are determined and the values of z_k are taken from (5). Given the coordinates x_i, y_j, and z_k, the n numerators of (6) and (7) may be calculated and stored on magnetic tape together with the weight which is determined, using the tabulated values of R. These steps are repeated until the numerators and weights for each of the m^3 points have been stored. The crystal volume, which is simply the sum of all the weights, is also determined in Part I.

In Part II the angles θ and χ for a particular reflection are substituted in (2) to obtain α and β, and these are used to calculate the $2n$ denominators of equations (6) and (7). Then, using the numerators from magnetic tape, the integrand for each point is evaluated as described above and summed with the appropriate weight (also from magnetic tape) to yield the integral A. The calculations of Part II are repeated for each reflection of interest.

The steps involved in obtaining the absorption correction factors may be summarized as follows: (1) A set of axes is fixed to the crystal and the equations of planes which approximate its faces are obtained. This can be done, for example, by measuring the oriented sample with a toolmaker's microscope to determine the coordinates of its corners, and solving for the coefficients of the planes which pass through these points. (2) The absorption coefficient, μ, is found experimentally or from tabulated cross-sections. (3) The Bragg angle, θ, and the interplanar angle, χ, are calculated for each reflection of interest. This step is a necessary preliminary to obtaining intensity data using counting techniques. (4) The above information is read into the computer, which calculates a value of A for each reflection.

With $m = 8$, the computing time for a crystal with six faces is 19 sec. for Part I and 31 sec. per reflection for Part II. The latter time should be approximately proportional to nm^3.

The precision of the integration was tested by constructing problems of simple but varied geometry which could be integrated analytically and which gave A's in the range from 0·50 to 0·62. Nineteen such calculations with $m = 8$ gave values of A which compared with the true values with a standard deviation of 0·2% and a maximum deviation of 0·5%. Rough tests indicate that an accuracy of 1 or 2% can be obtained with m as low as 4 or 5.

The accuracy of this routine is several times better than that of an earlier version, which averaged over points on an isometric lattice weighting them equally. It is believed that the inaccuracy of the older method was primarily due to the improper weighting of points near the sample surface.

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The Crystal and Molecular Structure of Pyrazine

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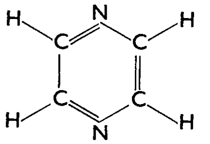
The crystal structure of pyrazine has been determined by three-dimensional Fourier methods. The crystals are orthorhombic, *Pmnn*, with two molecules in the unit cell. The molecular symmetry is $2/m$. The dimensions of the molecule are: C–N = 1·334 Å, C–C = 1·378 Å, N–C–N = 122·4°, C–N–C = 115·1°. The C–H distance was found to be 1·05 Å. Nine three-dimensional refinement cycles were carried out, the last five with the object of determining anisotropic thermal parameters for the carbon and nitrogen atoms. The motion of the molecule, considered as a rigid body, has been discussed in terms of the crystal packing. Corrections have been made to the bond lengths for the effect of the thermal motion. The final R factor is 8·3%.

Introduction

Cox, Cruickshank & Smith (1955) have shown that a systematic error may occur in the lengths of bonds as a result of anisotropic thermal motion. The effect is particularly important in simple planar molecules such as benzene and the azines. The correction to the C–C length in benzene was of the order of 0·015 Å, and this is confirmed by the discrepancy that exists in *s*-triazine. Wheatley (1955) found a value for the C–N length of 1·319 Å, whereas the figure obtained from an analysis of the rotational Raman spectrum (Stoicheff, 1956) is 1·338 Å. Since a similar error is likely to arise in pyrazine (I),



(I)

a large part of this investigation has been devoted to obtaining accurate thermal parameters. A knowledge of these parameters, apart from enabling an estimate of the requisite bond-length corrections to be made, helps to give an insight into the molecular motions at temperatures a little below the melting point. Since benzene and *s*-tetrazine (Bertinotti, Giacomello & Liquori, 1956) crystallize with the planes of adjacent molecules roughly at right angles, whereas *s*-triazine, pyrazine and pyrimidine pack with the planes of the molecules approximately parallel, significant differences in the principal thermal motions might be expected in the two types of structure.

Experimental

Pyrazine melts at 54° C. and sublimes, like all compounds consisting of small symmetrical molecules, very readily. The crystals used in this work were, therefore, grown in thin-walled Lindemann glass capillaries of 0·3 mm. internal diameter. Crystals grew preferentially with the shortest axis coinciding with the length of the tube, and the crystals were invariably twinned. A crystal grown by slow sublimation over a period of three months also showed twinning, so that all the intensities obtained were from crystals twinned on $\{101\}$. Cu $K\alpha$ radiation and the multiple-film technique were used to take Weissenberg photographs round [*b*] and [101] at 20° C. The relative intensities were estimated visually by comparison with a standard scale, and the different sets of photographs were placed on the same relative scale by correlation through common reflexions. A total of 207 independent reflexions were observed. The systematic absences of $h0l$ reflexions for $h+l = 2n+1$, and of $hk0$ reflexions for $h+k = 2n+1$, give *Pmnn* and *Pnn* as the possible space groups. The centrosymmetric *Pmnn* (No. 58) was employed from the start and this choice has been confirmed by the subsequent analysis.

The length of [*b*] was found from rotation photographs, and [*a*] and [*c*] were determined by the Straumanis method, yielding the following results:

$$a=9{\cdot}316\pm0{\cdot}003,\quad b=3{\cdot}815\pm0{\cdot}005,\quad c=5{\cdot}911\pm0{\cdot}003\ \text{Å}\,.$$

These cell constants give a calculated density of 1·266 g.cm.$^{-3}$, assuming two molecules in the unit cell.

The X-ray structure analysis

The molecule is required to have symmetry $2/m$. Thus there are seven positional parameters to be determined, one for the nitrogen atom and three each for the carbon and hydrogen atoms. The length of [*b*] implies that the molecule will lie roughly in the *ac* plane, and this is confirmed by the weighted reciprocal lattice shown in Fig. 1. The general disposition of the molecule required by the space-group symmetry is also confirmed. It should be possible to derive a value for the tilt of the molecule about [*a*] from Fig. 1, but a more reliable value can be obtained by considering the spacing in *s*-triazine. Here the molecules are stacked parallel at a distance of 3·64 Å apart, so that the tilt in pyrazine should be $\cos^{-1}(3{\cdot}64/3{\cdot}815) \sim 18°$. If the carbon and nitrogen atoms are assumed to lie at the same distance from the centre of the ring as in *s*-triazine, there is now sufficient information to obtain approximate values for the coordinates of the nitrogen and carbon atoms, as follows:

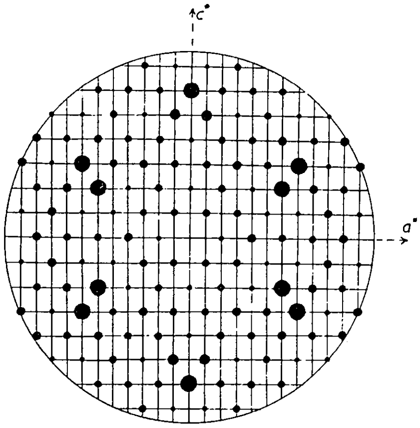


Fig. 1. The $h0l$ weighted reciprocal lattice.

	x/a	y/b	z/c
N	0·147	0	0
C	0·075	0·114	0·179

These coordinates were used immediately to calculate a set of three-dimensional structure factors. The scattering factors employed were those of Hoerni & Ibers (1954) modified by an assumed isotropic temperature factor of 3·7 Å^2. The reliability factor R was 17·5%. It was apparent that the reflexions 101, 110 and 011 suffered heavily from extinction, and these three reflexions have been omitted completely from all subsequent stages of the analysis.

The positional parameters were then refined by four successive cycles on the Manchester University electronic computer, using the programmes described by Ahmed & Cruickshank (1953). The R factor at this stage had dropped to 10·2%. The hydrogen atom was next located by means of a restricted three-dimensional synthesis, using the differences between the observed and calculated structure factors as coefficients in the Fourier synthesis. The height of the hydrogen peak was 0·39 e.Å^{-3}.

With the positions of the atoms established within narrow limits, anisotropic temperature factors were introduced for the carbon and nitrogen atoms, and an attempt made to refine them (Cruickshank, 1956*a*) by using only those planes with $\sin\theta > 0{\cdot}65$. As in the case of benzene (Cox, Cruickshank & Smith, 1957), this failed for a reason which is not apparent, but which is probably due to a systematic error in the estimation of the intensity of high-order reflexions. Consequently the thermal factors were refined by using all 204 reflexions and by making the observed and calculated peak heights and the observed and calculated curvatures equal. An isotropic temperature factor of 5·0 Å^2 was used for the hydrogen atoms, and the refinement of the positional parameters of the carbon and nitrogen atoms was continued as the refinement of the thermal parameters progressed. Table 1 shows the final values of the temperature factors for the carbon and nitrogen atoms, the temperature factor being defined by the expression

Table 1. *Final thermal parameters*

	N	C		N	C
B_{11}	2·70 Å^2	3·63 Å^2	B_{22}	5·78 Å^2	4·89 Å^2
B_{12}	0	−0·35	B_{23}	0·70	0·32
B_{13}	0	−0·32	B_{33}	4·69	3·81

$$f_i = f_0 \exp\left[-(B_{11}h^2+2B_{12}hk+2B_{13}hl + B_{22}k^2+2B_{23}kl+B_{33}l^2)\right].$$

The final peak heights and curvatures are shown in Table 2 and the final coordinates in Table 3.

Table 2. *Final curvatures and peak heights*

	Nitrogen		Carbon	
	Obs.	Calc.	Obs.	Calc.
A_{hh} (e.Å^{-5})	−83·60	−83·16	−65·49	−65·77
A_{kk} (e.Å^{-5})	−60·49	−60·37	−57·89	−58·03
A_{ll} (e.Å^{-5})	−71·50	−71·16	−65·34	−65·48
ϱ (e.Å^{-3})	7·55	7·53	6·51	6·51

Table 3. *Final atomic coordinates*

	x/a	y/b	z/c
N	0·1485$_0$	0	0
C	0·0728$_8$	0·1113$_5$	0·1733$_1$
H	0·130$_1$	0·192$_9$	0·317$_1$

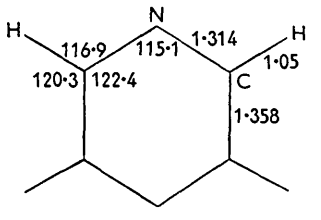


Fig. 2. Molecular dimensions, before corrections for the effect of thermal motion have been made.

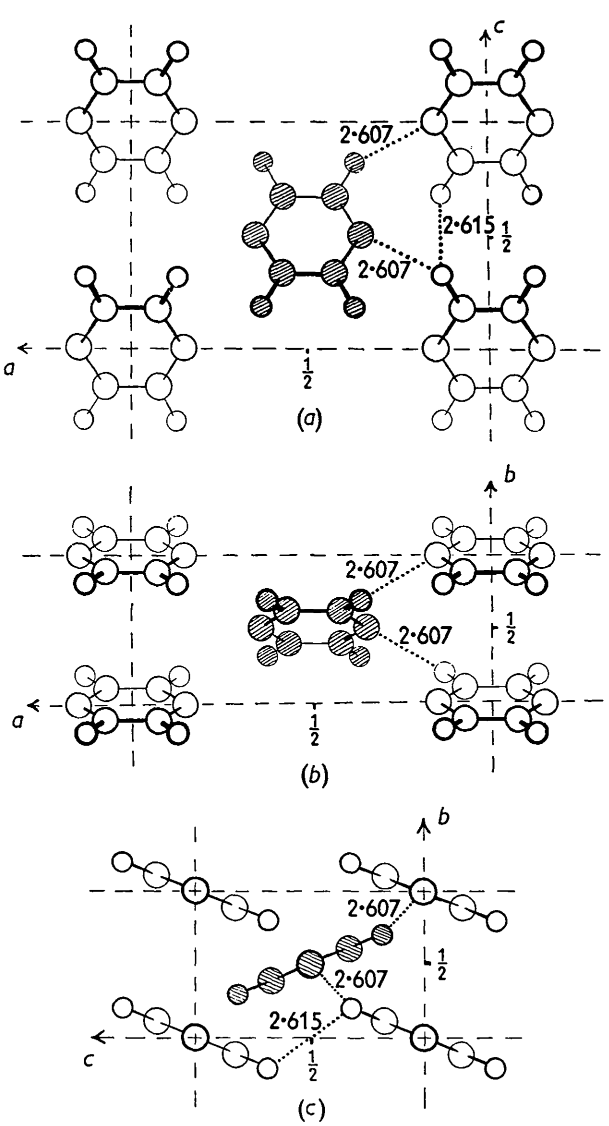


Fig. 3. Pyrazine: projections of the structure (*a*) on (010), (*b*) on (001) and (*c*) on (100). In each case a right-handed set of axes has been chosen, and the shaded molecule is displaced by half a translation in the positive vertical direction, i.e. upwards in (*a*) and (*c*), downwards in (*b*).

The molecular dimensions, before applying any correction for the thermal motion (Cruickshank, 1956*c*), are shown in Fig. 2, and the closest intermolecular contacts in Fig. 3. These contacts are between hydrogen and hydrogen atoms, and between hydrogen and nitrogen atoms. The position of the hydrogen atom is determined with much less certainty than that of the nitrogen atom. In particular, the hydrogen atom is drawn in towards the ring in the manner usually observed in X-ray analysis (Wheatley, 1953), so that the C–H distance is shorter than expected. In pyrazine, however, as opposed to benzene (Cox *et al.*, 1957), there is no need for the hydrogen atom to be located radially from the centre of the ring, so that, even though the C–H length were known accurately, the position of the hydrogen atom would still be in doubt. Moreover, there is no good reason to think that the intermolecular separation is determined by the position of the atomic *nuclei*, and the location of the hydrogen atom as determined by X-rays may give a more reliable figure for the separation. For these reasons it seems more realistic to retain the hydrogen atom coordinates given in Table 3, rather than to confuse the issue by selecting a more plausible value for the C–H internuclear separation. Despite the fact that the intermolecular distances are subject to greater errors than the bond lengths, the general nature of the packing is undoubtedly correct.

The standard deviations of the atomic coordinates, obtained by the method of Cruickshank (1949), are:

	$\sigma(x)$ (Å)	$\sigma(y)$ (Å)	$\sigma(z)$ (Å)
N	0·0024	0	0
C	0·0030	0·0028	0·0028

These figures give a standard deviation of 0·003 Å for the C–C and C–N bond lengths, and a standard deviation of 0·3° for the C–C–N and C–N–C angles.

The molecule is tilted by 22° 3′ around [*a*], and the equation of the plane through the molecule is

$$-0{\cdot}9237y+0{\cdot}3830z = 0\ ,$$

the coordinates being expressed in Ångström units. The hydrogen atom lies at a distance of 0·038 Å from this plane. The final reliability factor for the 204 planes was 8·3%.

After completion of the refinement of the positional and thermal parameters, an attempt was made to discover if any significant regions of electron density remained. Seven sections were computed with the differences between the observed and the final calculated structure factors as coefficients. The sections were chosen so that the effects of bonding electrons or of the lone pair of electrons on the nitrogen atom should appear. In these sections the electron density ranged from $-0{\cdot}19$ e.Å^{-3} to $+0{\cdot}18$ e.Å^{-3}, but no features of these difference maps could be associated with the molecule. At the centre of the ring, i.e. at the origin, the electron density had a value of $-0{\cdot}03$ e.Å^{-3}, so that no 'hole' appears in the middle of the molecule.

The thermal motion

The relevant values for the B_{ij} tensors for the carbon and nitrogen atoms are given in Table 1. These tensors are defined with respect to the original crystal axes, and to obtain information about the molecular motion it is necessary to transform to molecular axes. We define three new axes, therefore, (1) the axis (L) passing through the nitrogen atoms, which is coincident with the original [*a*], and has direction cosines (1, 0, 0); (2) the normal to the molecular plane (N), with direction cosines (0, 0·9238, −0·3830); and (3) the transverse axis (T), perpendicular to the other two, and with direction cosines (0, 0·3830, 0·9238). In order to maintain the necessary symmetry, and to give the requisite weights, we need to consider three atoms: two carbon atoms and one nitrogen atom. We select the original nitrogen atom, the original carbon atom, now labelled C_1, and the carbon atom, which we label C_2, related to C_1 by the twofold axis. The thermal parameters of these three atoms in the molecular axes (L, N and T) are given in Table 4.

Table 4. *Thermal parameters in terms of molecular axes*

	N	C_1	C_2
C_{LL}	2·70 Å^2	3·63 Å^2	3·63 Å^2
C_{LN}	0	−0·21	0·21
C_{LT}	0	−0·43	0·43
C_{NN}	5·12	4·51	4·51
C_{NT}	0·88	0·61	0·61
C_{TT}	5·35	4·19	4·19

For the nitrogen atom, the figures in Table 4 give directly the parameters for the motion tangentially to, radially in, and normal to, the ring. The values for the carbon atoms can be obtained after the necessary transformation. These figures are given in Table 5, together with the weighted mean. The last column in Table 5, therefore, gives the average values of the three important molecular motions.

Table 5. *Thermal parameters of the atoms relative to the ring*

	N	$C_{1,2}$	Weighted mean
Tangential	5·35 Å^2	4·17 Å^2	4·56 Å^2
Radial	2·70	3·66	3·34
Normal	5·12	4·51	4·72

Assuming that the molecule is a rigid body, we may now determine the rotational and translational vibration tensors (Cruickshank, 1956*b*). As in benzene (Cox *et al.*, 1957), it is not possible to determine all the coefficients. Table 6 lists the values that can be obtained. The absolute values of ω_{LL}, ω_{TT} and T_{NN} cannot be determined (although the sum of $\omega_{LL}\bar{r}^2$, $\omega_{TT}\bar{r}^2$ and T_{NN} must equal 4·72 Å^2, from Table 5), but the following relation exists:

$$1{\cdot}18\omega_{TT}-\omega_{LL} = 0{\cdot}497\ .$$

Table 6. *Coefficients of the translational and rotational vibration tensors*

T_{LL}	2·74 Å^2	ω_{LL}	—
T_{LN}	0	ω_{LN}	0
T_{LT}	0	ω_{LT}	0
T_{NN}	—	ω_{NN}	0·71
T_{NT}	0·48	ω_{NT}	−0·23
T_{TT}	3·91	ω_{TT}	—

These figures enable us to discuss the thermal motion in relation to the packing of the molecules in the crystal.

ω_{NN} corresponds to a r.m.s. angular displacement of 5·7°, assuming an average ring radius of 1·328 Å. Thus the oscillation about the normal to the molecular plane is not such a prominent part of the molecular motion as it is in benzene (Cox *et al.*, 1955). The motion along [L] is considerably more difficult than along [T], because the molecules can slide under each other in the [T] direction without causing much shortening in the N · · · H contact of 2·607 Å or the H · · · H contact of 2·615 Å (Fig. 3(a)). From the relation connecting ω_{TT} and ω_{LL} it follows that the rotation about [T] is considerably easier than about [L], though the absolute values of these motions are not known. Again this is physically reasonable, since (Fig. 3(c)) the hydrogen atom is sandwiched between the nitrogen and hydrogen atoms in the [b] direction, thus restricting rotation around [L].

The motion normal to the plane of the molecule cannot be resolved into rotational and translational components. If it could, then the rather large value of the cross-term T_{NT} would probably be very significant, as it would give the direction of maximum translation, which need not coincide with either [N] or [b]. The total motion normal to the plane, after allowance for the fact that the average temperature factor in benzene is higher than that in pyrazine by a factor of 5·57/4·21, is seen to be more important in pyrazine. Since the mode of packing in pyrazine is basically different from that in benzene, it seems unwise to split the motion into rotational and translational contributions using the method employed for benzene (Cox *et al.*, 1957). There seems to be no reason in pyrazine why the rotational and translational contributions to this motion should not be approximately of equal importance.

Correction to the bond lengths

From the difference in the values for motion radially in the ring and tangentially to the ring given in Table 5, we may obtain the correction to the bond lengths required for the motion round the normal to the molecular plane (Cruickshank, 1956*c*). The breadth parameter, given by the ratio of the peak height to the mean curvature of an atom, is $q^2 = 0{\cdot}104$ Å^2 in pyrazine. If we assume an average radius for the ring of 1·328 Å, the addition to be made to the bond lengths shown in Fig. 3 is 0·005 Å. For motion normal to the ring the maximum correction, assuming that the average figure 4·72 Å^2 is due entirely to rotation about axes in the plane of the molecule, is 0·029 Å. If, as suggested above, half the motion is assumed to be rotational, as seems reasonable, then the total correction to be added to the bond lengths will be $0{\cdot}005+0{\cdot}015 = 0{\cdot}020$ Å. The best estimates for the bond lengths are, therefore,

$$\text{C–C} = 1{\cdot}378 \text{ Å}, \quad \text{C–N} = 1{\cdot}334 \text{ Å} .$$

These bond lengths have a standard deviation of 0·003 Å due to random errors, and an error of ±0·015 Å due to uncertainty in the thermal motion. The values for the angles shown in Fig. 3 will be little affected by the thermal motion, and can probably be trusted to 1°.

Discussion of the molecular dimensions

The value of the C–N–C angle, 115° 8′, in pyrazine confirms the observation of Hameka & Liquori (1956) that, in heterocyclic rings containing nitrogen atoms, the bond angle of nitrogen is less than 120°.

The structure of pyrazine has been determined by electron diffraction (Schomaker & Pauling, 1939) with the following results:

$$\text{C–C} = 1{\cdot}39, \ \text{C–N} = 1{\cdot}35, \ \text{C–H} = 1{\cdot}09 \text{ Å}; \\ \text{all angles} = 120^\circ .$$

With the exception of the C–H distance, the present results are probably more reliable. The structure of tetramethylpyrazine has also been found by means of X-ray diffraction (Cromer, Ihde & Ritter, 1951), but the refinement was not carried as far as in the present investigation.

Reliable values of molecular dimensions are now available for benzene (Stoicheff, 1954; Cox *et al.*, 1955, 1957), pyridine (Liquori & Vaciago, 1956), *s*-triazine (Stoicheff, 1956), *s*-tetrazine (Bertinotti *et al.*, 1956) and pyrazine. Pyrimidine and pyridazine, the only other known azines, are under investigation in this laboratory.

Finally, two general conclusions may be drawn as a result of this and similar work. First, in simple cyclic molecules, X-ray determinations of the values of angles between bonds are more reliable than the bond lengths themselves. Secondly, if the main purpose of an X-ray analysis is the determination of accurate bond lengths, it would be better to work at low temperatures where the molecular motion is less important.

I would like to thank Prof. E. G. Cox for his interest in this work; Miss D. E. Pilling for carrying out much of the three-dimensional refinement; the Manchester University Computing Laboratory staff for use of the computer; Dr R. C. Hirt of the American Cyanamid Company for the gift of a sample; and Dr D. W. J. Cruickshank for much assistance, particularly with the analysis of the thermal motion.

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The Structure of Guinier–Preston Zones. II. The Room-Temperature Ageing of the Al–Cu Alloy

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In a previous paper the diffuse intensity diffracted by a Guinier–Preston (G.–P.) zone was expressed as a trigonometrical series with the coefficients dependent on the structure of the zone. The present paper deals with the determination of these coefficients from the variation of the diffuse intensity in relrod (00*l*) by the method of least squares for the natural aged Al–Cu alloy. From these the concentration of copper atoms in various atomic planes of the G.–P. zone, as well as the displacements of these planes with respect to the matrix, were determined.

1. Introduction

In the first part of this paper (Toman, 1955) the diffuse intensity of X-rays diffracted by a Guinier–Preston (G.–P.) zone was expressed as a trigonometrical series with coefficients determined by the structure of the zone; the possibility of determining these coefficients from the measured diffuse intensity distribution in relrod (00*l*) was shown. The knowledge of these coefficients enables us to evaluate structural features of the zone, namely the coppper-atom fraction in the various atomic planes (001) of the G.–P. zone and the displacements of these planes with respect to the matrix.

This method is applied in this paper in a modified form to the determination of the structure of G.–P. zones of the natural aged Al–Cu alloy.

2. Experimental part

High-purity materials were used for the preparation of the Al–Cu alloy with 3·95% Cu, the impurity content (Si, Fe) being below 0·03%. The wires, made from cast ingots, were 1 mm. in diameter and were converted to monocrystals by means of the strain annealing method. The monocrystals were quenched at 520° C. and aged naturally for 10 days. The aged crystals were etched in sodium hydroxide solution at 30° C. The etched crystals, prismatic in shape with cross-section 0·2 × 0·2 mm., were used for X-ray diffraction.

The Weissenberg patterns were obtained by using monochromatized Cu $K\alpha$ radiation. The bent quartz crystal monochromator was employed.

3. Intensity measurements

The diffuse intensity (00*l*) was measured on the microphotometer for the values of the continuous variable *l* ranging from 1 to 5. A series of photographs was taken with different exposure times under controlled conditions on account of the considerable variation of darkening in the measured interval. The measured intensities were corrected for polarization and Lorentz factors as usual. The absorption factor was determined by means of graphical integration. The diffuse intensity shows considerable maxima in the neighbourhood of the relps of the matrix with the streaks going out from the diffuse maxima in the direction of increasing Bragg angle. This observation is in agreement with that of Gerold (1954). The diffuse intensity in relps (002) and (004) and in their close neighbourhood cannot be directly measured because it overlaps the Bragg reflexion of the matrix. The interpolation in this range involves a certain degree of arbitrariness.

4. The determination of the structure

The diffuse intensity of X-rays diffracted by a G.–P. zone can be expressed as a trigonometrical series

$$I(l)_{\text{diff.}} = K\left[\sum_{-\infty}^{+\infty} A_n \cos \pi ln - \pi l \sum_{-\infty}^{+\infty} B_n \sin \pi ln + \pi^2 l^2 \sum_{-\infty}^{+\infty} C_n \cos \pi ln - \pi^3 l^3 \sum_{-\infty}^{+\infty} D_n \sin \pi ln\right], \quad (1)$$

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