

The Molecular and Crystal Structure of Cyclohexasulphur-1,3-diimide, $S_6(NH)_2$ Isomer III

By H. J. POSTMA, F. VAN BOLHUIS AND AAFJE VOS

Laboratorium voor Structuurchemie, Rijksuniversiteit Groningen, Zernikelaan Paddepoel,
Groningen, The Netherlands

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Crystals of $S_6(NH)_2$ -III have been studied by means of X-ray diffraction at room temperature. The crystals are orthorhombic with $a = 8.171 (\pm 1)$, $b = 12.815 (\pm 2)$, $c = 14.035 (\pm 2)$ Å, space group $Pbca$, $Z = 8$. The intensities of 3036 non-zero reflexions hkl were measured on a three circle Nonius diffractometer with Zr filtered Mo radiation. The structure was determined from a three-dimensional Patterson synthesis and refined by anisotropic least-squares techniques, $R = 0.043$. The molecules of $S_6(NH)_2$ -III are eight-membered rings with the nitrogen atoms in the 1,3 position, as expected from earlier spectral data. The molecules have the crown conformation also occurring in $S_6(NH)_2$ -I (nitrogen atoms in the 1,4 position) and in $S_6(NH)_2$ -II (nitrogen atoms in the 1,5 position). The molecules of $S_6(NH)_2$ -III do not show the expected mirror symmetry because of their asymmetric surroundings in the crystal. Strong $S \cdots S$ and $NH \cdots S$ interactions are present giving rise to two-dimensional layers of two molecules thick, perpendicular to the z axis. The average length of the S-S bonds, $2.055 (\pm 2)$ Å, compares nicely with the lengths observed in related molecules. The mean value of N-S, $1.672 (\pm 4)$ Å is equal to the value observed in $S_4(NH)_4$, but considerably smaller than the mean value of 1.724 Å reported for $S_6(NH)_2$ -I. The tentative assumption was made that the elongation of the N-S bonds observed for the latter compound is caused by disorder. The average values of the valence angles are $106.8 (\pm 1)$ for S-S-S, $123.9 (\pm 2)$ for S-N-S, $108.9 (\pm 1)$ for S-S-N and $110.0 (\pm 2)^\circ$ for N-S-N. The standard deviations in parentheses refer to the individual bond lengths or angles.

Introduction

In 1963 Tavs, Schulze-Steinen & Colchester (1963) and Heal (1963) showed that three compounds with formula $S_6(NH)_2$ can be obtained from a reaction mixture of S_2Cl_2 and NH_3 . The isomers are indicated as $S_6(NH)_2$ -I (m.p. $130^\circ C$), $S_6(NH)_2$ -II (m.p. $153^\circ C$) and $S_6(NH)_2$ -III (m.p. $123^\circ C$) (van de Grampel & Vos, 1969). X-ray diffraction studies of isomer I (van de Grampel & Vos, 1969) and isomer II (Weiss, 1960) have shown the NH-groups to be in the 1,4 and 1,5 positions respectively. On the basis of infrared and mass spectra of the three compounds van de Grampel & Vos (1969) concluded that $S_6(NH)_2$ -III is cyclohexasulphur-1,3-diimide rather than cyclohexasulphur-1,2-diimide. This conclusion is confirmed by the present X-ray study.

Experimental

Crystallographic data

A pure sample of $S_6(NH)_2$ -III, which according to its infrared spectrum did not contain more than 1% of isomer II, was kindly provided by Dr Heal, Department of Chemistry, Queen's University, Belfast, Ireland. Crystals were obtained from a solution in benzene by slow evaporation of the solvent. The crystals appeared to have orthorhombic symmetry. According to the systematic extinctions ($0kl$ absent for $k \neq 2n$, $h0l$ absent for $l \neq 2n$, $hk0$ absent for $h \neq 2n$), the space group $Pbca$ (D_{2h}^{15} in *International Tables for X-ray Crystallography*, 1952) was adopted. The cell dimensions were de-

termined from zero-layer line Weissenberg photographs taken with Ni filtered Cu radiation and calibrated with NaCl reflexion spots. The $\theta(K\alpha_1)$ and $\theta(K\alpha_2)$ values of 12 $0kl$ and 18 $hk0$ reflexions were measured. Least-squares adjustment of the cell dimensions to the $\sin^2 \theta$ values gave: $a = 8.171 (\pm 1)$, $b = 12.815 (\pm 2)$, $c = 14.035 (\pm 2)$ Å. $\lambda(Cu K\alpha_1) = 1.54051$, $\lambda(Cu K\alpha_2) = 1.54433$ Å, $a(NaCl) = 5.64006$ Å at $21^\circ C$. For $Z = 8$ the calculated density, 2.010 g.cm^{-3} , shows good agreement with the experimental value of 2.02 g.cm^{-3} , obtained by Tavs (1962).

Structure factors

The intensities of the reflexions hkl were measured at room temperature on an automatic Nonius three-circle diffractometer (Zr filtered Mo radiation, θ - 2θ scan method). A crystal measuring $0.40 \times 0.45 \times 0.44$ mm was used. The intensities of all independent reflexions with $2\theta < 70^\circ$ were collected. For 3036 reflexions, out of 3234 reflexions measured, I_{net} appeared to be larger than zero. Corrections for the Lorentz and polarization effects and for absorption were applied, $\mu(Mo) = 16.8 \text{ cm}^{-1}$. The latter corrections were calculated with a TR4 program written according to the Busing & Levy (1957) scheme; 14 bounding planes were taken into account, the location of which was determined as accurately as possible. The crystal was divided in $8 \times 8 \times 8$ volume fragments.

Individual weights $w_c(F)$ were calculated from the relation $w_c = 1/\sigma_c^2$, where σ_c is the standard deviation in the F -values due to counting statistics.

Structure determination and refinement

A rough model of the structure could be obtained from a three-dimensional Patterson map. It was assumed that the conformation of the molecules is analogous to the conformation of the molecules in S_8 (Caron & Donohue, 1965), in $S_6(NH)_2$ -I and in $S_6(NH)_2$ -II. The peaks around the origin of the Patterson map and the peaks in the Harker sections especially appeared to be of great help in the determination of the orientation and location of the molecules in the unit cell. When the positions of the heavy atoms had been found from the Patterson map, a [100] Fourier projection showed the nitrogen atoms to be in the 1,3 position. The structure was refined on a TR4 computer by use of a least-squares program working in block-diagonal approximation (Cruickshank & Pilling, 1961). An $[F_o - F_c(S, N)]$ -synthesis showed the hydrogen atoms to be in the S-N-S planes and the NH bonds were given a length of 0.99 Å (Worsham, Levy & Peterson, 1957) and were assumed to bisect the respective S-N-S angles. During the final cycles of the refinement the hydrogen atoms were kept fixed at positions obeying this geometry. Their temperature factors were assumed to be isotropic and equal to $\exp(-3.5 \sin^2 \theta / \lambda^2)$. For the nitrogen and sulphur atoms anisotropic temperature factors were used. The scattering factors were taken from Moore (1963) for nitrogen and sulphur and from Stewart, Davidson & Simpson (1965) for hydrogen. In the last few cycles corrections for extinction according to Zachariasen (1968) were made. For the strongest reflexion, 212, the correction amounted to 30% in F . The weights of the reflexions were calculated from the relation $w = (w_c^{-1} + p|F_o|^2)^{-1}$, with $p = 0.57 \times 10^{-4}$. At the end of the

refinement $\langle w|\Delta F|^2 \rangle$ hardly varied with $|F|$. The residual $R = (\sum |\Delta F|^2 / \sum |F_o|^2)^{1/2}$ decreased to 0.043. The final coordinates and thermal parameters are listed in Tables 1 and 2. The standard deviation as calculated by the least-squares program appeared to be approximately equal for all sulphur (nitrogen) atoms and nearly independent of direction. The average value is 0.0006 Å for sulphur and 0.0018 Å for nitrogen. These standard deviations give a lower estimate of the errors in the atomic positions, however, as errors will also occur due to uncertainties in the weighting scheme and in the positions of the hydrogen atoms. From the results of some least-squares refinements in which reasonable variations in the weighting scheme and in the hydrogen positions were adopted, root-mean-square shifts of 0.0009 and 0.0028 Å for sulphur and nitrogen respectively were calculated. By considering these values as standard deviations the total error in the atomic positions is represented by a standard deviation of 0.0011 Å for sulphur and of 0.0033 Å for nitrogen. The observed and calculated intensities are compared in Table 3.

Table 1. *Atomic coordinates*

For numbering of atoms, see Figs. 2 and 3.

	x	y	z
N(1)	0.0443	0.1000	0.3619
N(2)	0.1773	0.2350	0.4914
S(1)	0.06897	0.12636	0.47803
S(2)	0.09218	0.35158	0.51247
S(3)	0.09843	0.43739	0.38908
S(4)	-0.10617	0.40101	0.31002
S(5)	-0.03847	0.27686	0.22523
S(6)	-0.11030	0.14436	0.29481
H(1)	0.13	0.06	0.33
H(2)	0.30	0.23	0.50

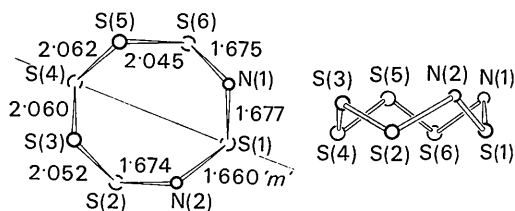


Fig. 1. Two perpendicular views of a molecule of $S_6(NH)_2$ -III, with bond lengths. The pseudo mirror plane of the molecule is given by 'm'.

Thermal motion

Analysis of the anisotropic thermal motion of the sulphur and nitrogen atoms (Cruickshank, 1956) with variable coordinates for the point of intersection of the libration axes (Pawley, 1963) showed that the molecules do not vibrate like rigid bodies. The root-mean-square difference between the observed thermal parameters U_{ii} for the sulphur atoms and the corresponding parameters based on the rigid body model is 0.0014 Å², which is approximately 6 times larger than the

Table 2. *Thermal parameters of the temperature factor*

$$\exp\{-2\pi^2(U_{11}h^2a^{*2} + U_{22}k^2b^{*2} + U_{33}l^2c^{*2} + 2U_{12}hka^*b^* + 2U_{23}klb^*c^* + 2U_{13}hla^*c^*)\}$$

Standard deviations as calculated by the least squares program are given in parentheses. All values have been multiplied by 10⁴.

	U_{11}	U_{22}	U_{33}	$2U_{12}$	$2U_{23}$	$2U_{13}$
N(1)	491 (10)	356 (9)	351 (9)	229 (16)	163 (14)	11 (16)
N(2)	293 (8)	354 (9)	414 (9)	71 (13)	-4 (14)	138 (14)
S(1)	446 (3)	280 (2)	325 (2)	37 (4)	-91 (4)	-1 (4)
S(2)	350 (2)	298 (2)	286 (2)	14 (4)	18 (4)	77 (4)
S(3)	377 (3)	303 (2)	355 (2)	-117 (4)	-88 (4)	-8 (4)
S(4)	387 (3)	317 (2)	356 (3)	107 (4)	-91 (4)	99 (4)
S(5)	482 (3)	441 (3)	253 (2)	20 (5)	1 (4)	-26 (4)
S(6)	427 (3)	339 (2)	371 (3)	-65 (4)	105 (4)	115 (5)

Table 3 (cont.)

Table 3 (cont.) - A large table of numerical data, likely representing bond lengths and angles for various molecules, organized in columns and rows with various labels and sub-headers.

the bond lengths and angles are listed in Table 4. The standard deviations calculated from the standard deviations in the coordinates are 0.004 for N-S, 0.002 Å for S-S, 0.2 for N-S-N, 0.2 for S-N-S, 0.1 for N-S-S and 0.1° for S-S-S. Fig. 1 shows that the molecules of

S₆(NH)₂-III have the crown conformation, earlier observed in S₈, S₆(NH)₂-I and in S₆(NH)₂-II. In S₆(NH)₂-III the NH-groups are in the 1,3 position. The atoms are arranged in two approximately parallel planes (dihedral angle 6.16°). If the 'best' planes through the respective atoms are subject to the condition that they must be parallel, the distance between the planes appears to be 0.925 Å. Fig. 1 and Table 4 show that the mirror symmetry, which one expects for a crown shaped S₆(NH)₂ molecule with the NH-groups in the 1,3 position, is only roughly present in S₆(NH)₂-III. The differences between the corresponding dihedral angles are especially large and Table 5 shows that there are also significant differences between corresponding bond lengths and corresponding valence angles. In the following section we shall see that this lack of symmetry is presumably due to the asymmetry in the surroundings of the molecule.

The four N-S bonds in the molecule average to 1.672 Å. The S-S bonds S(2)-S(3) and S(5)-S(6) next to the N-S bonds, are 2.048 Å on the average, whereas the remaining S-S bonds average to 2.061 Å. The slight decrease in length of the S-S bonds when moving to N-S, may be due to an increase of the double bond character because of a polarization of the sulphur *d*-orbitals by the nitrogen atoms (Hordvik, 1966; Craig & Zauli, 1962). For the comparison with the bond

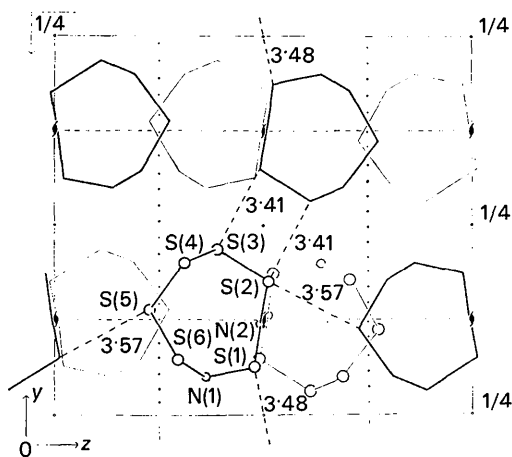


Fig. 2. The crystal structure of S₆(NH)₂-III in [100] projection. Dashed lines represent short intermolecular distances. The (positive) x axis points downwards. The 'thick' molecules lie around x=0 and the 'thin' ones around x=½.

Table 4. Molecular geometry

Bond lengths		Valence angles		Dihedral angles	
S(6)-N(1)	1.675 Å	N(1)-S(1)-N(2)	110.0°	S(6)N(1)S(1) / N(1)S(1)N(2)	89.0°
N(1)-S(1)	1.677	S(1)-N(2)-S(2)	123.2°	N(1)S(1)N(2) / S(1)N(2)S(2)	-98.4
S(1)-N(2)	1.660	S(6)-N(1)-S(1)	124.7°	S(5)S(6)N(1) / S(6)N(1)S(1)	-95.4
N(2)-S(2)	1.674	N(2)-S(2)-S(3)	108.6°	S(1)N(2)S(2) / N(2)S(2)S(3)	104.1
S(2)-S(3)	2.052	S(5)-S(6)-N(1)	109.5°	S(4)S(5)S(6) / S(5)S(6)N(1)	92.8
S(3)-S(4)	2.060	S(2)-S(3)-S(4)	108.3°	N(2)S(2)S(3) / S(2)S(3)S(4)	-86.2
S(4)-S(5)	2.062	S(3)-S(4)-S(5)	105.5°	S(3)S(4)S(5) / S(4)S(5)S(6)	-95.4
S(5)-S(6)	2.045	S(4)-S(5)-S(6)	106.7°	S(2)S(3)S(4) / S(3)S(4)S(5)	89.8

lengths in related molecules the average length of the S-S bonds (2.055 Å) will be used.

The packing of the molecules in $S_6(NH)_2$ -III

The structure in [100] and [010] projections is shown in Fig. 2 and Fig. 3. In Fig. 2 all intermolecular distances which are more than 0.1 Å shorter than the sum of the van der Waals radii (1.85 Å for S, 1.50 Å for N and 1.20 Å for H; Pauling, 1960) are given by dashed lines, except those involving hydrogen atoms. It is seen that especially around the inversion centres at $(0, \frac{1}{2}, \frac{1}{2})$ and $(0, 0, \frac{1}{2})$ short distances occur. Moreover, Fig. 4 shows that short N-H...S distances are present, approximately lying in the plane $z = \frac{1}{2}$. Due to these interactions, the molecules bordering upon the plane $z = \frac{1}{2}$ form a two-dimensional network, having a thickness of two molecules in the z direction. The interaction between successive layers is small, only one short independent intermolecular distance, *viz.* S(2)...S(5''); $x, \frac{1}{2} - y, \frac{1}{2} + z = 3.57$ Å, is observed between the layers. The interaction around the centre at $(0, \frac{1}{2}, \frac{1}{2})$ gives rise to the conformation shown in Fig. 5(a). From Figs. 5(a) and 4 we see that S(2) is surrounded by 5 atoms at short distances. Around the centre at $(0, 0, \frac{1}{2})$ we find

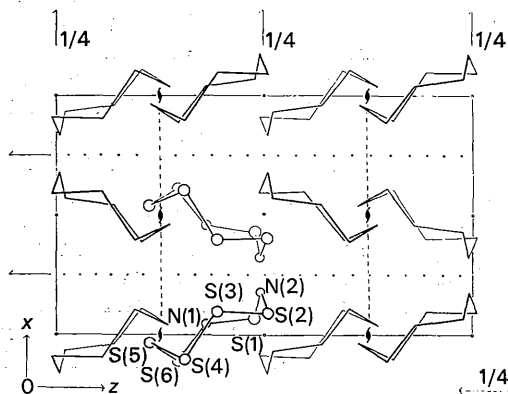


Fig. 3. The crystal structure of $S_6(NH)_2$ -III in [010] projection. The (positive) y axis points upwards. The molecules lying around $y = \frac{1}{4}$ (thick) and $y = -\frac{1}{4}$ (thin) are given.

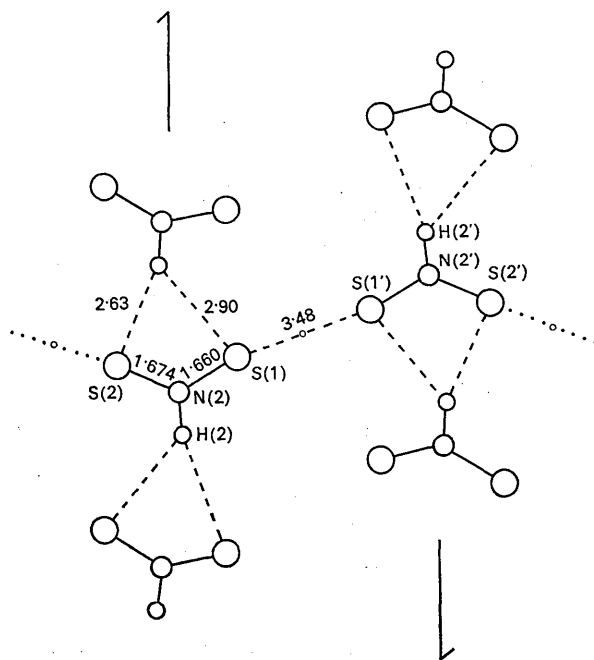


Fig. 4. Short S...H distances, approximately lying in the plane $z = \frac{1}{2}$. The dotted line connects S(2; x, y, z) with its symmetrically related atom S(2'; $-x, 1 - y, 1 - z$). See also Fig. 5(a).

the configuration shown in Fig. 5(b). A similar interaction across an inversion centre appears to occur in $S_6(NH)_2$ -I [Fig. 5(c)].

By comparison of Figs. 4 and 5 with Fig. 1 (left hand side) we see that the lower part of the molecule (atoms S(1) to S(3)) has a strong interaction with its neighbours, whereas for the upper part of the molecule the intermolecular interaction is small. In view of this difference between the surroundings of the pseudo-symmetrically related parts of the molecule, it seems reasonable to assume that the deviations from the mirror symmetry are caused by intermolecular interaction. No quantitative explanation has been found yet for the observed deviations. In contradistinction to $S_6(NH)_2$ -III, the molecules in $S_6(NH)_2$ -I and in $S_6(NH)_2$ -II have

Table 5. Comparison of bond lengths or valence angles, related by the pseudo-mirror plane in the $S_6(NH)_2$ -III molecule

The σ values refer to the considered differences Δ .			
Bonds / Angles	Δ	Δ/σ	Conclusion
S(4)–S(3) ; S(4)–S(5)	0.002 ₄ Å	1.0	not significant
S(3)–S(2) ; S(5)–S(6)	0.007 ₂	3.3	significant
S(2)–N(2) ; S(6)–N(1)	0.000 ₄	0.1	not significant*
N(2)–S(1) ; N(1)–S(1)	0.016 ₃	3.3	significant*
S(4)–S(3)–S(2) ; S(4)–S(5)–S(6)	1.51°	21.5	significant
S(3)–S(2)–N(2) ; S(5)–S(6)–N(1)	0.9 ₀	7.0	significant
S(2)–N(2)–S(1) ; S(6)–N(1)–S(1)	1.5 ₄	5.7	significant

* The N–S bonds can be compared in another way. The bonds S(2)–N(2) and N(2)–S(1) below 'm' in Fig. 1 can be assumed to be equal, as their difference Δ is only possibly significant [$2 \times \sigma(\Delta)$]; for the calculation of $\sigma(\Delta)$, see Cruickshank & Robertson, (1953)]. Also S(6)–N(1) and N(1)–S(1) lying above 'm' in Fig. 1 appear to have equal lengths. The two sets of N–S bonds are not equal, however, the difference between the respective average values being 4 times the standard deviation.

retained the expected symmetry in the crystalline state (presence of a twofold axis or a mirror plane). This is due to the fact that in these compounds the symmetry of the surroundings matches the symmetry of the molecules, because of the location of the molecules on special positions in the crystal.

Comparison with related compounds

Shape of the molecules, valence angles

In all $S_{8-x}(NH)_x$ compounds with known structure the molecules have the crown conformation (see Fig. 1, for instance). For each of the molecules we have calculated the 'best' planes for both the upper and lower four non-hydrogen atoms. Moreover, these planes were determined subject to the condition that they must be parallel. The distance between these parallel planes was considered as the 'best' height of the molecule. The sulphur atoms were given a weight 9 and the nitrogen atoms a weight 1, according to the relation $w(S):w(N)=\sigma^{-2}(S):\sigma^{-2}(N)\simeq 9:1$.

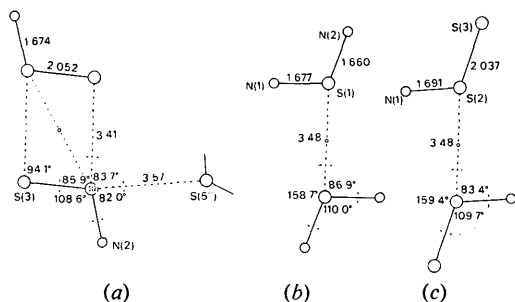


Fig. 5. Strong $S \cdots S$ interactions. (a) In $S_6(NH)_2$ -III around the centre at $(0, \frac{1}{2}, \frac{1}{2})$. The atoms are projected onto the plane $0.888X + 0.376Y + 0.265Z = 4.268$ Å through S(2), S(3) and the inversion centre. Atomic distances to the plane: N(2): -0.023 , S(5 *): -0.776 Å. (b) In $S_6(NH)_2$ -III around the centre at $(0, 0, \frac{1}{2})$. The atoms are projected onto the best plane through the considered atoms [$w(S):w(N)=9:1$]. Equation of the plane: $0.921X - 0.374Y - 0.110Z = -0.774$ Å. Atomic distances to the plane: N(1): 0.067 , S(1): -0.053 , N(2): 0.220 Å. (c) In $S_6(NH)_2$ -I.

The dihedral angles between the best planes and the best heights are listed in Table 6(a). The molecules $S_6(NH)_2$ -II are not considered as their structure is known only approximately. The Table shows that the best height is largest for S_8 and smallest for $S_4(NH)_4$. This is understandable from the fact that in S_8 both planes consist of sulphur atoms, whereas in $S_4(NH)_4$ one of the planes is composed out of relatively small nitrogen atoms. As to the dihedral angles, it is noteworthy that the angle in $S_6(NH)_2$ -III is by far the largest. This may be understood from Fig. 1, which shows that in $S_6(NH)_2$ -III both nitrogen atoms are at the right hand side of the molecule. Due to this the planes through the upper and lower atoms are not parallel, but relatively close to each other at the right hand side of the molecule. In $S_6(NH)_2$ -I where the nitrogen atoms are further apart than in $S_6(NH)_2$ -III, the heights and the dihedral angles are approximately equal to the corresponding values in S_8 .

The valence angles in the compounds $S_{8-x}(NH)_x$ are listed in Table 6(b). The large spread in the angles observed for each of the compounds, makes it difficult to detect systematic differences between the different compounds. We see that the angles S-S-S and S-S-N in $S_6(NH)_2$ -III tend to be relatively small, whereas S-N-S and N-S-N are relatively large. It is not known yet whether the variations in the angles are mainly due to intra- or to inter-molecular forces.

Bond lengths

The average value of the S-S bond lengths in $S_6(NH)_2$ -III, $2.055 (\pm 7)$ Å, compares nicely with the average values obtained for S_8 , $2.047 (\pm 3)$ Å, and for $S_6(NH)_2$ -I, $2.048 (\pm 9)$ Å. The values in parentheses are root-mean-square deviations from the respective average values. The quoted bond lengths are not corrected for libration effects; for S_8 this correction has been estimated to be as high as 0.013 Å (Caron & Donohue, 1965).

The mean value of the N-S bond lengths in $S_6(NH)_2$ -III, $1.672 (\pm 7)$ Å, is equal to the mean N-S bond length

Table 6. Conformation of different $S_{8-x}(NH)_x$ molecules

(a) Dihedral angles and 'best' heights

Compound	Dihedral angle	'Best' height
S_8	0.36°	0.986 Å
$S_4(NH)_4$	0.44	0.667
$S_6(NH)_2$ -I mol I*	0.12	0.974
$S_6(NH)_2$ -I mol II*	0.50	0.969
$S_6(NH)_2$ -III	6.16	0.925

* Two independent molecules per cell

(b) Valence angles. Average values are given with, in parentheses, the r.m.s. deviations observed for each of the compounds.

Compound	S-S-S	S-S-N	S-N-S	N-S-N
S_8	$108.1 (\pm 8)$			
$S_4(NH)_4$			$122.2 (\pm 12)$	$108.4 (\pm 6)$
$S_6(NH)_2$ -I	$107.3 (\pm 1)$	$110.1 (\pm 11)$	$119.1 (\pm 14)$	
$S_6(NH)_2$ -III	$106.8 (\pm 14)$	$108.9 (\pm 9)$	$123.9 (\pm 11)$	110.0^\dagger

† One observed value

of $1.675 (\pm 5) \text{ \AA}$ in $S_4(NH)_4$ (Sass & Donohue, 1958). In contradistinction to this most of the observed N-S bond lengths in $S_6(NH)_2$ -I, given in Fig. 6, are considerably longer. We have not been able to give a theoretical explanation for an elongation of the N-S bonds in $S_6(NH)_2$ -I. By looking at the crystal structure of $S_6(NH)_2$ -I, we think it reasonable to assume, however, that the observed elongation is not genuine, but due to disorder. This assumption is based on the following facts: (1) In $S_6(NH)_2$ -I the intermolecular interaction is much smaller than in $S_6(NH)_2$ -III; (2) No essential changes will occur in the packing by, for instance, a rotation of the $S_6(NH)_2$ -I molecules around the pseudo-twofold or pseudo-fourfold axes given in Fig. 6; (3) The assumed disorder will have a relatively small effect on the S-S bond lengths, as the maximum of the electron density for sulphur is approximately three times as high as that for nitrogen. For N-S the influence of the disorder is considerable. When replacing a fraction x of the S-N bonds by S-S in such a way that the directions and the sulphur atoms S of $S \rightarrow N$ and $S \rightarrow S$ coincide, an elongation of $3x[r(S-S) - r(S-N)] / (1 + 2x)$ is obtained for S-N (displacement of centre of gravity in electron density). If the sulphur atoms do not coincide smaller elongations are expected. In view of the fact that the elongation of the N-S bonds in $S_6(NH)_2$ -I is 0.052 \AA on the average in comparison with $S_6(NH)_2$ -III, we estimate that approximately 10% of the N-S bonds are replaced by S-S and *vice versa*. It is necessary, of course, to check this hypothesis. This can be done by studying crystals of $S_6(NH)_2$ -I by neutron diffraction. Moreover it would be interesting to compare the N-S bond lengths in $S_6(NH)_2$ -I and in $S_6(NH)_2$ -III with those in $S_6(NH)_2$ -II. As the structure of $S_6(NH)_2$ -II as determined by Weiss is too inaccurate for this purpose, a refinement of this compound is in progress.

We are grateful to Dr J. C. van de Grampel for valuable discussions and for his interest during the course of the investigation. The calculations were done at the Computing Centre of the University of Groningen.

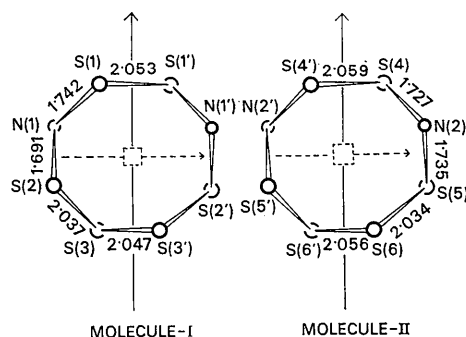


Fig. 6. Reported values of the bond lengths in the two independent molecules of $S_6(NH)_2$ -I. Each molecule has a two-fold axis. Pseudo-twofold and pseudo-fourfold axes are indicated by dashed lines.

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