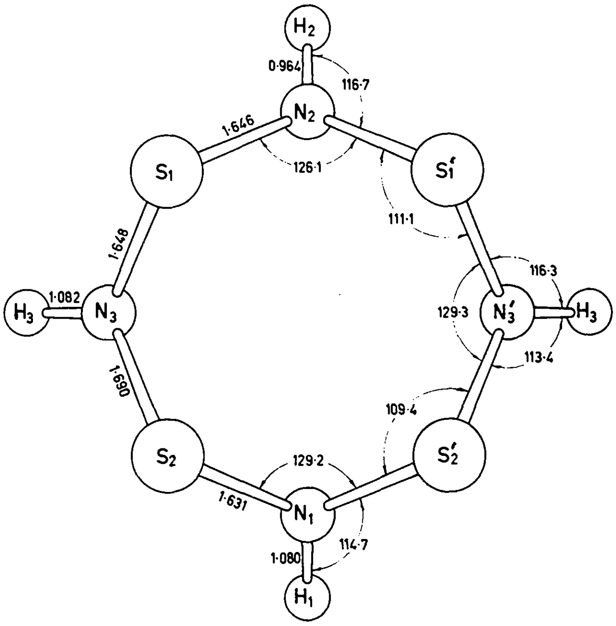

Fig. 1. The $S_4N_4H_4$ molecule.

Table 2. *Atomic parameters and their standard deviations*

Positional parameters are expressed as fractions of the lattice parameters.
Thermal parameters are in the form $\exp -[\beta_{11}h^2+\beta_{22}k^2+\beta_{33}l^2+\beta_{12}hk+\beta_{13}hl+\beta_{23}kl]$.
All parameters are given $\times 10^4$.

	x	$\sigma(x)$	y	$\sigma(y)$	z	$\sigma(z)$	β_{11}	$\sigma(\beta_{11})$	β_{22}	$\sigma(\beta_{22})$	β_{33}	$\sigma(\beta_{33})$	β_{12}	$\sigma(\beta_{12})$	β_{13}	$\sigma(\beta_{13})$	β_{23}	$\sigma(\beta_{23})$
S(1)	473	27	1297	12	1843	20	329	57	57	15	13	32	−13	21	36	29	17	17
S(2)	1447	23	1292	12	7510	21	213	41	28	11	52	32	−3	17	3	30	−4	17
N(1)	2280	18	2500	0	7196	11	161	32	36	6	110	19	0	0	28	21	0	0
N(2)	1067	15	2500	0	2698	17	128	25	65	7	151	26	0	0	−23	20	0	0
N(3)	1617	11	923	5	9923	10	177	21	47	5	122	13	32	9	−25	12	9	7
H(1)	3625	45	2500	0	7075	31	104	73	85	21	148	54	0	0	13	55	0	0
H(2)	1949	55	2500	0	3672	46	352	106	118	29	147	79	0	0	−31	71	0	0
H(3)	2772	31	501	16	259	30	230	56	96	15	369	58	121	27	−90	45	48	28

Table 3. *Intramolecular distances and angles*

Standard deviations are shown in brackets.

Distances	
S(1) -N(2)	1·646 (0·018) Å
S(1) -N(3)	1·648 (0·018)
S(2)- N(3)	1·690 (0·016)
S(2) -N(1)	1·631 (0·017)
N(1)-H(1)	1·080 (0·05)
N(2)-H(2)	0·964 (0·05)
N(3)-H(3)	1·082 (0·03)
S(1) -S(1′)	2·935 (0·03)
S(2) -S(2′)	2·947 (0·03)
S(1) -S(2)	3·017 (0·02)
N(1)-N(3)	2·711 (0·01)
N(2)-N(3)	2·717 (0·01)

Angles	
N(3)-S(1) -N(2)	111·1 (1)°
S(1) -N(3)-S(2)	129·3 (1)
N(3)-S(2) -N(1)	109·4 (1)
S(2) -N(1)-S(2′)	129·2 (1)
S(1) -N(2)-S(1′)	126·1 (1)
S(1) -S(2) -S(2′)	89·9 (0·5)
N(1)-N(3)-N(2)	89·7 (0·2)
N(3)-N(1)-N(3′)	90·4 (0·4)
N(3)-N(2)-N(3′)	90·2 (0·4)
S(1)- N(2)-H(2)	116·7 (0·8)
S(1) -N(3)-H(3)	116·3 (1)
S(2) -N(3)-H(3)	113·4 (1)
S(2) -N(1)-H(1)	114·7 (0·7)

Table 4. *R.M.S. component of thermal displacement along principal axes and directions of principal axes*

The directions of the principal axes are referred to a cartesian coordinate system so that axis 1 is the plane $y=\frac{1}{4}$ at $-20°$ to **c**, axis 2 lies in the xz plane perpendicular to 1 and axis 3 is along **b**.

	i	$(\overline{u_i^2})^{\frac{1}{2}}$	α_{i1}	α_{i2}	α_{i3}
S(1)	1	0·026	13·6	98·6	79·6
	2	0·209	99·5	83·4	11·6
	3	0·329	99·6	169·1	85·0
S(2)	1	0·108	17·6	104·0	100·4
	2	0·146	79·5	90·9	10·5
	3	0·263	104·0	165·9	88·3
N(1)	1	0·152	0·1	90·1	90·0
	2	0·165	90·0	90·0	0·0
	3	0·233	90·1	179·9	90·0
N(2)	1	0·176	45·1	135·1	90·0
	2	0·213	44·9	45·1	90·0
	3	0·222	90·0	90·0	180·0
N(3)	1	0·144	124·8	56·2	127·2
	2	0·184	138·5	92·8	48·6
	3	0·258	109·7	146·1	116·4
H(1)	1	0·174	148·7	121·3	90·0
	2	0·193	121·3	31·3	90·0
	3	0·253	90·0	90·0	180·0
H(2)	1	0·181	20·6	110·6	90·0
	2	0·299	90·0	90·0	0·0
	3	0·340	110·6	159·4	90·0
H(3)	1	0·041	100·2	43·1	131·3
	2	0·310	161·3	86·8	71·6
	3	0·366	105·5	132·9	133·0

Thermal vibrations

The r.m.s. components of the thermal displacements along the principal axes of the vibration ellipsoids, and the angles that these axes make with a cartesian coordinate system in the crystal, are given in Table 4. This cartesian system is such that axis 1 is in the plane $y=\frac{1}{4}$ at an angle of $-20°$ to **c**, axis 2 lies in the xz plane perpendicular to 1 and axis 3 is along **b**; thus axes 1 and 3 are in the plane of the molecule while 2 is normal to it. Examination of Table 4 shows that the largest component of thermal vibration for the non-hydrogen atoms is normal to the plane of the molecule. The observation by Sass & Donohue that the vibration amplitudes are least along **c** is confirmed. No explanation can be given for the very small r.m.s. vibration of S(1) and H(1) along one of their principal axes.

Re-analysis of X-ray data

An attempt was made to see whether inclusion of the hydrogen atoms would improve the agreement between the observed and calculated X-ray structure factors. An R value of 0·27 was obtained when X-ray structure factors using neutral atom scattering factors, and neutron diffraction atomic parameters, were compared with the observed structure factors of Sass & Donohue.

As soon as the atomic positions and thermal parameters were varied the refinement diverged with high negative temperature factors on the hydrogen atoms. A difference Fourier section in the plane S(1)S(1′) N(2)H(2) with the hydrogen atoms omitted from the calculated structure factors showed no evidence whatever for the presence of a hydrogen atom. This result again is in accord with Sass & Donohue's observation that the X-ray data insensitive to the hydrogen atom position.

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