lumpiness in the early diffraction lines and intensity aberrations would result.

A further possibility is the interference of oxide lines. In Thewlis's pattern of β-uranium from the pure metal one can find nearly every line of the UO_2 and UO patterns and these patterns are about as intense as the β-uranium pattern. While the number of cases in which there is interference is small there are such cases. For example, the β-phase (202) reflection and the UO (200) reflection are very close together. The difference in oxidation resistance for the pure metal and the chromium alloy reported by Wilson & Rundle (1948) could produce intensity differences in the case of interfering oxide lines.

The point previously made (Tucker, 1952*a*), but not discussed by Thewlis, is worth repeating, namely, that decreasing the chromium content from 4 to 0·3 atomic% produces no significant change in the intensities of the β-phase reflections. If the β-phase structure in the low chromium alloys were really different from that in the pure metal, then there must be some transition region for going from one structure to the other. It hardly seems reasonable that less than 0·3 atomic% chromium would have any important effect on the crystal structure.

In view of the preceding discussion in this section and the numerous difficulties of working out a crystal structure as complex as that of β-uranium from powder patterns, we are led to retain our previous view that the crystal structures of β-uranium in the low chromium alloys and in the pure metal are identical.

It is a pleasure to acknowledge the interest and encouragement of Drs J. E. Burke, J. P. Howe and J. R. Low in this work.

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On the Crystal and Molecular Structure of N_2O_2

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(*Received* 30 *March* 1953)

The crystal structure of nitric oxide dimer is monoclinic, $P2_1/a$ with two N_2O_2 molecules in a unit cell of dimensions, $a = 6{\cdot}68$, $b = 3{\cdot}96$, $c = 6{\cdot}55$ Å and $\beta = 127{\cdot}9°$. Each dimer has two possible orientations, thus accounting for the observed residual entropy of 1·5 e.u. per mole of dimer. Average atomic parameters of one molecule, referred to the center of symmetry as the origin, are $x_1 = 0{\cdot}228$, $y_1 = 0{\cdot}121$, $z_1 = 0{\cdot}194$, $x_2 = 0{\cdot}160$, $y_2 = -0{\cdot}101$ and $z_2 = 0{\cdot}241$. The statistically-averaged dimer is a rectangular planar molecule with a short edge of $1{\cdot}12 \pm 0{\cdot}02$ Å and long edges of 2·40 Å. The X-ray evidence cannot distinguish between parallel or antiparallel orientation of NO groups within the dimer. Although the average of the two long edges of the molecule is $2{\cdot}40 \pm 0{\cdot}01$ Å, models in which these distances are different from one another by any value between 0 and almost 0·5 Å are compatible with the data, but the quantitative agreement is insignificantly better when these distances are equal.

Introduction

A number of phenomena of peculiar interest are associated with the molecular structure of nitric oxide. The monomer in the gas phase is paramagnetic (Bauer & Picard, 1920). However, in condensed phases nitric oxide is diamagnetic (Lips, 1935); this and other general properties of the liquid (Rice, 1936; Eucken & d'Or, 1932) have led to the natural assumption that polymerization takes place upon condensation.

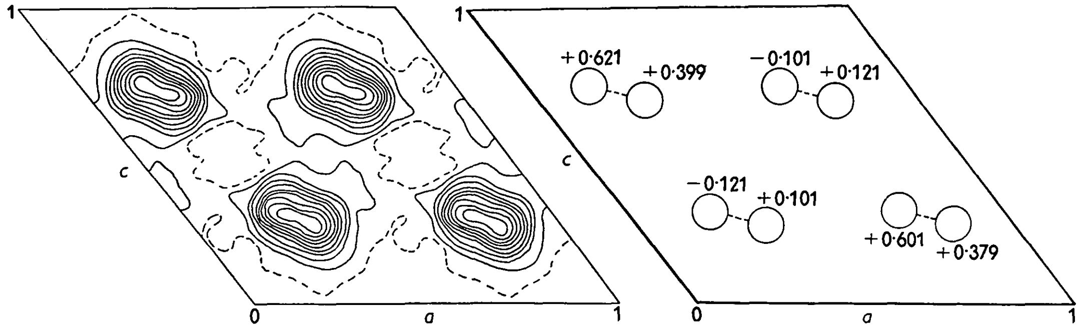

Fig. 1. Fig. 2.

Fig. 1. Electron-density projection along the b axis. Contours are in intervals of 1 e.Å^{-2}. The contour for 1 e.Å^{-2} is broken.
Fig. 2. Line drawing showing out-of-plane coordinates in Fig. 1.

Further evidence has shown conclusively that only a dimer is present (Smith, Keller & Johnston, 1951), as was earlier suggested by the residual entropy of $1{\cdot}5\pm0{\cdot}2$ e.u. per mole of dimer (Johnston & Giauque, 1929) which is consistent with a dimer with two possible orientations or alternatively a solid solution of two isomers.

The present investigation was undertaken in order to examine the structural basis for the residual entropy, and to obtain more information concerning the structure of the dimer molecule. A preliminary account of this work (Dulmage, Meyers & Lipscomb, 1951) has indicated some of the unusual features of this structure. The unusually long distance between NO groups seems consistent with the low value of 3710 cal./mole for the heat of dissociation of the dimer (Smith & Johnston, 1952).

Determination of the structure

A sample of nitric oxide gas, obtained from the Matheson Chemical Company, East Rutherford, N. J., was distilled on a vacuum line into a pyrex bulb to which a capillary about 1 mm. in diameter and 10 mm. in length was attached. The bulb was sealed off after the nitric oxide was frozen by use of liquid nitrogen, and then mounted on a Buerger precession camera. Single crystals were then grown at the freezing point of −164° C. by use of a cold air stream obtained by evaporation of liquid nitrogen (Reed & Lipscomb, 1953). The samples were maintained at about −175 °C. during photography of the $\{h0l\}$, $\{0kl\}$, $\{hk0\}$, $\{hhl\}$, $\{hk\bar{k}\}$, $\{hk\bar{h}\}$ and $\{h,k,\overline{2k}\}$ zones with the use of Mo $K\alpha$ radiation and precession angles of 20° to 28°. The observed structure factors listed in Table 2 were obtained from visual estimation of intensities, with the aid of a standard scale, which were then corrected for the Lorentz and polarization factors (Waser, 1951).

The symmetry of the reciprocal lattice is $2/m$, and the monoclinic unit cell has the parameters

$$a = 6{\cdot}68\pm0{\cdot}03,\ b = 3{\cdot}96\pm0{\cdot}01,\ c = 6{\cdot}55\pm0{\cdot}03\ \text{Å}$$
$$\text{and}\quad \beta = 127{\cdot}9\pm0{\cdot}2^\circ.$$

No systematic extinction of the general hkl reflections occurred, but the presence of $h0l$ reflections only with h even and $0k0$ reflections only with k even uniquely determines the space group $P2_1/a$, and the volume of this unit indicates two molecules of the dimer N_2O_2. Calculation of the density of the solid yields 1·46 g.cm.$^{-3}$, which seems reasonable. The dimer is thus indicated to occupy a position at a center of symmetry, but in view of the disorder indicated by the residual entropy, the molecule itself may or may not be centrosymmetric.

Because of the unusually short b axis the Patterson projection along b was analyzed to give x and z parameters. Although the corresponding Fourier projection refined satisfactorily (Figs. 1 and 2), the resolution

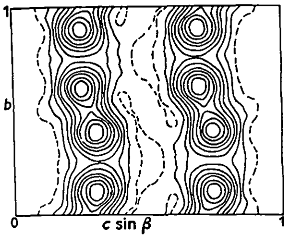

Fig. 3.

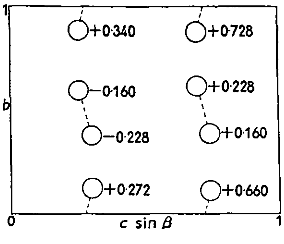

Fig. 4.

Fig. 3. Electron-density projection along the a axis. Contours are in intervals of 1 e.Å^{-2}. The contour for 1 e.Å^{-2} is broken.
Fig. 4. Line drawing showing out-of-plane coordinates for Fig. 3.

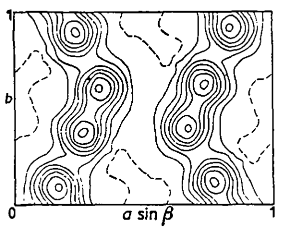

Fig. 5.

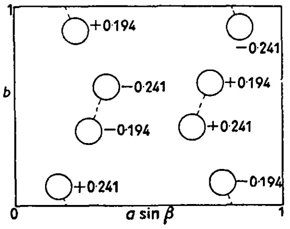

Fig. 6.

Fig. 5. Electron-density projection along the c axis. Contours are in intervals of 1 e.Å^{-2}. The contour for 1 e.Å^{-2} is broken.
Fig. 6. Line drawing showing out-of-plane coordinates for Fig. 5.

was so poor that the Fourier projections along a (Figs. 3 and 4) and c (Figs. 5 and 6) were then refined from these x and z parameters and from y parameters obtained by trial and error. Backshift corrections were applied to each of these projections, by the usual procedure of comparison with Fourier series containing the corresponding calculated structure factors, and the final average parameters thus obtained are listed in Table 1.

It is worth pointing out that the peaks on all our Fourier projections are equal well within experimental error, a result which seems to confirm the completely random disorder indicated by the residual entropy. The structure factors were calculated with the use of an arithmetic average of the scattering factors for N and O (*Internationale Tabellen*, vol. 2, p. 571). On the basis of our previous experience we would have expected an observable difference between the maximum electron densities of the two atoms not related by a center of symmetry if, because of incomplete disorder, a physical distinction really did exist. That these electron densities are indeed equal is further suggested by calculations that indicated only one sign change, either (120) or ($20\bar{3}$), when the two possible ordered structures with separate scattering curves for nitrogen and oxygen were assumed.

A least-squares treatment (Hughes, 1941) was then applied to the data, first on the assumption of a randomly disordered molecule with equal long bonds. Temperature and scale factors were included in the treatments of each of the zones, $\{h0l\}$, $\{hk0\}$ and $\{0kl\}$, and weighting factors

$$w = 1/I_o \quad \text{for} \quad F_o \geq 4F_{\text{min.}},$$
$$\text{and} \quad w = I_o/256\,I^2_{o,\,\text{min.}}, \quad \text{for} \quad F_o \leq 4F_{\text{min.}}$$

were employed. The final parameters including their calculated probable errors, and the weighted mean with the Fourier parameters are listed in Table 1.

Table 1. *Parameters for the model with equal long bonds*

	Fourier	Least squares	Weighted mean	Probable error
x_1	0·227	0·228	0·228	$0{\cdot}001_4$
y_1	0·117	0·124	0·121	$0{\cdot}003_6$
z_1	0·194	0·194	0·194	$0{\cdot}001_1$
x_2	0·159	0·160	0·160	$0{\cdot}001_2$
y_2	−0·100	−0·101	−0·101	$0{\cdot}004_0$
z_2	0·242	0·241	0·241	$0{\cdot}001_2$

These final parameters were then used to calculate all structure factors listed in Table 2. The temperature factor $\exp(4{\cdot}0 \sin^2/\lambda^2)$, determined by the least-squares treatment has been multiplied into the F_c. Excluding unobserved reflections the 'reliability factor' $\Sigma||F_o|-|F_c||\div\Sigma|F_o|$ is 0·10. For the model with equal long bonds,

$$\begin{array}{ccc} x_2 & \cdots\cdots & x_3 \\ | & & | \\ x_1 & \cdots\cdots & x_4 \end{array} \quad (A),$$

the molecular parameters* are $x_1x_2 = x_3x_4 = 1{\cdot}12\pm 0{\cdot}02$ Å, $x_2x_3 = x_1x_4 = 2{\cdot}40\pm0{\cdot}01$ Å and $\angle\, x_2x_3x_4 = 91{\cdot}2\pm1{\cdot}6°$. Thus this model is rectangular within experimental error.

A further least-squares treatment was then made for randomly disordered molecules of the planar trapezoidal type,

$$\begin{array}{cccc} & x_2 \cdots x_3 & & \\ / & & \backslash & \\ x_1 & \cdots\cdots & & x_4 \end{array} \quad (B),$$

on the assumption that the average of the x_2x_3 and x_1x_4 distances is 2·40 Å. The disorder required by the residual entropy was also included, so that the average structure consists essentially of elongated peaks each of which has an average position at the maximum of the rectangular model. The parameter describing the distortion $x_1x_4-x_2x_3$ would be expected to interact rather strongly with the scale and temperature factors, which were therefore included in this least-squares treatment. The results are that the sums of squares of residuals of the $h0l$ zone are 30·8, 30·2, 32·6 and 243·2 for respective values of $x_1x_4-x_2x_3$ equal to 0, 0·24, 0·48, and 0·96 Å. However, because the $h0l$ zone is unresolved, a similar treatment was made of the $0kl$ data, for which the sums of squares of residuals† are 5·9, 6·3 and 6·8 for values of $x_1x_4-x_2x_3$ equal, respectively, to 0, 0·30 and 0·48 Å. While it is difficult to be sure that these sums are significantly unfavorable for the smaller distortions, it seems certain that a difference $x_1x_4-x_2x_3$ greater than 0·5 Å is in disagreement with the data, and therefore $x_2x_3 > 2{\cdot}15$ Å and $x_1x_4 < 2{\cdot}65$ Å. Actually, we feel, chiefly on the basis of the results for the better-resolved $0kl$ zone, that the strictly rectangular model (A) might be slightly favored over the distorted model (B), or over another distorted model (C),

$$\begin{array}{cccc} & x_2 \cdots\cdots & x_3 & \\ / & & / & \\ x_1 \cdots\cdots & x_4 & & \end{array} \quad (C),$$

which gives the same averaged structure as (B) when the randomness required by the residual entropy is introduced.

Discussion of the molecular structure

The various molecular structures which can be derived from these general results are all planar. Although there are more, the six most obvious ones are derived from (A), (B), and (C) by introducing either parallel or antiparallel arrangements of NO groups. Of these, the three most reasonable are probably

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* The closest approaches between atoms in different N_2O_2 molecules are 3·1, 3·3, 3·3, 3·5, 3·6 and 3·7 Å.

† The weighting factor used in the least-squares treatment actually influences these results very little, for the corresponding unweighted sums of squares of residuals are 7·0, 8·8 and 11·1 for the $0kl$ zone.

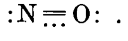
Table 2. *Comparison of calculated and observed structure factors*

hkl	F_c	F_o
001	+10·0	10·6
002	−28·8	29·5
003	−10·4	10·9
004	+ 6·6	6·6
005	+ 4·1	3·6
200	−23·6	23·2
201	−17·7	17·3
$20\bar{1}$	+16·4	18·9
202	+ 6·5	7·8
$20\bar{2}$	+26·9	25·6
203	+ 8·0	8·9
$20\bar{3}$	− 1·6	2·5
204	− 0·2	*a*
$20\bar{4}$	− 9·5	8·8
205	− 2·7	2·8
$20\bar{5}$	− 1·4	0·8
400	+ 1·3	1·8
401	+ 6·3	7·0
$40\bar{1}$	− 8·1	9·1
402	+ 1·1	*a*
$40\bar{2}$	− 4·2	4·2
403	− 2·3	2·8
$40\bar{3}$	+ 3·6	3·7
$40\bar{4}$	+ 1·6	*a*
$40\bar{5}$	+ 0·6	*a*
$40\bar{6}$	+ 1·3	2·6
$40\bar{7}$	+ 0·3	*a*
$40\bar{8}$	− 1·3	1·8
$60\bar{3}$	+ 1·2	2·2
$60\bar{4}$	+ 0·6	*a*
$60\bar{5}$	− 2·9	4·6
$60\bar{6}$	− 2·0	2·6
$60\bar{7}$	+ 1·8	2·6
$60\bar{8}$	+ 2·0	2·8
$80\bar{3}$	− 1·2	1·3
$80\bar{4}$	+ 0·9	1·2
$80\bar{5}$	+ 2·4	3·0
$80\bar{6}$	0·0	*a*
$80\bar{7}$	− 2·4	3·4
011	− 1·3	0·8
012	− 5·3	6·0
013	+ 2·0	2·5
014	+ 2·9	2·6
015	+ 2·1	2·2
110	+10·4	11·0
111	−18·9	20·6
$11\bar{1}$	+27·7	24·4
112	−10·4	10·7
$11\bar{2}$	+ 0·5	*a*
113	+ 5·3	6·1
$11\bar{3}$	−13·0	12·6
114	+ 4·6	4·6
$11\bar{4}$	− 2·8	*a*
$11\bar{5}$	+ 2·9	3·0
210	+ 4·7	4·8
$21\bar{1}$	− 5·2	6·6
$21\bar{2}$	−12·1	12·4
310	− 9·6	9·6
$31\bar{1}$	− 7·4	6·9
$31\bar{2}$	+ 6·9	6·5
$31\bar{3}$	+ 6·5	5·7
410	− 0·5	*a*
$41\bar{1}$	+ 7·2	7·0
$41\bar{2}$	+ 4·9	4·8
$41\bar{4}$	− 7·3	6·9
510	+ 2·3	2·5
$51\bar{1}$	− 0·4	*a*
610	− 1·0	1·5
$61\bar{1}$	− 3·0	2·4
$1\bar{1}1$	−18·9	20·6
$1\bar{1}2$	−10·4	10·7
$2\bar{1}1$	− 3·5	2·9
$3\bar{1}1$	− 2·2	2·4
$3\bar{1}2$	+ 5·7	5·1
$4\bar{1}1$	+ 3·0	2·5
$5\bar{1}1$	+ 0·9	0·8
020	+ 3·8	3·8
021	+ 0·2	*a*
022	− 2·5	2·8
023	− 0·4	*a*
024	+ 1·1	1·4
025	+ 0·2	1·8
120	− 2·1	2·4
$12\bar{1}$	− 7·8	8·6
$12\bar{2}$	− 0·1	*a*
220	− 1·2	1·4
$22\bar{2}$	+ 1·6	1·4
320	+ 6·2	6·6
$32\bar{2}$	− 7·5	6·5
$32\bar{3}$	−10·7	9·8
420	− 0·7	1·6
$42\bar{2}$	+ 0·8	0·7
$42\bar{4}$	− 0·6	*a*
520	− 2·7	3·7
$52\bar{2}$	+ 7·1	6·3
$52\bar{4}$	− 5·8	4·6
$52\bar{5}$	− 4·6	4·6
$62\bar{2}$	− 0·7	*a*
$72\bar{2}$	− 2·5	2·1
$1\bar{2}4$	− 1·7	2·2
031	+ 1·5	*a*
032	− 1·7	2·7
033	− 2·0	1·9
034	+ 1·2	1·9
035	+ 1·6	2·4
130	− 1·6	1·5
$13\bar{1}$	− 5·6	4·9
230	+ 3·4	3·9
$23\bar{2}$	− 5·3	5·6
330	+ 2·1	2·1
$33\bar{3}$	− 2·2	*a*
430	− 0·9	1·6
$43\bar{4}$	− 3·4	3·5
040	− 5·2	4·9
041	− 1·1	1·8
042	+ 3·7	3·9
043	+ 1·8	1·9
044	− 1·3	1·7
140	+ 1·2	1·4
$14\bar{1}$	+ 0·8	*a*
240	+ 2·5	2·6
$24\bar{2}$	− 3·3	2·4

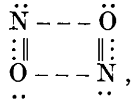
$\begin{matrix}N&O\\O&N\end{matrix}$ (*A'*); $\begin{matrix}&N&N&\\O&&&O\end{matrix}$ (*B'*); and $\begin{matrix}&N&&O\\O&&N&\end{matrix}$ (*C'*).

In (*A'*) we assume equivalent long bonds between N and O, and in (*B'*) and (*C'*) we assume that if one long bond is shorter than the other it is an N N bond.*

The electronic structures cannot be greatly different for any of these models because the long bonds can hardly be of very large order. The most extreme situation is represented by structure (*B'*), with N–N as short as 2·15 Å. Use of the equation, $d = 1{\cdot}46 - 0{\cdot}6 \log n$ (Pauling, 1947), yields $n = 0{\cdot}08$ electron pair. Even though this equation may yield values of n low by a factor of about 2 in this region of bond order, it must remain clear that the binding between the two NO groups amounts to much less than a single bond for all of the possible structures. This conclusion is, of course, quite consistent with the low value of the heat of dissociation of slightly less than 4 kcal./mole of N_2O_2. Now the bond order in the monomer is about 2·5, and the structure is usually represented as a three-electron bond plus a double bond:



It is, of course, understood that one of the three electrons is in an antibonding orbital (π^*2p_z) so that only one of the three electrons is a binding electron, and therefore this formulation does not violate the octet rule. A formal, and not very informative, description of the dimerization can be indicated schematically for structure (*A'*) as an example:



where it is understood that the dashed lines indicate the weak interactions and complete pairing of electrons. A resonance formulation is

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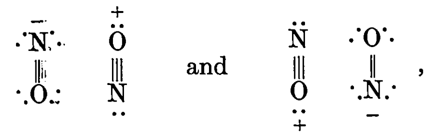
* Although a model similar to (*B'*) was pointed out previously (Dulmage, Meyers & Lipscomb, 1951), a more highly distorted model was suggested to us by Professors L. Pauling and V. Schomaker.



with much smaller contributions from structures involving an electron pair in the appropriate bond. Direct support for this interpretation is found from the short interatomic distances, which are 1·12 Å, appropriately between the double-bond and triple-bond distances of 1·19 Å and 1·07 Å, respectively (Dulmage, Meyers & Lipscomb, 1951), and rather near to the observed value of 1·14 Å for NO in the gas phase, for which the bond order is certainly close to 2·5.

The very small dipole moment of NO, 0·16 debye (Watson, Rao & Ramanaswamy, 1934), makes it seem very likely that even if the dimer had a non-centrosymmetric structure such as (B'), its dipole moment would be small. Of course, the experimental establishment of a permanent moment, provided it could be distinguished satisfactorily from an expected atom polarization, would eliminate the centrosymmetric structures. The difficulty of obtaining such unambiguous results for N_2O_2 is obvious.

However, another argument lends very slight weight to a centrosymmetric model. The residual entropy of N_2O, which has a dipole moment of 0·17 debye (Schulman, Dailey & Townes, 1948), is 0·90±0·32 e.u. The residual entropy of CO, which has a dipole moment of 0·10 debye (Watson, Rao & Ramanaswamy, 1934), is 1·12±0·1 e.u. (Clayton & Giauque, 1932). Both of these values of the residual entropies are significantly below the theoretical value of 1·38 e.u. expected for random disorder, and it is possible that the small dipole moments as well as the slight differences in sizes of the atoms forming these molecules cause a partial ordering in their crystal structures. The fact that the residual entropy* per mole of N_2O_2 is 1·50±0·20 e.u. indicates completely random disorder, and suggests that the difference in sizes of atoms is not important, and that the dipole moment (if any) of N_2O_2 is probably less than that of CO or N_2O.

The strongest evidence in favor of a non-centrosymmetric model is the spectroscopic studies of Smith, Keller & Johnston (1951) and of Mayence (1952). In view of the weak coupling between NO groups, however, the coincidence between the infra-red and Raman spectra may be accidental; its observation does not therefore necessarily eliminate a centrosymmetric model. The detailed interpretation of these spectra is certainly not obvious, and in the absence of a complete assignment of frequencies we feel that the conclusions in the spectroscopic studies to date are not unambiguous.

Further evidence slightly in favor of either structure (B') or (C'), as compared with structure (A'), is that N_2O_4 is planar with a weak bond between nitrogen atoms. The observed bond distance (Broadley & Robertson, 1949) of 1·64 Å indicates a bond order of about 0·5 if one assumes Pauling's equation. This N–N bond is weaker than a single bond because of the repulsions of the formal charges on the nitrogen atoms (Chalvet & Daudel, 1952), and it therefore seems remarkable that the dimerization of NO_2 occurs through an N–N bond rather than through an N–O bond (Pauling, 1948, p. 271).

Thus in our attempts to weigh the evidence concerning these possible molecular structures we are not able to decide among them, but we feel that the number of possible models for N_2O_2 has been very distinctly limited by our results.

It is a pleasure to acknowledge support of the major part of this investigation by the Office of Naval Research.

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* The equality of peaks in our Fourier projections seems to require the randomness associated with random positions of N and O. Hence further randomness associated with an additional explanation in terms of solid solution of two isomers seems very unlikely, because the predicted residual entropy would then be high by a factor of two.