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Accurate Determination of Hydrogen Positions from X-ray Data. I. The Structure of *s*-Diformohydrazide at 85 K

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The crystal structure of diformohydrazide, $C_2H_4N_2O_2$, space group $P2_1/c$, $a = 3{\cdot}4829\ (3)$, $b = 6{\cdot}1855\ (6)$, $c = 8{\cdot}9383\ (7)$ Å, $\beta = 112{\cdot}06\ (1)°$ (85 K), $Z = 2$, has been reinvestigated for the purpose of developing a method for the accurate determination of H positional parameters. 2693 selected Mo $K\alpha$ reflections ($2\theta_{\max} = 163°$) were measured with the crystal at 85 K; nearly 2600 of these had $I/\sigma(I) > 15$. Intensities were corrected for scan truncation errors. Full-matrix least-squares refinements were performed with minimum $\sin\theta/\lambda$ values of 0, 0·25, 0·40, 0·50, 0·65, 0·75 and 0·85 Å^{-1}. H atoms were not refined for cut-offs above 0·75 Å^{-1}. The heavy-atom parameters converged to their final values for the 0·75 Å^{-1} cut-off. The maximum deviation for most parameters occurred with the 0·50 Å^{-1} cut-off. The final bond distances are: C—H 1·070 (18), N—H 1·021 (19), C=O 1·2385 (2), C—N 1·3313 (2) and N—N 1·3799 (2) Å; angles: N—C—H 115·4 (0·9), C—N—H, 120·7 (1·1)°. For the 0·50 Å^{-1} cut-off C—H became 0·87 Å and N—H 0·79 Å, N—C—H 113·0° and C—N—H 120·8°. R indices are in the range 0·019 to 0·029. The successful determination of the H positions can be attributed to the large number of precise, high-order reflections: 2090 reflections ($R = 0{\cdot}019$) above 0·75 Å^{-1}, and 1842 ($R = 0{\cdot}020$) above 0·85 Å^{-1}.

Introduction

H positions determined by standard X-ray methods differ systematically from those determined by neutron methods, often by as much as a few tenths of an ångström. This results from the use of a spherically symmetric atom model to describe a grossly asymmetric electron distribution around the proton. The extraction of well defined, accurate positional parameters for H atoms from X-ray data has been a hitherto unsolved problem. However, through considerations outlined below, we have arrived at a possible solution.

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Centered at the proton there exists a small region within which the electron distribution is essentially spherically symmetric. This region can be referred to as the pseudo-core. The density and size of the symmetric region presumably depend on the atom to which the H is bonded.

In an X-ray diffraction pattern scattering from the pseudo-core, virtually free of scattering from outer, less symmetric regions, would be found at high values of $s = \sin\theta/\lambda$. In order to determine H parameters without asphericity bias two key criteria must therefore be satisfied: (1) data must be obtained at s values where asphericity effects are insignificant; (2) there must be sufficient scattering from H to permit a satisfactory determination of parameters.

Preliminary experiments in this laboratory on compounds containing a number of CH groups in various environments had suggested that useful results might be obtained. There were clear indications that data below $s = 0{\cdot}65$ Å^{-1} could not be used, and that the observations/parameters ratio must be much greater than the ratio of 10/1 often used in standard structure determinations.

We have used the structure of *s*-diformohydrazide, $C_2H_4N_2O_2$, as a test case in an effort to determine the requirements for a successful determination of H parameters. The structure had been studied earlier with X-ray methods, both at room temperature and at 110 K (Tomiie, Koo & Nitta, 1958; Ottersen, 1974). More recently, after our work was completed, another room-temperature study has been reported (Tanaka, 1978).

The compound crystallizes with the symmetry of the monoclinic space group $P2_1/c$, with two molecules in the unit cell; *i.e.* the asymmetric unit contains only the moiety –NHCHO. This small asymmetric unit results in an easily manageable number of reflections, while still giving us the opportunity to observe possible differences in refinement behavior as a result of H being bonded to different elements. Such differences were indicated in a study of sucrose (Hope, Poling & Ottersen, 1976).

Experimental

Crystals of diformohydrazide were grown by slow evaporation of a water solution. A crystal of dimensions $0{\cdot}40 \times 0{\cdot}40 \times 0{\cdot}25$ mm was selected for intensity measurements on a Picker automated four-circle diffractometer equipped with an incident-beam graphite monochromator and a locally modified Enraf–Nonius gas-stream low-temperature apparatus. The temperature was kept constant within $\pm 0{\cdot}2$ K at approximately 85 K throughout the experiments. The angular coordinates $(2\theta,\omega,\varphi,\chi)$ of 14 Mo $K\alpha_1$ reflections ($\lambda = 0{\cdot}70926$ Å) with 2θ values from 81 to 110° were used in a least-squares calculation to determine the crystal orientation and cell dimensions. The cell dimensions (85 K) were found to be: $a = 3{\cdot}4829\ (3)$, $b = 6{\cdot}1855\ (6)$, $c = 8{\cdot}9383\ (7)$ Å, $\beta = 112{\cdot}06\ (1)°$, $V = 178{\cdot}47\ (3)$ Å^3.

In order to optimize diffractometer usage, reflections for which observable intensities were expected, as well as a suitable scan speed and background time, were determined prior to data collection by computer simulation. Predictions were based on positional parameters and adjusted thermal parameters already determined at 110 K (Ottersen, 1974), together with sample scan and background measurements. Adjustment of the thermal parameters consisted of simply multiplying each parameter by the temperature ratio (85/110). Simulation of the data collection was performed with the program *REFLEC* devised by one of us (HH).

In order to obtain also data for a subsequent study of the valence-electron deformation density all reflections below $s = 0{\cdot}65$ Å^{-1} were measured, whereas for higher-order reflections only those which had predicted $I/\sigma(I) > 15$ were measured. In this way 2693 reflections ($2\theta_{\max} = 163°$) were selected to be scanned.

Intensities were measured with a $\theta/2\theta$ scan at a speed of 2° min^{-1} (2θ). Background counts were taken for 20 s at each end of the scan range, defined by $[2\theta(\alpha_1) - 0{\cdot}9°] - [2\theta(\alpha_2) + 0{\cdot}9°]$. Coincidence losses were minimized by adding an attenuator filter when the count rate exceeded 9500 counts s^{-1}.

Two check reflections, remeasured after every 150 reflections, showed no systematic variations during the data collection.

Data reduction

Each recorded number of counts, N, was assigned a standard deviation $\sigma(N) = [N + (0{\cdot}0026N)^2]^{1/2}$; the factor 0·0026 reproduces the observed variance in the check reflections. Based on the values of $\sigma(N)$ for scan and background counts e.s.d.'s for the net intensities were calculated. Of the 2693 reflections measured the 2683 which had $I > \sigma(I)$ were used in the refinement procedures.

Lorentz and polarization corrections were applied to the net intensities. The polarization factor includes the polarization by the monochromator crystal (Hope, 1971).

Initial refinements showed a systematic trend in the ratio F_o/F_c, which decreased progressively with increasing 2θ. We could find no computational cause for this behavior, but the effect seemed to fit the description of truncation errors given by Denne (1977). A correction of the form $I_{\text{corr}} = I_o/(1 - T)$ was therefore applied. Using the notation of Denne, $1 - T$ is given by: $1 - T = A_1C_1 + A_2C_2 + (\lambda_a - \lambda_b)A_1[I_1(\lambda_a) + I_1(\lambda_b)] + A_2[I_2(\lambda_a) + I_2(\lambda_b)]$. The line-widths ($W_1 = 2{\cdot}9 \times 10^{-4}$ Å and $W_2 = 3{\cdot}2 \times 10^{-4}$ Å) and intensity ratio ($A_2/A_1 =$

0·499) were obtained from Compton & Allison (1935), and the wavelength values used are $\lambda_1 = 0{\cdot}70926$, $\lambda_2 = 0{\cdot}71354$ Å.

We found a very satisfactory correlation between the calculated correction factors and the observed discrepancies. All refinements described here were performed with the truncation-corrected data. The correction was computed with a program written by one of us (TO).

Refinements

The quantity $\sum w(F_o - K|F_c|)^2$ was minimized by full-matrix least-squares procedures; w is $1/\sigma^2(F_o)$. We used the form factors calculated by Doyle & Turner (1968) for C, N and O, and the contracted spherical form factor of Stewart, Davidson & Simpson (1965) for H.

The starting parameters were those used in the prediction of observable intensities (see above).

The s cut-off value was varied systematically. Refinements were performed with lower cut-offs of 0, 0·25, 0·40, 0·50, 0·65, 0·75 and 0·85 Å^{-1}, and one series also with data *below* $s = 0{\cdot}70$ Å^{-1}.

The isotropic thermal parameters found for the H atoms in refinements using low-order data were relatively large, and the H atoms were therefore refined with anisotropic thermal parameters in the refinements with $s_{\min}$ up to 0·40 Å^{-1}. H parameters could not be successfully refined for the 0·85 Å^{-1} cut-off.

Results

A summary of the results from the various refinements is presented in Table 1. Atomic parameters are given in Table 2,* and the molecular geometry is shown in Fig. 1.

The refinement behavior of the H atoms was of primary interest in this study. All refinements with $s_{\min} < 0{\cdot}65$ Å^{-1} resulted in shortened C—H and N—H distances. Surprisingly, the shortest distances occur with $s_{\min} = 0{\cdot}50$ Å^{-1} (C—H = 0·87, N—H = 0·79 Å). At $s_{\min} = 0{\cdot}65$ Å^{-1} the C—H distance settled at a value near 1·10 Å, whereas N—H was still somewhat short at 0·98 Å. With the data above 0·75 Å^{-1} both distances are close to commonly accepted values. Neutron and electron diffraction results for other compounds indicate that C—H should be near 1·08 and N—H near 1·02 Å in diformohydrazide.

The isotropic thermal parameters for the H atoms (Table 2) seem reasonable when compared with the C and N thermal parameters. R.m.s. amplitudes are about 0·13 Å for H and 0·09 Å for C and N. However, there was no good indication that a steady value had been reached, and although an isotropic temperature factor

Table 2. *Parameters from the various refinement schemes*

$s_{\min}$ (Å^{-1}) is the minimum value for $\sin\theta/\lambda$. The anisotropic temperature factor is given by $\exp[-\frac{1}{4}(B_{11}h^2a^{*2} + \ldots + 2B_{23}klb^*c^*)]$. E.s.d.'s (in parentheses) are in units of the last digit.

	$s_{\min}$	x	y	z	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
O	.85	.38048(4)	.14611(2)	.26208(1)	.864(3)	.722(2)	.547(2)	-.027(2)	-.084(2)	-.057(2)
	.75	.38056(4)	.14608(2)	.26211(1)	.855(3)	.721(2)	.541(2)	-.028(2)	-.089(2)	-.054(2)
	.65	.38064(4)	.14600(2)	.26215(1)	.850(3)	.725(2)	.540(2)	-.028(2)	-.092(2)	-.051(2)
	.50	.38093(5)	.14593(3)	.26226(2)	.827(4)	.713(4)	.525(3)	-.032(3)	-.095(3)	-.046(3)
	.40	.38090(6)	.14599(3)	.26223(2)	.826(4)	.712(4)	.527(3)	-.038(3)	-.092(3)	-.050(3)
	.25	.38072(7)	.14603(4)	.26216(2)	.837(6)	.729(5)	.536(4)	-.040(4)	-.092(4)	-.046(4)
	.0	.38081(8)	.14603(4)	.26214(5)	.841(6)	.735(6)	.535(5)	-.040(5)	-.092(4)	-.046(4)
N	.85	-.06488(4)	.10465(2)	.00325(1)	.672(3)	.566(2)	.475(2)	.057(2)	-.002(2)	-.031(2)
	.75	-.06492(4)	.10465(2)	.00326(1)	.671(2)	.555(2)	.472(2)	.059(2)	-.003(1)	-.032(2)
	.65	-.06495(4)	.10469(2)	.00326(1)	.674(3)	.549(2)	.471(2)	.061(2)	-.005(2)	-.035(2)
	.50	-.06518(5)	.10477(3)	.00320(1)	.658(4)	.529(3)	.459(3)	.063(3)	-.012(2)	-.034(3)
	.40	-.06502(6)	.10476(3)	.00326(2)	.656(4)	.531(4)	.460(3)	.061(3)	-.011(3)	-.030(3)
	.25	-.06497(7)	.10480(4)	.00335(3)	.668(5)	.535(5)	.468(4)	.069(4)	-.013(4)	-.029(4)
	.0	-.06502(8)	.10486(4)	.00332(3)	.675(6)	.536(5)	.476(5)	.071(5)	-.013(4)	-.033(4)
C	.85	.10702(5)	.21815(2)	.13872(1)	.697(3)	.561(2)	.552(2)	-.004(2)	.058(2)	-.065(2)
	.75	.10703(4)	.21817(2)	.13868(2)	.689(3)	.559(2)	.545(2)	-.003(2)	.054(2)	-.063(2)
	.65	.10697(4)	.21817(2)	.13866(2)	.687(3)	.558(3)	.544(2)	-.004(2)	.054(2)	-.065(2)
	.50	.10701(6)	.21826(3)	.13875(2)	.668(4)	.545(4)	.536(4)	-.016(3)	.063(3)	-.067(3)
	.40	.10715(7)	.21820(4)	.13876(3)	.659(5)	.544(4)	.541(4)	-.017(4)	.068(3)	-.063(4)
	.25	.10715(8)	.21819(4)	.13876(3)	.670(6)	.560(6)	.553(5)	-.022(5)	.077(4)	-.061(5)
	.00	.10725(9)	.21822(5)	.13880(3)	.675(6)	.564(6)	.557(6)	-.025(6)	.081(5)	-.065(5)
HC	.75	-.0128(54)	.3730(29)	.1360(21)	1.27(16)					
	.65	-.0115(46)	.3547(24)	.1362(18)	1.60(15)					
	.50	.0053(39)	.3485(21)	.1320(19)	1.65(16)					
	.40	.0113(24)	.3510(15)	.1301(8)	1.5(3)	1.4(3)	.2(2)	-.1(2)	-.6(2)	-.3(2)
	.25	.0006(17)	.3586(9)	.1309(6)	.8(2)	.5(2)	.1(2)	-.2(2)	-.4(2)	-.6(2)
	.0	.0004(17)	.3608(9)	.1346(6)	.6(2)	.7(2)	.0(2)	-.3(2)	-.2(2)	-.6(2)
HN	.75	-.2906(59)	.1710(31)	-.0953(23)	1.55(16)					
	.65	-.2737(51)	.1747(28)	-.0900(19)	1.79(16)					
	.50	-.2395(30)	.1565(15)	-.0737(12)	1.37(12)					
	.40	-.2573(26)	.1607(13)	-.0793(9)	2.1(3)	1.1(2)	.7(2)	.1(2)	.3(2)	-.4(2)
	.25	-.2601(23)	.1558(10)	-.0800(8)	3.3(3)	1.3(3)	1.1(2)	.0(3)	.7(2)	-1.4(2)
	.0	-.2602(23)	.1612(11)	-.0876(8)	4.0(4)	1.2(3)	1.5(3)	.2(3)	.2(3)	-1.5(3)

Table 1. *Some results from refinements using various parts of the data set*

Estimated standard deviations are given in parentheses.

$\sin\theta/\lambda$ cut-off (Å^{-1})	Number of reflections	Number of parameters	R (%)	R_w (%)	Goodness of fit	Scale for F_o	C—H (Å)	N—H (Å)	∠N—C—H (°)	∠C—N—H (°)
<0·70	493	46	2·93	3·26	8·70	0·0359 (1)	0·959 (11)	0·922 (14)	113·5 (6)	122·9 (8)
–	2683	46	2·68	3·31	4·46	0·0365 (1)	0·953 (6)	0·911 (7)	114·6 (3)	122·7 (4)
>0·25	2657	46	2·56	3·03	3·94	0·0366 (1)	0·937 (6)	0·858 (7)	112·9 (3)	122·3 (4)
>0·40	2587	46	2·37	2·56	3·06	0·0369 (1)	0·879 (8)	0·861 (8)	112·7 (4)	120·5 (5)
>0·50	2496	36	2·29	2·39	2·54	0·0369 (1)	0·873 (13)	0·794 (10)	113·0 (8)	120·8 (7)
>0·65	2281	36	1·91	1·77	1·59	0·0363 (1)	1·107 (15)	0·981 (16)	115·8 (8)	118·7 (10)
>0·75	2090	36	1·88	1·70	1·37	0·0362 (1)	1·070 (18)	1·021 (19)	115·4 (9)	120·7 (11)
>0·85	1842	28	1·96	1·80	1·29	0·0360 (1)				

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* A list of structure factors has been deposited with the British Library Lending Division as Supplementary Publication No. SUP 33806 (9 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

probably does not give a good description of the thermal motion, a possible implication is that the H form factor is not satisfactory in the *s* ranges involved.

All heavy-atom parameters converged to their final values with the 0·85 Å^{-1} cut-off. Most of the C and N parameters had already converged with the 0·65 Å^{-1} cut-off, in agreement with observations for hexamethylenetetramine (Stevens & Hope, 1975). The O, however, shifted about 2σ on changing $s_{\min}$ from 0·65 to 0·75 Å^{-1}. The low e.s.d.'s for the final heavy-atom parameters are noteworthy. Paralleling the results for H is the observation that the largest deviations from converged high-order parameters occurred with $s_{\min} = 0{\cdot}50$ Å^{-1}.

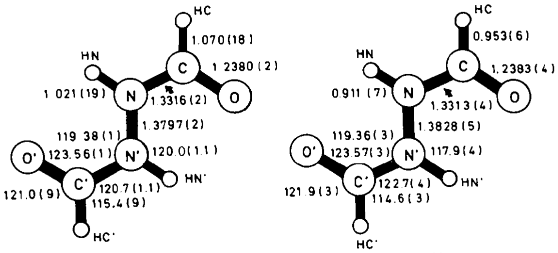
The refinements with $s_{\min} > 0{\cdot}50$ Å^{-1} resulted in increasing b_{ii} values with increasing $s_{\min}$. It is possible that this effect is the result of incomplete correction of truncation errors. Variations in the scale factor were generally small. The trend is toward a lower scale factor ($KF_o = F_c$) with increasing $s_{\min}$. The correlation coefficients between the scale factor and the B_{ii}'s for C, N and O in the high-order refinements are of the order of −0·4 to −0·5. The absolute values of other correlation coefficients are all less than 0·3.

The goodness-of-fit parameter shows a steady decrease with increasing $s_{\min}$, indicating that model errors gradually decrease. The same trend can be the result of increasingly poor counting statistics, but our data collection strategy (see above) prevents this effect. The lowest goodness of fit is about 1·3, indicating the presence of errors not accounted for.

The lowest goodness-of-fit parameter obtained before truncation correction was 1·6. The most apparent effect of using data not corrected for truncation errors is an increase in B_{ii} values. In our case it corresponds to an increase of about 0·04 Å^2 in B_{ii}.

Conclusion

This study indicates that acceptable positional parameters for H atoms can be derived from X-ray data by the use of a spherical pseudo-core model in conjunction with high-order data. The estimated standard deviations in the C—H and N—H bond lengths are still too large (0·02 Å) to claim unqualified success; nevertheless, we find the results very encouraging. We anticipate that with more experience substantial improvement will take place.



(*a*) (*b*)

Fig. 1. Bond lengths (Å) and bond angles (°) with estimated standard deviations. Results are refinements using (*a*) only data with sin θ/λ values above 0·85 Å^{-1} and (*b*) all data.

The large number of reflections per parameter (60:1) is probably necessary. Further work in this area is underway in order to determine the *s* ranges to be emphasized in data collection, as well as the relative importance of counting statistics and number of measurements.

Different refinement behavior is indicated for the two H atoms, bonded to C and N respectively, although the magnitudes of the positional e.s.d.'s preclude a sharp distinction. Stronger indications of a difference related to bonding environment has been found in a study of sucrose (Hope *et al.*, 1976), where H atoms bonded to C could be successfully refined, but not those bonded to O. The results are in accord with the idea that the electronegativity of the atom to which H is bonded plays a large role in determining the shape of the electron distribution around the proton. Work on the positioning of H bonded to O is in progress in this laboratory.

The use of high-order X-ray data in accurate structure work is becoming well accepted. However, our finding that the *maximum* deviations from the final high-order structure occurred when data below 0·50 Å^{-1} were excluded shows that the outcome can be highly dependent on the way 'high-order' is defined. It is likely that the limit must be set at least as high as 0·70 Å^{-1}, perhaps even higher for some cases.

Although the method outlined in this report requires very large data sets, the total effort expended can still be less than that in a comparable neutron study. However, the attainable precision cannot be expected to match that of neutron determinations. The wider availability of X-ray diffraction equipment would still make the X-ray method a preferred one in many cases.

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