PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Mixed Crystals of Phenazine and N-oxyphenazine: Refinement of Crystal Structures

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IV. MIXED CRYSTALS OF PHENAZINE AND *N*-OXYPHENAZINE: REFINEMENT OF CRYSTAL STRUCTURES

By A. M. GLAZER

CONTENTS

Compositions (accurate to about 1%) of mixed crystals of phenazine (P) and *N*-oxyphenazine (NOP) were determined from their u.v. absorption spectra. Densities, habit and unit-cell dimensions were found at 20 °C for crystals containing respectively 0, 8, 52, 81 and 100 mole % NOP relative to P; and X-ray diffraction data (F_{obs}) were obtained for all these and also at −90 °C for NOP. All are isostructural (NOP being pseudocentrosymmetric) in $P2_1/a$ with two molecules per average unit cell. Atomic coordinates, bond lengths and anisotropic temperature factors are listed, site-occupation factors for the oxygen atoms being used for the 8, 52 and 81 mole % compositions. A qualitative explanation, in terms of structure, is offered for the anisotropic thermal expansion of NOP.

Introduction

In 1961, Curti, Riganti & Locchi (1961) investigated the crystal structure of *N*-oxyphenazine. The space group was found to be $P2_1/c$ with two molecules in the unit cell. Since the molecules of *N*-oxyphenazine are not centrosymmetric (figure 1*a*), it was decided that they must pack in the crystal in two possible orientations to produce an average centre of symmetry. Such arrangements are fairly common in crystals, for example 1,4-bromochlorobenzene (Hendricks 1933), azulene (Robertson, Shearer, Sim & Watson 1962), anthrone (Srivastava 1964; Flack 1968). The crystal structure of *N*-oxyphenazine was therefore refined by using a phenazine nucleus to which were attached oxygen atoms with half-weight, as in figure 1*b*. The disordered structure was thus dealt with as if it were truly centrosymmetric. Refinement was carried out for the fractional coordinates and isotropic temperature factors. The final results ($R = 0.17$ approx.) seemed to indicate that the initial assumption of an average centre of symmetry was correct. Other evidence for this was provided by polarity tests and the $N(z)$ test (Howells, Phillips & Rogers 1950). This was further tested by growing mixed crystals of *N*-oxyphenazine and phenazine. The results of the structure analysis of phenazine (Herbstein & Schmidt 1955) showed that this centrosymmetric molecule ($C_{12}H_8N_2$) packed in the crystal in the same space group as *N*-oxyphenazine with two molecules in the unit cell. According to the theory of Kitaigorodskii (1957), which deals with the conditions for solubility in the solid state, complete miscibility between the two components is possible if the symmetry of the distribution of the molecules in the end-members is the same. Since Curti, Riganti and Locchi obtained a series of mixed crystals showing a gradual change in cell dimensions and intensities of reflexions, they considered this to be sufficient evidence for the existence of an average centre of symmetry. However, as will be shown in the following paper (part V) there is considerable evidence that there is a region of immiscibility in the mixed-crystal series.

In the present paper, further refinement of the crystal structures of phenazine and *N*-oxyphenazine will be described, together with the structures of some of the mixed crystals. In later papers, the variation of physical properties with changing composition and the short-range-order diffuse scattering will be discussed.

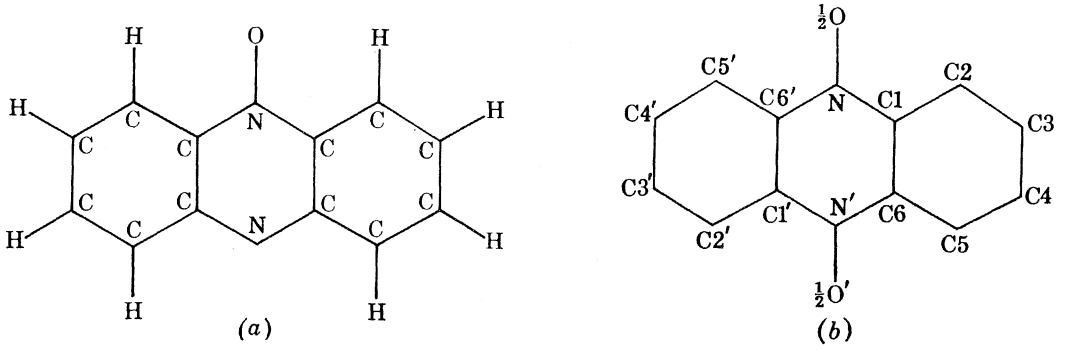

FIGURE 1. (*a*) Formula for *N*-oxyphenazine. (*b*) Formula assumed for purposes of calculation, showing numbering adopted in this paper.

GROWTH AND COMPOSITION OF MIXED CRYSTALS

Definite amounts of phenazine and *N*-oxyphenazine were weighed out, mixed and dissolved together in methyl ethyl ketone, in small glass vessels made especially for this purpose. The solutions were then heated over a water-bath and excess methyl ethyl ketone was driven off, until some deposition of solid appeared on the walls of the glass vessels. These containers were sealed with ground-glass stoppers and allowed to cool in vacuum flasks. In this manner, small crops of needle or lath-like crystals could be grown in about 20 min. The crystals were removed from the liquor with a spatula, care being taken to avoid having much of the solution drying on their surfaces. This ensured that the outer portions of the crystals did not assume compositions differing from the interior.

Preliminary X-ray photographs of the crystals, taken with the needle axis vertical, showed that there was no significant difference within any single batch and also that the crystals were all monoclinic, the *b* axis being the needle axis in each case.

Ultraviolet absorption peaks were obtained at 250 nm for phenazine and 266 nm for *N*-oxyphenazine. Typical spectra are shown in figure 2. The molar extinction coefficients were measured:

	ϵ	$\lg \epsilon$	
phenazine	1.05×10^5	5.02	at 250 nm
N-oxyphenazine	0.86×10^5	4.94	at 266 nm

By using these values and the relation $D = \epsilon c l$, where D is the optical density, ϵ the molar extinction coefficient, c the concentration/mol l^{-1} and l the path length/cm, it was possible to determine the compositions of the mixed crystals to about 1 % accuracy.

DATA COLLECTION

The unit-cell dimensions, densities and habit were examined. The detailed results are given in table 1 and in part V of this paper. For convenience, a and c were taken so that the space group $P2_1/a$ was assigned to *N*-oxyphenazine (rather than $P2_1/c$ as given by Curti *et al.* 1961)

and to the mixed crystals, to agree with that already given by Herbstein & Schmidt (1955) for phenazine.

Any attempts at cutting the crystals caused splitting, usually parallel to the length of the needles ([010]) and therefore it was found best to collect the necessary intensity data about the *b* axis only. To do this, the inclined-beam oscillation method of Milledge (1963) was used, with Cu Kα radiation. In this method a Weissenberg camera is employed, with the camera inclined at an

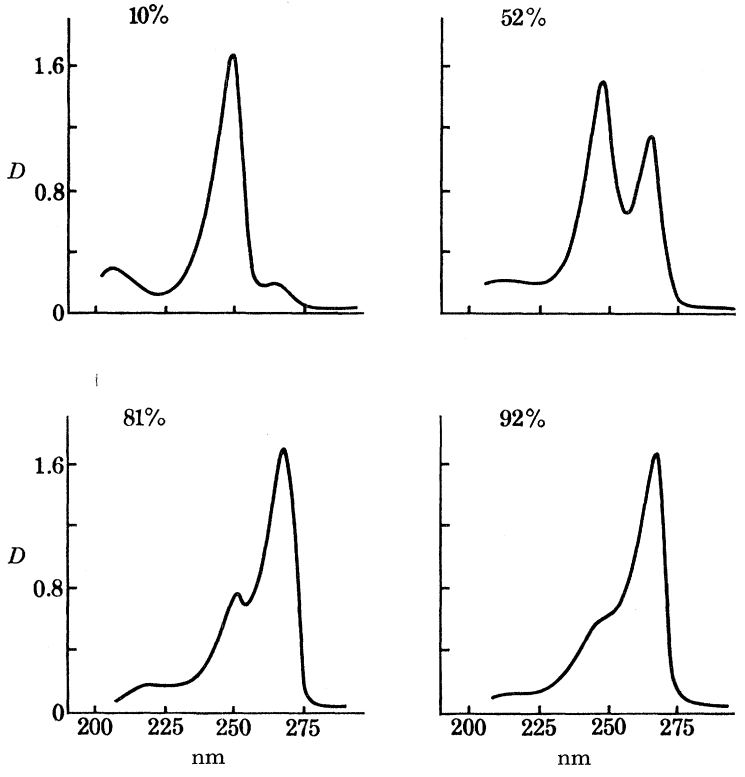

FIGURE 2. Typical u.v. spectra of solutions (in methyl ethyl ketone) of the mixed crystals, used in the determination of composition.

TABLE 1. UNIT-CELL DIMENSIONS (AT 20 °C) AND DENSITIES (g cm^{-3}; D_m AT 20 °C) OF THE END MEMBERS OF THE MIXED-CRYSTAL SERIES

substance	a/Å	b/Å	c/Å	β/deg	v_{cell}/Å^3	D_m	D_x
phenazine	13.197	5.05	7.084	109.2	445.8	1.330	1.341
N-oxyphenazine	14.29	4.60	7.439	108.8	462.8	1.410	1.408

angle to the incident X-ray beam. This angle is best taken to be 45°, although, in practice, 43° was used on account of a limitation imposed by the X-ray tube. The advantages of this method were given by Milledge. During the exposures the camera was moved, at a fixed rate, in a direction parallel to [010] (the oscillation axis). This had the effect of elongating the reflexions so as to allow easy tracking through by a photometer, and to obtain a form of integrated intensity measurement. The intensities were measured in two ways: (*a*) by a photometric cell, (*b*) by Isodensitracer (I.d.t.) (Milledge & Graeme-Barber 1969).

The data were collected at 20 °C for crystals containing respectively 0, 8, 52, 81 and 100 mole % *N*-oxyphenazine relative to phenazine. In addition, a set of low-temperature data were collected

(−90 °C) for 100 mole % (i.e. pure) *N*-oxyphenazine, the crystal being surrounded by a stream of cold nitrogen gas. It was not necessary to collect low-temperature data for phenazine, since this has already been done (at −190 °C) by Hirshfeld & Schmidt (1957), their refinement being of reasonable accuracy. It was thought best to refine the end-member data to improve on the published results and to be consistent with the mixed-crystal refinements.

The I.d.t. was used for measurement of the low-temperature *N*-oxyphenazine and room-temperature phenazine data. For comparison a set of phenazine data was also measured by the photometric cell and refined independently. All $|F_{obs}|$ and F_{calc} are given in table 2.

TABLE 2. LIST OF $|F_{obs}|$ AND F_{calc} FOR THE MIXED CRYSTALS

Symbols used are as follows:

> too weak to measure

. outside range of sphere of reflexion

→ reflexion produced near edge of oscillation ranges

? overlapping reflexions, not resolved by eye

?? close reflexions, not resolved by the measuring instrument but resolved by the eye; here, the I.d.t. was rather better than the photometer

† reflexions omitted for a variety of reasons, such as spreading of the intensity at high angles, poor spot shape, etc.

* extinction-affected reflexions

– low-order reflexions omitted by the geometry of the inclined-beam method

			mole % *N*-oxyphenazine											
			photometer 0%		I.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
2	0	0	*	17.89	*	17.65	16.55	17.95	*	19.04	*	20.07	*	21.23
4	0	0	4.58	−3.70	4.68	−4.07	5.04	−4.20	6.52	−5.46	7.49	−7.15	8.54	−7.07
6	0	0	4.06	3.91	4.57	3.47	3.74	3.17	1.91	1.88	>	0.26	>	−0.05
8	0	0	8.39	−8.52	8.97	−9.31	8.40	−9.36	8.10	−8.46	9.51	−9.29	8.32	−8.26
10	0	0	5.84	−5.67	5.35	−6.36	5.06	−5.80	4.01	−3.84	4.50	−4.17	3.24	−3.37
12	0	0	.	.	.	.	>	−0.33	>	0.29	>	0.32	>	0.44
−12	0	1	→	2.44	1.61	2.21	2.27	2.98	2.34	3.01	3.19	3.53	2.16	2.37
−10	0	1	2.28	−1.78	2.15	−1.90	>	−1.13	>	0.16	>	0.05	>	−0.36
−8	0	1	2.12	−1.74	2.28	−1.66	2.70	−1.47	>	0.68	>	0.75	1.29	1.42
−6	0	1	2.37	1.82	2.17	1.65	2.46	1.77	2.82	2.59	3.43	2.71	3.42	3.07
−4	0	1	†	−3.78	4.92	−3.97	5.54	−4.91	8.03	−6.91	8.42	−8.35	9.73	−8.38
−2	0	1	*	28.32	*	28.46	*	27.59	*	26.24	*	25.88	22.31	24.61
0	0	1	*	16.36	*	16.38	*	16.33	*	16.94	17.12	17.46	16.98	16.84
2	0	1	10.14	−11.82	*	−12.11	11.29	−11.63	9.30	−9.83	9.09	−8.32	8.32	−7.44
4	0	1	7.67	7.08	7.42	7.13	7.64	7.67	8.51	8.64	9.57	9.70	9.94	9.69
6	0	1	2.73	−2.05	2.81	−2.24	3.29	−2.53	5.02	−5.05	6.57	−6.14	6.35	−6.46
8	0	1	5.75	−5.75	5.95	−5.96	6.45	−6.40	8.46	−8.68	10.08	−10.44	8.93	−9.71
10	0	1	>	−0.29	>	−0.05	>	−0.04	>	−0.24	>	−0.79	>	−0.38
−12	0	2	†	6.32	†	7.05	†	7.42	3.50	5.11	4.53	5.80	2.87	3.58
−10	0	2	1.24	−1.31	1.13	−1.08	>	−1.25	1.89	−1.86	2.85	−2.94	2.06	−2.88
−8	0	2	>	1.69	1.42	1.46	2.00	1.70	1.49	1.39	1.80	1.73	1.29	1.21
−6	0	2	5.79	−5.38	5.96	−5.70	6.42	−5.58	4.72	−4.00	4.39	−3.57	3.45	−3.29
−4	0	2	*	−21.91	*	−21.99	*	−21.96	18.16	−18.73	16.65	−18.32	15.22	−17.14
−2	0	2	6.76	−8.24	6.92	−8.04	9.40	−8.37	9.42	−8.49	10.55	−9.32	10.74	−10.04
0	0	2	1.13	−0.92	1.16	−0.93	1.96	−1.54	5.23	−4.33	7.67	−6.42	9.12	−8.04
2	0	2	6.99	6.85	6.78	7.03	8.18	6.99	7.40	6.58	8.56	7.10	8.16	7.33

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TABLE 2 (*cont.*)

			mole % *N*-oxyphenazine											
			photometer 0%		I.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
4	0	2	>	−0.76	0.75	−0.51	>	−0.12	2.10	2.24	3.83	4.16	4.11	4.44
6	0	2	>	−0.54	>	−0.47	>	−0.28	>	−0.11	>	0.35	>	0.54
8	0	2	>	1.53	†	1.76	>	1.57	>	0.93	>	0.36	>	0.74
10	0	2	.	.	.	.	.	.	>	−0.58	>	−0.96	>	−0.62
−12	0	3	>	−0.19	>	−0.33	>	−0.80	>	−0.23	>	−0.18	>	−0.36
−10	0	3	>	−0.07	>	−0.37	>	−0.32	>	−0.90	>	−1.11	>	−1.06
−8	0	3	>	0.99	1.20	1.00	>	0.98	1.59	−1.42	3.16	−2.33	3.12	−3.06
−6	0	3	†	−13.67	11.55	−13.79	14.06	−13.96	11.76	−13.31	13.43	−15.15	13.11	−14.11
−4	0	3	8.22	−8.44	7.68	−8.75	8.69	−8.26	6.56	−5.77	5.17	−4.68	3.63	−3.24
−2	0	3	2.17	1.76	2.26	1.50	2.50	1.86	4.44	3.78	6.03	5.28	5.98	5.53
0	0	3	>	0.86	1.29	0.62	>	1.02	>	1.21	>	0.92	>	0.81
2	0	3	8.65	−9.91	9.07	−10.29	11.25	−10.82	10.46	−11.29	11.51	−12.67	11.93	−12.58
4	0	3	7.93	−8.43	8.32	−8.60	8.81	−8.85	6.46	−6.47	6.36	−6.18	6.03	−6.12
6	0	3	>	1.41	†	1.69	>	1.34	2.06	2.13	2.87	2.85	2.65	3.02
8	0	3	†	−1.87	1.35	−1.65	†	−1.94	>	−1.08	>	−1.20	>	−0.93
10	0	3	>	−0.04	>	0.45	>	0.39	>	0.31	>	0.55	>	−0.06
−12	0	4	.	.	.	.	>	−0.81	>	0.27	>	0.53	>	0.34
−10	0	4	>	0.82	1.03	1.21	>	1.52	1.28	1.50	2.53	2.23	1.95	2.16
−8	0	4	1.33	−1.25	1.78	−1.33	?	−1.57	2.15	−1.62	2.82	−2.54	?	−1.57
−6	0	4	>	0.35	>	−0.11	>	0.18	2.46	−2.25	4.19	−3.47	3.00	−2.14
−4	0	4	3.41	3.22	3.24	3.11	2.99	2.96	1.15	1.16	2.67	1.84	?	2.00
−2	0	4	2.92	−2.39	3.05	−2.44	2.80	−2.29	1.18	−0.88	>	0.83	>	−0.40
0	0	4	5.14	−4.63	4.97	−4.61	5.54	−4.92	5.51	−5.44	7.27	−6.29	6.15	−6.37
2	0	4	†	−8.95	8.19	−9.08	9.46	−9.76	8.83	−9.36	10.61	−10.86	9.61	−10.29
4	0	4	>	−1.21	1.41	−1.04	>	−1.04	>	−0.69	>	−0.84	1.29	−0.70
6	0	4	>	0.24	>	0.39	>	0.07	>	−0.84	>	−1.30	>	−1.37
8	0	4	>	−0.63	>	−0.36	>	−0.28	.	.	.	.	.	.
−10	0	5	>	−2.65	2.07	−2.77	†	−3.15	>	−1.11	>	−0.37	>	0.33
−8	0	5	>	0.20	>	0.06	>	0.20	>	1.41	>	2.11	†	3.10
−6	0	5	>	0.66	1.77	1.03	>	1.09	>	0.05	>	−0.08	>	0.94
−4	0	5	?	−2.74	†	−2.26	?	−2.90	?	−2.90	?	−3.03	→	−3.97
−2	0	5	?	5.68	6.83	6.21	?	6.16	?	2.94	?	2.54	>	1.39
0	0	5	4.78	4.97	5.32	5.28	4.82	4.81	→	1.66	→	0.96	>	1.23
2	0	5	>	−0.62	>	−0.89	>	−0.61	>	0.32	>	0.99	>	0.90
4	0	5	>	1.07	>	0.70	>	1.10	>	0.66	>	0.73	>	0.38
−8	0	6	>	−0.01	>	0.41	>	0.61	>	0.51	>	0.46	>	1.01
−6	0	6	>	−1.61	1.48	−1.45	>	−2.15	>	−0.55	>	−0.33	>	−0.02
−4	0	6	6.40	6.27	†	6.51	5.62	6.64	??	6.41	7.23	7.57	†	7.31
−2	0	6	†	12.15	†	12.57	†	12.96	??	8.11	7.18	8.05	†	7.49
0	0	6	1.86	1.67	1.77	1.46	>	1.40	>	−0.04	>	−0.27	>	−1.36
2	0	6	>	0.07	>	−0.22	>	0.01	>	0.01	>	0.32	>	−0.23
4	0	6	>	0.43	>	0.14	>	0.23	>	0 09	>	−0.33	>	0.05
1	1	0	–	4.48	–	4.54	–	4.94	–	6.24	–	7.29	–	8.42
2	1	0	*	−24.29	*	−24.26	*	−24.19	*	−23.83	*	−23.66	*	−24.15
3	1	0	*	31.19	*	31.25	*	30.93	*	28.50	25.50	27.91	*	26.96
4	1	0	*	−15.23	*	−15.59	15.25	−15.51	12.51	−14.74	13.10	−15.24	13.62	−15.24
5	1	0	3.36	−3.14	3.66	−3.46	4.22	−3.99	6.20	−6.11	8.09	−7.47	8.09	−8.11
6	1	0	6.55	−7.01	7.65	−7.43	8.21	−7.32	7.26	−7.09	7.24	−7.01	6.91	−6.62
7	1	0	5.70	−5.82	6.10	−6.26	7.50	−6.30	6.78	−6.27	7.66	−7.33	7.49	−6.95
8	1	0	3.79	−3.65	3.48	−3.61	3.85	−3.50	→	−2.93	→	−2.86	2.85	−2.52
9	1	0	>	−0.08	>	−0.10	>	−0.05	>	0.80	>	0.60	>	0.95
10	1	0	1.20	1.26	1.78	1.57	>	1.73	1.80	1.33	2.20	1.75	1.93	1.57
11	1	0	1.40	−1.78	1.92	−1.79	>	−1.57	>	−0.72	>	−0.53	>	−0.40
12	1	0	1.71	1.75	2.07	2.15	>	1.99	>	0.74	>	1.00	>	0.98
13	1	0	.	.	.	.	>	−0.80	>	−0.27	>	−0.29	>	−0.04

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TABLE 2 (*cont.*)

			mole % *N*-oxyphenazine									
			photometer 0%		I.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
14	1	0	.	.	.	.	>	−0.61	>	−0.79	>	−1.15	>	−0.69
−14	1	1	.	.	.	.	.	.	>	1.58	>	1.92	>	1.29
−13	1	1	>	0.67	>	0.98	>	1.28	>	0.53	>	0.70	>	0.12
−12	1	1	>	−0.20	>	−0.06	>	0.05	>	0.60	>	0.35	>	0.20
−11	1	1	>	−0.67	>	−0.49	>	−0.51	>	−0.11	>	−0.44	>	−0.43
−10	1	1	>	0.06	>	0.28	>	0.10	>	0.10	>	0.21	>	0.27
−9	1	1	>	0.03	>	−0.18	>	0.34	>	1.34	>	1.58	1.31	1.18
−8	1	1	>	2.21	?	2.60	?	2.10	?	1.72	>	1.85	1.93	1.62
−7	1	1	5.96	−6.20	6.87	−6.50	7.06	−6.29	5.48	−4.94	5.95	−5.07	4.91	−4.68
−6	1	1	1.20	0.71	1.15	1.01	>	0.28	→	−1.68	→	−2.10	2.54	−2.66
−5	1	1	>	−0.09	>	−0.14	>	−0.70	→	−1.72	→	−2.51	2.58	−2.53
−4	1	1	1.56	1.88	1.61	1.73	1.26	1.07	>	−1.17	>	−1.47	2.07	−2.22
−3	1	1	?	3.62	3.91	3.53	?	2.61	>	−0.51	>	−2.28	3.38	−3.11
−2	1	1	>	−0.61	>	−0.57	>	−0.41	>	0.02	>	0.13	†	−1.10
−1	1	1	2.85	−2.84	†	−2.95	3.14	−3.18	3.08	−3.18	3.56	−3.68	2.87	−3.68
0	1	1	*	−12.97	*	−12.90	11.96	−12.54	*	−13.07	11.10	−13.16	→	−13.65
1	1	1	*	27.62	*	27.96	*	28.67	*	32.55	*	34.64	*	35.60
2	1	1	*	−15.73	*	−15.68	*	−16.08	*	−17.78	18.81	−18.43	16.43	−18.26
3	1	1	7.35	7.92	†	8.01	7.97	8.35	8.93	10.21	12.01	12.06	12.51	12.36
4	1	1	>	−0.43	>	−0.30	>	−0.47	?	−2.20	?	−2.61	?	−2.70
5	1	1	5.39	−4.90	5.41	−5.15	5.38	−5.18	6.22	−5.81	7.28	−6.22	6.69	−6.18
6	1	1	1.08	−1.15	1.45	−0.83	1.47	−1.19	2.75	−2.27	3.45	−2.61	3.56	−2.78
7	1	1	?	2.19	2.20	2.14	?	2.10	>	1.13	>	0.03	>	−0.05
8	1	1	>	0.00	>	0.25	>	0.24	>	0.96	>	1.04	>	0.99
9	1	1	1.03	−1.50	1.78	−1.65	>	−1.84	2.42	−2.35	3.53	−2.98	2.65	−2.54
10	1	1	2.08	2.20	2.04	2.31	†	2.46	2.20	3.02	3.91	3.74	2.58	3.54
11	1	1	2.65	−2.53	2.63	−2.91	†	−2.97	2.29	−2.69	3.29	−3.20	1.91	−2.16
12	1	1	>	0.07	>	−0.08	>	−0.29	>	−0.04	>	−0.11	>	0.32
13	1	1	.	.	.	.	.	.	>	−1.13	>	−1.75	>	−0.86
−15	1	2	.	.	.	.	.	.	.	.	.	.	>	1.49
−14	1	2	>	1.30	>	1.65	>	1.61	>	1.05	>	1.13	>	0.29
−13	1	2	>	−0.28	>	−0.27	>	−0.47	>	−0.35	>	−0.37	>	0.01
−12	1	2	2.34	−2.19	2.65	−2.22	†	−2.31	2.20	−2.33	3.06	−3.01	3.62	−2.63
−11	1	2	4.33	4.46	4.58	4.57	4.39	4.80	3.14	3.32	3.52	3.60	2.62	2.68
−10	1	2	4.39	−4.05	4.53	−4.41	4.89	−4.72	3.62	−3.82	4.19	−4.10	2.98	−2.99
−9	1	2	2.13	2.13	2.15	2.26	>	2.50	>	1.09	>	0.57	>	−0.40
−8	1	2	5.10	−4.69	5.45	−4.85	5.62	−5.03	4.71	−4.04	4.46	−4.22	3.42	−3.39
−7	1	2	7.26	−8.58	8.53	−8.72	8.75	−8.65	7.30	−6.78	7.16	−7.20	6.41	−6.34
−6	1	2	6.44	−6.61	7.02	−6.68	6.63	−6.71	6.62	−5.87	6.17	−5.79	5.58	−5.47
−5	1	2	3.62	−3.17	3.97	−3.50	3.76	−3.26	1.48	−1.40	>	−0.08	>	0.51
−4	1	2	1.68	−1.45	1.63	−1.25	1.75	−1.60	2.06	−1.74	2.40	−1.76	2.27	−2.43
−3	1	2	3.47	−3.18	3.72	−3.43	3.80	−3.69	3.94	−3.31	3.39	−3.29	3.62	−3.42
−2	1	2	8.92	9.43	8.86	9.66	→	9.57	8.31	7.68	7.71	6.74	6.33	5.65
−1	1	2	??	−2.64	2.28	−2.65	??	−2.56	→	−1.68	2.47	−2.43	2.98	−2.73
0	1	2	2.71	2.22	2.67	2.26	2.04	1.69	>	−0.62	>	−1.65	2.44	−1.95
1	1	2	4.02	−3.77	4.17	−3.55	3.64	−3.82	3.21	−3.18	4.08	−3.49	3.85	−3.65
2	1	2	2.74	−2.24	2.69	−2.28	2.35	−2.10	>	−0.78	>	0.05	>	0.35
3	1	2	3.28	−2.74	3.38	−2.82	3.14	−2.90	2.86	−2.54	2.62	−1.97	1.69	−1.77
4	1	2	>	0.67	>	0.74	>	0.95	2.00	1.75	2.31	1.85	2.54	2.30
5	1	2	3.25	3.40	3.59	3.49	4.13	3.67	4.57	4.28	5.92	5.14	6.18	5.15
6	1	2	1.37	−1.30	1.40	−1.23	†	−1.68	†	−1.66	3.47	−2.53	2.54	−1.95
7	1	2	>	−1.15	>	−0.98	>	−0.87	>	−0.31	>	−0.31	>	−0.26
8	1	2	>	−0.50	>	−0.54	>	−0.62	>	0.02	>	0.05	>	0.01
9	1	2	>	−0.67	>	−0.55	>	−0.80	??	−1.81	2.31	−2.29	1.91	−2.27
10	1	2	.	.	.	.	>	0.96	>	0.96	>	1.29	>	0.77
11	1	2	.	.	.	.	.	.	>	−0.26	>	−0.83	>	−0.53

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

TABLE 2 (*cont.*)

			mole % *N*-oxyphenazine									
			photometer 0%		I.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
12	1	2	.	.	.	.	.	.	>	−0.65	>	−0.97	>	−0.90
−15	1	3	.	.	.	.	.	.	.	.	.	.	>	0.17
−14	1	3	>	−1.23	>	−1.39	>	−1.38	>	−0.90	>	−1.25	>	−1.25
−13	1	3	1.93	2.44	2.59	2.53	†	2.57	1.98	2.46	3.03	3.46	2.98	2.84
−12	1	3	4.04	−4.34	4.35	−4.79	4.53	−4.72	3.31	−3.77	4.41	−4.68	3.62	−3.53
−11	1	3	4.93	5.30	5.44	5.73	5.18	5.77	3.79	3.96	4.38	4.67	3.83	3.47
−10	1	3	3.56	−3.28	3.76	−3.46	3.92	−3.42	3.21	−2.82	3.81	−3.35	?	−2.31
−9	1	3	2.54	−2.91	2.88	−3.01	2.91	−3.34	3.11	−3.56	4.79	−4.85	4.00	−4.38
−8	1	3	2.71	−2.52	2.69	−2.41	1.99	−2.09	2.28	−1.54	3.79	−1.71	1.93	−1.13
−7	1	3	>	−1.35	1.74	−1.66	>	−1.40	2.00	−2.05	2.87	−2.45	2.56	−2.41
−6	1	3	1.65	−1.42	1.84	−1.40	>	−1.33	>	−0.12	>	0.46	>	1.26
−5	1	3	>	0.54	>	0.55	>	0.54	>	0.17	>	1.13	>	0.46
−4	1	3	5.64	6.01	5.50	5.97	6.77	6.12	6.80	6.21	7.52	7.47	7.20	7.51
−3	1	3	?	−10.43	?	−10.54	?	−10.21	?	−8.11	?	−7.18	7.09	−6.99
−2	1	3	?	6.19	?	6.25	?	6.16	?	4.59	?	4.37	3.62	3.87
−1	1	3	?	−10.87	?	−11.10	?	−11.01	?	−10.24	?	−10.85	10.46	−10.33
0	1	3	6.61	−7.75	7.68	−7.72	8.57	−8.35	7.92	−8.68	10.02	−9.11	9.82	−9.59
1	1	3	7.12	−7.45	7.07	−7.54	8.32	−7.73	7.62	−7.42	9.08	−8.29	9.17	−8.33
2	1	3	4.96	−4.16	5.00	−4.04	4.75	−4.22	3.83	−3.22	4.69	−3.13	4.01	−3.34
3	1	3	?	1.49	?	1.57	?	1.49	?	1.10	?	1.00	>	1.28
4	1	3	>	1.05	?	1.40	>	1.07	>	1.02	>	0.64	>	0.54
5	1	3	2.97	−2.95	3.12	−2.92	3.35	−3.10	2.22	−2.15	2.39	−1.79	2.02	−1.45
6	1	3	>	0.90	1.39	1.21	>	1.06	>	0.31	>	0.06	>	0.07
7	1	3	>	−1.58	>	−1.23	>	−1.16	>	0.26	>	1.26	>	0.82
8	1	3	2.50	2.58	2.79	3.03	2.85	3.21	2.22	2.49	3.82	3.07	1.35	2.28
9	1	3	?	4.35	†	4.98	3.64	4.89	†	3.94	4.24	4.86	3.02	3.91
10	1	3	†	3.03	†	3.42	>	3.49	†	2.14	†	2.58	>	1.78
−14	1	4	.	.	.	.	.	.	>	0.01	>	−0.22	>	−0.06
−13	1	4	.	.	.	.	.	.	>	0.85	>	1.40	>	0.79
−12	1	4	>	−1.17	>	−1.08	>	−0.90	>	−0.56	>	−0.70	>	−0.04
−11	1	4	>	−0.57	>	−0.62	>	−1.08	>	0.20	>	0.58	>	0.69
−10	1	4	>	0.71	>	0.78	>	0.94	>	0.47	>	0.47	>	0.96
−9	1	4	>	0.53	>	−0.40	>	−0.15	>	0.07	>	0.11	>	0.64
−8	1	4	>	−0.20	>	0.32	>	0.36	>	−0.23	>	−0.61	>	0.06
−7	1	4	2.19	1.86	2.21	2.06	>	2.25	>	0.21	>	−0.04	>	−0.08
−6	1	4	>	0.01	>	−0.12	>	−0.38	>	−0.12	>	0.60	>	0.54
−5	1	4	3.54	−3.21	3.36	−3.27	4.37	−3.57	5.02	−4.97	6.06	−5.64	6.74	−6.02
−4	1	4	2.48	2.09	2.85	2.23	3.20	2.44	3.89	3.26	4.46	4.67	4.27	3.55
−3	1	4	>	−1.01	1.33	−1.35	>	−1.14	?	3.25	?	−2.64	?	−2.11
−2	1	4	?	−2.33	2.51	−1.93	2.13	−2.09	?	−1.93	?	−2.42	?	−2.62
−1	1	4	?	−1.62	2.73	−1.71	?	−1.77	?	−1.13	?	−0.59	>	0.33
0	1	4	3.28	−3.19	3.20	−2.97	3.47	−3.18	3.61	−3.15	4.84	−3.81	3.49	−3.14
1	1	4	1.65	−1.70	2.02	−1.69	>	−1.36	>	0.14	>	0.76	>	0.88
2	1	4	2.31	1.80	2.33	1.96	2.39	2.09	2.04	1.76	2.64	1.96	2.40	1.83
3	1	4	1.34	−1.97	2.37	−2.22	→	−2.47	3.27	−3.34	5.06	−4.43	3.98	−4.53
4	1	4	3.33	3.17	3.17	3.30	3.61	3.49	2.71	3.01	4.16	3.27	2.85	2.76
5	1	4	4.13	−3.86	4.03	−4.06	4.51	−4.33	3.40	−3.55	4.58	−4.15	?	−4.03
6	1	4	2.10	2.34	2.21	2.49	2.55	2.84	†	3.02	2.42	3.67	1.94	3.34
7	1	4	?	2.18	?	2.58	?	2.73	†	3.58	4.08	4.88	3.04	4.39
8	1	4	1.86	2.52	2.04	2.75	2.26	3.15	†	3.47	3.64	4.50	?	3.88
9	1	4	.	.	.	.	.	.	>	1.71	†	2.33	>	1.78
−14	1	5	>	−0.37	>	−0.27	>	−0.27	>	−0.48	>	−0.62	>	−0.44
−13	1	5	>	−1.68	†	−1.76	†	−1.89	→	−1.41	→	−1.66	>	−1.26
−12	1	5	>	−1.09	1.20	−1.20	>	−1.32	>	−0.78	>	−0.63	>	−0.01
−11	1	5	>	−0.52	>	−0.52	>	−0.49	>	0.11	>	0.27	>	0.47
−10	1	5	>	0.22	>	0.08	>	0.13	>	0.39	>	0.43	>	0.99

TABLE 2 (*cont.*)

			photometer 0%		I.d.t. 0%		mole % *N*-oxyphenazine 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
−9	1	5	>	−0.20	>	−0.04	>	0.28	>	0.45	>	0.89	>	1.36
−8	1	5	>	−0.24	>	−0.22	>	−0.40	>	−0.66	>	−1.15	>	−0.90
−7	1	5	>	−1.79	1.59	−1.76	>	−2.17	>	−0.54	>	−0.09	>	0.33
−6	1	5	>	0.85	1.33	1.06	>	0.94	>	0.51	>	0.89	>	0.04
−5	1	5	4.82	4.64	4.65	4.62	5.26	4.82	3.61	3.14	4.81	3.10	?	3.24
−4	1	5	?	4.96	?	5.07	?	5.17	?	3.94	?	3.86	1.96	2.72
−3	1	5	?	6.13	?	6.46	?	6.39	?	3.10	?	2.57	3.15	2.71
−2	1	5	?	2.89	?	2.74	?	2.82	?	1.86	?	1.53	>	1.47
−1	1	5	>	−1.01	>	−0.82	>	−0.99	>	−0.65	>	−0.26	>	−1.15
0	1	5	>	0.15	>	−0.30	>	0.06	>	0.50	>	1.05	1.56	1.54
1	1	5	>	1.31	1.43	1.29	>	1.35	>	0.35	>	0.34	>	−0.29
2	1	5	>	−0.32	>	−0.79	>	−0.28	>	0.76	>	1.35	?	1.51
3	1	5	>	−0.35	>	−0.67	>	−0.92	?	−1.61	?	−2.62	?	−2.25
4	1	5	>	−0.43	>	−0.72	>	−0.49	>	0.24	>	0.07	>	0.54
5	1	5	?	−1.58	1.44	−1.82	?	−1.59	>	−0.27	>	−0.34	>	0.05
6	1	5	>	−0.84	>	−0.85	>	−0.85	>	0.21	>	0.42	>	0.46
7	1	5	>	0.17	>	0.17	>	0.28	>	0.49	>	0.71	>	0.28
−12	1	6	>	−0.38	>	−0.38	>	−0.43	>	−0.18	>	−0.29	>	0.23
−11	1	6	>	−0.47	>	−0.35	>	−0.08	>	−0.60	>	−1.33	>	−1.04
−10	1	6	>	1.53	1.63	1.67	>	1.63	>	1.44	>	1.37	>	1.33
−9	1	6	3.07	−2.46	2.60	−2.54	†	−2.95	>	−1.76	>	−1.91	>	−1.36
−8	1	6	2.50	2.24	2.37	2.32	2.50	2.18	1.98	1.70	1.85	1.73	>	1.51
−7	1	6	>	0.85	>	1.05	>	0.85	2.67	2.59	3.24	3.87	?	4.03
−6	1	6	4.82	4.94	†	4.82	4.73	4.90	3.29	4.01	?	4.57	2.98	3.98
−5	1	6	6.36	5.57	4.65	6.08	?	6.28	?	4.77	?	5.39	?	4.92
−4	1	6	?	4.71	4.96	4.47	?	4.72	?	2.96	?	2.96	?	1.97
−3	1	6	>	−0.14	>	0.07	>	−0.37	>	−0.67	>	−1.08	?	−2.20
−2	1	6	>	−0.87	>	−1.18	>	−1.05	?	−1.35	?	−1.64	?	−1.97
−1	1	6	?	2.42	?	2.66	3.18	2.78	>	1.63	>	1.40	>	0.99
0	1	6	3.90	−3.99	3.95	−4.30	4.13	−4.27	†	−2.52	†	−2.54	>	−2.36
1	1	6	4.44	4.67	4.65	4.85	4.36	4.97	†	2.28	1.77	1.93	>	2.25
2	1	6	2.70	−2.67	2.65	−2.88	2.42	−2.74	>	−1.45	>	−1.76	>	−1.18
3	1	6	>	−1.18	?	−1.53	?	−1.43	>	−1.20	>	−1.42	>	−1.26
4	1	6	>	−1.79	1.04	−1.82	†	−1.93	>	−1.51	†	−2.06	>	−1.36
−10	1	7	.	.	.	.	.	.	>	0.50	>	0.42	>	0.43
−9	1	7	>	−1.50	1.29	−1.50	>	−1.72	>	−0.91	>	−1.29	>	−0.61
−8	1	7	>	0.38	>	0.33	>	0.14	>	0.47	>	0.89	>	0.66
−7	1	7	>	0.49	>	0.77	>	0.88	>	0.93	>	1.31	>	1.10
−6	1	7	>	0.40	>	0.49	>	0.43	>	0.87	>	1.64	>	0.59
−5	1	7	>	0.09	>	0.04	>	−0.28	>	−0.22	>	−0.04	>	−0.86
−4	1	7	>	−0.69	>	−0.48	>	−0.39	>	−0.71	>	−0.97	>	−1.72
−3	1	7	>	0.57	>	0.55	>	0.46	>	1.61	?	2.17	?	2.53
−2	1	7	2.23	−2.40	1.79	−2.36	†	−2.51	†	−2.57	3.00	−3.60	2.91	−3.00
−1	1	7	3.28	3.73	†	4.14	3.14	4.52	†	3.57	3.00	4.01	†	4.27
0	1	7	>	−1.76	?	−1.92	†	−1.93	>	−1.76	>	−2.11	>	−1.65
1	1	7	>	0.25	>	0.17	>	0.27	>	−0.74	>	−1.13	>	−1.99
0	2	0	–	7.10	–	7.28	–	7.52	–	8.12	–	9.28	–	9.25
1	2	0	–	1.36	–	1.35	–	0.85	–	−0.13	–	−0.62	–	−0.70
2	2	0	–	−7.61	–	−7.56	–	−7.41	–	−6.15	–	−5.50	–	−5.88
3	2	0	5.35	−5.67	5.92	−5.93	6.07	−6.13	6.43	−6.67	7.02	−6.68	7.29	−7.01
4	2	0	1.63	−1.82	2.69	−2.00	3.02	−2.33	3.82	−3.54	4.94	−4.41	4.93	−4.97
5	2	0	*	−12.51	12.30	−12.58	?	−12.26	10.13	−9.74	9.93	−9.36	9.44	−9.44
6	2	0	??	7.92	8.25	8.10	7.63	7.55	?	4.88	?	3.85	?	3.44
7	2	0	3.17	−3.13	3.21	−3.21	2.89	−2.67	>	−0.50	>	0.07	>	0.80
8	2	0	>	0.58	>	0.87	>	0.67	>	0.09	>	−0.08	>	−0.03
9	2	0	>	0.25	>	0.11	>	0.46	>	0.50	>	1.00	?	1.84

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

TABLE 2 (*cont.*)

			photometer 0%		I.d.t. 0%		mole % *N*-oxyphenazine 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
10	2	0	>	−0.36	>	−0.47	>	−0.34	>	0.10	>	0.03	>	0.54
11	2	0	>	−1.97	2.26	−2.16	>	−2.14	>	−1.98	†	−2.41	1.15	−1.38
12	2	0	>	−1.82	2.08	−1.94	>	−2.04	>	−0.80	>	−1.79	>	−1.10
13	2	0	>	−0.71	>	−0.87	>	−0.95	>	−1.03	>	−1.28	>	−0.89
14	2	0	>	−0.96	>	−1.00	>	−1.00	>	−0.40	>	−0.57	>	−0.80
−15	2	1	.	.	.	.	.	.	.	.	.	.	>	−0.09
−14	2	1	>	−0.79	>	−0.44	>	−0.64	>	−0.47	>	−0.81	>	−0.37
−13	2	1	>	0.32	>	0.32	>	0.30	>	0.32	>	0.33	>	0.03
−12	2	1	>	0.62	>	0.85	>	0.98	>	0.27	>	0.18	>	0.11
−11	2	1	>	−0.45	>	−0.59	>	−0.79	>	0.10	>	0.01	>	0.06
−10	2	1	>	−0.88	>	−0.60	>	−0.59	?	−1.03	?	−1.10	1.07	−1.33
−9	2	1	?	−2.33	?	−2.46	?	−2.78	?	−2.63	?	−3.12	2.78	−2.90
−8	2	1	?	−2.87	2.98	−2.92	3.24	−3.15	2.86	−2.74	3.31	−2.90	2.53	−2.66
−7	2	1	>	0.18	>	0.00	>	−0.52	1.86	−1.87	2.34	−1.92	2.27	−2.00
−6	2	1	3.17	−4.03	4.56	−4.67	5.19	−4.82	4.74	−4.93	4.87	−4.97	4.47	−4.97
−5	2	1	5.69	5.68	6.39	6.05	6.07	5.33	5.63	4.18	4.98	3.85	3.92	3.36
−4	2	1	6.50	−7.43	7.71	−7.87	7.81	−7.78	8.07	−7.48	8.30	−8.13	8.29	−8.71
−3	2	1	4.22	4.36	4.66	4.50	4.84	4.64	4.44	4.53	4.09	4.21	?	3.93
−2	2	1	3.37	3.90	†	4.11	–	4.18	–	3.71	–	2.61	–	2.06
−1	2	1	>	0.86	>	0.73	–	0.92	–	1.73	–	1.92	–	2.42
0	2	1	>	0.03	>	0.43	–	−0.02	–	−0.65	–	−0.51	>	−0.21
1	2	1	1.13	0.97	>	0.94	>	1.32	1.46	1.68	2.09	2.31	†	2.45
2	2	1	2.88	2.61	3.09	2.86	3.48	2.95	4.35	3.93	4.80	4.69	5.46	5.50
3	2	1	?	−10.41	?	−10.65	10.53	−10.68	9.98	−11.80	12.25	−12.26	10.77	−12.65
4	2	1	??	15.60	??	15.82	13.43	16.15	??	16.71	15.92	17.01	16.12	17.12
5	2	1	→	−9.70	9.25	−9.91	10.19	−10.33	10.44	−10.66	11.68	−11.97	12.20	−11.69
6	2	1	3.83	3.24	3.94	3.27	3.90	3.18	3.86	3.30	4.06	3.12	3.20	2.74
7	2	1	>	−0.37	>	−0.45	>	−0.48	>	−0.67	>	−0.62	>	−0.27
8	2	1	>	−1.32	1.86	−1.67	>	−1.77	>	−1.52	>	−2.01	2.06	−1.73
9	2	1	>	−1.13	>	−1.08	>	−1.38	>	−1.26	>	−1.08	>	−0.70
10	2	1	>	−0.62	1.63	−1.05	>	−1.20	>	−1.08	>	−1.79	>	−0.93
11	2	1	>	−1.43	1.72	−1.56	>	−1.59	>	−1.05	>	−1.18	>	−0.79
12	2	1	→	−2.06	2.15	−2.39	→	−2.54	1.77	−2.01	2.08	−2.47	1.94	−2.05
13	2	1	>	1.24	>	1.33	>	1.29	>	1.03	>	1.42	>	0.90
14	2	1	.	.	.	.	.	.	.	.	.	.	>	−0.66
−15	2	2	.	.	.	.	.	.	>	−0.22	>	−0.43	>	−0.44
−14	2	2	>	0.41	>	0.43	>	0.40	>	0.16	>	0.42	>	0.35
−13	2	2	>	−0.18	>	−0.40	>	−0.40	>	−0.34	>	−0.65	>	−0.33
−12	2	2	>	−0.04	>	−0.04	>	−0.05	>	−0.79	>	−0.86	>	−0.67
−11	2	2	>	−1.28	>	−1.34	>	−1.38	>	−1.09	>	−1.63	1.28	−1.32
−10	2	2	3.02	−3.52	3.09	−3.61	3.31	−3.70	2.55	−2.86	3.08	−3.54	3.29	−2.84
−9	2	2	5.52	−5.46	6.70	−5.72	?	−5.55	?	−4.18	?	−4.09	?	−3.41
−8	2	2	?	1.28	?	1.17	?	1.51	>	1.39	?	1.51	>	0.97
−7	2	2	4.01	−3.76	→	−3.83	4.37	−4.11	?	−2.26	?	−1.14	>	−0.47
−6	2	2	1.34	1.39	1.65	1.39	>	1.47	1.43	1.02	1.98	1.71	>	1.02
−5	2	2	2.68	2.10	2.72	2.37	2.71	2.13	2.47	1.52	2.42	1.83	1.97	1.73
−4	2	2	>	−0.72	1.02	−0.73	>	−0.54	>	0.06	>	0.09	>	0.14
−3	2	2	1.63	−1.24	1.54	−1.24	2.01	−1.53	3.74	−3.20	4.84	−4.24	5.12	−5.03
−2	2	2	3.98	−4.09	4.06	−4.15	4.92	−4.47	5.17	−5.17	??	−5.84	5.98	−5.75
−1	2	2	5.81	−5.85	5.66	−5.73	6.31	−5.97	5.57	−6.03	6.88	−6.19	6.17	−7.18
0	2	2	6.48	−6.04	6.37	−6.13	6.88	−6.47	5.77	−6.23	6.14	−6.47	6.41	−6.09
1	2	2	0.96	1.09	1.25	1.17	>	1.32	6.12	6.65	>	−0.34	>	−0.64
2	2	2	3.63	3.36	3.40	3.36	4.13	3.92	5.11	−5.22	8.18	7.43	9.32	8.58
3	2	2	3.46	−3.04	3.60	−2.98	3.85	−3.50	2.49	2.16	6.76	−6.36	7.33	−6.12
4	2	2	>	−0.20	>	−0.04	>	0.04	>	−1.15	3.88	2.99	4.09	3.64
5	2	2	>	−1.40	1.36	−1.15	>	−1.13	>	−0.66	>	−1.18	>	−0.95

TABLE 2 (*cont.*)

			photometer 0%		I.d.t. 0%		mole % *N*-oxyphenazine 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
6	2	2	>	−0.89	>	−0.73	>	−0.90	?	2.19	>	−0.04	>	−0.66
7	2	2	2.38	2.64	?	3.17	?	2.96	3.16	2.96	?	2.58	?	1.91
8	2	2	3.49	4.01	3.83	4.22	3.92	4.21	>	0.24	2.97	3.18	2.80	2.36
9	2	2	>	1.12	1.82	1.35	>	0.94	>	−0.89	>	0.11	>	−0.11
10	2	2	>	−0.02	>	0.02	>	0.01	>	1.07	>	−1.30	>	−1.14
11	2	2	>	0.57	>	0.44	>	0 60	>	−1.07	>	1.53	>	1.36
12	2	2	>	−0.56	0.86	−0.69	>	−0.70	.	.	.	.	.	.
−15	2	3	.	.	.	.	.	.	>	0.03	>	0.13	>	0.10
−14	2	3	>	0.40	>	0.38	>	0.43	>	0.35	>	0.52	>	0.54
−13	2	3	>	−0.02	>	0.14	>	0.14	>	0.15	>	0.17	>	−0.07
−12	2	3	?	−1.08	0.79	−1.17	?	−1.36	>	−0.59	>	−0.51	>	0.49
−11	2	3	3.49	−2.75	2.97	−2.78	2.91	−2.61	2.31	−2.40	2.96	−3.12	3.06	−2.97
−10	2	3	4.33	4.03	3.38	4.21	3.83	4.61	3.40	3.68	4.06	4.10	?	3.99
−9	2	3	6.10	−6.52	7.03	−7.00	6.94	−6.93	5.07	−5.55	5.99	−5.93	5.03	−4.86
−8	2	3	5.93	6.63	6.49	7.04	6.89	7.06	4.33	4.30	4.49	4.43	3.11	3.35
−7	2	3	3.05	−2.48	3.08	−2.55	2.67	−2.57	1.97	−1.73	>	−0.72	>	−0.17
−6	2	3	>	−0.99	>	−1.22	>	−1.37	3.06	−1.80	2.33	−1.47	1.97	−1.80
−5	2	3	>	−1.10	0.84	−1.01	>	−0.56	>	−0.27	>	0.64	>	0.83
−4	2	3	3.28	−2.73	3.40	−3.15	2.90	−2.83	??	−2.67	??	−2.12	2.53	−1.93
−3	2	3	?	−7.25	?	−7.16	8.81	−7.26	??	−6.31	??	−6.48	7.06	−5.92
−2	2	3	?	−8.27	8.41	−8.18	?	−8.36	??	−7.23	??	−6.79	7.55	−6.55
−1	2	3	>	1.19	?	1.43	>	1.37	>	1.02	>	0.65	>	1.09
0	2	3	7.94	−7.04	6.91	−6.99	8.68	−6.83	??	−5.18	??	−5.20	5.30	−5.08
1	2	3	6.10	6.04	5.99	6.42	6.16	5.94	4.03	4.13	3.53	3.51	3.08	2.90
2	2	3	3.05	−2.86	3.15	−2.89	3.16	−2.86	2.85	−2.48	3.67	−3.30	3.06	−3.13
3	2	3	>	−0.47	>	−0.25	>	−0.54	>	−0.20	>	0.13	>	−0.58
4	2	3	>	−0.41	>	−0.08	>	−0.16	>	−0.12	>	−0.04	>	0.01
5	2	3	2.27	2.49	2.80	3.00	3.05	3.24	?	4.28	?	5.31	4.39	4.80
6	2	3	5.32	6.28	5.96	6.80	6.53	7.10	6.02	6.53	7.21	8.07	6.75	7.47
7	2	3	5.00	4.93	4.91	5.53	4.68	5.42	4.19	4.42	4.40	4.64	3.65	4.15
8	2	3	1.66	1.91	1.93	2.09	→	2.46	→	2.26	→	3.05	?	2.66
9	2	3	1.02	1.15	1.44	1.14	>	1.22	>	0.31	>	0.08	>	0.28
10	2	3	>	−0.97	0.63	−0.97	>	−0.84	>	−0.65	>	−0.68	>	−0.61
11	2	3	>	0.04	>	−0.08	>	0.03	>	0.20	>	0.28	>	0.05
−15	2	4	.	.	.	.	.	.	>	−0.48	>	−0.33	>	−0.31
−14	2	4	>	−0.51	0.61	−0.62	>	−0.84	>	−0.57	>	−0.74	>	−0.18
−13	2	4	>	−0.33	>	−0.21	>	−0.15	>	0.13	>	0.19	>	0.27
−12	2	4	>	0.92	>	0.77	>	0.82	>	1.05	>	1.77	1.78	1.95
−11	2	4	2.41	−1.91	2.14	−2.20	>	−1.88	1.89	−1.85	2.61	−2.68	1.64	−1.71
−10	2	4	4.21	3.26	2.33	3.42	3.85	3.54	3.10	2.78	4.65	3.54	3.46	3.11
−9	2	4	??	−2.33	?	−2.61	??	−2.56	??	−2.75	3.14	−3.15	?	−2.24
−8	2	4	>	−0.79	>	−0.93	>	−1.48	>	−1.16	>	−1.04	>	−1.33
−7	2	4	1.97	1.51	1.91	1.49	>	1.74	>	0.96	>	1.57	>	1.37
−6	2	4	1.80	1.53	1.87	1.24	>	1.55	>	0.67	>	0.65	>	0.08
−5	2	4	>	1.07	0.73	0.97	>	1.04	>	1.29	>	1.35	>	1.73
−4	2	4	1.48	1.14	2.09	1.57	>	1.74	>	0.47	>	0.12	>	−0.22
−3	2	4	>	0.39	>	0.46	>	0.44	?	2.02	?	2.64	?	3.56
−2	2	4	??	−4.47	4.66	−4.24	?	−4.31	?	−3.97	?	−3.90	?	−3.73
−1	2	4	?	3.64	4.12	3.92	4.57	4.17	5.24	4.60	5.86	5.36	5.00	5.38
0	2	4	1.65	−1.05	1.52	−1.51	>	−1.26	1.70	−1.71	1.65	−1.69	>	−1.49
1	2	4	>	−0.19	>	−0.07	>	−0.33	>	−0.68	>	−1.10	>	−1.90
2	2	4	>	−0.19	>	−0.55	>	−0.38	>	−0.01	>	−0.47	>	−0.20
3	2	4	>	−0.75	>	−0.75	>	−0.84	>	−0.29	>	−0.70	>	−0.97
4	2	4	>	1.19	0.83	1.21	>	1.51	??	2.53	??	2.95	3.20	3.13
5	2	4	?	2.74	?	3.16	2.62	3.29	?	3.92	?	4.57	4.35	3.89
6	2	4	>	0.11	>	0.01	>	0.10	>	0.39	>	0.84	>	0.64

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TABLE 2 (*cont.*)

			photometer 0%		I.d.t. 0%		mole % *N*-oxyphenazine 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
7	2	4	2.12	2.31	2.38	2.63	†	2.83	>	2.07	>	2.41	1.47	1.90
8	2	4	1.60	−2.21	1.64	−2.28	†	−2.31	>	−1.60	>	−1.55	>	−1.50
9	2	4	>	0.31	>	0.37	>	0.57	>	0.14	>	−0.04	>	0.01
10	2	4	.	.	.	.	.	.	.	.	.	.	>	0.52
−14	2	5	.	.	.	.	.	.	>	−1.15	>	−1.53	>	−0.97
−13	2	5	.	.	.	.	.	.	>	0.93	>	0.94	>	1.15
−12	2	5	>	−0.83	1.22	−1.01	>	−0.82	>	−0.40	>	−0.58	>	−0.33
−11	2	5	>	0.13	>	0.10	>	0.10	>	0.17	>	0.01	>	0.43
−10	2	5	1.97	−0.98	>	−0.95	>	−1.33	>	−0.04	>	0.07	>	−0.02
−9	2	5	>	0.99	>	1.08	>	0.97	>	1.08	>	1.62	>	1.70
−8	2	5	3.69	2.89	3.15	2.92	3.33	2.94	3.33	3.06	3.85	3.84	3.64	3.28
−7	2	5	5.08	5.55	5.02	5.61	5.61	5.60	3.51	4.22	3.05	4.18	3.22	3.78
−6	2	5	5.98	6.05	6.35	6.24	6.42	6.65	4.52	4.52	4.71	4.69	3.92	3.91
−5	2	5	3.66	3.33	3.89	3.39	?	3.12	>	1.67	>	1.11	>	0.30
−4	2	5	>	0.18	>	0.23	>	0.07	>	−0.59	>	−1.30	?	−1.83
−3	2	5	>	−0.12	>	−0.05	>	0.00	>	0.69	>	1.28	>	0.29
−2	2	5	>	0.68	>	0.38	>	0.59	>	−0.78	>	−0.97	>	−0.48
−1	2	5	>	−0.45	>	−0.54	>	−0.36	>	0.38	>	0.48	>	0.01
0	2	5	>	0.77	>	0.45	>	0.65	>	0.19	>	0.28	>	1.21
1	2	5	>	−1.32	1.72	−1.67	>	−1.51	>	−1.33	>	−2.13	>	−1.32
2	2	5	2.90	−2.91	2.74	−3.01	>	−2.93	>	−1.26	>	−1.32	>	−0.97
3	2	5	>	−1.57	1.59	−1.78	>	−1.60	>	−0.70	>	−0.64	>	−0.10
4	2	5	1.75	−1.42	1.54	− 1.41	>	−1.68	>	−1.51	?	−1.93	?	−2.43
5	2	5	>	0.16	>	0.25	>	0.49	>	1.12	?	1.95	>	1.24
6	2	5	>	−0.98	>	−1.04	>	−1.36	>	−1.76	?	−2.34	?	−2.78
7	2	5	>	0.90	0.56	0.98	>	1.15	>	0.85	>	1.03	>	0.86
8	2	5	.	.	.	.	.	.	.	.	.	.	>	0.36
−13	2	6	.	.	.	.	.	.	>	0.45	>	0.21	>	0.08
−12	2	6	>	−0.26	>	−0.21	>	−0.12	>	−0.10	>	−0.57	>	−0.37
−11	2	6	>	−0.12	>	−0.05	>	−0.50	>	−0.18	>	−0.31	>	−0.15
−10	2	6	>	0.95	1.39	1.17	>	1.03	>	1.24	>	1.37	>	1.14
−9	2	6	2.56	2.15	2.37	2.29	2.48	2.05	2.44	2.16	2.19	2.88	2.18	2.64
−8	2	6	3.14	2.77	2.76	2.79	2.67	2.96	1.90	2.03	2.48	2.68	>	2.08
−7	2	6	4.38	3.96	3.91	4.10	3.92	4.03	2.38	2.60	2.79	2.71	>	1.70
−6	2	6	>	−0.92	>	−1.11	>	−1.37	>	−0.52	>	−0.05	>	−0.75
−5	2	6	>	0.74	>	0.71	>	0.72	>	−0.54	>	−1.08	>	−1.75
−4	2	6	>	−1.10	>	−0.95	>	−0.91	>	−0.01	>	−0.01	>	0.26
−3	2	6	>	0.27	>	0.07	>	0.22	>	0.07	>	−0.47	>	−0.40
−2	2	6	>	0.75	>	1.11	>	1.24	>	0.28	>	−0.04	>	0.52
−1	2	6	>	0.64	>	0.22	>	0.57	>	−0.03	>	−0.36	>	−0.18
0	2	6	→	−2.28	1.61	−2.18	→	−2.32	1.50	−2.11	?	−2.60	3.45	−2.83
1	2	6	1.83	−1.88	2.09	−2.42	>	−2.23	>	−1.90	>	−1.97	>	−1.38
2	2	6	>	0.21	>	0.38	>	0.21	>	−0.41	>	−0.77	>	−0.81
3	2	6	2.70	−2.65	2.87	−3.03	>	−3.04	>	−1.20	>	−0.72	>	−0.45
4	2	6	?	2.15	?	2.22	>	2.03	>	0.69	>	0.67	>	0.55
5	2	6	>	−0.30	>	−0.45	>	−0.30	>	0.27	>	0.62	>	0.82
6	2	6	.	.	.	.	.	.	.	.	.	.	>	0.48
−11	2	7	.	.	.	.	.	.	>	−0.83	>	−1.10	>	−0.59
−10	2	7	>	−0.90	0.95	−0.82	>	−0.83	>	−0.54	>	−0.83	>	−1.06
−9	2	7	>	0.42	>	0.44	>	0.32	>	0.46	>	1.12	>	0.62
−8	2	7	>	−1.40	1.76	−1.51	>	−1.70	>	−1.61	>	−1.71	>	−2.15
−7	2	7	1.11	1.38	1.61	1.38	>	1.37	>	0.93	>	1.23	>	0.59
−6	2	7	1.13	−1.48	?	−1.41	>	−1.54	>	−0.41	>	−0.19	>	0.16
−5	2	7	>	0.80	>	0.68	>	0.72	>	0.64	>	0.45	>	0.98
−4	2	7	>	0.41	>	0.79	>	0.90	>	0.80	>	0.85	>	1.36
−3	2	7	>	0.48	>	0.33	>	0.51	>	0.35	>	0.39	>	0.40

TABLE 2 (*cont.*)

			mole % *N*-oxyphenazine									
			photometer 0%		l.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\lvert F_o \rvert$	F_c	$\lvert F_o \rvert$	F_c	$\lvert F_o \rvert$	F_c	$\lvert F_o \rvert$	F_c	$\lvert F_o \rvert$	F_c	$\lvert F_o \rvert$	F_c
−2	2	7	>	−0.36	>	−0.50	>	−0.67	>	−0.90	>	−1.01	>	−1.64
−1	2	7	>	−0.42	>	−0.69	>	−0.41	>	−1.29	>	−1.64	>	−2.01
0	2	7	→	1.21	→	1.03	>	1.13	>	0.82	>	0.75	>	1.01
1	2	7	1.60	−2.23	†	−2.56	†	−2.80	>	−2.26	>	−2.89	>	−2.42
2	2	7	†	2.94	†	3.14	†	3.24	>	2.16	>	2.35	†	3.01
1	3	0	–	−4.41	–	−4.22	–	−4.21	–	−3.02	–	−2.23	–	−2.53
2	3	0	†	−3.36	†	−3.61	†	−3.54	–	−3.40	–	−3.68	†	−4.30
3	3	0	2.55	2.14	2.20	2.00	2.43	2.24	2.67	2.38	2.31	2.13	1.94	1.71
4	3	0	1.09	0.76	1.00	0.77	>	0.33	>	−0.44	>	−0.89	>	−0.54
5	3	0	>	−1.11	1.16	−1.14	>	−1.01	>	−0.58	>	−0.91	>	−0.68
6	3	0	>	0.03	>	−0.10	>	0.20	>	0.38	>	0.78	?	1.62
7	3	0	>	0.65	>	0.28	>	0.07	>	−0.72	>	−1.11	>	−0.17
8	3	0	4.68	−4.64	5.37	−4.86	4.59	−4.41	2.12	−2.19	2.08	−1.97	1.92	−1.02
9	3	0	>	−0.66	>	−0.94	>	−1.39	?	−2.18	?	−2.90	1.97	−2.20
10	3	0	2.66	−2.53	2.57	−2.88	>	−2.61	>	−1.17	>	−1.10	>	−0.49
11	3	0	>	−1.14	0.88	−1.01	>	−1.36	>	−1.34	→	−1.69	→	−2.13
12	3	0	>	0.22	>	0.06	>	0.06	>	−0.12	>	0.04	>	−0.29
13	3	0	>	0.47	>	0.56	>	0.42	>	0.37	>	0.35	>	−0.28
14	3	0	.	.	.	.	.	.	.	.	.	.	>	−0.68
−14	3	1	>	0.01	>	−0.07	>	−0.06	>	−0.02	>	−0.14	>	0.13
−13	3	1	>	−0.54	>	−0.35	>	−0.54	>	−0.72	>	−0.99	>	−0.92
−12	3	1	>	−0.74	0.83	−0.86	>	−0.86	>	−0.72	>	−0.86	>	−0.79
−11	3	1	2.64	−3.03	2.76	−3.10	?	−3.36	2.31	−2.87	3.20	−3.61	2.53	−2.87
−10	3	1	>	−0.60	0.92	−0.93	>	−0.98	>	−0.60	>	−0.18	>	−0.58
−9	3	1	>	−0.76	>	−1.08	>	−1.06	>	−1.43	>	−1.30	>	−0.91
−8	3	1	>	−0.72	>	−0.91	>	−1.37	>	−0.81	>	−0.51	>	−0.90
−7	3	1	>	0.45	>	0.58	>	0.65	>	0.39	>	0.87	>	1.11
−6	3	1	>	0.50	>	0.39	>	0.31	>	−0.20	>	−0.65	>	−1.20
−5	3	1	>	0.60	0.79	0.62	>	0.82	>	0.84	>	0.35	>	0.40
−4	3	1	3.17	2.84	3.30	3.02	3.08	2.92	1.73	1.67	1.18	0.92	>	0.84
−3	3	1	5.29	−5.70	5.96	−5.99	6.37	−6.10	6.09	−6.45	5.92	−6.79	7.09	−7.41
−2	3	1	4.40	5.11	5.12	5.67	5.01	5.38	4.13	4.42	3.94	4.21	3.91	4.63
−1	3	1	–	−5.52	–	−5.51	–	−5.41	–	−5.22	–	−5.11	–	−5.37
0	3	1	–	2.41	–	2.19	–	2.78	–	2.88	–	2.99	†	3.50
1	3	1	2.35	2.32	2.44	2.31	2.69	2.60	2.28	2.53	2.57	2.53	2.63	2.65
2	3	1	>	−0.33	0.88	−0.70	>	−0.62	>	−0.29	>	−0.83	>	−0.26
3	3	1	2.27	−2.02	2.10	−1.90	2.43	−2.16	1.97	−1.75	1.97	−1.68	2.40	−1.74
4	3	1	>	−0.33	0.69	−0.64	>	−0.61	>	−0.92	>	−0.86	>	−0.68
5	3	1	?	2.03	2.46	2.28	2.43	2.07	2.62	2.24	2.75	2.35	2.66	2.21
6	3	1	5.61	−5.58	5.66	−5.90	?	−6.02	5.51	−5.43	4.85	−5.26	5.89	−5.54
7	3	1	?	6.68	?	6.84	5.64	6.67	4.64	5.50	5.67	5.42	5.28	5.27
8	3	1	4.68	−5.41	4.64	−5.83	5.53	−5.87	4.53	−4.63	6.13	−4.78	4.26	−4.16
9	3	1	>	0.40	>	0.41	>	0.21	>	−0.01	>	−0.17	>	−0.33
10	3	1	>	−0.19	>	−0.40	>	−0.29	>	−0.21	>	0.26	>	0.57
11	3	1	>	0.34	>	0.17	>	0.14	>	0.38	>	0.47	>	0.20
12	3	1	>	0.07	>	0.09	>	−0.04	>	−0.19	>	0.03	>	−0.05
13	3	1	>	0.13	>	−0.04	>	−0.06	>	0.11	>	−0.13	>	−0.04
−15	3	2	.	.	.	.	.	.	>	−0.28	>	−0.29	>	0.00
−14	3	2	>	−0.26	>	−0.38	>	−0.36	>	−0.26	>	−0.45	>	−0.47
−13	3	2	†	−1.74	1.77	−1.63	†	−1.84	>	−1.14	>	−1.33	>	−0.51
−12	3	2	??	−1.58	1.53	−1.89	??	−1.57	>	−0.97	>	−1.25	>	−1.18
−11	3	2	>	−0.47	>	−0.42	>	−0.19	>	−0.36	>	−0.19	>	−0.17
−10	3	2	>	−0.98	1.10	−1.22	>	−1.24	>	−0.29	>	−0.08	>	0.16
−9	3	2	>	0.63	>	0.88	>	0.89	>	−0.04	>	0.57	>	0.12
−8	3	2	>	0.75	>	0.74	>	0.65	>	0.33	>	0.99	1.22	1.12
−7	3	2	>	−0.20	>	−0.09	>	−0.11	>	−0.13	>	0.09	>	0.12

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TABLE 2 (*cont.*)

			photometer 0%		I.d.t. 0%		mole % *N*-oxyphenazine: 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
−6	3	2	2.64	−2.15	2.70	−2.38	3.01	−2.12	2.53	−2.22	2.31	−1.71	>	−1.51
−5	3	2	2.50	−2.25	2.50	−2.51	2.92	−2.44	3.52	−3.24	4.04	−3.55	3.57	−3.89
−4	3	2	4.42	−4.83	4.81	−4.61	5.14	−4.95	4.50	−4.18	4.42	−4.09	4.18	−3.70
−3	3	2	5.49	−5.82	5.98	−5.94	6.36	−6.09	6.92	−6.48	6.63	−6.62	7.41	−7.16
−2	3	2	>	0.49	>	0.62	>	0.61	>	0.15	>	−0.15	>	0.24
−1	3	2	>	−0.11	>	0.00	>	0.25	>	0.47	>	0.02	>	0.14
0	3	2	>	0.12	>	−0.15	>	−0.04	>	0.06	>	−0.39	>	−0.44
1	3	2	→	−1.14	1.24	−0.90	1.15	−1.16	1.08	−1.02	1.51	−1.37	→	−1.04
2	3	2	1.74	−1.37	1.64	−1.31	>	−1.12	>	−0.16	>	0.38	>	0.19
3	3	2	>	0.18	>	0.70	>	0.46	1.26	1.60	1.99	2.41	2.90	2.73
4	3	2	1.82	2.10	2.39	2.42	1.91	2.35	>	0.97	>	1.26	>	0.51
5	3	2	†	9.26	8.26	9.79	†	10.21	9.96	11.02	10.74	12.48	12.63	12.41
6	3	2	>	0.53	>	0.77	>	0.33	>	−1.79	>	−2.59	3.67	−3.04
7	3	2	3.31	3.13	3.23	3.26	3.50	3.39	3.83	3.56	4.40	4.07	3.73	3.87
8	3	2	>	−0.03	>	−0.03	>	0.11	>	−0.56	>	−0.76	>	−0.34
9	3	2	>	−0.58	>	−0.86	>	−0.84	>	−0.53	>	−0.55	>	−0.71
10	3	2	>	0.41	>	0.50	>	0.41	>	0.32	>	0.67	>	0.51
11	3	2	>	0.78	>	0.45	>	0.57	>	0.41	>	0.29	>	0.26
12	3	2	>	−0.25	>	−0.42	>	−0.47	>	−0.11	>	−0.11	>	−0.15
−15	3	3	.	.	.	.	.	.	>	−0.74	>	−0.82	>	−0.47
−14	3	3	>	0.18	>	0.32	>	0.49	>	0.31	>	0.31	>	0.49
−13	3	3	>	−0.25	>	−0.30	>	−0.19	>	0.06	>	0.30	>	0.35
−12	3	3	>	−0.26	>	−0.42	>	−0.11	>	−0.26	>	−0.54	>	−0.11
−11	3	3	>	0.39	>	0.37	>	0.43	>	−0.30	>	−0.07	>	0.10
−10	3	3	>	0.48	>	0.46	>	0.39	>	−0.23	>	−0.23	>	−0.37
−9	3	3	>	−0.52	>	−0.68	>	−0.90	>	−0.83	>	−0.46	>	0.20
−8	3	3	??	−2.43	2.95	−2.83	2.20	−2.54	1.82	−2.37	>	−1.77	2.39	−2.01
−7	3	3	4.40	4.08	4.30	4.02	4.33	4.50	3.45	3.40	3.44	3.48	3.06	3.53
−6	3	3	7.64	−8.29	8.04	−8.72	8.30	−8.48	6.76	−5.50	5.45	−4.72	4.43	−3.98
−5	3	3	1.74	1.46	1.61	1.74	>	1.79	>	0.78	>	0.67	>	0.42
−4	3	3	4.46	−4.11	3.99	−3.76	→	−3.65	2.66	−1.95	2.05	−1.38	>	−0.33
−3	3	3	>	−1.11	?	−1.00	>	−1.15	>	−0.55	>	−0.35	>	−0.21
−2	3	3	>	0.75	1.22	1.09	>	1.11	>	0.52	>	0.21	>	0.14
−1	3	3	>	0.81	1.52	0.71	>	0.63	>	0.46	>	0.25	>	0.02
0	3	3	>	−1.11	0.90	−0.96	>	−1.23	1.20	−1.05	>	−1.72	>	−2.65
1	3	3	2.35	−1.76	2.24	−1.55	1.92	−1.88	1.28	−1.15	>	−1.34	>	−1.72
2	3	3	4.99	4.78	4.96	5.17	5.08	5.19	4.70	4.76	4.92	4.63	4.58	4.30
3	3	3	?	2.40	2.35	2.60	2.92	2.97	5.02	4.56	6.03	5.35	6.32	5.70
4	3	3	7.12	6.97	?	7.52	5.65	7.32	5.38	5.41	5.08	5.49	4.52	4.87
5	3	3	2.33	2.56	2.81	2.60	2.64	3.00	??	3.28	??	3.78	4.39	4.31
6	3	3	>	0.89	0.92	1.00	>	1.18	>	0.64	>	0.76	>	0.64
7	3	3	>	−1.05	→	−1.07	>	−1.01	>	−1.14	>	−1.18	>	−1.04
8	3	3	>	−0.40	>	−0.31	>	−0.17	>	0.21	>	0.18	>	0.14
9	3	3	>	0.83	0.67	0.76	>	0.93	>	0.67	>	0.92	>	0.32
10	3	3	>	0.29	>	0.32	>	0.02	>	−0.17	>	−0.51	>	−0.80
11	3	3	.	.	.	.	.	.	>	−0.43	>	−0.55	>	−0.78
−13	3	4	>	0.38	>	0.17	>	0.39	>	1.10	>	0.09	>	0.41
−12	3	4	>	−0.53	0.69	−0.58	>	−0.52	>	1.16	>	−0.04	>	−0.16
−11	3	4	>	−0.34	>	−0.68	>	−1.01	>	1.88	>	−0.23	>	0.42
−10	3	4	>	0.07	>	0.10	>	0.15	>	1.92	>	0.04	>	−0.58
−9	3	4	4.12	4.53	4.29	4.39	4.73	4.60	4.34	4.30	5.15	5.35	4.96	5.24
−8	3	4	1.31	−1.29	1.61	−1.59	>	−1.43	>	−1.47	?	−2.08	1.86	−2.17
−7	3	4	6.73	6.75	6.33	7.28	7.22	7.47	5.91	5.95	5.98	6.00	5.73	5.18
−6	3	4	1.94	−1.73	1.91	−1.60	>	−1.56	>	−1.03	>	−1.20	>	−1.03
−5	3	4	>	−0.33	>	0.00	>	−0.44	>	−0.11	>	−0.41	1.55	−0.96

TABLE 2 (*cont.*)

			photometer 0%		l.d.t. 0%		mole % *N*-oxyphenazine 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
−4	3	4	>	0.59	>	0.97	>	1.23	>	1.25	>	1.92	2.43	2.14
−3	3	4	>	0.43	>	0.26	>	0.42	>	−0.01	>	0.17	>	−0.08
−2	3	4	>	−0.58	>	−0.70	>	−0.78	>	−0.36	>	−0.33	>	0.06
−1	3	4	>	−0.77	>	−0.72	>	−0.67	>	−1.04	>	−1.34	>	−1.60
0	3	4	>	0.71	>	0.49	>	0.40	>	1.39	>	0.95	>	1.80
1	3	4	4.23	−3.87	3.99	−3.90	3.66	−3.94	3.18	−2.87	3.94	−3.18	?	−3.39
2	3	4	3.56	3.07	3.34	3.33	3.55	3.31	3.29	3.46	3.63	3.67	3.81	3.43
3	3	4	>	−0.79	0.92	−1.06	>	−0.86	>	−0.61	>	−0.61	>	−0.90
4	3	4	>	0.39	>	0.66	>	0.43	>	0.26	>	0.70	>	−0.57
5	3	4	>	−0.32	0.79	−0.29	>	−0.14	>	−0.47	>	−0.61	>	−0.50
6	3	4	>	−0.59	>	−0.45	>	−0.43	>	−0.11	>	−0.15	>	−0.45
7	3	4	>	0.53	0.69	0.65	>	0.80	>	0.62	>	0.82	>	1.12
8	3	4	>	0.82	>	1.21	>	1.06	>	0.88	>	0.67	>	0.56
9	3	4	>	−0.38	>	−0.48	>	−0.39	>	−0.33	>	−0.04	>	−0.09
−14	3	5	.	.	.	.	.	.	>	−0.12	>	−0.28	>	−0.28
−13	3	5	>	−0.02	>	−0.06	>	−0.24	>	0.06	>	−0.29	>	−0.12
−12	3	5	>	0.64	>	0.98	>	0.82	>	1.07	>	1.33	>	0.97
−11	3	5	>	0.96	>	0.90	>	0.65	>	1.17	>	1.74	1.12	1.47
−10	3	5	3.76	3.82	3.92	4.09	3.55	4.10	2.26	2.72	2.44	2.82	2.07	2.45
−9	3	5	3.86	4.07	4.10	4.18	4.06	4.28	3.49	3.57	3.89	4.28	3.03	3.33
−8	3	5	2.91	2.64	2.64	2.60	→	2.53	>	0.85	>	0.50	>	0.11
−7	3	5	>	0.32	0.83	0.50	>	0.09	>	0.32	>	0.10	>	−0.95
−6	3	5	>	0.28	>	0.35	>	0.47	>	−0.08	>	−0.23	>	−0.40
−5	3	5	>	−0.52	>	−0.65	>	−0.51	>	−0.46	>	−0.66	>	−1.10
−4	3	5	>	0.32	>	0.24	>	0.24	>	0.21	>	−0.14	>	0.37
−3	3	5	>	0.69	0.83	0.61	>	1.17	>	0.20	>	−0.04	>	0.10
−2	3	5	>	−0.90	>	−1.19	>	−1.21	>	−0.59	>	−0.72	>	−0.01
−1	3	5	3.17	−3.10	2.89	−3.16	2.91	−3.09	2.83	−3.13	3.46	−3.76	2.56	−3.30
0	3	5	>	−1.82	1.68	−1.87	>	−1.76	>	−0.16	>	0.30	>	0.52
1	3	5	1.97	−1.78	2.11	−2.08	>	−2.10	2.03	−2.25	2.95	−2.23	>	−1.90
2	3	5	>	−1.10	0.79	−0.90	>	−1.17	>	−0.45	>	−0.16	>	−1.05
3	3	5	>	0.67	>	0.51	>	0.57	>	0.37	>	0.80	>	0.74
4	3	5	>	0.38	>	0.38	>	0.30	>	−0.21	>	−0.47	>	−0.84
5	3	5	>	0.31	>	0.34	>	0.38	>	0.59	>	0.60	>	0.91
6	3	5	>	0.62	>	0.73	>	0.74	>	0.68	>	0.59	>	0.84
7	3	5	>	−0.55	>	−0.68	>	−0.90	>	−0.96	>	−1.34	>	−0.97
−12	3	6	1.65	1.77	1.53	2.07	>	1.93	>	1.11	>	1.36	>	1.12
−11	3	6	>	−0.17	>	−0.33	>	−0.34	>	−0.22	>	−0.20	>	−0.32
−10	3	6	2.04	2.08	?	2.11	>	1.84	>	0.92	>	1.40	>	0.70
−9	3	6	>	0.10	>	−0.08	>	−0.08	>	−0.08	>	0.00	>	−0.47
−8	3	6	>	−0.29	>	−0.40	>	−0.58	>	−0.47	>	−0.18	>	−0.44
−7	3	6	>	−0.27	>	−0.33	>	−0.19	>	0.21	>	0.14	>	0.23
−6	3	6	>	0.05	>	0.20	>	0.22	>	0.54	>	0.18	>	0.80
−5	3	6	>	0.45	>	0.32	>	0.75	>	0.38	>	0.44	>	0.76
−4	3	6	>	0.89	>	1.00	>	0.99	>	−0.05	>	−0.71	>	−0.48
−3	3	6	>	−1.56	2.71	−1.80	>	−1.70	>	−1.37	>	−1.48	>	−1.72
−2	3	6	>	−0.60	>	−0.82	>	−0.72	>	−1.32	>	−1.58	>	−2.04
−1	3	6	>	−1.01	0.88	−1.10	>	−1.12	>	−1.13	>	−1.33	>	−0.83
0	3	6	>	−0.51	>	−0.76	>	−0.87	>	−0.17	>	−0.22	>	−0.23
1	3	6	>	0.57	>	0.62	>	0.49	>	0.28	>	0.61	>	1.30
2	3	6	>	0.87	>	0.58	>	0.64	>	0.45	>	0.67	>	0.96
3	3	6	>	−0.29	>	−0.22	>	−0.32	>	−0.01	>	0.28	>	0.35
4	3	6	>	−0.03	>	−0.38	>	−0.23	>	−0.26	>	−0.03	>	0.48
5	3	6	>	0.10	.	.	>	−0.10	>	−0.58	>	−1.13	>	−1.08
−10	3	7	>	−0.13	>	−0.18	>	−0.49	>	−0.44	>	−0.40	>	−0.68
−9	3	7	>	0.28	>	0.28	>	0.37	>	0.52	>	0.64	>	0.50

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

TABLE 2 (*cont.*)

			mole % *N*-oxyphenazine									
			photometer 0%		I.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
−8	3	7	>	0.21	>	0.24	>	0.12	>	0.19	>	0.30	>	0.60
−7	3	7	>	−0.12	>	−0.10	>	0.02	>	−0.02	>	−0.24	>	−0.03
−6	3	7	0.81	0.91	?	1.04	>	1.00	>	0.90	>	1.13	>	1.05
−5	3	7	>	−1.54	?	−1.80	>	−1.95	>	−1.40	>	−1.22	>	−1.47
−4	3	7	>	1.21	?	1.03	>	1.22	>	0.14	>	−0.02	>	−0.29
−3	3	7	>	−1.01	†	−1.11	>	−1.10	>	−0.40	>	−0.35	>	−0.06
−2	3	7	>	0.64	>	0.24	>	0.37	>	0.01	>	−0.61	>	0.13
−1	3	7	>	0.07	>	0.33	>	0.28	>	0.21	>	−0.01	>	0.63
0	3	7	>	0.21	>	0.04	>	0.08	>	0.24	>	0.22	>	0.65
1	3	7	>	−0.73	>	−0.58	>	−0.85	>	−0.54	>	−0.40	>	−0.86
0	4	0	–	0.90	–	0.35	–	0.80	–	0.81	–	0.60	–	0.21
1	4	0	0.81	1.04	†	1.03	>	0.60	>	−0.06	>	−0.47	>	−0.18
2	4	0	1.41	−1.32	1.35	−1.31	1.42	−1.26	1.15	−1.05	1.12	−0.99	>	−0.80
3	4	0	>	0.64	>	0.24	>	0.28	>	−0.01	>	0.15	>	0.47
4	4	0	0.80	−1.10	1.46	−1.18	1.81	−1.15	1.76	−1.08	1.57	−0.99	>	−0.52
5	4	0	2.35	−2.24	2.48	−2.59	2.41	−2.56	1.57	−1.72	1.70	−1.44	>	−0.90
6	4	0	1.02	−1.45	→	−1.86	>	−1.56	>	−1.01	>	−1.21	>	−0.92
7	4	0	2.35	−2.29	2.47	−2.43	2.54	−2.71	1.83	−2.24	1.89	−2.06	1.38	−1.66
8	4	0	0.96	−1.62	1.75	−1.75	>	−1.82	>	−1.33	>	−1.26	>	−1.26
9	4	0	>	0.00	>	0.06	>	0.17	>	0.24	>	0.49	>	0.07
10	4	0	>	0.84	>	0.61	>	0.28	>	−0.26	>	−0.15	>	−0.44
11	4	0	>	−0.47	>	−0.34	>	−0.22	>	0.37	>	0.44	.	.
12	4	0	>	−0.25	>	−0.33	>	−0.63	>	−0.92	>	−1.36	.	.
−13	4	1	>	−0.54	>	−0.72	>	−0.60	>	−0.43	>	−0.38	>	−0.18
−12	4	1	>	−0.54	>	−0.79	>	−0.79	>	−0.67	>	−0.56	>	−0.95
−11	4	1	>	−0.27	>	−0.49	>	−0.51	>	−0.20	>	0.14	>	0.67
−10	4	1	>	−0.18	>	0.02	>	−0.18	>	−0.51	>	−0.19	>	−0.67
−9	4	1	1.38	1.60	→	1.57	>	1.71	>	1.31	>	1.77	?	1.80
−8	4	1	?	−1.65	1.56	−1.68	?	−1.68	?	−1.85	?	−2.19	1.38	−1.75
−7	4	1	>	1.07	1.03	1.14	>	1.22	>	0.66	>	0.68	>	0.02
−6	4	1	1.78	−1.77	1.95	−2.06	1.82	−1.93	2.80	−2.67	2.97	−3.03	2.08	−2.42
−5	4	1	1.65	−1.96	1.80	−1.70	1.95	−2.22	2.49	−2.50	2.94	−2.81	2.78	−3.79
−4	4	1	>	−0.83	0.81	−0.65	>	−0.54	>	−1.21	>	−1.28	>	−0.49
−3	4	1	>	−0.29	0.39	−0.39	>	−0.09	>	−0.39	>	−0.60	>	−0.99
−2	4	1	>	0.49	>	0.14	>	0.64	>	0.67	>	0.39	>	0.55
−1	4	1	1.90	1.82	1.87	1.82	2.05	2.00	2.22	1.83	>	1.47	2.39	2.00
0	4	1	4.42	−4.46	4.47	−4.91	4.65	−4.84	3.77	−3.82	?	−3.61	4.87	−4.13
1	4	1	2.85	2.60	2.60	2.70	2.95	2.58	2.69	2.30	2.95	2.42	3.22	2.97
2	4	1	2.61	−2.12	2.53	−1.89	2.60	−2.08	1.27	−1.12	>	−0.37	>	−0.44
3	4	1	2.46	2.03	2.16	1.91	2.60	2.05	??	2.17	2.59	2.41	2.54	2.47
4	4	1	1.90	2.60	2.93	2.71	3.00	2.80	3.13	2.93	3.20	3.38	2.85	3.13
5	4	1	>	0.62	0.85	0.55	>	0.38	>	0.32	>	0.11	>	0.46
6	4	1	>	−1.11	0.88	−1.07	>	−1.15	>	−0.64	>	−0.62	>	−0.77
7	4	1	>	−0.01	>	−0.26	>	−0.17	>	−0.30	>	0.06	>	0.37
8	4	1	1.44	1.63	1.65	1.68	>	1.46	>	1.04	>	1.07	>	1.21
9	4	1	?	−2.21	1.95	−2.47	?	−2.38	>	−1.40	>	−0.97	>	−1.04
10	4	1	?	2.70	?	2.71	2.15	2.53	1.51	1.40	>	1.21	>	1.19
11	4	1	>	−1.87	?	−2.25	>	−2.00	>	−0.94	>	−0.92	>	−0.54
−13	4	2	>	−0.02	>	−0.15	>	−0.07	>	−0.01	>	0.09	>	0.19
−12	4	2	>	0.05	>	0.21	>	0.24	>	−0.33	>	−0.08	>	−0.01
−11	4	2	>	0.45	>	0.28	>	0.29	>	−0.08	>	0.22	>	0.12
−10	4	2	>	−0.62	>	−0.42	>	−0.61	>	−0.93	>	−0.74	>	−0.15
−9	4	2	>	−0.55	1.23	−0.99	>	−0.60	>	−0.30	>	0.25	>	0.11
−8	4	2	0.96	−1.12	?	−1.30	>	−0.98	>	−1.48	>	−1.36	>	−1.13
−7	4	2	3.28	−3.45	3.56	−3.64	3.72	−3.78	2.62	−2.45	2.26	−2.19	1.84	−2.03
−6	4	2	2.52	−2.30	2.38	−2.15	2.38	−2.04	2.30	−2.06	2.44	−1.85	2.14	−1.66

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

TABLE 2 (*cont.*)

			mole % *N*-oxyphenazine											
			photometer 0%		I.d.t. 0%		8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
−5	4	2	1.61	−1.37	1.76	−1.40	1.84	−1.48	1.89	−1.54	1.77	−1.56	1.53	−1.44
−4	4	2	>	−0.24	>	−0.04	>	−0.01	>	0.24	>	0.02	>	0.46
−3	4	2	>	0.57	>	0.55	>	0.77	>	0.72	>	0.39	>	0.46
−2	4	2	??	−1.43	1.75	−1.72	??	−1.81	?	−1.90	?	−2.42	3.12	−3.26
−1	4	2	>	0.77	?	1.24	>	1.07	?	2.07	?	1.97	2.82	2.19
0	4	2	2.35	−1.92	2.08	−1.70	2.55	−1.98	1.55	−1.51	2.04	−1.46	2.76	−2.29
1	4	2	4.81	4.94	4.64	5.17	5.45	5.32	5.30	5.46	5.66	5.82	6.58	6.29
2	4	2	5.21	5.56	5.51	6.14	5.82	6.18	5.36	5.75	5.71	5.99	6.07	6.20
3	4	2	4.87	4.95	4.59	4.96	5.15	5.14	4.72	4.65	4.61	4.66	4.87	5.03
4	4	2	2.69	2.45	2.81	2.85	2.96	2.67	2.22	2.05	2.20	1.97	2.24	2.15
5	4	2	>	0.48	>	0.25	>	0.57	>	0.24	>	0.28	>	0.59
6	4	2	>	0.09	>	0.33	>	0.24	>	0.75	>	0.84	>	0.88
7	4	2	1.11	−1.82	2.16	−2.11	2.27	−2.15	2.58	−2.57	2.70	−2.73	2.97	−3.23
8	4	2	3.08	3.36	2.93	3.43	2.60	3.62	3.26	3.71	3.46	4.18	?	3.84
9	4	2	1.75	−2.26	†	−2.60	1.87	−2.76	2.31	−2.84	2.77	−3.36	2.97	−3.53
10	4	2	0.68	0.49	>	0.28	>	0.35	>	0.64	>	0.60	>	0.28
11	4	2	>	−0.85	>	−1.15	>	−0.97	>	−0.81	>	−0.99	.	.
−13	4	3	>	0.57	>	0.53	>	0.49	>	0.18	>	0.25	>	0.27
−12	4	3	>	−0.82	>	−1.03	>	−1.24	>	−0.78	>	−0.51	>	−0.12
−11	4	3	>	0.60	>	0.28	>	0.55	>	0.28	>	0.41	>	0.42
−10	4	3	>	0.58	>	0.46	>	0.66	>	0.72	>	1.29	>	1.19
−9	4	3	>	−0.41	1.02	−0.74	>	−0.42	>	0.40	>	0.31	>	0.59
−8	4	3	>	−0.48	>	−0.24	>	−0.06	>	0.13	>	0.35	>	0.04
−7	4	3	>	−0.45	>	−0.19	>	−0.22	>	0.24	>	0.32	>	0.40
−6	4	3	>	−0.70	>	−0.17	>	−0.44	>	0.28	>	0.20	>	0.57
−5	4	3	1.56	−1.43	1.85	−1.39	>	−1.28	>	−0.97	>	−0.30	>	−0.46
−4	4	3	3.74	3.53	4.18	3.82	3.85	3.67	2.58	2.28	2.27	1.69	1.53	1.57
−3	4	3	3.65	−3.78	3.88	−3.71	4.11	−3.84	2.49	−1.97	1.67	−1.75	1.50	−1.79
−2	4	3	2.21	1.49	1.73	1.71	>	1.30	>	0.32	>	−0.40	>	−1.43
−1	4	3	>	0.30	>	0.54	>	0.55	>	1.23	>	0.88	>	1.31
0	4	3	2.61	2.24	2.85	2.41	2.46	2.26	2.15	2.44	2.18	2.24	1.81	1.96
1	4	3	3.85	3.52	3.46	3.60	3.86	3.65	2.76	2.96	2.89	2.78	2.57	2.64
2	4	3	3.23	2.88	2.95	3.02	3.07	3.01	2.55	2.36	2.39	2.42	2.67	2.54
3	4	3	>	0.50	>	0.48	>	0.70	>	0.43	>	0.58	>	0.21
4	4	3	>	−1.39	1.23	−1.23	>	−1.24	>	−0.81	>	−0.70	>	−0.53
5	4	3	>	0.23	>	0.35	>	0.52	>	−0.18	>	−0.43	>	−0.72
6	4	3	>	0.23	>	0.26	>	0.60	2.06	1.82	2.25	2.35	2.79	2.62
7	4	3	>	0.42	>	0.63	>	0.32	1.49	−1.00	1.79	−1.61	>	−2.36
8	4	3	>	0.26	>	0.14	>	0.28	>	0.67	>	0.97	>	0.89
9	4	3	>	−0.45	>	−0.51	>	−0.49	>	−0.97	>	−1.30	>	−1.38
−13	4	4	>	1.21	>	1.42	>	1.31	>	1.10	>	1.44	.	.
−12	4	4	>	1.02	>	0.89	>	0.85	>	1.16	>	1.58	>	1.16
−11	4	4	>	2.28	?	2.28	>	2.47	>	1.88	>	2.03	>	1.82
−10	4	4	2.17	2.45	†	2.64	>	2.66	>	1.92	>	1.99	>	1.54
−9	4	4	?	1.22	?	1.55	>	1.57	>	1.28	>	0.96	>	0.22
−8	4	4	>	−0.01	>	0.23	>	−0.18	>	0.14	>	0.01	>	0.15
−7	4	4	>	−1.26	1.03	−0.94	>	−0.95	>	−1.08	>	−1.29	>	−2.19
−6	4	4	3.56	3.26	3.46	3.48	3.39	3.59	2.71	3.04	2.83	3.11	3.06	3.59
−5	4	4	4.30	−4.33	4.78	−4.58	4.49	−4.61	3.37	−3.65	3.36	−3.60	3.52	−3.70
−4	4	4	3.40	3.65	3.75	3.91	3.79	4.04	2.62	2.64	2.42	2.25	2.54	2.44
−3	4	4	?	−3.21	?	−3.31	?	−3.15	?	−2.16	>	−2.19	?	−1.49
−2	4	4	?	−1.44	1.51	−1.52	?	−1.76	?	−1.19	>	−1.52	?	−1.61
−1	4	4	>	−0.89	>	−0.79	>	−0.65	>	−0.44	>	−0.26	>	−0.01
0	4	4	>	−0.25	>	−0.49	>	−0.53	>	−0.39	>	−0.09	>	−0.59
1	4	4	>	−0.50	>	−0.49	>	−0.70	>	−0.60	>	−0.41	>	−1.08

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

TABLE 2 (*cont.*)

			photometer 0%		I.d.t. 0%		mole % *N*-oxyphenazine 8%		52%		81%		100%	
h	*k*	*l*	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c	$\|F_o\|$	F_c
2	4	4	>	−0.54	>	−0.42	>	−0.51	>	−0.78	>	−0.61	>	−1.38
3	4	4	1.27	1.51	1.50	1.56	>	1.71	>	1.45	>	1.29	>	1.39
4	4	4	1.16	−1.40	1.44	−1.37	>	−1.30	>	−0.70	>	−0.86	>	−0.74
5	4	4	2.38	2.07	2.20	2.48	?	2.31	?	1.63	>	1.47	>	1.49
6	4	4	>	−0.50	>	−0.65	>	−0.48	>	−0.36	>	−0.45	>	0.17
7	4	4	>	0.10	>	0.28	>	0.14	>	−0.26	>	−0.10	>	−0.47
8	4	4	>	−0.33	>	−0.37	>	−0.34	.	.	.	.	.	.
−12	4	5	>	2.15	†	2.15	>	2.10	>	1.31	>	1.43	.	.
−11	4	5	0.74	1.26	?	1.32	>	1.22	>	0.70	>	0.99	>	0.25
−10	4	5	>	0.71	>	0.55	>	0.27	>	−0.03	>	−0.08	>	−0.14
−9	4	5	>	−0.10	>	0.12	>	0.17	>	0.00	>	−0.19	>	−0.82
−8	4	5	>	−0.07	>	−0.29	>	−0.25	>	0.50	>	0.78	>	1.11
−7	4	5	0.93	−0.56	>	−0.59	>	−0.46	>	−1.15	>	−1.96	>	−1.81
−6	4	5	>	1.58	1.51	1.63	>	2.01	1.67	1.97	1.71	2.08	2.05	2.29
−5	4	5	>	−1.18	1.52	−1.44	>	−1.28	1.69	−1.90	2.34	−2.36	1.56	−2.04
−4	4	5	?	−1.69	?	−1.67	>	−1.80	>	−1.23	>	−1.57	?	−1.88
−3	4	5	>	−1.43	?	−1.47	>	−1.23	>	−1.09	>	−0.90	>	−0.43
−2	4	5	1.78	−1.76	?	−2.01	>	−2.00	>	−1.48	>	−1.39	>	−1.68
−1	4	5	1.19	−0.70	>	−0.73	>	−1.09	>	−0.43	>	−0.08	>	0.30
0	4	5	>	0.49	>	0.48	>	0.60	>	0.03	>	0.36	>	0.13
1	4	5	1.61	1.05	0.97	0.83	>	0.72	>	0.93	>	1.26	>	1.77
2	4	5	>	−0.47	>	−0.47	>	−0.53	>	−1.03	>	−1.23	>	−1.29
3	4	5	0.99	1.03	0.88	1.03	>	1.03	>	1.28	>	1.36	>	1.78
4	4	5	>	−0.72	>	−1.01	>	−1.04	>	−1.29	>	−1.71	>	−1.35
5	4	5	>	−0.83	1.02	−0.70	>	−1.01	>	−0.68	>	−0.70	>	−1.13
6	4	5	>	−0.54	>	−0.71	>	−0.63	>	−0.62	>	−0.66	.	.

List of Structure Factors for *N*-oxyphenazine at −90 °C

h	*k*	*l*	$\|F_o\|$	F_c	*h*	*k*	*l*	$\|F_o\|$	F_c	*h*	*k*	*l*	$\|F_o\|$	F_c
4	0	0	7.48	−7.20	−6	0	4	3.36	−3.15	9	1	1	3.08	−2.92
8	0	0	9.67	−9.58	0	0	4	6.94	−6.24	10	1	1	3.33	4.02
10	0	0	4.03	−4.03	2	0	4	12.06	−11.29	11	1	1	3.38	−3.10
−12	0	1	3.22	3.08	−8	0	5	3.37	3.50	−12	1	2	2.57	−2.72
−6	0	1	4.02	3.51	−4	0	5	4.97	−5.09	−11	1	2	3.21	3.33
−4	0	1	7.63	−8.18	−2	0	5	2.21	2.17	−10	1	2	4.18	−3.89
2	0	1	7.29	−7.46	−4	0	6	8.13	8.41	−8	1	2	4.50	−3.80
4	0	1	9.64	9.99	−2	0	6	7.77	8.16	−7	1	2	6.77	−6.77
6	0	1	7.29	−7.01						−6	1	2	5.82	−5.05
8	0	1	10.59	−10.56	5	1	0	7.56	−8.07	−4	1	2	2.54	−2.45
−12	0	2	5.27	5.21	6	1	0	7.06	−7.29	−3	1	2	2.15	−2.27
−10	0	2	3.34	−3.07	7	1	0	7.46	−7.06	−2	1	2	5.30	6.03
−6	0	2	3.00	−2.78	8	1	0	2.57	−2.50	−1	1	2	2.70	−2.71
−2	0	2	8.03	−9.68	−9	1	1	2.16	1.50	0	1	2	1.95	−2.08
0	0	2	6.49	−6.70	−8	1	1	1.38	1.83	1	1	2	3.97	−3.87
2	0	2	6.65	7.60	−7	1	1	4.91	−4.98	4	1	2	3.01	2.54
4	0	2	4.10	4.17	−6	1	1	2.34	−2.39	5	1	2	6.10	5.67
−8	0	3	3.01	−3.59	−5	1	1	2.62	−2.50	6	1	2	2.96	−1.97
−6	0	3	12.91	−14.55	−4	1	1	1.77	−2.03	9	1	2	3.00	−2.73
−4	0	3	3.62	−3.47	−3	1	1	2.91	−2.36	−13	1	3	3.55	3.96
−2	0	3	5.93	5.69	−1	1	1	3.02	−3.57	−12	1	3	4.61	−4.93
2	0	3	12.20	−13.46	3	1	1	11.85	12.51	−11	1	3	5.16	4.51
4	0	3	7.17	−6.75	4	1	1	3.58	−3.43	−9	1	3	5.21	−4.58
6	0	3	3.74	3.40	5	1	1	6.14	−6.23	−7	1	3	2.79	−2.90
−10	0	4	3.02	2.43	6	1	1	2.98	−2.13	−4	1	3	6.82	6.63

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TABLE 2 (*cont.*)

h	k	l	$\|F_o\|$	F_c	h	k	l	$\|F_o\|$	F_c	h	k	l	$\|F_o\|$	F_c
−3	1	3	6.47	−7.28	−2	2	3	6.25	−6.71	5	3	3	4.47	4.22
−1	1	3	9.55	−11.37	0	2	3	4.98	−5.22	−9	3	4	6.60	6.59
0	1	3	8.88	−10.25	1	2	3	3.23	3.09	−8	3	4	3.00	−2.55
1	1	3	8.69	−8.61	2	2	3	3.65	−3.22	−7	3	4	6.85	6.44
2	1	3	3.57	−3.62	5	2	3	6.81	5.98	−6	3	4	2.42	−2.22
8	1	3	3.28	3.05	6	2	3	8.28	8.31	−4	3	4	2.96	2.48
9	1	3	5.49	5.04	7	2	3	5.83	4.91	2	3	4	3.48	4.34
−5	1	4	6.77	−6.76	8	2	3	2.41	2.66	−10	3	5	3.40	3.14
−4	1	4	4.84	4.62	−11	2	4	3.03	−2.93	−9	3	5	4.77	4.22
0	1	4	4.31	−3.96	−10	2	4	4.25	3.48	−1	3	5	4.52	−4.11
3	1	4	4.91	−4.61	−2	2	4	4.76	−4.52	1	3	5	3.14	−2.55
4	1	4	3.90	3.37	−1	2	4	5.94	5.73					
5	1	4	5.15	−4.25	4	2	4	3.07	3.83	4	4	0	1.89	−1.51
6	1	4	4.51	4.07	5	2	4	4.67	4.53	6	4	0	1.95	−1.76
7	1	4	5.30	5.38	7	2	4	2.34	1.99	7	4	0	1.54	−1.99
8	1	4	4.31	4.38	−9	2	5	2.34	1.75	−9	4	1	1.55	2.01
−5	1	5	4.64	3.78	−8	2	5	4.81	4.87	−8	4	1	2.84	−3.10
−4	1	5	3.25	3.73	−7	2	5	4.35	4.17	−9	4	1	4.40	−4.24
−7	1	6	4.75	5.01	−6	2	5	4.95	4.44	−5	4	1	3.55	−3.64
−6	1	6	4.69	4.44	6	2	5	2.90	−2.79	−2	4	1	1.31	0.87
−5	1	6	6.22	5.80	−9	2	6	3.19	3.17	−1	4	1	1.26	0.57
0	1	6	2.81	−2.64	−8	2	6	3.00	2.63	0	4	1	3.96	−4.26
−2	1	7	2.96	−3.63	−7	2	6	2.82	2.68	1	4	1	3.71	3.56
−1	1	7	3.60	4.78	0	2	6	3.11	−3.46	3	4	1	3.67	3.57
					−7	2	7	2.17	1.58	4	4	1	4.01	3.73
6	2	0	4.99	5.20	−4	2	7	2.23	1.48	−7	4	2	3.09	−2.70
11	2	0	1.78	−2.20	1	2	7	2.51	−2.57	−6	4	2	2.19	−2.31
−9	2	1	2.65	−2.80						−5	4	2	2.00	−1.80
−8	2	1	3.42	−2.98	2	3	0	2.50	−2.89	−2	4	2	3.95	−2.88
−7	2	1	2.26	−1.80	3	3	0	2.76	2.69	−1	4	2	2.75	2.43
−6	2	1	4.54	−4.50	−11	3	1	3.08	−3.63	1	4	2	7.53	7.26
−5	2	1	4.03	4.15	−3	3	1	6.97	−7.70	2	4	2	6.45	7.37
−3	2	1	4.95	4.70	−2	3	1	4.55	4.17	3	4	2	5.06	4.66
1	2	1	2.50	2.53	0	3	1	2.50	3.19	4	4	2	1.65	2.01
2	2	1	3.86	4.16	1	3	1	2.38	2.50	7	4	2	3.19	−3.15
5	2	1	11.70	−12.19	3	3	1	1.92	−1.68	8	4	2	4.19	4.27
6	2	1	4.07	3.97	6	3	1	6.04	−5.36	9	4	2	4.20	−4.77
8	2	1	1.59	−2.00	7	3	1	6.36	6.26	−10	4	3	1.45	1.61
−10	2	2	3.86	−3.80	8	3	1	6.18	−5.10	−4	4	3	2.14	1.52
−9	2	2	3.17	−3.02	−8	3	2	1.62	1.08	−3	4	3	2.56	−2.45
−5	2	2	1.61	1.37	−5	3	2	4.71	−4.73	0	4	3	2.66	2.79
−3	2	2	5.18	−5.42	−4	3	2	5.51	−4.87	1	4	3	2.95	3.32
−1	2	2	5.97	−7.73	−3	3	2	6.75	−7.04	2	4	3	2.65	2.47
0	2	2	5.88	−6.19	1	3	2	1.50	−1.26	6	4	3	3.05	2.58
2	2	2	6.58	7.90	3	3	2	3.09	2.82	7	4	3	1.96	−3.01
3	2	2	6.58	−7.04	4	3	2	2.89	2.13	−11	4	4	2.14	2.56
4	2	2	4.11	3.74	5	3	2	13.74	13.46	−10	4	4	1.88	1.93
7	2	2	2.90	2.85	6	3	2	4.59	−3.37	−6	4	4	3.48	3.82
8	2	2	3.42	3.10	7	3	2	5.89	4.86	−5	4	4	4.28	−4.74
−11	2	3	3.37	−3.25	−7	3	3	3.53	3.75	−4	4	4	2.79	2.89
−9	2	3	6.45	−6.55	−6	3	3	5.59	−5.40	−3	4	4	3.75	−2.81
−8	2	3	3.81	4.00	0	3	3	2.58	−2.31	5	4	4	2.01	1.59
−6	2	3	1.85	−2.27	2	3	3	6.18	5.41	−7	4	5	2.26	−2.27
−4	2	3	2.61	−3.16	3	3	3	6.48	6.52	−6	4	5	2.82	2.64
−3	2	3	6.77	−7.22	4	3	3	6.77	5.18	−5	4	5	3.42	−3.25
										3	4	5	2.37	2.31

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Refinement and results

Refinement was carried out, starting with the room-temperature results of Herbstein & Schmidt for phenazine and those of Curti *et al.* for *N*-oxyphenazine and for the mixed crystals.

Hydrogen atoms were included in the calculation of structure factors, but were not refined. They were assigned isotropic temperature factors of 3.5 Å² for the room-temperature structures, 2.0 Å² for the *N*-oxyphenazine data at −90 °C.

Refinement of the fractional coordinates x, y, z and anisotropic temperate factors b_{ij} was carried out by the diagonal-least-squares program of Milledge & Milledge (1961). The scale factors between layer lines were also refined, half shifts being applied.

Site-occupation factors (Q) were used for the oxygen atoms to specify the different compositions. In the case of the 8 mole % crystal, it was not reasonable to refine the position of the oxygen atom, since the site-occupation factor is very small. In table 3, the R-factors are given after the final cycles of refinement.

Table 3. Refinement of the mixed crystals of phenazine and *N*-oxyphenazine

x	Q	n	cycles	R	R_w
0 (photometer)	0.000	248	6	0.1009	0.1066
0 (I.d.t)	0.000	335	5	0.1025	0.1211
8	0.004	192	5	0.0898	0.0988
52	0.260	203	5	0.0935	0.1017
81	0.405	209	6	0.1020	0.1133
100 (20 °C)	0.500	227	11	0.0983	0.1041
100 (−90 °C)	0.500	237	7	0.0885	0.0922

Notation: x, composition in mole % *N*-oxyphenazine; Q, site-occupation factor as used for the oxygen atoms; n, number of reflexions used in the refinement; R_w, R-factor for weighted ΔF values (Glazer 1968).

In table 4 are listed the final fractional coordinates in each case. The anisotropic temperature factors were converted to thermal ellipsoids. These are represented diagrammatically in figure 3. Bond lengths and bond angles are given in table 5 (cf. figure 1*b*). More detailed results for each composition are given by Glazer (1968).

(*a*) 0 *mole* % *N-oxyphenazine* (*i.e. phenazine*)

From table 4 it can be seen that the fractional coordinates agree within their standard deviations as given by the photometer and the I.d.t. results. These results also agree reasonably well with those of Herbstein & Schmidt (1955). In figure 4, two projections of the unit cell are shown.

In table 6 are listed the orientations of the molecular inertia axes (L, long; M, medium; N, normal) with respect to the mutually orthogonal crystal axes, a, b, c^*.

The temperature factors appear to be more or less isotropic. This accounts for the large estimated errors given for the orientations of the ellipsoid axes.

A rigid-body thermal-motion analysis calculation, by the method of Cruickshank (1956) was carried out. The T and ω tensors obtained in this way are given in table 7, with the differences between observed and calculated atomic mean square amplitudes (ϵ) in table 8. It should be noted that while the T_{ij} are in fairly good agreement for the two sets of data, some large discrepancies are observed for the ω_{ij}. This is especially true for ω_{11}, ω_{12} and ω_{13}. All that can really be said is that ω_{11} is large.

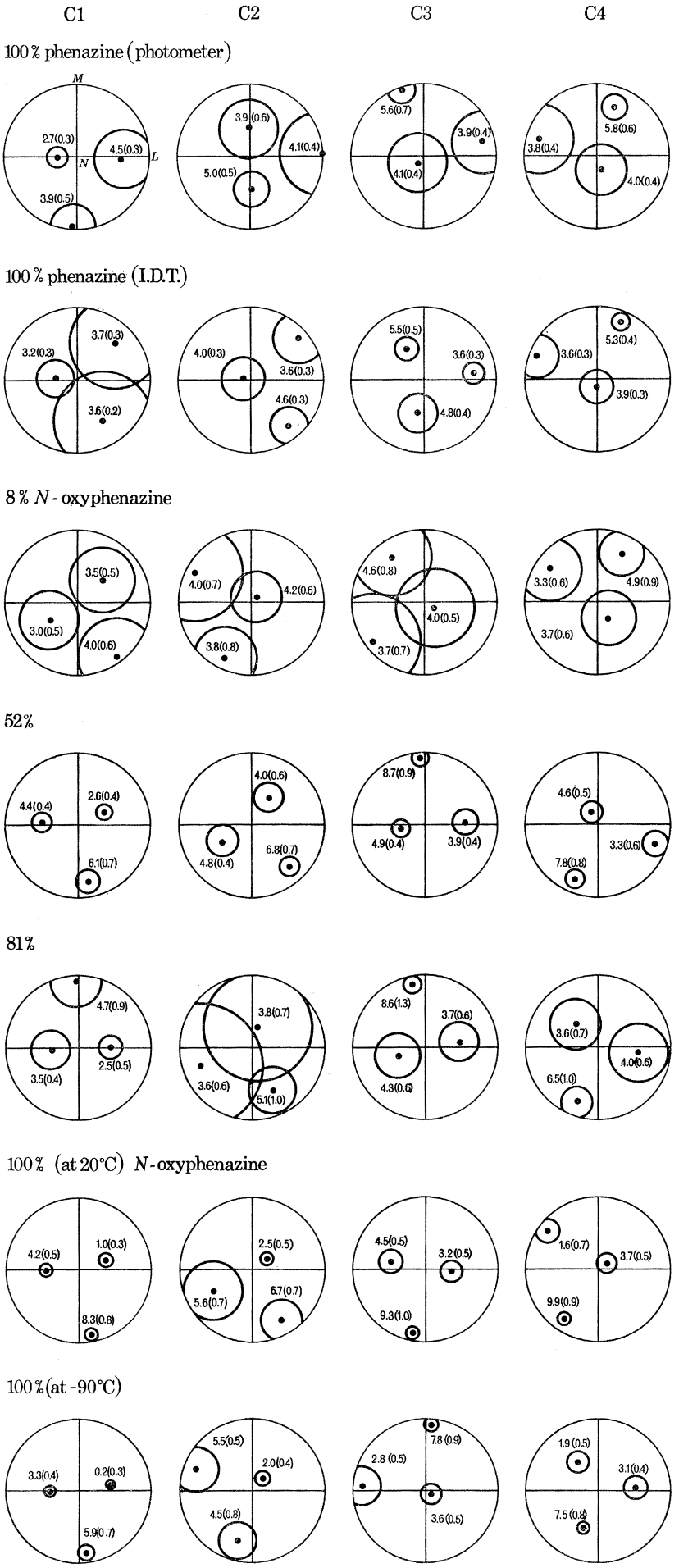
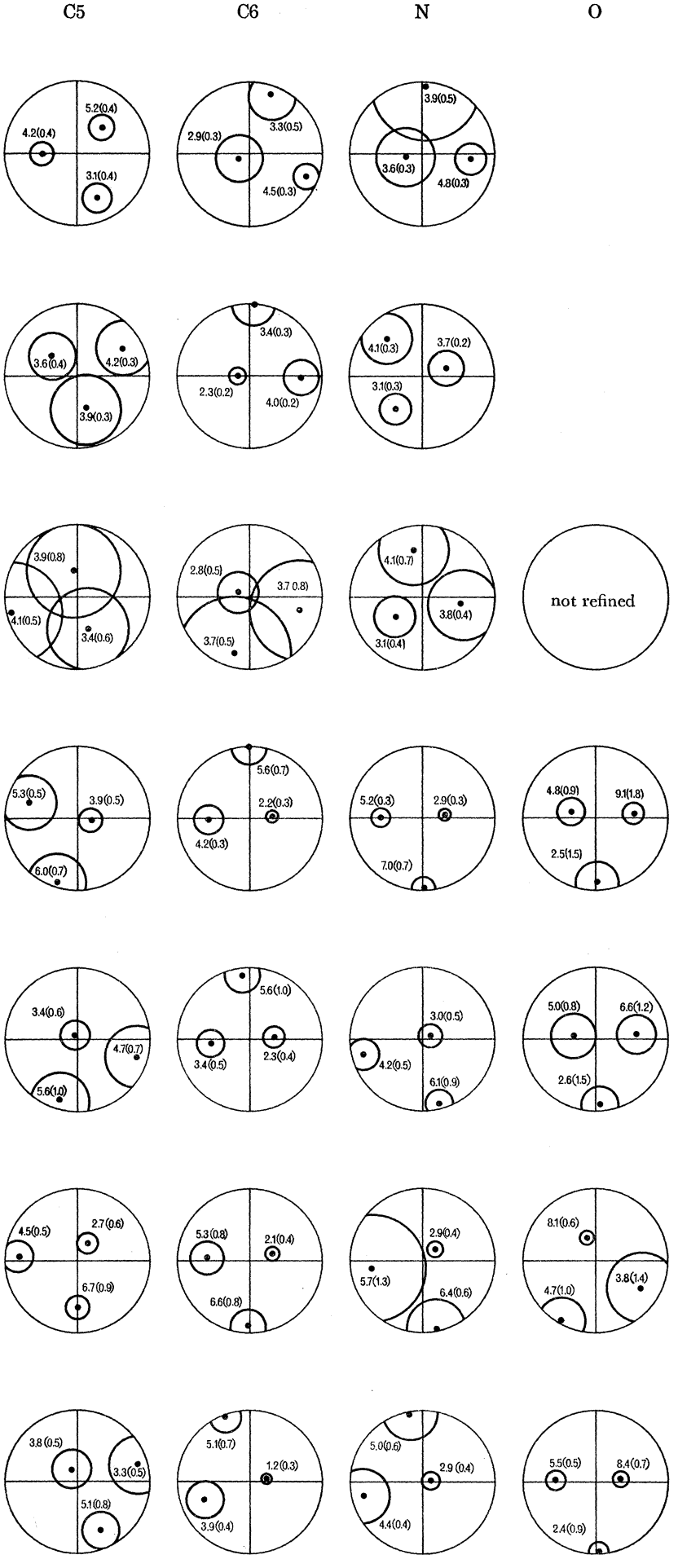


FIGURE 3. Anisotropic temperature factors for all C, N and O atoms in the mixed-crystal series, shown in stereographic projection relative to the L (long), M (medium), N (normal) axes of the molecules. The information thus provided shows the directions of the principal axes of the thermal ellipsoids, with standard deviations in the form of circles. (In fact the standard deviation for each direction cosine varies, but this is a minor detail



which could confuse the diagram.) Where the circle is large, the direction given has very little significance. The thermal amplitudes corresponding with the directions are shown as a number which is the calculated value of B (with e.s.d.) in each case. Units of B are Å^2.

Table 4. Fractional coordinates from refinements (with e.s.d. in parentheses), percentages are of *N*-oxyphenazine

	phenazine					*N*-oxyphenazine	
	0% (photometer)	0% (I.d.t.)	8%	52%	81%	100% (20 °C)	100% (−90 °C)
x							
C 1	0.07661 (62)	0.07734 (45)	0.07791 (87)	0.08029 (81)	0.07968 (92)	0.07809 (75)	0.07968 (68)
C 2	0.15660 (61)	0.15709 (45)	0.15795 (83)	0.16174 (75)	0.16111 (95)	0.16073 (77)	0.16244 (73)
C 3	0.18861 (68)	0.18904 (51)	0.18891 (93)	0.18795 (80)	0.18907 (113)	0.18757 (83)	0.18720 (78)
C 4	0.13920 (74)	0.14029 (52)	0.13991 (87)	0.13677 (79)	0.13527 (108)	0.13396 (91)	0.13460 (80)
C 5	0.06232 (59)	0.06303 (42)	0.06174 (81)	0.05639 (76)	0.05697 (99)	0.05488 (75)	0.05524 (71)
C 6	0.02867 (60)	0.02937 (45)	0.02744 (85)	0.02631 (71)	0.02575 (99)	0.02421 (80)	0.02683 (69)
N	0.04738 (51)	0.04796 (36)	0.04918 (65)	0.05199 (59)	0.05278 (81)	0.05270 (63)	0.05136 (56)
O	—	—	not refined	0.09539 (162)	0.09652 (152)	0.09689 (102)	0.09633 (103)
y							
C 1	0.18726 (135)	0.18587 (122)	0.18452 (212)	0.18353 (181)	0.18162 (239)	0.17667 (190)	0.18670 (132)
C 2	0.38233 (171)	0.38455 (137)	0.37984 (224)	0.37350 (233)	0.37411 (277)	0.37323 (208)	0.38226 (187)
C 3	0.51603 (169)	0.51768 (150)	0.51983 (257)	0.51810 (255)	0.52077 (317)	0.51969 (268)	0.52817 (197)
C 4	0.46159 (159)	0.46448 (140)	0.46535 (223)	0.47356 (222)	0.47760 (321)	0.47210 (233)	0.48459 (171)
C 5	0.27541 (152)	0.27709 (135)	0.27655 (225)	0.28944 (204)	0.29576 (283)	0.29219 (234)	0.29925 (202)
C 6	0.13121 (150)	0.13090 (120)	0.13112 (224)	0.13744 (194)	0.14023 (256)	0.14459 (203)	0.14156 (163)
N	0.05567 (114)	0.05629 (100)	0.05476 (162)	0.04568 (149)	0.03935 (214)	0.03519 (179)	0.03463 (163)
O	—	—	not refined	0.07477 (690)	0.06697 (522)	0.06912 (355)	0.07780 (392)
z							
C 1	0.02797 (123)	0.02872 (88)	0.02846 (167)	0.03610 (158)	0.03798 (200)	0.04407 (166)	0.04267 (150)
C 2	0.06447 (119)	0.06578 (90)	0.06711 (158)	0.08262 (158)	0.08910 (204)	0.09172 (174)	0.08755 (154)
C 3	0.24093 (150)	0.24097 (96)	0.24318 (198)	0.25244 (175)	0.25569 (244)	0.25822 (178)	0.25923 (183)
C 4	0.39083 (122)	0.38953 (92)	0.38820 (163)	0.38587 (149)	0.38382 (207)	0.38676 (151)	0.38829 (138)
C 5	0.35500 (107)	0.35753 (82)	0.35641 (154)	0.34821 (148)	0.34549 (204)	0.34492 (160)	0.34727 (155)
C 6	0.17565 (119)	0.17811 (87)	0.17556 (164)	0.17244 (155)	0.17250 (220)	0.17496 (168)	0.17081 (152)
N	−0.14846 (95)	−0.14680 (68)	−0.14719 (123)	−0.13337 (119)	−0.12961 (163)	−0.12729 (129)	−0.12621 (124)
O	—	—	not refined	−0.23612 (348)	−0.22917 (316)	−0.23015 (225)	−0.22938 (217)

Table 5. Bond lengths from refinements (e.s.d. in parentheses) (Å); percentages given are of *N*-oxyphenazine

	phenazine					*N*-oxyphenazine	
	0% photometer	0% (I.d.t.)	8%	52%	81%	100% (20 °C)	100% (−90 °C)
C 1–C 2	1.404 (11)	1.414 (9)	1.405 (16)	1.403 (15)	1.409 (18)	1.438 (15)	1.422 (13)
C 1–C 6	1.419 (12)	1.429 (9)	1.443 (16)	1.450 (15)	1.454 (20)	1.431 (17)	1.398 (14)
C 1–N	1.354 (11)	1.343 (8)	1.348 (15)	1.346 (14)	1.348 (18)	1.372 (15)	1.370 (13)
C 2–C 3	1.349 (13)	1.350 (10)	1.373 (18)	1.365 (17)	1.352 (23)	1.352 (18)	1.374 (17)
C 3–C 4	1.445 (13)	1.429 (10)	1.424 (18)	1.402 (16)	1.413 (23)	1.422 (18)	1.400 (16)
C 4–C 5	1.344 (12)	1.354 (9)	1.361 (16)	1.376 (15)	1.348 (20)	1.353 (16)	1.358 (14)
C 5–C 6	1.403 (12)	1.408 (9)	1.418 (16)	1.418 (15)	1.412 (21)	1.376 (17)	1.429 (15)
C 6–N′	1.344 (11)	1.354 (8)	1.346 (15)	1.351 (14)	1.342 (18)	1.328 (16)	1.318 (15)
N–O	—	—	not refined	1.118 (25)	1.116 (25)	1.149 (18)	1.156 (17)
Bond angles from refinements (in degrees)							
C 1–C 2–C 3	120.3	120.7	120.8	120.1	122.3	120.5	117.7
C 2–C 3–C 4	120.3	120.3	119.7	121.0	118.8	119.0	121.8
C 3–C 4–C 5	119.4	120.4	121.2	121.4	122.3	122.0	121.2
C 4–C 5–C 6	121.7	120.8	120.5	119.2	120.3	120.5	119.1
C 5–C 6–C 1	118.6	118.8	118.4	119.0	118.5	119.5	119.3
C 6–C 1–C 2	119.7	119.0	119.4	119.3	117.8	118.5	120.9
C 6–C 1–N	121.5	121.9	121.7	118.6	121.0	123.0	118.7
C 1–C 6–N′	123.0	120.4	121.9	121.0	120.0	117.6	120.6
C 1–N–C 6′	115.9	117.5	116.4	118.9	119.0	119.4	120.7
O–N–C 1	—	—	not refined	117.5	120.3	121.2	116.7
O–N–C 6′	—	—	not refined	123.6	120.7	119.4	122.2

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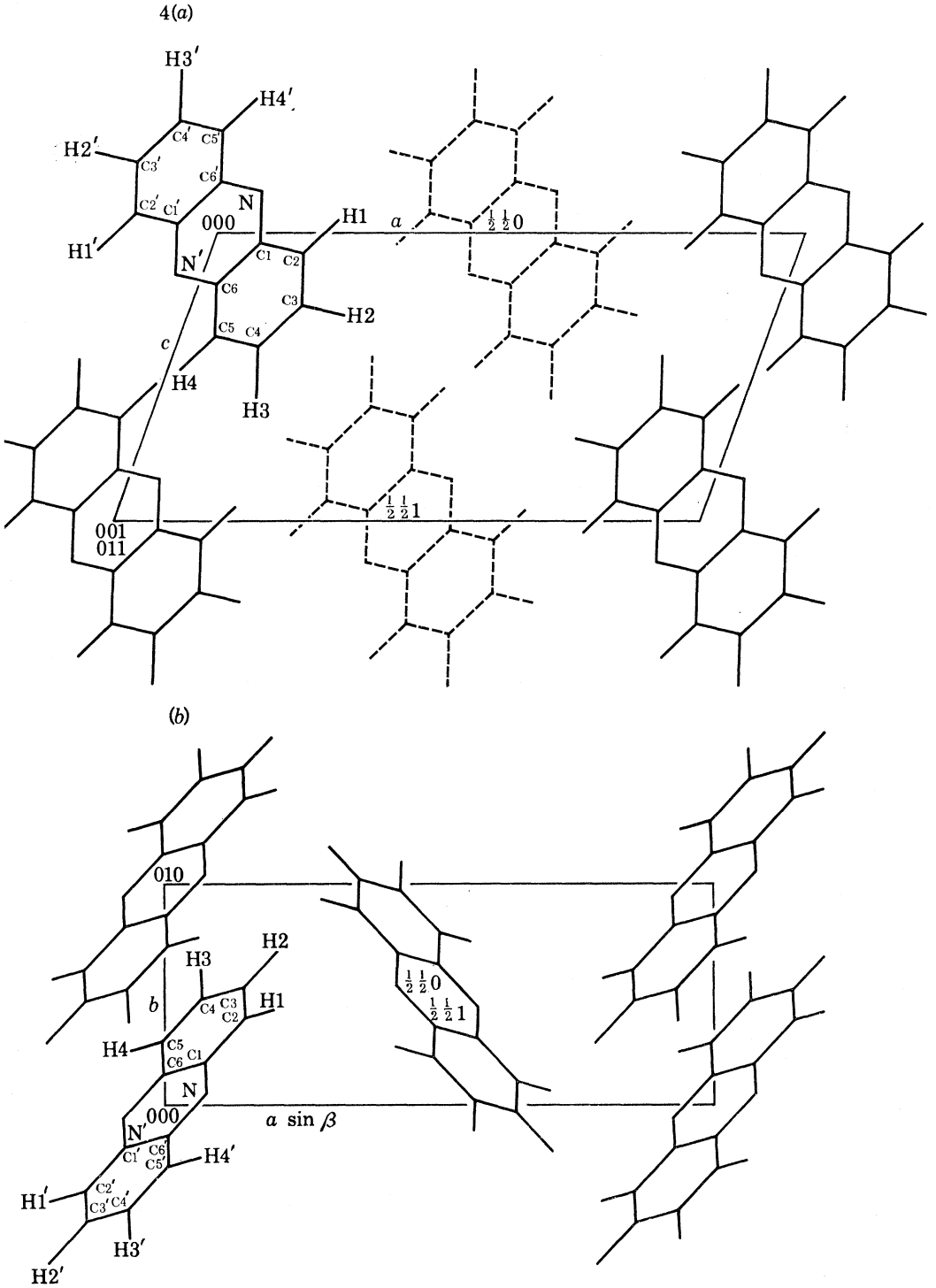

FIGURE 4. Projections of the unit cell of phenazine. (*a*) along [010], (*b*) along [001].

TABLE 6. DIRECTIONS OF INERTIA AXES L, M, N, WITH RESPECT TO a, b, c^* (IN DEGREES) FROM REFINEMENTS, AT 20 °C EXCEPT WHERE OTHERWISE STATED; PERCENTAGES ARE OF *N*-OXYPHENAZINE

	0% (photometer)	0% (I.d.t)	0%† (−190 °C)	8%	52%	81%	100%	100% (−90 °C)
$L:a$	66.40	66.30	66.53	66.05	65.58	64.97	65.00	65.88
$L:b$	45.75	45.75	45.53	46.20	48.13	49.38	50.25	48.77
$L:c^*$	53.55	53.65	53.93	53.33	51.73	50.93	50.05	50.88
$M:a$	46.30	46.35	47.47	45.70	42.00	40.90	40.30	40.65
$M:b$	78.68	78.78	77.97	79.25	81.63	82.55	82.43	82.05
$M:c^*$	134.01	134.18	134.90	133.70	130.75	129.92	129.28	129.53
$N:a$	127.00	127.00	128.32	126.12	121.72	119.97	119.30	120.45
$N:b$	46.45	46.43	47.11	45.78	43.01	41.60	40.75	42.33
$N:c^*$	113.80	113.80	113.00	114.07	115.85	115.93	115.60	116.32

† Calculated from atomic coordinates given by Hirshfeld & Schmidt (1957).

TABLE 7. T_{ij}, ω_{ij} FOR PHENAZINE AT 20 °C

(1) *Photometer*

$$T\begin{pmatrix}5.12 & 0.06 & 0.17\\ & 4.50 & -0.09\\ & & 3.28\end{pmatrix}10^{-2}\text{Å}^2 \qquad \omega\begin{pmatrix}26.50 & 0.13 & -4.76\\ & 4.14 & -1.07\\ & & 5.81\end{pmatrix}\text{deg}^2$$

(2) *I.d.t.*

$$T\begin{pmatrix}4.68 & -0.09 & 0.10\\ & 4.33 & -0.03\\ & & 3.31\end{pmatrix}10^{-2}\text{Å}^2 \qquad \omega\begin{pmatrix}15.11 & -2.23 & -0.01\\ & 4.92 & -0.75\\ & & 5.15\end{pmatrix}\text{deg}^2$$

TABLE 8. DIFFERENCES (ϵ) BETWEEN OBSERVED AND CALCULATED ATOMIC MEAN SQUARE AMPLITUDES FOR THE T_{ii}, ω_{ii} IN TABLE 7 (PURE PHENAZINE)

		photometer	I.d.t.	
C 1	L	−0.06	−0.27	10^{-2} Å^2
	M	+0.25	+0.12	
	N	+0.07	+0.37	
C 2	L	−0.22	+0.10	
	M	+0.22	+0.03	
	N	+0.04	−0.47	
C 3	L	−0.14	+0.15	
	M	+0.22	+0.12	
	N	−0.01	+0.33	
C 4	L	−0.11	+0.08	
	M	+0.13	+0.02	
	N	−0.06	−0.16	
C 5	L	+0.05	+0.09	
	M	−0.85	−0.33	
	N	+0.10	+0.13	
C 6	L	+0.11	+0.14	
	M	−0.37	−0.22	
	N	−0.02	−0.54	
N	L	+0.38	−0.29	
	M	+0.40	+0.28	
	N	−0.12	+0.34	
$\langle\lvert\epsilon\rvert\rangle = \frac{\Sigma\lvert\epsilon_{ii}\rvert}{21}$		0.19	0.22	
average e.s.d of $\langle u^2\rangle$		0.52×10^{-2} Å^2	0.39×10^{-2} Å^2	

Thermal-expansion measurements have been made for phenazine. It is found (part V) that the direction of maximum thermal expansion is along a and the minimum along c^*. This is explained by considering figure 4 with respect to table 6. It is seen that the direction of the L molecular axis is hardly affected by the change in temperature. Therefore the molecule can be thought of as rotating about the L axis as the temperature changes. The direction of rotation with increase in temperature allows for closer packing of the molecules along [001] (and hence the small thermal expansion is approximately along this direction), but the reverse is true with respect to molecule 000 and molecule $\frac{1}{2}\frac{1}{2}0$. Therefore the greatest thermal expansion is along a.

(*b*) 100 *mole* % *N-oxyphenazine*

In figure 5, two projections of the unit cell of *N*-oxyphenazine are shown and in figure 6 the asymmetric units have been plotted both at room and low temperature. From these and the results in table 6, one can offer an explanation of thermal-expansion results.

At 20 °C, the molecular planes are inclined at 49.25° (*N* axis at 40.75°) to the b axis and at −90 °C they are inclined at 47.67° (*N* axis at 42.33°). This change of 1.58° is accompanied by a similar reorientation of the L axis with respect to b of 1.48°. These being the largest changes, it can be seen that on reducing the temperature, the molecules have rotated more or less about the M axis (cf. phenazine). Since at low temperature the planes of the molecules are inclined at a smaller angle to the b axis, it would be expected that b should be *longer* at low temperature. This is indeed found to be the case. Also, the orientations of the thermal ellipsoids with respect to a, b, c^* show that the maximum thermal vibration for the oxygen atom is more closely parallel to [010] at low temperature than at room temperature (51° at 20 °C; 8° at −90 °C). This would also help to account for the negative thermal expansion along [010], especially as the actual magnitude of the maximum thermal ellipsoid axis for O is hardly altered by the temperature change. In the case of thermal expansion in the $\{h\,0\,l\}$ zone, it is found that the smallest thermal expansion lies along the direction corresponding to what would seem the most likely direction for intermolecular bonding (along c). In a direction at right angles to this, but still in the $\{h\,0\,l\}$ zone, the expansion is a maximum. This corresponds to a change in intermolecular distance of as much as 0.08 Å between H 2 for the molecule at 000 and H 4 for the molecule at $\frac{1}{2}\frac{1}{2}1$ (see Glazer 1968, pp. 104–106). Some of this is due to a rotational change of the molecule as the temperature is altered. It is interesting to note that the two smaller thermal expansions lie along directions where short-range order has been found (see later). Short-range order suggests some closer interaction between molecules and it might be expected, therefore, that the smaller expansions would occur along directions where interactions are important.

Examination of the change in temperature factors with temperature shows an interesting phenomenon. The temperature factors B were summed at each temperature and $\Sigma B(L)/\Sigma B(R)$ was plotted against $T(L)/T(R)$ where L signifies low, R room, and T absolute temperature. According to El Sayed (1965) and to Lonsdale & El Sayed (1965), for substances free from disorder and zero-point energy one obtains a straight line passing through the fixed points (0, 0) (1, 1). Accurate data for hexamine, however, show some curvature towards very low temperature to give an intercept on the $\Sigma B(L)/\Sigma B(R)$ axis. It was found that our single point $\Sigma B_{-90\,^\circ\mathrm{C}}/\Sigma B_{20\,^\circ\mathrm{C}}$ lies significantly higher than the line through (0, 0) (1, 1). This indicates that considerable disorder is included in B for this crystal.

A thermal-motion-analysis calculation was made: (1) for the whole 'averaged' molecule, (2) for the phenazine nucleus alone.

If the whole molecule vibrates as a rigid body, removal of the oxygen atom should not affect the T, ω tensors (Lonsdale, Walley & El Sayed 1966*b*). The results of this analysis are given in tables 9 and 10. The following observations can be made from these results:

(1) On removal of the oxygen atom, large changes occur in the angular libration tensor ω,

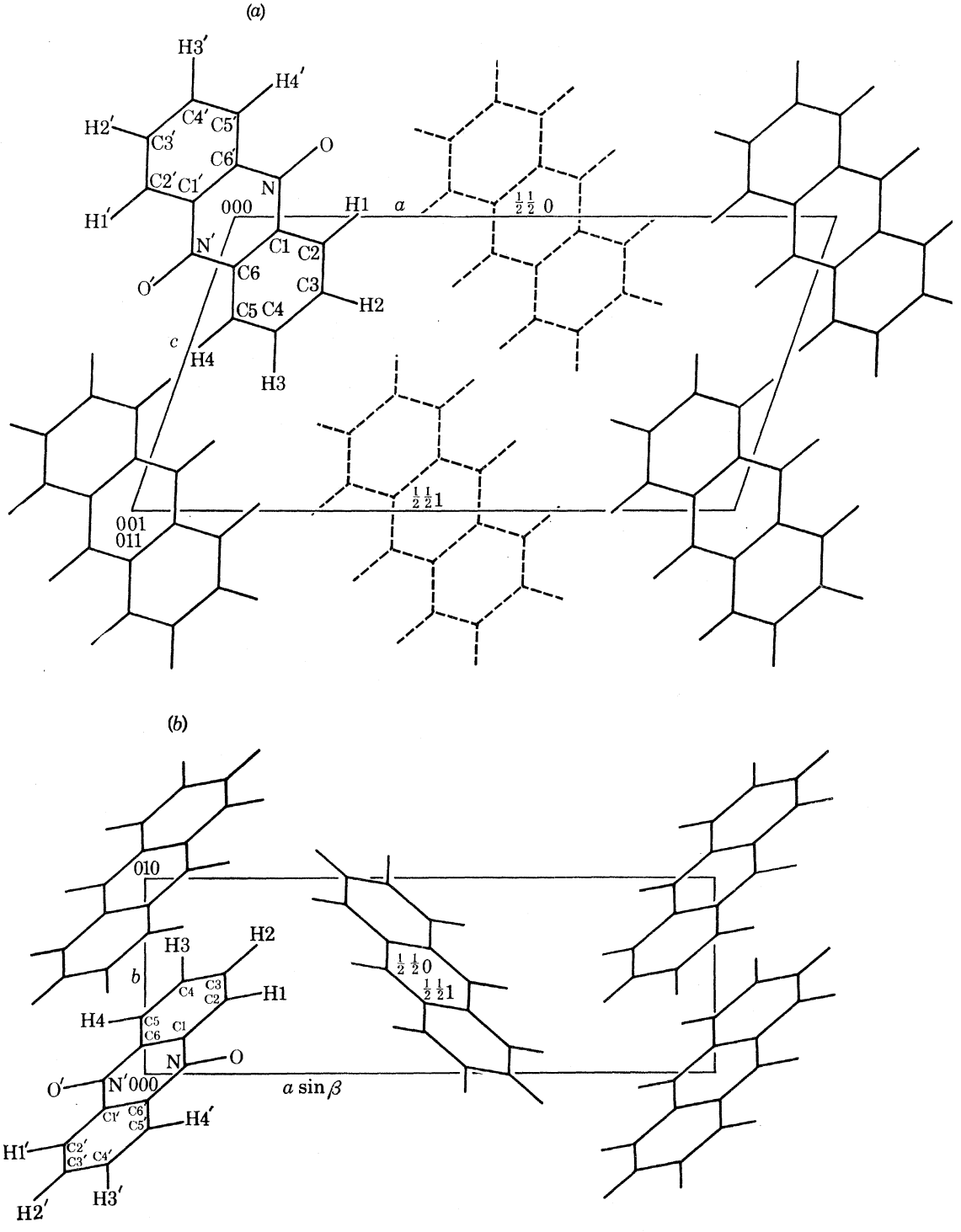

FIGURE 5. Projections of the unit cell of pseudosymmetric *N*-oxyphenazine (*a*) along [010], (*b*) along [001].

especially for ω_{11} and ω_{13}. The importance of this effect must be weighed against the fact that, as table 7 showed, different methods of measurement may also affect ω very drastically. Nevertheless, the changes in ω_{11} are consistent with those found for other compounds with substituted oxygen, and indicate independent out-of-plane vibration of the oxygen atom. A similar effect is shown by anthrone (Flack 1968) and to a slightly lesser extent by anthraquinone (Lonsdale, Milledge & El Sayed 1966*a*).

(2) Lowering of the temperature seems to have produced an increase in nearly all of the ω_{ij}, especially for ω_{11}, whereas the T_{ij} have decreased. It is impossible to explain this on thermal grounds.

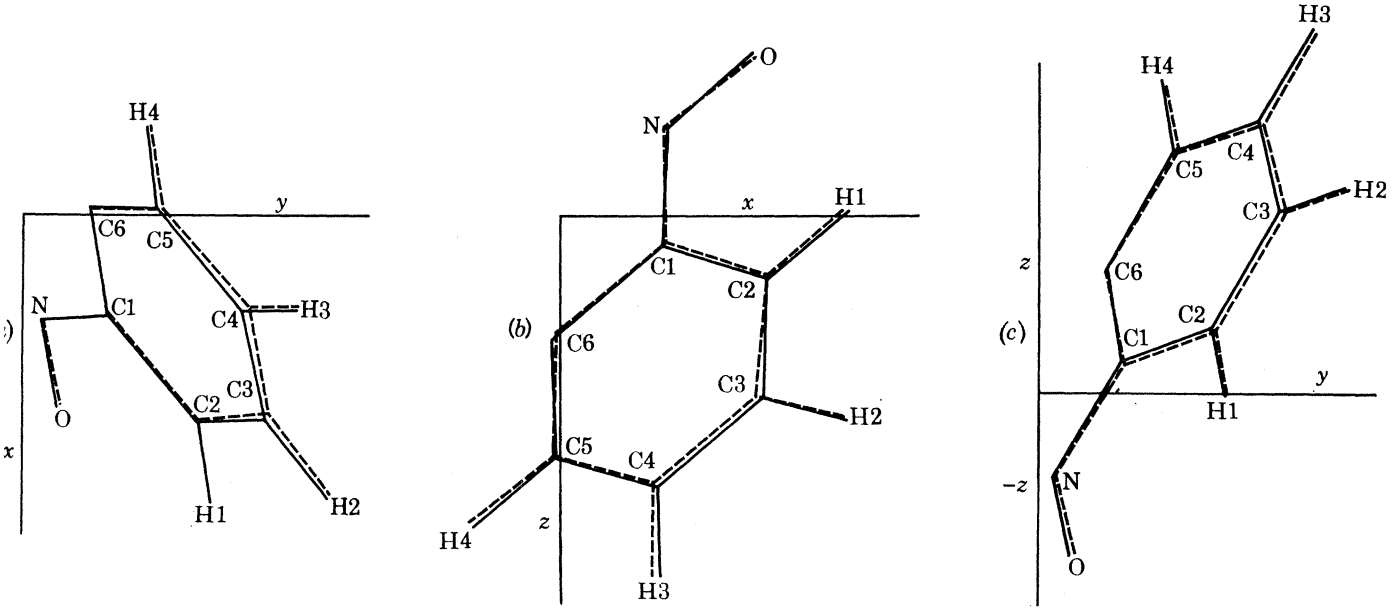

FIGURE 6. The 'asymmetric unit' of the *N*-oxyphenazine molecule plotted from the 20 °C (——) and −90 °C (– – –) data for the (*a*) *x*, *y* coordinates, (*b*) *x*, *z* coordinates, (*c*) *y*, *z* coordinates, to show the molecular movement.

TABLE 9. T_{ij}, ω_{ij} FOR *N*-OXYPHENAZINE

(1) *All atoms* (20 °C)

$$T\begin{pmatrix}4.96 & 0.65 & -0.87\\ & 7.17 & -0.79\\ & & 2.20\end{pmatrix}10^{-2}\text{Å}^2 \qquad \omega\begin{pmatrix}34.39 & 1.81 & 1.67\\ & 4.56 & 1.67\\ & & 6.79\end{pmatrix}\text{deg}^2$$

(2) *Molecule less oxygen atoms* (20 °C)

$$T\begin{pmatrix}5.15 & 0.67 & -0.87\\ & 7.43 & -0.15\\ & & 3.12\end{pmatrix}10^{-2}\text{Å}^2 \qquad \omega\begin{pmatrix}9.00 & 1.74 & 3.48\\ & 3.88 & 0.29\\ & & 6.56\end{pmatrix}\text{deg}^2$$

(3) *All atoms* (−90 °C)

$$T\begin{pmatrix}4.43 & 0.03 & -0.51\\ & 5.21 & 0.00\\ & & 1.24\end{pmatrix}10^{-2}\text{Å}^2 \qquad \omega\begin{pmatrix}40.20 & 3.09 & -2.86\\ & 6.16 & 2.95\\ & & 8.06\end{pmatrix}\text{deg}^2$$

(4) *Molecule less oxygen atoms* (−90 °C)

$$T\begin{pmatrix}4.24 & 0.00 & -0.51\\ & 6.06 & -0.18\\ & & 1.88\end{pmatrix}10^{-2}\text{Å}^2 \qquad \omega\begin{pmatrix}22.00 & 3.06 & 4.00\\ & 5.77 & 2.43\\ & & 4.95\end{pmatrix}\text{deg}^2$$

Examination of the errors given in table 10 also shows that there is out-of-plane vibration of the oxygen atom.

The results found for *N*-oxyphenazine seem to indicate that the structure is not straightforward and that the abnormal results are due either to short-range order or to elements of disorder other

TABLE 10. DIFFERENCES (ϵ) BETWEEN OBSERVED AND CALCULATED ATOMIC MEAN SQUARE AMPLITUDES FOR THE T_{ii}, ω_{ii} IN TABLE 9 (PURE *N*-OXYPHENAZINE)

		20 °C		−90 °C		
		whole molecule	subtract oxygen	whole molecule	subtract oxygen	
C 1	*L*	−1.31	−1.49	−2.47	−2.22	10^{-2} Å^2
	M	+2.12	+1.87	+1.76	+1.05	
	N	+0.11	−0.43	+0.48	+0.13	
C 2	*L*	+1.16	+0.99	+1.72	+2.11	
	M	−0.87	−1.08	−1.00	−1.30	
	N	−0.81	−0.11	−1.23	−0.68	
C 3	*L*	−0.07	−0.25	−0.97	−0.73	
	M	+1.36	+1.21	+1.47	+1.84	
	N	+0.36	+0.11	+0.81	+0.59	
C 4	*L*	+0.01	−0.17	−0.50	−0.25	
	M	−0.95	−1.11	−1.33	−0.97	
	N	+0.36	+0.08	−0.07	−0.27	
C 5	*L*	+0.03	−0.14	−0.56	−0.17	
	M	−1.15	−1.37	−0.55	−0.85	
	N	−1.00	−0.28	−0.46	+0.08	
C 6	*L*	+0.28	+0.10	+0.00	+0.24	
	M	+0.39	+0.15	+0.70	−0.01	
	N	+0.73	+0.28	−0.18	−0.53	
N	*L*	+1.13	+0.96	+0.65	+1.03	
	M	+0.58	+0.32	+1.09	+0.24	
	N	−0.24	+0.35	+0.28	+0.68	
O	*L*	−1.22	−1.35	+2.14	+2.94	
	M	−1.48	−1.74	−2.17	−3.02	
	N	+0.48	+4.55	+0.38	+3.27	
$\langle\|\epsilon\|\rangle = \frac{\Sigma\|\epsilon_{ii}\|}{24}$		+0.76	+0.85	+0.96	+1.05	
average e.s.d. of $\langle u^2\rangle$		0.8×10^{-2} Å^2		0.7×10^{-2} Å^2		

than pseudocentrosymmetry. (The term 'disorder' is taken here to exclude thermal disorder, i.e. vibration.) The evidence can be summed up as follows:

(1) Curti & Riganti (1960) determined the structure of phenazine-5,10-dioxide and found that this refined to $R = 0.14$ for $\{h\,0\,l\}$, whereas they had obtained $R = 0.19$ for $\{h\,0\,l\}$ for 'centrosymmetric' *N*-oxyphenazine. They suggest that this is due to the statistical nature of the latter structure.

(2) From table 3 it can be seen that lowering the temperature of the crystal has resulted in some but not in a great improvement in the *R*-factor, such as is often found for crystals where thermal vibration constitutes the only major form of disorder.

(3) The molecular geometry of the 'average' *N*-oxyphenazine molecule shows significant departure from *mmm* symmetry at both 20 and $-90\,^{\circ}$C. There seems to be poor agreement between the two sets of results (see table 5, last two columns).

(4) Decreasing the temperature of the crystal seemed to have only a small effect on the temperature factors.

(5) Decreasing the temperature actually appears to have increased the values of ω_{ij}.

(6) The ellipsoids representing 'thermal vibrations' show anisotropy that seems unlikely for a completely ordered structure. The orientations of the maximum ellipsoid axes at room and low temperatures suggest a spread of electron density in the *LM* plane (apart from the oxygen atom).

This could be interpreted as being due to the superposition of molecules in slightly different positions. To test whether this is a reasonable hypothesis, it was decided to refine the data in terms of two molecules replacing the original single 'average' molecule, i.e. each atom is replaced by two atoms. Figure 7 (*a*) shows such an arrangement. For simplicity it has been assumed that the centre of symmetry is retained.

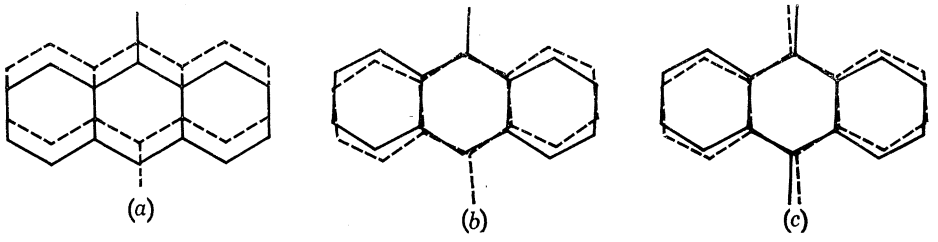

FIGURE 7. Possible arrangements of the *N*-oxyphenazine molecules to produce pseudosymmetric effects.

Also, if any significance can be placed on the actual directions of the thermal ellipsoid axes in the molecular plane, then these can be interpreted as in figure 7 *b*.

It was thought that refinement of this latter case might be of interest. The two molecules were given coordinates corresponding to the two orientations that best fitted the thermal-ellipsoid results. In addition, the two molecules were separated along [010] by 0.04 Å. It was not thought reasonable to refine both complete molecules (and omit the centre of symmetry) since this would require refinement of 125 parameters. In order to keep the number of parameters as low as possible, the centre of symmetry was retained and only the two asymmetric units were put into the refinement (figure 7 *c*). This would mean that in the refinement of the anisotropic temperature factors, the largest component of the 'thermal' ellipsoid for the oxygen ought to be in the molecular plane. This was found not to be the case. It was not known how much of this was due to real out-of-plane vibration of the oxygen atom. Therefore the structure was refined with only a single oxygen atom (using anisotropic temperature factors for this atom). For the other atoms, refinement was carried out with isotropic temperature factors. The final *R*-factor was 0.108.

There was no tendency for the two molecules to approach one another to re-form the original single molecule and hence the adopted model seemed to fit the data well.

(*c*) *Mixed crystals*

From table 4 it can be seen that there is a general change in the fractional coordinates as the composition is varied. Similarly, a smooth change in the orientations of the molecular inertia

axes is found (table 6). In figure 8 is plotted the molecule in the positions of the end-members. The molecules in the mixed crystals occupy orientations between these two extreme positions.

From figure 3, it can be seen that the temperature factors for 0 and 8 mole % *N*-oxyphenazine are smaller and more isotropic than for 52, 81 and 100 mole %. In other words, as far as the temperature factors are concerned, the crystal containing 8 % *N*-oxyphenazine is similar to pure phenazine, whereas the crystals of composition 52 and 81 % *N*-oxyphenazine are similar

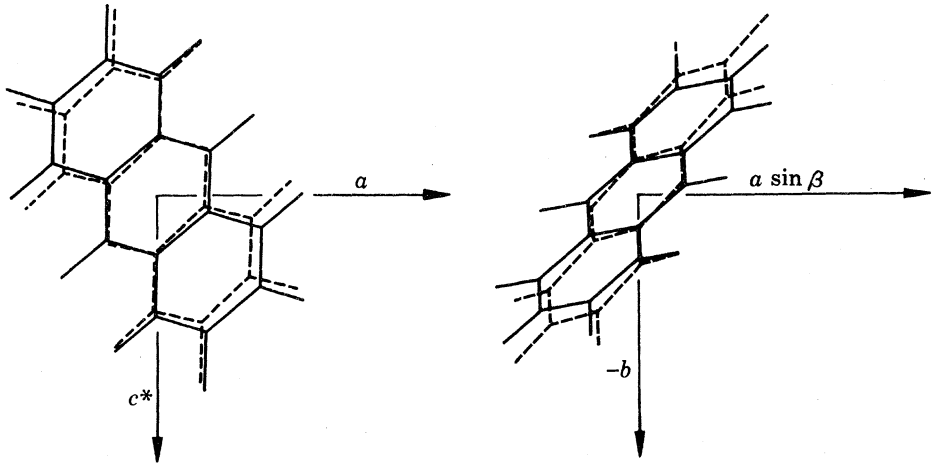

FIGURE 8. Molecular orientations for phenazine (– – –) and pseudosymmetric *N*-oxyphenazine (——).

to pure *N*-oxyphenazine. This may be an indication of the presence of a miscibility gap, somewhere between 8 and 52 % *N*-oxyphenazine (see part V). The 81 % crystal seems to have lower temperature factors than either the 52 or 100 % crystals. It is difficult to assess whether this is a real effect or not, since the errors are large. If this is a real effect, then this too may indicate a region of immiscibility, since this would imply a discontinuous change in temperature factors across the series.

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