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International Union of Crystallography Commission on Crystallographic Apparatus Single Crystal Intensity Measurement Project Report I. Inter-Experimental Agreement*

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Seventeen sets of measurements of structure factors of D(+)-tartaric acid, within the range $(\sin\theta)/\lambda < 0{\cdot}5$ Å^{-1}, were provided by the participants in the International Union of Crystallography Single Crystal Intensity Measurement Project. Each participant used a different crystal, all being derived from a single crystallization batch. The results in the Project are representative of those from a wide variety of currently used diffractometers and techniques. The instruments included four-circle, normal-beam and equi-inclination diffractometers. Cu and Mo radiations were used – unfiltered, with single and balanced filters, and with crystal monochromators.

The aims of the project were twofold: (*a*) to provide an estimate of the spread of F values associated with the range of variables involved in the project and (*b*) to locate, if possible, the sources of error. A number of agreement indices were used to measure the spread of F values both for equivalent reflections within any one experiment and for comparisons between experiments. In an attempt to allocate errors to certain plausible sources, an analysis-of-variance was applied to the weighted deviations of individual values of F from the set of mean values. The variables specified were intensity I, a θ angle factor d^* and the Miller indices h,k,l. From the values of the agreement indices and the interaction curves from the analysis-of-variance, it was possible to recognize outlier sets that differ considerably from the mean and to isolate these, where necessary, before arriving at an estimate of the error spread of the main group.

In this project, there is no one simple figure of merit which provides a ready assessment of the accuracy of measurement of structure factors. Rather, there are several ways of indicating the probable accuracy. One way is to present the spread of values of

$$R_{ij}(\equiv\Sigma\,(|F_i|-|F_j|)/\Sigma\,\tfrac{1}{2}(|F_i|+|F_j|)\,.$$

This shows that two scaled *experimental* sets of structure factors, measured under circumstances similar to those of the project, will most probably differ by 6%, agree no better than 3%, and usually no worse than 10% except in cases of extreme systematic error where it may rise to 50% or more. From the analysis-of-variance, inferences are drawn concerning the concordance of results derived from the different types of diffractometer, on features of technique associated with the diffractometers and on other aspects, including λ dependence, monochromaticity, count rates and extinction in the crystals. It is concluded that other sources of error may be present and that future projects should be designed to reveal these.

Introduction

In 1959 the Commission on Crystallographic Apparatus of the International Union of Crystallography held an Inter-Congress Meeting in Stockholm, one section of which dealt with Counter Methods for Crystal Structure Analysis (I.U.Cr., 1959). Subsequently, during the 6th Congress of the Union in Rome in 1963, two Open Sessions of the Commission were devoted to 'Automatic Single-Crystal Diffractometers for X-rays and Neutrons' (I.U.Cr., 1964*a*). Since the main features regarding the design and technical aspects of the various types of instruments appeared to have been adequately covered by these meetings, the Commission's interest was transferred to a study of the measurement of integrated X-ray intensities of single crystals.

The method chosen to investigate this complex problem involved seeking the cooperation of crystallographers, on an international basis, in a series of measurements on the same material. A project of this type would then provide crystallographers with a measure of the possible accuracy as opposed to the individual precision of a set of measurements of structure factors,

* A preliminary report on the I.U.Cr. Project was made by S.C.A. and W.C.H. in an open session of the Commission on Crystallographic Apparatus at the 7th Congress of the I.U.Cr. at Moscow, July 1966.

F. The Commission therefore extended an invitation to all interested crystallographers to participate in a Single Crystal Intensity Project (I.U.Cr., 1964*b*). The accuracy of the integrated intensities and resultant structure factors measured by current diffractometer methods, with data to be collected by the participant's normal routine procedure, could thereby be assessed. It was also hoped that analysis of the data supplied would indicate the major sources of error so that structure factor measurement of improved accuracy might be attained in the future.

For the I.U.Cr. project, a low-symmetry, low-absorption organic material was considered most suitable because the American Crystallographic Association (1962) had initiated a Single Crystal Intensity Project with similar aims, using a high-symmetry inorganic compound, CaF_2, as test specimen.* With two such projects in operation, it was clearly desirable to avoid unnecessary overlap. The A.C.A project, by its national character, could involve measurement on the same standard sphere of CaF_2, but in the I.U.Cr. project, with prospective participants from many countries, measurements were necessarily made on a different crystal in each laboratory. By this procedure, exploration of a region of variation additional to that considered in the A.C.A project was possible (Mathieson, 1969). The material chosen for the I.U.Cr. project was D(+)-tartaric acid. All crystals were from a single crystallization batch, grown by A. McL. Mathieson. Each participant was supplied, by air mail, with approximately 12 well-developed small crystals.

The lattice constants were remeasured for the I.U.Cr. project by Cooper (1966), using Bond's (1960) method. $a=7{\cdot}7290\pm1$, $b=6{\cdot}0004\pm1$, $c=6{\cdot}2126\pm1$ Å and $\beta=100{\cdot}153°\pm2$ at 25°C.† The space group of tartaric acid is $P2_1(C_2^2)$.

Each participant was asked to measure the integrated intensity of every *hk*0 reflection, including all

* The results of the A.C.A project have been published (Abrahams, Alexander, Furnas, Hamilton, Ladell, Okaya, Young & Zalkin, 1967) and discussed (Mackenzie & Maslen, 1968; Abrahams, Alexander, Furnas, Hamilton, Ladell, Okaya, Young & Zalkin, 1969).

† Standard deviations are given in units of the least significant digit.

Table 1. *Participants in the I.U.Cr. Project*

Crystallographer	Laboratory
Abrahams, S. C. & Bernstein, J. L.	Bell Telephone Laboratories, Inc., Murray Hill, New Jersey, U.S.A.
Ferguson, G.	Chemistry Department, The University, Glasgow, Scotland. (Present address: University of Guelph, Ontario, Canada.)
Gabe, E.	Institute for Cancer Research, Fox Chase, Philadelphia, Pennsylvania, U.S.A. (Present address: Mines Branch, Dept. of Mines and Technical Surveys, 555 Booth Street, Ottawa, Canada.)
Gomes de Mesquita, A. H.	Philips Research Laboratories, Eindhoven, The Netherlands.
Kheiker, D. M., Nekrasov, Ju. V. & Mimrin, V. A.	Institute of Crystallography, Academy of Science of USSR, Moscow, USSR.
Lenhert, P. G.	Dept. of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee, U.S.A.
McGandy, E. L. & Ševčík, J.	Dept. of Biological Sciences, Purdue University, Lafayette, Indiana, U.S.A. (Present address: Dept. of Biochemistry and Nutrition, University of Pittsburgh, Pittsburgh, Pennsylvania, U.S.A.)
Okaya, Y.	International Business Machines Corp. Yorktown Heights, New York, U.S.A. (Present address: Dept. of Chemistry, State University of New York, Stony Brook, New York, U.S.A.)
Palmer, K. J.	U.S. Dept. of Agriculture, Albany, California, U.S.A.
Przybylska, M., Bevan, J. & Saunderson, C.	National Research Council, Ottawa 2, Canada.
Shibata, A., Yoshimatsu, M., Hori, T., Sata, M. & Araki, H.	Rigaku-Denki Co. Ltd., Akishima-Shi, Tokyo, Japan.
Smolin, G. Y.	Institute of Silicate Chemistry, Academy of Sciences of USSR, Leningrad, USSR.
Suvorov, E. V. & Kozlovsky, V. F.	Physics Department, Moscow State University, Moscow, USSR.
Townes, W. D.	U.S. Army Electronic Command, Fort Monmouth, New Jersey, U.S.A.
Wallwork, S. C.	Dept. of Chemistry, University of Nottingham, England.
Young, R. A.	Georgia Institute of Technology, Atlanta, Georgia, U.S.A.

equivalent reflections, within the range $(\sin\theta)/\lambda \leq 0{\cdot}5$ Å^{-1}; also, all reflections with positive k and l within the same $(\sin\theta)/\lambda$ range. Any X-radiation could be used. A comprehensive questionnaire was sent to each participant, replies to which gave the relevant details for each experiment (see following section).

Approximately 60 laboratories in 10 countries expressed interest in the Commission's invitation to take part in the I.U.Cr. project. Of these, 44 agreed to participate, with a total of 16 ultimately providing measurements and completed questionnaires for analysis. The participants are listed alphabetically, with their location at the time, in Table 1. One participant submitted a second set of data (experiment 11*b* in Table 2) before analysis was completed. He suggested that his first set (experiment 11*a*) might be in error due to the crystal not being completely bathed in the incident X-ray beam. The results from one active participant who used neutrons as his radiation have been omitted from analysis in this part of the Report.

With a multi-parameter structure, such as that of D(+)-tartaric acid, there is an obvious interest in comparing the various sets of experimental data with theoretical structure factors. The calculated values are however dependent on the particular model selected, not only for the molecular conformation but also for the individual atomic scattering factors. This refinement and comparison of each set against a currently acceptable theoretical model forms part II (Hamilton & Abrahams, 1969) of the Report.

A comparison against theoretical values does not necessarily provide an estimate of experimental accuracy of the measurement sets. For this purpose, the experimental data sets may be compared solely in terms of their internal and mutual consistency. This approach constitutes part I of this Report.

Experimental procedures

The questionnaires sent to the participants contained 22 major questions, of which some were sub-divided into as many as six parts. A summary of some of the more important items of information, which varied in completeness, is collected in Table 2, where each experiment has been given an identification number; further information is available on request.

The stability values given in the second and third columns are defined as $100(I_{\max}-I_{\min})/I_{\text{mean}}$ where the I values are the integrated intensities of a standard reflection measured repeatedly throughout the time interval stated. These values do not necessarily indicate a corresponding uncertainty in the final integrated intensities since some experimenters used the standard reflection values to compensate measured intensities for this variation.* The fourth column indicates the radiation used and the method of filtering or monochromatization. The fifth column gives the maximum count rate, while the method used to ensure an apparent linear response from the counting system is indicated in the sixth column.

The basic type of diffractometer is described under 'Geometry' in the seventh column, where '4-circle' refers to an instrument in which the angles φ, χ, ω and θ (see Furnas, 1957; Arndt & Willis, 1966 for terminology) may be varied, with all intensity measurements being made in the equatorial plane. 'Equi-inclination' indicates variation only of ω and υ (Buerger, 1960) within a given reciprocal lattice layer; in 'normal beam' instruments, the ω axis remains normal to the incident X-ray beam for all layers measured.

The shape of the crystal used, together with its dimensions, is given in the eighth column. Some crystals, described as 'natural', were used exactly as supplied. Others were shaped either by grinding or by cutting. The ratio of the maximum to minimum absorption correction is given in column nine for the cases where this correction was made (in four experiments); the error in individual reflections due to absorption is as much as ± 10 per cent in some of the remaining experiments. It should be emphasized here that each participant was asked to measure and report the integrated intensities and structure factors of D(+)-tartaric acid by his normal, routine method. In some participating laboratories, normal procedures involved use of spherical or cylindrical crystals; the difficulty in grinding D(+)-tartaric acid without damage to the crystal, together with lack of facilities to make absorption corrections for the natural-shaped crystal, resulted in these participants deviating from their 'normal' method. Assessment of the mosaic spread was requested. The method suggested was measurement, with a fine receiving slit, of the crystal rotation angle (ω) from 5% of the maximum intensity on one side of the peak to 5% on the other side of the peak for a reflection in the region of $2\theta \simeq 30°$. The values, in the tenth column, indicate some variation in the apparent mosaic spread of the crystal.*

The method used to determine the 'background' for each reflection is indicated in column eleven. B_1 and B_2 are the extreme positions of the scan made across the reciprocal lattice point in experiments 1, 5, 6, 7, 8, 10, 11*a*, 11*b*, 12 and 16. The 'background' count is sampled at a single angular setting in experiments 2, 3, 9 and 13. In experiment 15, the background was sampled at every $2{\cdot}5°$ interval in ω on each layer. The procedures used in experiments 4 and 14 are given explicitly in Table 2. The angles varied in each intensity measurement are indicated in column twelve, while the expression used to derive the integrated intensity is given in the final column. C_T is the total number of counts

* No evidence was presented to indicate a monotonic decrease or increase in intensity with time which might be consistent with a simple process of radiation damage to the crystals.

* The measurement gives the convolution of the mosaic spread of the crystal with the resolution function of the instrument (*e.g.* Cooper & Nathans, 1968).

Table 2. *Experimental variables in the*

Experiment	Short term stability*	Long term stability*	Radiation monochromaticity	Maximum counts per second	Method used to attain linearity	Geometry	Crystal shape (mm)
1	1·5% (4 hours)	14·4% (17 days)	Mo *K* No filter	20,000	None	4-circle	Natural $0{\cdot}81\times0{\cdot}21\times0{\cdot}20$
2	2·6% (15 mins)	3·1% (14 days)	Cu *K* Ni filter	9,200	Ni attenuator	4-circle	Ellipsoid $0{\cdot}25\times0{\cdot}23\times0{\cdot}21$
3	0·6% (2 mins)	–	Cu *K* Ni filter	20,000	Tube current reduced	4-circle	Cut $0{\cdot}30\times0{\cdot}21\times0{\cdot}19$
4	4·4% (30 mins)	4·4% (11 hours)	Cu *K* Ni filter	2,000	Attenuators	4-circle	Ellipsoid $0{\cdot}72\times0{\cdot}61\times0{\cdot}56$
5	1·6% (3 hours)	4·4% (5 days)	Mo *K* Balanced Zr, Y filters	6,000	Brass-foil attenuators	4-circle	Natural $0{\cdot}31\times0{\cdot}27\times0{\cdot}19$
6	1·4% (50 mins)	6·7% (6 days)	Mo *K* Balanced Zr, Y_2O_3 filters	2,500	Al attenuators	4-circle zero layer	Natural $0{\cdot}69\times0{\cdot}61\times0{\cdot}28$
7	1·6% (1 hour)	12·0% (7 days)	Mo *K* Balanced Zr, Y filters	11,000	Attenuators	4-circle	Cut $0{\cdot}22\times0{\cdot}22\times0{\cdot}22$
8	3·1%	6·8%	Cu *K* Balanced Co_2O_3, Ni filters	10,000	Ni attenuator	4-circle	Sphere $R=0{\cdot}117$
9	1·2% (1 hour)	3·8% (6 days)	Cu *K* ($l=0,1,2$) Mo *K* ($l=3,4,5$) Balanced Ni, Co; Zr, Sr filters	6,000	Ni attenuator	Normal beam	Natural $0{\cdot}50\times0{\cdot}25\times0{\cdot}25$
10	1·2% (45 mins)	3·2% (2 days)	Cu *K* Ni filter	6,000	Tube current reduced	Normal beam zero layer	Ellipsoid 0·18 diameter
11*a*	1·4% (9 mins)	13·8% (8 days)	Mo *K* Zr filter	100,000	None	Equi-inclination	Natural $1{\cdot}20\times0{\cdot}90\times0{\cdot}25$
11*b*	2·8% (9 mins)	3·1% (8 days)	Mo *K* Zr filter	100,000	None	Equi-inclination	Natural $0{\cdot}70\times0{\cdot}54\times0{\cdot}08$
12	–	4·1% (2·5 days)	Mo *K* Zr filter	100,000	Tube current reduced	Equi-inclination	Cut $0{\cdot}50\times0{\cdot}25\times0{\cdot}25$
13	1·8% (1 day)	8·4% (26 days)	Mo *K* Balanced $SrSO_4$, $Zr(NO_3)_4$ filters	10,000	Tube current reduced	Equi-inclination	Cut $0{\cdot}50\times0{\cdot}26\times0{\cdot}25$
14	1·6% (90 mins)	2·2% (14 days)	Mo *K* Balanced Zr, Y filters	52,000	Tabular interpolation correction	Equi-inclination	Natural $0{\cdot}41\times0{\cdot}38\times0{\cdot}22$
15	–	0·7%	Mo *K* NaCl (200)	1,500	None	Equi-inclination	Sphere $R=0{\cdot}10$
16	0·9% (2 hours)	5·2% (22 days)	Mo *K* LiF (200)	40,000	None	Equi-inclination	Ellipsoid $0{\cdot}23\times0{\cdot}20\times0{\cdot}15$

* $[100(I_{max}-I_{min})]/[I_{mean}]$.
† Absorption correction not applied but estimated as 1·19 for Cu.
‡ Scan range extreme.

participating diffractometer techniques

Max/min absorption correction	Mosaic spread	Background	Sampling	Integrated intensity
Not made	0·82°	S.R.E.‡ at B_1 and B_2	Continuous ω, 2θ	$C_T - t(C_{B_1} + C_{B_2})$
Not made	0·23	S.R.E.‡ at B on high 2θ side of peak	Continuous ω, 2θ	$C_T - tC_B$
Not made	0·21	B at high 2θ side of peak	Fixed crystal, fixed counter	$C_T - tC_B$
1·13	Not reported	First and last $3C_j$	Stepped ω, 2θ	$\sum_1^{24} C_j - 4(\sum_1^{3} C_j + \sum_{22}^{24} C_j)$
Not made	0·22	S.R.E.‡ at B_1 and B_2	Continuous ω, 2θ	$(C_{T\beta} - C_{T\alpha}) - t[(C_{B_1} + C_{B_2})_\beta - (C_{B_1} + C_{B_2})_\alpha]$
Not made	0·51	S.R.E.‡ at B_1 and B_2	Continuous ω, 2θ	$C_T - t(C_{B_1} + C_{B_2})$
Not made	0·095	S.R.E.‡ at B_1 and B_2	Continuous ω, 2θ	$(C_{T\beta} - C_{T\alpha}) - t[(C_{B_1} + C_{B_2})_\beta - (C_{B_1} + C_{B_2})_\alpha]$
1·02	0·20	S.R.E.‡ at B_1 and B_2	Continuous 2θ	$(C_{T\beta} - C_{T\alpha}) - t[(C_{B_1} + C_{B_2})_\beta - (C_{B_1} + C_{B_2})_\alpha]$
Not made†	0·29	S.R.E.‡ at B	Continuous ω	$(C_{T\beta} - C_{T\alpha}) - (C_{B\beta} - C_{B\alpha})$
Not made	0·5	S.R.E.‡ at B_1 and B_2	Continuous ω	$C_T - t(C_{B_1} + C_{B_2})$
Not made	0·75	S.R.E.‡ at B_1 and B_2	Continuous ω	$C_T - t(C_{B_1} + C_{B_2})$
Not made	0·67	S.R.E.‡ at B_1 and B_2	Continuous ω	$C_T - t(C_{B_1} + C_{B_2})$
Not made	0·33	S.R.E.‡ at B_1 and B_2	Continuous ω	$C_T - t(C_{B_1} + C_{B_2})$
Not made	Not reported	S.R.E.‡ at B	Continuous ω	$(C_{T\beta} - C_{T\alpha}) - (C_{B\beta} - C_{B\alpha})$
1·01	0·76	5-point plateaux at α- and β-edges, B_1 and B_2	Stepped ω	$\sum_{B_1}^{B_2} C_j - (B_2 - B_1)/10 \left[\sum_{B_1}^{B_1+5} C_j + \sum_{B_2-5}^{B_2} C_j\right]$
Not made	0·66	Function of υ on each layer	Continuous ω	$C_T - C_B$
1·00	0·12	S.R.E.‡ at B_1 and B_2	Stepped ω	$C_T - t(C_{B_1} + C_{B_2})$

obtained by the sampling technique, C_i is the count at the ith point and t is the ratio of total measurement time to that used in the background measurement. $C_{i\alpha,\beta}$ is the count obtained using the α,β member of a balanced filter pair. In experiment 14, the count measured at each ith point is the difference between those obtained using each member of a pair of balanced filters.

The integrated intensities measured in the seventeen experiments were reduced to unscaled structure factor (F) values by use of the appropriate Lorentz and polarization factors; absorption corrections were made for the four experiments indicated in column nine.

Preliminary treatment of the data

The basic data, h,k,l and unscaled F, were either supplied on punched tabulating cards or were on data sheets from which cards were then punched, each carrying a number identifying the experiment. A small number of values, reported erroneously as zero, or with a negative sign, or with an obvious error in decimal point position as judged by reference to the other sets, were eliminated. The remaining data consisted of 5641 individual structure factors lying within the limit $(\sin\theta)/\lambda=0{\cdot}5$ Å^{-1} specified in the questionnaire.

The participants were requested to provide an estimate of the standard deviation of F based on counting statistics alone; few did so however. Although counting statistics rarely reflect the true error in a measured structure factor, this information would have been valuable in establishing the minimum possible error in the reported values of F. Weights used in the scaling program and in calculation of the weighted R factors were based upon standard deviations estimated on the assumption $\sigma(F)=kF$.

Data tests

For testing the data, three procedures were used. The first procedure determined measures of internal consistency for each experiment from the data for equivalent $hk0$ reflections and for general reflections measured more than once. The experimenters had not been asked to carry out the latter measurements, and the sampling of such measurements was thus rather non-uniform. Each group of equivalent reflection data was then replaced by a single average value, the 17 sets of data were mutually scaled, and the second procedure was applied. This was an overall comparison of the data sets in pairs and of each set relative to the set of mean values. This provided a second measure of the agreement in the project data (see Mathieson, 1969). The final test procedure was an application of analysis-of-variance methods to the deviations of individual sets from the set of mean values of F in an attempt to allocate the main errors in the data among certain specified variables (see Abrahams *et al.*, 1967).

Internal consistency

For each experiment, the agreement between values of the structure factors, F_{he}, for equivalent reflections may be used to assess the degree of internal consistency. Two measures for experiment i are*

$$R_i=\sum_h\sum_e |F_{he}-\bar{F}_h|/\sum_h\sum_e |F_{he}| \quad (1)$$

and

$$wR_i=\left[\sum_h\sum_e \frac{1}{\sigma^2}(F_{he}-\bar{F}_h)^2/\sum_h\sum_e \frac{1}{\sigma^2_{he}}F^2_{he}\right]^{1/2}.\dagger \quad (2)$$

The values of R_i and wR_i are presented in Table 3.

Table 3. *Internal consistency of individual experiments as measured by agreement among equivalent and replicate reflections*

The data for experiments 4 and 14 were submitted with averaging among equivalent reflections already completed.

Experiment	No. of HKL's	No. of observations	R_i	wR_i
6	36	112	0·0552	0·0677
11*b*	42	131	0·0283	0·0592
11*a*	36	120	0·0401	0·0483
10	31	94	0·0243	0·0454
8	36	70	0·0139	0·0279
13	38	131	0·0186	0·0262
5	40	314	0·0099	0·0236
12	39	134	0·0098	0·0209
9	38	105	0·0111	0·0209
15	37	126	0·0094	0·0193
7	40	138	0·0120	0·0179
1	17	42	0·0083	0·0177
3	39	135	0·0095	0·0126
16	39	128	0·0074	0·0119
2	36	72	0·0075	0·0104

Scaling the data

When the data for each group of repeated or equivalent reflections were replaced by a single average value and when a number of obviously anomalous observations were eliminated, there resulted 4265 structure factor values representing 332 non-equivalent hkl reflections. These were placed on a common scale by the method of Hamilton, Rollett & Sparks (1965) and the data, so scaled, are listed in Table 4. This least-squares procedure assigns as much as possible of the discrepancy between F values to differences in scale factor, and it is assumed here that this is appropriate because all structure factors were derived on a relative scale. Table 4

* $\bar{F}_h$ is the mean over all experiments.

† It will be noted that with the assumption $\sigma(F)=kF$ (2) simplifies considerably. Thus

$$wR_i=\left[\sum_h\sum_e \frac{(F_{he}-\bar{F}_h)^2}{F_{he}^2}\Big/ r\right]^{1/2} \simeq \left\langle\left(\frac{\Delta F}{F}\right)^2\right\rangle^{1/2} \quad (2a)$$

where r is the number of reflections and $\langle\ \rangle$ denotes 'average value'.

Table 4. *Structure factor values, on a common scale, from the different data sets*

H	K	L	[illegible]	1	2	3	4	5	6	7	8	9	10	11b	12	13	14	15	16	11a
1	0	0	760	0	717	770	721	735	805	742	734	716	804	0	752	0	0	783	754	844
2	0	0	1034	0	927	977	980	986	955	946	924	952	1054	1109	918	1740	973	988	945	1075
3	0	0	2298	0	1813	2310	1891	2357	2307	2056	2020	2082	2441	2729	1909	4046	2006	2540	2243	2019
4	0	0	266	0	224	215	230	236	0	189	226	233	248	500	413	244	170	231	204	424
5	0	0	300	0	287	283	299	275	274	285	273	301	278	334	274	456	297	281	264	337
6	0	0	239	0	232	225	242	223	230	226	222	240	212	256	219	369	239	206	217	260
7	0	0	252	0	251	228	256	227	237	237	236	263	0	258	219	387	247	232	236	268
1	1	0	1192	0	1039	1182	1043	1107	1145	1140	1119	1226	1201	0	1063	2016	0	1198	1116	1095
2	1	0	1075	0	967	1038	1004	995	1024	1020	961	1060	1061	1264	927	1772	993	1026	987	1095
3	1	0	1049	0	952	1012	987	976	963	989	934	1029	998	1163	898	1778	1054	994	962	1099
4	1	0	715	680	686	694	701	666	649	682	657	696	606	780	608	1168	670	687	653	776
5	1	0	608	579	585	592	595	571	553	581	569	612	568	646	521	984	580	573	560	664
6	1	0	483	288	479	472	492	475	429	483	462	508	446	514	423	808	482	464	449	540
7	1	0	923	889	857	854	866	870	886	895	0	936	0	930	783	1449	916	866	866	982
0	2	0	3567	0	2352	3425	2754	3581	3722	3063	3149	3843	3824	3766	2635	6494	0	4110	3221	0
1	2	0	2524	0	1929	2445	1851	2417	2621	2329	2269	2737	2722	2906	2044	4624	0	2751	2387	1828
2	2	0	605	0	591	581	642	537	562	576	590	587	580	658	513	973	580	575	542	587
3	2	0	1455	902	1347	1441	1374	1392	1455	1432	1298	1495	1459	1666	1266	2390	1486	1470	1396	1461
4	2	0	1010	964	954	985	962	925	947	976	927	984	937	1106	864	1681	1013	971	940	1040
5	2	0	724	702	690	695	689	665	629	703	691	718	665	780	617	1170	773	698	668	754
6	2	0	479	467	466	456	472	441	437	475	449	491	416	498	409	774	468	463	454	509
7	2	0	701	680	673	641	667	647	693	685	653	722	0	695	587	1130	686	666	667	727
1	3	0	463	444	452	456	462	409	444	448	433	467	447	502	406	743	0	452	425	416
2	3	0	1544	1460	1408	1508	1460	1419	1561	1501	1447	1574	1521	1698	1324	2469	0	1544	1464	1341
3	3	0	1185	1045	1121	1167	1113	1090	1149	1152	1108	1207	1169	1209	1024	1930	0	1157	1125	1104
4	3	0	575	565	568	563	526	527	561	564	543	572	553	622	504	942	495	557	550	563
5	3	0	929	916	912	900	818	872	881	921	880	940	844	1004	819	1510	818	923	902	935
6	3	0	1133	1111	1094	1050	963	1070	1126	1095	1049	1159	1123	1187	986	1843	1062	1109	1092	1147
0	4	0	1416	1373	1318	1392	1228	1344	1392	1392	1356	1473	1381	1567	1229	2215	0	1427	1352	1223
1	4	0	1056	1036	1011	1042	942	960	1048	1023	996	1066	1043	1150	923	1688	0	1051	1000	923
2	4	0	116	0	125	90	172	103	0	104	143	108	0	109	99	140	102	0	100	93
3	4	0	138	0	150	93	284	131	0	119	202	116	0	106	103	177	107	0	129	84
4	4	0	437	423	429	421	393	387	415	409	416	447	394	484	397	685	427	431	417	449
5	4	0	523	510	517	492	466	475	500	506	497	525	469	552	457	847	536	516	506	517
1	5	0	814	796	792	777	710	735	802	794	837	820	789	876	719	1260	0	802	789	724
2	5	0	170	0	156	160	147	154	189	173	157	165	0	167	150	250	0	0	193	150
3	5	0	531	512	519	511	459	477	540	515	503	533	490	561	485	863	0	533	506	495
4	5	0	408	392	403	378	346	365	308	391	387	410	0	424	372	667	0	399	396	385
0	6	0	372	0	363	338	314	348	381	354	387	0	0	402	347	541	0	363	359	342
-7	0	1	1117	920	1047	0	1052	1084	0	1046	0	1126	0	0	959	1781	1091	1107	1075	0
-6	0	1	1685	0	1558	0	1526	1777	0	1711	1598	1744	0	0	374	2901	1738	1873	1739	0
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-4	0	1	210	0	198	0	317	169	0	175	241	179	0	0	168	307	165	0	172	214
-3	0	1	3613	0	2162	0	2630	4084	0	3068	3014	3068	0	0	2952	7105	3402	4563	3700	0
-2	0	1	132	0	122	0	130	116	0	116	120	132	0	0	0	221	118	0	104	142
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0	0	1	77	0	72	79	90	69	0	78	92	0	0	0	0	0	0	0	59	0
1	0	1	1711	0	1292	1685	1362	1681	0	1512	1440	1386	0	2049	1710	3194	1594	1888	1620	1544
2	0	1	1337	0	1136	1305	1171	1268	0	1216	1184	1192	0	1520	1198	2278	1243	1427	1233	1353
3	0	1	315	272	300	306	319	288	0	296	293	302	0	344	286	495	312	287	284	345
4	0	1	101	0	72	81	77	63	0	55	0	283	0	80	116	109	90	0	105	76
5	0	1	53	0	34	36	53	49	0	19	72	71	0	46	81	58	0	0	62	0
6	0	1	122	0	110	114	121	124	0	118	126	127	0	108	117	193	111	0	93	129
7	0	1	799	768	743	751	767	748	0	771	0	796	0	806	688	1219	783	808	739	799
-7	1	1	137	0	122	0	128	108	0	118	0	141	0	98	144	201	0	0	174	0
-6	1	1	531	0	515	0	519	493	0	501	478	514	0	494	460	835	542	528	491	0
-5	1	1	640	596	617	0	625	586	0	605	590	627	0	634	548	1021	601	614	589	714
-4	1	1	1264	965	1161	0	1205	1208	0	1220	1161	1240	0	1309	1093	2087	1264	1249	1181	1353
-3	1	1	1853	0	1554	0	1596	1778	0	1733	1658	1857	0	0	1585	3126	1787	1959	1751	0
-2	1	1	965	0	857	0	905	893	0	877	855	957	0	0	867	1637	968	939	863	966
-1	1	1	1494	0	1174	0	1217	1401	0	1310	1301	1513	0	0	1334	2911	1457	1504	1316	0
0	1	1	2805	0	1833	2696	1840	3015	0	2328	2308	2894	0	0	2552	5430	2744	3404	2622	0
1	1	1	1960	0	1440	1909	1421	1945	0	1806	1719	2003	0	0	1856	3418	2019	2133	1852	0
2	1	1	766	0	718	752	715	718	0	720	706	716	0	0	668	1253	766	749	703	779
3	1	1	402	378	376	396	390	378	0	390	376	386	0	427	362	648	389	367	352	413
4	1	1	2087	1916	1795	2093	1780	2088	0	1929	1928	2097	0	0	1801	3477	1986	2226	2014	0
5	1	1	577	549	553	568	565	547	0	556	534	561	0	581	500	909	559	559	528	587
6	1	1	971	931	895	951	921	930	0	922	892	961	0	974	826	1544	973	938	926	981
7	1	1	593	565	566	558	572	557	0	563	0	0	0	583	499	910	617	558	567	0
-7	2	1	617	640	642	0	647	609	0	619	0	632	0	145	562	1018	626	638	626	0
-6	2	1	1953	0	1672	0	1679	1828	0	1838	1728	1918	0	0	0	3072	1934	2004	1857	0
-5	2	1	229	0	236	0	244	198	0	208	221	212	0	0	189	342	218	0	218	0
-4	2	1	704	646	659	0	893	615	0	635	622	669	0	0	586	1093	688	0	623	720
-3	2	1	1975	1875	1698	0	1694	1923	0	1869	1937	2022	0	0	1679	3293	1769	2117	1915	1881
-2	2	1	782	0	747	0	747	734	0	743	711	801	0	0	695	1281	675	784	715	755
-1	2	1	848	0	798	0	824	814	0	815	790	862	0	0	770	1379	726	862	785	756
0	2	1	499	0	494	477	511	459	0	492	448	500	0	538	471	822	352	501	469	448
1	2	1	1959	0	1554	1958	1499	1929	0	1847	1863	2121	0	0	1781	3338	0	2126	1899	1597
2	2	1	1626	792	1470	1658	1439	1605	0	1580	1561	1678	0	1882	1474	2823	1660	1715	1605	1443
3	2	1	286	266	274	278	281	263	0	274	269	278	0	301	257	446	273	305	249	272
4	2	1	877	837	823	863	824	827	0	825	813	853	0	934	775	1415	861	858	808	839
5	2	1	60	0	59	33	122	0	0	48	75	28	0	47	68	58	0	0	58	0
6	2	1	769	746	731	736	725	733	0	737	701	768	0	774	668	1215	760	757	738	741
-6	3	1	192	0	190	0	185	147	0	170	172	189	0	0	172	340	175	190	177	0
-5	3	1	1012	992	975	0	916	921	0	931	952	985	0	0	888	1599	946	1008	958	1079
-4	3	1	619	603	604	0	581	560	0	584	581	613	0	0	555	989	530	600	586	656
-3	3	1	1204	1185	1123	0	1112	1109	0	1131	1133	1225	0	0	1072	1941	1047	1231	1157	1187
-2	3	1	576	557	545	0	556	536	0	539	534	584	0	0	521	920	0	569	527	529
-1	3	1	940	0	883	0	896	890	0	884	878	943	0	0	843	1487	0	942	878	820
0	3	1	1497	1459	1377	1468	1324	1454	0	1456	1458	1263	0	1639	1364	2449	0	1560	1444	1240
1	3	1	1082	1046	1001	1065	1006	998	0	1058	1026	1089	0	0	978	1781	0	1061	1029	933
2	3	1	379	362	380	381	364	339	0	357	365	386	0	396	361	642	304	346	370	336
3	3	1	1117	1081	1052	1098	998	1057	0	1066	1054	1117	0	0	991	1812	0	1131	1073	995
4	3	1	656	639	655	652	611	616	0	628	627	646	0	679	582	1051	586	633	629	600
5	3	1	770	0	735	753	696	733	0	739	720	768	0	786	697	1265	655	768	745	717
6	3	1	225	0	228	216	210	212	0	211	227	0	0	221	203	358	210	0	185	0
-5	4	1	827	788	798	0	743	735	0	807	752	801	0	0	720	1301	824	830	787	859
-4	4	1	473	447	469	0	434	418	0	456	443	462	0	0	418	722	455	496	452	472
-3	4	1	442	412	452	0	532	406	0	441	437	419	0	0	368	653	413	417	395	407
-2	4	1	1094	1083	1022	0	1003	1041	0	1078	1031	990	0	0	976	1726	0	1142	1045	990
-1	4	1	245	236	236	0	231	220	0	231	255	240	0	0	223	374	0	0	235	209
0	4	1	501	493	490	493	462	462	0	490	478	495	0	520	452	818	0	450	481	422
1	4	1	1256	1223	1140	1229	1094	1154	0	1231	1198	1240	0	1331	1121	2037	0	1313	1226	1047
2	4	1	320	274	291	281	314	257	0	279	869	278	0	294	259	434	153	287	283	247
3	4	1	385	374	367	376	346	351	0	378	362	382	0	395	336	628	0	354	384	352
4	4	1	446	431	431	434	398	407	0	435	419	438	0	451	413	710	460	442	429	393
5	4	1	239	0	236	225	217	235	0	233	226	226	0	238	227	380	238	197	228	0
-4	5	1	511	489	494	0	437	458	0	502	478	0	0	498	472	764	0	538	504	496
-3	5	1	595	573	579	0	523	545	0	574	562	589	0	608	569	862	0	607	591	558
-2	5	1	416	402	405	0	366	392	0	398	392	410	0	424	390	618	0	423	406	384
-1	5	1	753	749	747	0	444	731	0	749	715	763	0	783	704	1216	0	769	754	664

Table 4 (*cont.*)

H	K	L	μ	1	2	3	4	5	6	7	8	9	10	11b	12	13	14	15	16	11a
0	5	1	1274	1268	1178	1212	1092	1193	0	1243	1208	1268	0	1323	1165	2022	0	1335	1264	1069
1	5	1	351	343	342	337	313	316	0	333	338	344	0	362	333	541	0	353	364	299
2	5	1	319	312	325	309	281	284	0	303	316	311	0	338	291	456	0	303	323	0
3	5	1	626	0	617	584	533	559	0	588	585	608	0	628	564	978	0	643	621	0
-7	0	2	254	0	249	0	249	239	0	229	242	259	0	0	0	321	232	0	267	0
-6	0	2	420	389	393	0	414	390	0	402	389	424	0	0	0	556	423	410	391	457
-5	0	2	217	0	215	0	221	208	0	182	206	212	0	0	0	295	226	0	160	244
-4	0	2	1458	1355	1326	0	1348	1440	0	1403	1328	1381	0	0	0	2138	1431	1461	1392	1488
-3	0	2	961	0	895	0	916	919	0	887	873	886	0	0	0	1395	956	938	888	1020
-2	0	2	646	0	604	0	631	601	0	601	581	599	0	0	0	936	641	627	582	704
-1	0	2	717	0	680	0	728	708	0	712	678	0	0	0	0	803	749	0	678	0
0	0	2	2579	0	1719	2371	1745	2440	0	2225	2048	0	0	0	0	6005	2322	0	2340	0
1	0	2	1764	0	1432	1773	1523	1735	0	1661	1602	1546	0	0	2184	2846	1760	1422	1696	1751
2	0	2	559	0	553	560	573	539	0	549	530	174	0	639	586	862	579	521	526	639
3	0	2	675	595	633	650	670	638	0	644	618	660	0	734	652	992	661	624	615	734
4	0	2	165	0	160	144	307	131	0	160	172	127	0	169	142	184	134	0	134	175
5	0	2	160	0	148	151	161	147	0	154	144	160	0	158	154	223	173	0	137	169
6	0	2	440	421	414	422	442	410	0	426	406	452	0	454	416	568	428	441	429	466
-7	1	2	150	0	152	0	220	125	0	125	151	146	0	119	137	190	0	0	136	0
-6	1	2	170	0	160	0	185	151	0	178	157	174	0	161	142	228	196	0	120	184
-5	1	2	263	250	253	0	255	246	0	226	248	250	0	275	259	372	272	249	222	300
-4	1	2	398	374	389	0	395	365	0	376	364	382	0	411	387	573	393	366	354	448
-3	1	2	1491	0	1305	0	1370	1457	0	1385	1375	1480	0	0	1355	2214	1515	1530	1389	1516
-2	1	2	2551	0	1837	0	1930	2640	0	2299	2206	2663	0	0	2583	4093	2508	2829	2469	0
-1	1	2	552	0	528	0	554	529	0	516	506	537	0	0	0	844	564	497	494	503
0	1	2	844	0	769	778	813	802	0	809	762	858	0	0	977	1261	836	801	771	739
1	1	2	2791	0	1913	2781	1939	2888	0	2517	2448	2795	0	0	3087	4561	2670	3218	2671	0
2	1	2	672	595	642	661	645	641	0	656	613	646	0	748	617	1000	689	621	621	688
3	1	2	1063	930	988	1057	998	1014	0	1008	975	1136	0	1167	976	1550	1072	1013	990	1076
4	1	2	805	762	769	805	778	766	0	766	743	796	0	863	724	1169	811	733	761	823
5	1	2	519	0	503	511	512	486	0	494	478	509	0	539	472	724	532	486	487	529
6	1	2	399	0	389	383	392	378	0	400	362	401	0	408	375	550	390	368	384	409
-7	2	2	131	0	127	0	134	131	0	116	125	165	0	0	120	178	0	0	81	0
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-5	2	2	918	902	887	0	879	877	0	827	850	914	0	0	817	1355	787	936	879	1018
-4	2	2	481	464	465	0	589	452	0	452	483	467	0	0	433	656	411	440	446	499
-3	2	2	481	470	481	0	477	468	0	455	474	481	0	0	459	700	411	453	441	491
-2	2	2	1214	0	1115	0	1153	1182	0	1155	1105	1268	0	0	1167	1818	1082	1260	1152	1110
-1	2	2	474	0	456	0	475	444	0	455	469	471	0	482	464	679	442	475	422	409
0	2	2	430	405	405	595	548	490	0	387	678	358	0	366	349	519	0	304	311	299
1	2	2	444	418	431	397	575	447	0	431	585	401	0	435	401	576	426	379	387	364
2	2	2	896	830	849	885	843	857	0	867	827	891	0	0	848	1324	916	873	839	0
3	2	2	1115	1047	1035	1098	1035	1062	0	1064	1026	1089	0	1214	1022	1658	1103	1098	1059	0
4	2	2	675	647	646	655	647	636	0	650	622	669	0	713	619	961	690	666	646	653
5	2	2	595	583	570	583	569	571	0	571	553	594	0	621	545	842	600	560	568	588
6	2	2	560	0	545	534	542	532	0	535	516	566	0	578	500	788	553	538	557	0
-6	3	2	1153	1143	1090	0	1046	1087	0	1126	1064	1126	0	0	1034	1672	1029	1199	1130	1250
-5	3	2	729	715	722	0	682	681	0	675	683	731	0	0	672	1044	642	743	702	787
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-3	3	2	1014	996	984	0	958	976	0	944	961	1027	0	1025	938	1533	888	1044	959	970
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-1	3	2	615	606	608	0	606	592	0	561	604	632	0	639	583	915	530	615	580	539
0	3	2	550	539	545	539	539	538	0	522	511	584	0	583	511	812	480	543	529	472
1	3	2	561	542	557	569	559	523	0	527	539	561	0	615	519	793	499	565	557	491
2	3	2	810	769	781	803	762	761	0	775	752	792	0	0	749	1300	769	834	780	710
3	3	2	442	0	439	441	415	413	0	415	427	438	0	463	411	627	425	449	429	399
4	3	2	236	0	241	246	226	231	0	240	241	245	0	248	221	335	198	144	265	221
5	3	2	587	0	579	577	535	569	0	571	553	589	0	605	560	835	531	569	585	554
-5	4	2	522	504	515	0	493	472	0	504	497	509	0	0	495	725	510	512	517	533
-4	4	2	662	650	642	0	607	618	0	649	604	646	0	648	613	948	650	691	646	657
-3	4	2	468	458	465	0	439	443	0	456	449	476	0	466	439	668	462	466	430	434
-2	4	2	879	0	845	0	813	851	0	870	822	872	0	886	821	1245	859	909	860	781
-1	4	2	339	335	329	0	327	318	0	327	329	349	0	350	329	467	324	0	327	292
0	4	2	360	0	355	375	365	343	0	343	415	339	0	352	340	477	329	375	339	288
1	4	2	382	0	367	355	390	354	0	353	385	561	0	372	347	490	357	354	359	307
2	4	2	268	0	266	262	253	263	0	265	264	278	0	278	265	373	224	257	271	231
3	4	2	456	448	456	449	411	435	0	471	438	452	0	478	435	571	414	489	444	0
4	4	2	284	0	287	270	256	272	0	274	273	278	0	281	268	400	268	0	284	0
-3	5	2	759	753	743	0	659	712	0	745	706	745	0	0	705	1060	0	778	743	0
-2	5	2	160	0	152	0	151	137	0	169	152	156	0	0	152	213	0	0	173	143
-1	5	2	254	0	241	0	229	253	0	255	239	254	0	0	251	343	0	0	248	224
0	5	2	338	338	342	324	314	307	0	335	317	330	0	0	319	470	0	372	344	283
1	5	2	502	508	507	480	458	471	0	494	478	504	0	0	487	698	0	0	503	430
2	5	2	323	325	329	308	297	307	0	321	330	325	0	0	316	416	0	0	326	280
-7	0	3	279	0	312	0	316	278	0	267	294	273	0	151	0	296	306	0	295	0
-6	0	3	150	0	135	0	151	183	0	83	148	184	0	0	0	167	165	0	116	172
-5	0	3	673	693	693	0	705	690	0	681	664	665	0	279	0	764	733	706	677	797
-4	0	3	498	500	503	0	538	493	0	481	502	481	0	388	0	560	516	475	470	571
-3	0	3	942	944	899	0	936	942	0	940	896	928	0	739	0	1115	974	957	909	1061
-2	0	3	1449	0	1292	0	1364	1464	0	1429	1347	1428	0	0	0	1767	1546	1441	1407	0
-1	0	3	1266	0	1305	0	1312	1493	0	1473	1393	0	0	162	0	0	1569	0	1420	0
0	0	3	280	0	274	266	303	272	0	284	261	0	0	324	0	0	0	0	258	0
1	0	3	740	664	718	733	769	708	0	728	701	726	0	538	917	873	769	707	708	842
2	0	3	336	301	342	202	365	334	0	332	336	330	0	359	393	407	357	273	325	389
3	0	3	82	0	72	72	75	70	0	52	92	80	0	86	113	86	109	0	59	96
4	0	3	554	521	545	550	568	520	0	521	534	518	0	591	545	610	566	561	543	612
5	0	3	950	918	921	939	946	924	0	945	924	910	0	994	899	1048	937	966	943	1032
6	0	3	626	620	621	608	641	608	0	616	604	0	0	642	597	651	0	0	639	666
-7	1	3	210	0	224	0	213	199	0	205	220	193	0	171	206	226	217	0	203	242
-6	1	3	494	496	393	0	503	487	0	492	488	471	0	469	473	583	529	0	470	567
-5	1	3	1271	1257	1212	0	1211	1260	0	1249	1189	1221	0	1263	1207	1488	1310	1302	1229	1399
-4	1	3	576	0	557	0	577	553	0	546	539	542	0	576	589	676	586	569	539	643
-3	1	3	726	725	705	0	725	704	0	710	687	688	0	0	792	870	749	584	692	810
-2	1	3	1574	1550	1368	0	1438	1571	0	1549	1198	1546	0	0	1883	1978	1638	1645	1523	0
-1	1	3	608	585	570	0	592	574	0	582	553	566	0	0	809	712	592	0	555	0
0	1	3	1400	1332	1254	1304	1214	1364	0	1340	1277	1353	0	0	2189	1647	1429	1402	1303	1198
1	1	3	747	713	722	742	742	728	0	726	711	735	0	0	895	897	793	598	713	748
2	1	3	465	437	465	473	469	447	0	462	446	443	0	0	489	556	471	415	445	490
3	1	3	285	283	291	294	308	280	0	286	275	273	0	298	302	337	307	178	268	303
4	1	3	406	393	405	408	409	390	0	388	381	377	0	418	415	459	420	0	400	417
5	1	3	347	0	351	346	353	341	0	329	342	325	0	359	327	391	360	353	324	359
-6	2	3	386	391	389	0	385	379	0	390	368	377	0	0	348	464	339	371	371	448
-5	2	3	947	957	937	0	922	930	0	936	896	914	0	0	864	1114	869	992	911	1072
-4	2	3	320	321	317	0	337	309	0	299	313	302	0	0	316	375	320	304	293	355
-3	2	3	477	475	477	0	491	465	0	469	483	452	0	0	481	555	508	431	445	475
-2	2	3	978	995	933	0	976	991	0	967	961	961	0	0	1063	1175	902	943	950	899
-1	2	3	563	570	570	0	610	578	0	550	608	542	0	0	633	645	492	532	516	468
0	2	3	1095	1115	1060	1075	1079	1100	0	1091	1050	1103	0	0	1225	1323	848	1082	1081	0
1	2	3	958	945	925	957	924	947	0	923	920	947	0	0	1040	1135	1005	938	931	882
2	2	3	619	606	608	621	612	611	0	593	576	599	0	0	644	732	639	639	593	589
3	2	3	547	0	545	551	528	536	0	541	516	533	0	575	555	635	553	520	533	541
4	2	3	268	269	266	262	290	261	0	255	256	245	0	271	263	299	266	278	277	266

Table 4 (*cont.*)

H	K	L	μ	1	2	3	4	5	6	7	8	9	10	11b	12	13	14	15	16	11a
5	2	3	717	723	718	711	712	706	0	706	692	683	0	721	659	818	723	743	719	715
-6	3	3	278	0	291	0	281	280	0	287	274	273	0	0	280	322	221	0	267	0
-5	3	3	592	606	604	0	579	579	0	583	562	556	0	0	578	691	529	579	591	662
-4	3	3	461	0	481	0	467	466	0	463	460	452	0	0	485	550	435	325	451	499
-3	3	3	217	0	211	0	209	218	0	221	215	217	0	0	239	235	191	0	211	215
-2	3	3	500	504	503	0	497	482	0	481	483	481	0	0	517	604	459	537	498	458
-1	3	3	858	873	861	0	830	859	0	846	845	848	0	0	888	1063	778	878	839	747
0	3	3	742	750	743	746	738	736	0	739	715	735	0	755	769	873	658	789	735	642
1	3	3	483	490	490	487	488	481	0	473	474	485	0	498	497	576	439	467	471	430
2	3	3	1270	1277	1212	1283	1181	1263	0	1238	1240	1244	0	1360	1264	1521	1194	1367	1277	1136
3	3	3	488	492	503	492	488	480	0	488	469	471	0	508	501	559	448	459	495	464
4	3	3	119	0	114	105	126	119	0	94	134	141	0	0	105	131	120	0	131	108
-5	4	3	367	366	363	0	342	354	0	362	0	349	0	0	348	417	375	0	376	380
-4	4	3	390	391	401	0	371	375	0	396	378	358	0	0	385	441	396	392	390	396
-3	4	3	325	325	338	0	315	320	0	346	323	297	0	0	320	369	339	319	315	300
-2	4	3	547	554	545	0	531	538	0	559	539	523	0	0	556	642	563	533	544	479
-1	4	3	254	261	262	0	247	254	0	250	266	245	0	0	252	286	252	0	262	208
0	4	3	480	488	490	487	459	470	0	485	474	0	0	496	501	541	482	467	476	409
1	4	3	411	417	427	413	389	397	0	417	407	372	0	410	404	477	422	418	427	365
2	4	3	351	357	355	345	330	350	0	341	346	339	0	347	352	435	360	0	346	306
3	4	3	310	307	317	291	281	289	0	298	305	353	0	304	294	368	319	0	309	0
-2	5	3	298	302	300	0	274	304	0	306	243	283	0	0	313	368	0	0	286	0
-1	5	3	726	0	731	0	658	720	0	751	706	716	0	735	723	835	0	751	750	634
0	5	3	657	674	680	621	597	652	0	677	660	655	0	673	670	762	0	639	686	553
1	5	3	492	511	503	460	450	487	0	502	488	485	0	503	506	568	0	494	515	420
-6	0	4	78	0	76	0	78	91	0	87	106	75	0	0	0	36	0	0	74	0
-5	0	4	466	484	477	0	495	471	0	487	460	457	0	0	0	304	495	445	468	550
-4	0	4	807	835	807	0	821	827	0	819	780	811	0	0	0	542	882	812	810	941
-3	0	4	462	477	465	0	491	454	0	467	452	452	0	0	0	333	492	456	455	551
-2	0	4	263	477	249	0	255	248	0	232	242	0	0	0	0	184	256	0	224	0
-1	0	4	1381	1418	1280	0	1345	1414	0	1396	1333	0	0	0	0	0	1497	0	1368	0
0	0	4	1693	1567	1440	1623	1470	1627	0	1624	1523	0	0	0	3030	1447	1663	0	1611	0
1	0	4	526	483	507	509	548	494	0	519	506	518	0	0	696	433	538	0	491	599
2	0	4	941	859	921	941	947	907	0	951	892	914	0	982	1074	797	995	940	928	1066
3	0	4	937	889	921	936	941	916	0	935	906	910	0	987	1013	791	949	968	936	1060
4	0	4	850	831	857	858	866	836	0	860	813	825	0	874	904	715	865	828	872	950
-6	1	4	348	361	367	0	365	356	0	350	340	344	0	310	355	248	363	0	346	425
-5	1	4	118	0	114	0	119	78	0	99	122	104	0	123	127	103	0	0	138	175
-4	1	4	609	620	608	0	616	602	0	631	590	584	0	0	689	456	632	578	599	707
-3	1	4	400	402	393	0	413	382	0	404	379	415	0	0	489	329	429	383	386	0
-2	1	4	616	616	608	0	626	604	0	622	585	584	0	0	841	521	659	542	584	0
-1	1	4	1016	1013	959	0	987	1002	0	1003	966	914	0	0	1521	837	1076	962	949	0
0	1	4	382	375	376	370	394	369	0	384	373	363	0	0	540	359	391	343	362	348
1	1	4	164	0	165	152	161	169	0	165	167	160	0	0	200	145	165	0	156	162
2	1	4	1216	0	1161	1239	1157	1220	0	1206	1152	1197	0	0	1373	1131	1304	1245	1212	0
3	1	4	769	762	760	781	759	766	0	764	748	749	0	0	840	697	819	748	761	812
4	1	4	553	555	566	559	562	557	0	564	534	547	0	545	577	489	598	491	562	597
-6	2	4	184	0	198	0	194	190	0	204	181	0	0	167	198	129	152	0	191	222
-5	2	4	686	716	705	0	689	701	0	703	669	669	0	655	691	520	634	748	709	800
-4	2	4	587	606	608	0	612	598	0	584	585	575	0	569	616	484	530	610	593	648
-3	2	4	649	672	667	0	666	653	0	666	636	636	0	0	732	540	606	640	658	665
-2	2	4	352	368	376	0	375	367	0	370	364	358	0	0	429	282	347	262	348	332
-1	2	4	1085	0	1060	0	1073	1108	0	1102	1064	1070	0	0	1309	915	1008	1157	1070	0
0	2	4	837	844	819	862	867	847	0	840	841	815	0	0	979	723	787	842	814	0
1	2	4	997	739	1013	1035	1005	1035	0	1037	999	1027	0	0	1167	989	807	1077	1029	0
2	2	4	1097	1105	1085	1106	1044	1093	0	1123	1054	1065	0	0	1167	978	1158	1164	1112	0
3	2	4	739	756	756	732	738	741	0	752	720	726	0	746	757	664	759	734	758	0
4	2	4	685	713	709	668	680	693	0	700	678	665	0	676	668	582	717	715	725	0
-5	3	4	745	780	769	0	727	760	0	744	734	735	0	0	753	591	687	799	772	839
-4	3	4	516	538	532	0	522	529	0	511	516	500	0	0	571	432	492	478	529	554
-3	3	4	384	0	401	0	391	382	0	387	378	372	0	0	433	321	359	414	386	384
-2	3	4	212	0	224	0	215	208	0	210	216	203	0	0	251	184	208	0	217	193
-1	3	4	626	656	655	0	633	644	0	638	585	622	0	0	693	571	588	664	643	548
0	3	4	285	300	304	291	296	298	0	294	309	278	0	0	330	251	280	211	298	255
1	3	4	450	473	477	456	447	463	0	446	452	448	0	0	519	410	430	403	476	401
2	3	4	794	835	807	798	786	814	0	810	785	792	0	0	860	726	785	796	820	735
3	3	4	422	440	431	419	432	426	0	426	427	419	0	0	463	380	399	383	449	408
-3	4	4	258	0	274	0	257	242	0	277	260	254	0	0	273	217	265	0	271	252
-2	4	4	457	472	469	0	449	463	0	474	454	452	0	0	492	401	490	0	462	402
-1	4	4	575	602	595	0	560	578	0	609	567	561	0	0	609	502	611	583	599	499
0	4	4	656	688	684	649	613	671	0	677	655	641	0	0	703	558	696	676	692	575
1	4	4	313	332	338	303	304	328	0	322	325	311	0	0	335	291	352	255	314	274
-5	0	5	218	0	215	0	219	198	0	221	203	222	0	0	0	0	216	0	190	276
-4	0	5	1263	1401	1237	0	1285	1370	0	1356	1259	1287	0	0	0	472	1424	0	1353	1447
-3	0	5	818	883	836	0	852	854	0	867	831	0	0	0	0	355	912	0	833	958
-2	0	5	592	652	629	0	655	629	0	660	613	0	0	0	0	200	674	0	613	0
-1	0	5	478	463	469	0	496	465	0	487	462	0	0	0	0	0	510	0	469	0
0	0	5	275	274	274	269	287	253	0	234	256	0	0	0	538	105	285	0	248	0
1	0	5	34	0	21	0	0	0	0	36	0	0	0	0	50	33	0	0	33	0
2	0	5	146	0	148	150	151	145	0	135	0	146	0	132	202	79	157	0	139	169
3	0	5	564	546	579	554	582	559	0	580	557	561	0	0	671	345	585	0	571	642
-5	1	5	114	0	114	0	120	119	0	114	115	118	0	102	141	46	126	0	121	135
-4	1	5	1365	1493	1313	0	1339	1452	0	1426	1338	1381	0	0	1772	560	1510	0	1442	0
-3	1	5	709	741	718	0	717	716	0	718	692	665	0	0	1023	329	780	0	697	0
-2	1	5	340	338	338	0	364	339	0	324	323	0	0	0	517	166	368	0	318	0
-1	1	5	802	0	756	0	767	768	0	791	748	0	0	0	1419	406	824	0	741	0
0	1	5	421	419	431	403	435	413	0	404	408	0	0	0	671	236	453	0	404	369
1	1	5	141	0	148	140	141	141	0	147	153	141	0	126	202	67	139	0	156	138
2	1	5	216	0	224	215	230	214	0	189	0	222	0	190	281	136	218	0	234	235
3	1	5	433	439	452	424	446	438	0	443	417	429	0	404	497	292	471	0	452	465
-4	2	5	524	564	557	0	618	545	0	509	525	0	0	0	626	228	483	0	532	580
-3	2	5	842	900	857	0	854	883	0	876	831	834	0	0	1095	447	789	0	862	869
-2	2	5	780	0	794	0	792	798	0	787	766	763	0	0	1047	477	854	0	791	714
-1	2	5	225	0	236	0	252	228	0	233	238	217	0	0	315	144	182	0	227	200
0	2	5	390	401	401	380	398	386	0	375	396	377	0	0	522	241	471	0	387	337
1	2	5	520	550	528	501	520	524	0	496	511	504	0	0	639	345	600	0	520	0
2	2	5	413	423	427	399	410	423	0	408	425	410	0	0	495	289	447	0	425	389
-3	3	5	455	479	481	0	455	466	0	480	457	452	0	0	557	287	418	0	472	0
-2	3	5	476	500	477	0	481	495	0	508	478	467	0	0	586	299	453	0	493	0
-1	3	5	399	418	418	0	407	399	0	406	392	382	0	0	490	282	380	0	413	0
0	3	5	152	0	173	147	159	147	0	147	160	156	0	0	208	106	135	0	135	0
1	3	5	708	741	743	675	707	721	0	738	715	702	0	0	862	478	668	0	747	0
-3	0	6	262	0	266	0	279	263	0	279	266	0	0	0	410	44	282	0	265	0
-2	0	6	35	0	21	0	22	0	0	40	0	0	0	0	0	0	0	0	58	0
-1	0	6	77	0	84	0	171	0	0	0	81	0	0	0	0	14	0	0	37	0
0	0	6	499	477	503	457	492	465	0	510	478	0	0	0	1028	81	506	0	494	0
-2	1	6	561	574	579	0	567	561	0	568	548	0	0	0	891	167	609	0	545	0
0	1	6	457	482	494	427	487	484	0	479	353	0	0	0	747	181	0	0	466	422

also contains, in the column headed μ, the mean values ($\bar{F}_h$) averaged over all experiments.

In the case of set 3, no $\bar{h}$ reflections were reported; the total listed for this set in Table 4 is therefore only 179. For 11*a*, 251 are listed but only 164 for 11*b*; again mainly $\bar{h}$ reflections were omitted. For experiments 6 and 10, only *hk*0 data were submitted.

Mutual consistency

Two measures of mutual consistency of the data of sets *i* and *j* are the quantities

$$R_{ij}=\sum_h |F_{hi}-F_{hj}|/\tfrac{1}{2}\sum_h (F_{hi}+F_{hj})\equiv\sum_h |\Delta F_{hij}|/\sum_h |F_{hij}| \quad (3)$$

and

$$wR_{ij}=\left[\sum_h \frac{1}{\sigma^2}(\Delta F_{hij})^2/\sum_h \frac{1}{\sigma^2}(F_{hij})^2\right]^{1/2*}. \quad (4)$$

Corresponding measures of agreement between the *i*th experiment and the set of mean values are

$$R_{i\mu}=\sum_h |F_{ih}-\bar{F}_h|/\sum_h \bar{F}\equiv\sum_h |\Delta F_{hi\mu}|/\sum_h |\bar{F}_h| \quad (5)$$

and

$$wR_{i\mu}=\left[\sum_h \frac{1}{\sigma^2}(\Delta F_{hi\mu})^2/\sum_h \frac{1}{\sigma^2}\bar{F}_h^2\right]^{1/2\dagger}. \quad (6)$$

The arrays of R_{ij} and $R_{i\mu}$, wR_{ij} and $wR_{i\mu}$ are given in Table 5. Moving averages‡ of R_{ij} values are shown in Fig.1, as also are histograms of R_{ij} for each value of *i*. Inferences to be drawn from these Figures will be treated in the Discussion section.

Analysis-of-variance

For each structure factor value F_{hi} in Table 4, a quantity y_{hi} is defined. It is

$$y_{hi}=(F_{hi}-\bar{F}_h)/\sigma_{hi} \quad (7)$$

and is a weighted difference between the value of the structure factor observed in experiment *i* and the mean value over all experiments. These quantities, y_{hi}, constitute the primary observations in the analysis-of-variance.

Because the types of instrument involved in the project included four-circle ('equatorial') devices whose angular dependence was likely to be mainly on θ and 'equi-inclination' devices operating layer by layer, the choice of factors had to reflect these conditions. Six factors were therefore considered: the experiment number (n) with effect $E(n)$, the intensity range (I) with effect $I(I)$, the angular range in which the reflection was observed (d^*) with effect $A(d^*)$ and each of the Miller indices h,k,l with effects $H(h)$, $K(k)$, $L(l)$. The level of each factor and the number of observations for each level are given in Fig.2.

The analysis-of-variance model used was similar to that in the A.C.A. single crystal intensity report (Abrahams *et al.*, 1967). It is assumed that

$$y_{hi}\equiv yE=\bar{y}+M+EI+EA+EH+EK+EL+\varepsilon \quad (8)$$

where $\bar{y}$ is the overall mean, which will be approximately zero because of definition (7),
M is the sum of the main effects E, I, A, H, K, L and also approximates to zero as a result of the scaling procedure,
EI is an experiment-intensity interaction effect,†
EA is an experiment-angle interaction effect,
EH, EK, EL are experiment–Miller-index interaction effects, and ε is a random error, assumed to be normally distributed with zero mean.

The standard analysis-of-variance technique determines the parameters in (8), both under general and specified linear hypotheses, using a least-squares method. Small changes in scale which may arise from different weighting schemes or by omission of individual data – including whole experiments – will have practically no influence on the interaction effects.‡

It is important for the reader to understand that the analysis-of-variance model used determines the various effects independent of one another; for example, the significance of an EH effect is not at all dependent on whether or not there is an EL effect. If the model is incomplete, however, EH and EL might both depend on some source of systematic error which was not considered and thus show an apparent correlation.

The analysis-of-variance was carried out for the group comprising all experiments, with the exception of 6 and 10 which are two-dimensional experiments. The F-ratios [see Abrahams *et al.* (1967) for terminology] calculated for the five hypotheses that the interaction effects are zero are compared (Table 6) with the significant value of F at the 0·005 level. Where this value, $F_{n2,n1}$ exceeds the tabulated value of $F_{n2,n1,\alpha}$ the

$$* \simeq \left\langle\left(\frac{\Delta F_{hij}}{F_{hij}}\right)^2\right\rangle^{1/2} \text{ if } \sigma(F)=kF. \quad (4a)$$

$$\dagger \simeq \left\langle\left(\frac{\Delta F_{hi\mu}}{F_{hi}}\right)^2\right\rangle^{1/2} \text{ if } \sigma(F)=kF. \quad (6a)$$

‡ These are histograms smoothed by convolution with a rectangular distribution function of width 0·025.

†The intensity was defined here as $[F^2(1+\cos^2 2\theta)]/\sin 2\theta$. The Lorentz factor, $1/\sin 2\theta$, is exactly appropriate only for the four-circle instruments. Even for the other experiments, I defined in this way is likely to be a more meaningful variable than F^2. For the equi-inclination experiments, I should be multiplied by a further factor of $\sin\theta/[(\sin^2\theta-\sin^2\mu)^{1/2}]$. Furthermore for monochromatized radiation, the polarization factor differs from $\frac{1}{2}(1+\cos^2 2\theta)$. Neglect of these factors would result in a few reflections being grouped in different intensity classes.

‡This was checked by arbitrarily altering the scale of experiment 1 by 50%. The interaction effects and F ratios remain the same to 1 part in 10,000. The expected large differences in the effect E and the corresponding F ratios were however evident.

Table 5(*a*). *Inter-experimental R_{ij} factors*

R_{ij} and $R_{i\mu}$ array $\times 10^3$

$j \backslash i$	13	12	11*b*	11*a*	14	15	16	1	5	7	2	4	8	3	9	6	10	Diffractometer geometry		λ	$R_{i\mu} \times 10^3$
13	–	497	359	416	430	383	443	401	435	448	482	480	460	434	406	520	508			Mo	478
12	497	–	164	126	138	151	123	128	121	120	135	136	130	130	139	140	162			Mo	141
11*b*	359	164	–	143	140	093	133	128	135	128	153	171	156	102	100	114	116	Equi-inclination	ω traverse	Mo	092
11*a*	416	126	143	–	086	118	104	111	100	096	098	093	113	099	109	116	125			Mo	093
14	430	138	140	086	–	098	071	085	070	065	106	099	092	059	076	069	088			Mo	062
15	383	151	093	118	098	–	075	071	069	085	126	135	110	066	066	042	034			Mo	058
16	443	123	133	104	071	075	–	044	038	034	070	087	057	040	060	048	071			Mo	048
1	401	128	128	111	085	071	044	–	048	041	046	072	063	062	062	072	083			Mo	053
5	435	121	135	100	070	069	038	048	–	044	048	086	060	038	061	051	057			Mo	046
7	448	120	128	096	065	085	034	041	044	–	060	071	046	043	057	059	080		$\omega, 2\theta$	Mo	049
2	482	135	153	098	106	126	070	046	085	060	–	047	061	090	091	120	145	4-circle	traverse	Cu	084
4	480	136	171	093	099	135	087	072	086	071	047	–	068	097	099	128	137			Cu	091
8	460	130	156	113	092	110	051	063	060	046	061	068	–	067	073	074	094			Cu	071
3	434	130	102	099	059	066	040	062	037	043	090	097	067	–	056	042	048	Fixed crystal/fixed counter		Cu	031
9	406	139	100	109	076	066	060	062	061	057	091	099	073	056	–	052	051	Normal beam ω		Cu/Mo	039
6	520	140	114	116	069	042	048	072	051	059	120	128	074	042	052	–	033	Zero-layer only		Mo	040
10	508	162	116	125	088	034	071	083	057	080	145	138	094	048	051	033	–			Cu	049

Table 5(*b*). *Inter-experimental wR_{ij} factors*

wR_{ij} and $wR_{i\mu}$ array $\times 10^3$.

$j \backslash i$	13	12	11*b*	11*a*	14	15	16	1	5	7	2	4	8	3	9	6	10	Diffractometer geometry		λ	$wR_{i\mu} \times 10^3$
13	–	500	373	416	433	399	435	421	437	435	447	460	443	428	406	496	499			Mo	460
12	500	–	189	176	210	177	208	196	205	218	214	240	244	231	214	124	176			Mo	204
11*b*	373	189	–	167	221	166	197	171	190	191	190	259	248	152	237	133	213	Equi-inclination	ω traverse	Mo	154
11*a*	416	176	167	–	134	146	150	133	141	144	134	185	204	144	202	120	177			Mo	122
14	433	210	221	134	–	127	110	123	097	108	116	161	180	104	167	079	114			Mo	096
15	399	177	166	146	127	–	090	105	094	099	132	147	178	107	103	047	047			Mo	082
16	435	208	197	150	110	090	–	095	085	092	117	206	153	102	153	046	072			Mo	089
1	421	196	171	133	123	105	095	–	093	094	088	115	171	103	086	116	134			Mo	092
5	437	205	190	141	097	094	085	093	–	077	092	144	143	067	158	070	058			Mo	070
7	435	218	191	144	108	099	092	094	077	–	082	172	155	080	178	061	087		$\omega, 2\theta$	Mo	080
2	447	214	190	134	116	132	117	088	092	082	–	156	139	106	171	110	130	4-circle	traverse	Cu	087
4	460	240	259	185	161	147	206	115	144	172	156	–	176	206	238	125	125			Cu	163
8	443	244	248	204	180	178	153	171	143	155	139	176	–	193	167	072	084			Cu	131
3	428	231	152	144	104	107	102	103	067	080	106	206	193	–	197	058	055	Fixed crystal/fixed counter		Cu	080
9	406	214	237	202	167	103	153	086	158	178	171	238	167	197	–	076	078	Normal beam ω		Cu/Mo	138
6	496	124	133	130	079	047	046	116	070	061	110	125	072	058	076	–	044	Zero-layer only		Mo	059
10	499	176	213	177	114	047	072	134	058	087	130	125	084	055	078	044	–			Cu	064

hypothesis may be rejected.* The probability of rejecting a true hypothesis is less than 100α per cent.

* $n1$ and $n2$ are the number of degrees of freedom; $n1$ is the number of observations minus the number of parameters determined (including the 332 means for the 332 independent reflections. n_2 is the dimension of the linear hypothesis, *i.e.* the number of independent linear relationships among the parameters of (8) specified by the hypothesis.

Estimates of the interaction effects and their standard deviations, for each of the five variables, were also derived and are given in Fig.2; for example, for the *EI* effect, the quantity plotted is $\bar{y}+E+I+EI$. It should be noted that only the difference between shapes of curves in any column is significant. That a particular curve is horizontal does not mean that there is no corresponding systematic error in the experiment but only that this

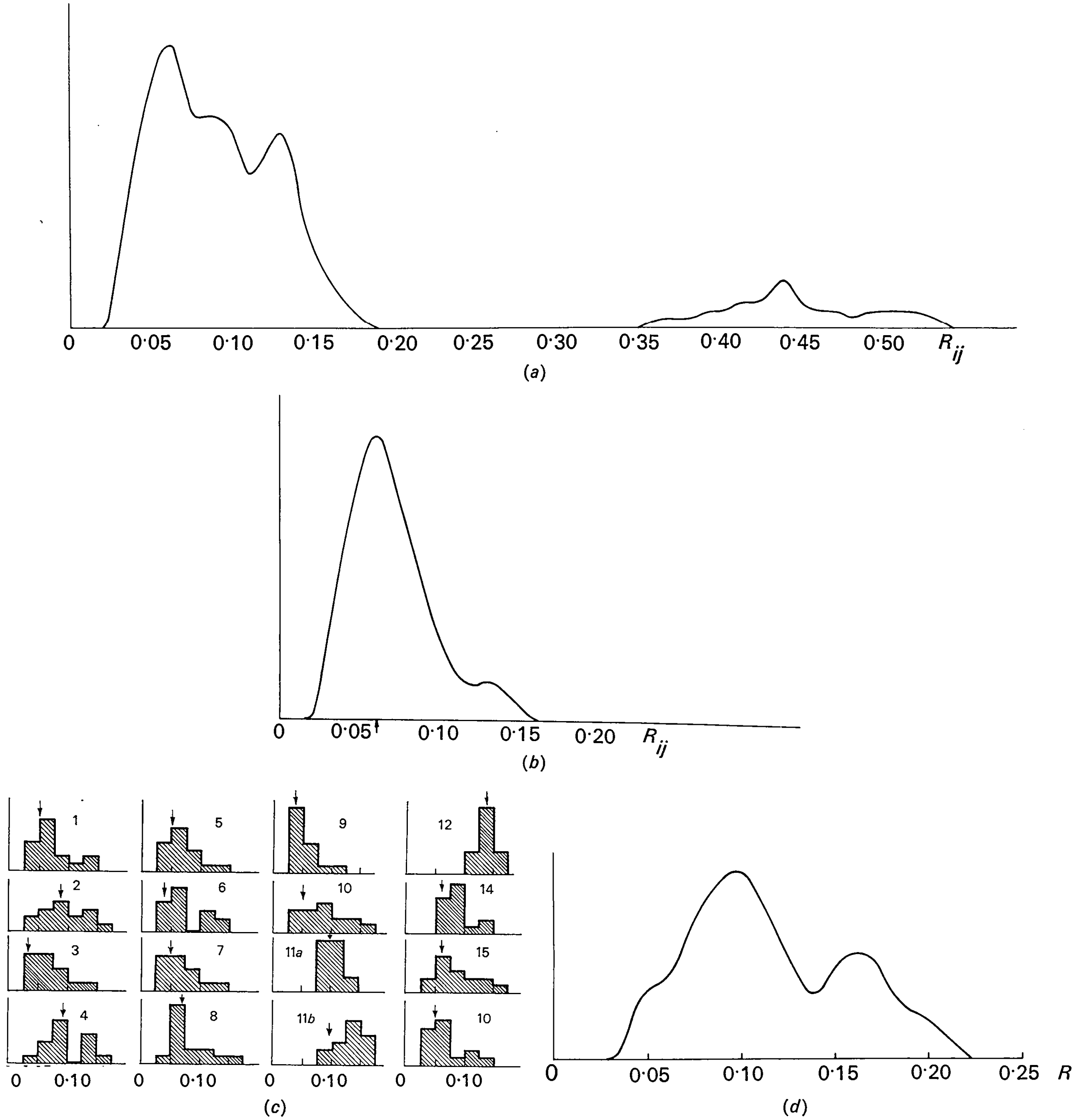

Fig. 1. Moving average curves of interexperimental R_{ij} and wR_{ij} values together with histograms of R_{ij}. (*a*) The R_{ij} curve for all sets. (*b*) As in (*a*) but with exclusion of sets 11*a*, 11*b*, 12 and 13. (*c*) The histograms of R_{ij} for individual sets *i* with that for set 13 excluded. The arows indicate the position of $R_{i\mu}$ for each *i*. (*d*) The wR_{ij} curve for the same group of sets as in (*b*).

experiment agrees well with the average insofar as its dependence on the variable of that column is concerned. Inferences from the deviation in these curves are presented in the following Discussion section.

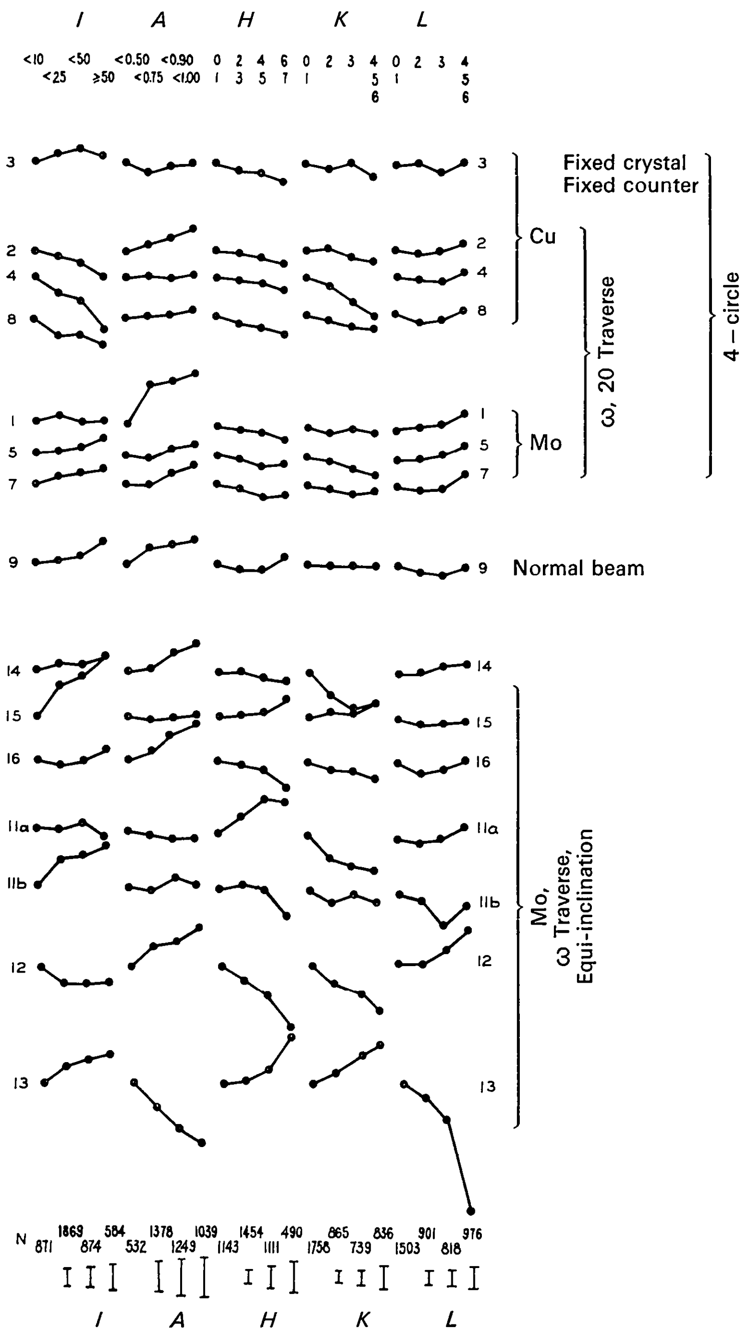

Fig. 2. Interaction effects derived by analysis-of-variance on all sets except 6 and 10. There were four levels for each factor as indicated at the top of the Figure. The numbeı N of observations at each level is indicated at the bottom. The experiment numbers are on the right and left-hand margins. Immediately below N are error bars 2σ in length where σ is the estimated standard deviation of the corresponding effect as derived from the analysis-of-variance least-squares program. Since it is only differences between the effects that can be determined by this procedure, the effect for the first level of each factor was arbitrarily set to zero, and there is therefore no associated estimated standard deviation. The error bars can thus be used only to indicate significant trends in one experiment with respect to the average or between any two experiments. The reader is cautioned to bear this in mind in interpretation of the Figure. For example, the strong downward trend with L in experiment 13 is compensated foı by an upward trend in all the other experiments. That the trend is up or down for one experiment is unimportant and reveals nothing. That the trend is different for two experiments is revealing.

Table 6. *Tests of the hypotheses that each of the five interaction effects is zero*

All experiments but 6, 10: Significant value $F_{42,3626,0\cdot005}=1\cdot65$

H_1: Experiment-*I* interaction effects are zero
$F_{42,3626}=5\cdot36$
H_2: Experiment-*A* interaction effects are zero
$F_{42,3626}=2\cdot58$
H_3: Experiment-*H* interaction effects are zero
$F_{42,3626}=3\cdot70$
H_4: Experiment-*K* interaction effects are zero
$F_{42,3626}=4\cdot50$
H_5: Experiment-*L* interaction effects are zero
$F_{42,3626}=11\cdot18$

Thus all hypotheses can be rejected with a high degree of confidence.

Discussion

As indicated in the Introduction, there are two main aspects of part I of the project – (i) assessment of the probable accuracy by reference to the spread of measured F values and (ii) the attempted allocation of some of the error to specific error-sources.

We treat (i) on the basis of the values of the various R indices already defined in the text. With the weighting procedure chosen, the wR indices give an estimate of the root-mean-square *percentage* deviation [see equations 2(*a*), 4(*a*), and 6(*a*)]. The R indices give a measure of the overall mean deviation. If the weighting scheme chosen is appropriate,* the wR is a better quantity to use than R in any arguments as to trends in the data.

First consider the estimates of internal consistency given for each set by wR_i and R_i, values of which are given in Table 3 in order of decreasing wR_i and R_i. They range from $\sim 1\%$ to $\sim 7\%$ for the former and $<1\%$ to $5\frac{1}{2}\%$ for the latter. There is a possible indication of subdivision into two groups – those below 3% in wR_i (2% in R_i) and those above. The latter group includes sets 6 and 10 which provide only zero-layer data.

It may thus be inferred from Table 3 that the *precision* in the estimate of F by an individual experimenter making measurements on *one* crystal is typically in the range of a few per cent when judged by either R or wR.†

The question of inter-set consistency is conveniently combined with that of mutual consistency by reference to the values of $wR_{i\mu}$ and wR_i, $R_{i\mu}$ and R_i which are compared in Table 7. Certain simple observations may be made from these comparisons. With the exception of the zero-layer sets 6 and 10, all $wR_{i\mu}$ ($R_{i\mu}$) are greater than the corresponding wR_i (R_i). These increases indicate the presence of errors in the data which were not evident in the tests of self-consistency. These errors must be associated with the additional variables of experimental technique and crystal specimen which occur in the inter-set comparisons. Two sets, 12 and 13, show an extreme change relative to their wR_i values – which themselves lie in an acceptable range. The overall pattern given by R_i and $R_{i\mu}$ is similar, although there are slight differences in sequence arising from different weighting given to data in different intensity ranges.

Having established certain broad features concerning the consistency of the sets, it is now appropriate to look more closely at the results for individual sets in relation to the data from the other participants. This can be done in two ways: by comparison in pairs with other individual sets, and with the set of mean values derived from all sets by a simple process of averaging. The resultant simple and weighted indices are recorded in Table 5(*a*) and (*b*) respectively. The grouping together of experiments with diffractometers of the same basic design is an obvious simplification for such tabulation. It allows comparison within each subgroup and between different techniques; also those experiments within a sub-group using a particular radiation are readily distinguished. All equi-inclination instruments used Mo radiation while, in the case of the 4-circle instruments, four used Cu and three Mo radiation. Experiment 3 used the stationary-crystal stationary-counter procedure. Experiments 9 and 10 used the normal beam procedure while sets 6 and 10 were restricted to zero-layer data.

Consider the values of $R_{i\mu}$ and $wR_{i\mu}$ in Table 5(*a*) and (*b*). For the equi-inclination devices, they may be broken into two groups, between 0·048 and 0·062 in R ($\sim 0\cdot09$ in wR) and the others $>0\cdot09$ in R($>0\cdot12$ in wR). For the 4-circle devices, the Mo sets group around 0·05 in R($\sim 0\cdot085$ in wR) while those with Cu radiation group around 0·08 in R but show wide variation, 0·087–0·163 in wR. Set 3 differs from the other Cu sets, as noted above. There is an indication that the lower R-valued group in both basic designs of diffractometer involves Mo radiation and lies in the range 0·046–0·062 in R and 0·070–0·096 in wR.

For convenient consideration of the rather large array of R_{ij} and wR_{ij} values, the alternative presentation of Fig. 1 (see footnote‡ page 10) is useful. Fig. 1(*a*) presents the curve derived from the values of R_{ij} for the complete group of data sets. It is obviously not the single-peaked function to be expected from a Gaussian distribution of errors. Rather it suggests a main group plus certain *outliers*. Identification of specific outliers, where an outlier can only be identified as different from the remaining group, and not necessarily as better or worse, is facilitated by reference to the individual histograms in Fig. 1(*c*). Supporting evidence is provided later in the analysis-of-variance results. One extreme outlier, set 13, produces the broad peak in Fig. 1(*a*) near $R=0\cdot48$. The main peak at $R=0\cdot06$ is accompanied by a partly isolated peak at $R=0\cdot14$, mainly due

* Most statistical procedures in common use depend for their validity on proper weights having been used. The weighting scheme used in the present data analysis is one that is frequently used and appears generally applicable, except for very weak reflections where counting statistics dominate.

† The precision range indicated here may be compared with that in the A.C.A. project (Abrahams *et al.* 1967; Mackenzie & Maslen, 1968). This comparison is permissible since the present test refers to measurements on *one* particular crystal by each participant and the A.C.A. project also dealt with measurements on *one* crystal.

to set 12 and a shoulder, mainly but not wholly, due to sets 11*a* and 11*b*. The corresponding R_{ij} curve after exclusion of 11*a*, 11*b*, 12 and 13 is shown in Fig.1(*b*), and this single-peaked distribution may be interpreted as due to a broad spread of error-sources throughout the sets of data involved.

Certain inferences may be made from this R_{ij} curve concerning how data from two crystallographers are probably related. Thus, from Fig.1(*b*), we may infer that if two crystallographers each independently measure, on different diffractometer assemblies, different crystals of the same low-absorption compound, then the data sets, as assessed by R_{ij}, are most likely to differ by 6%, that they are not very likely to agree better than 3% nor usually worse than 10%, but may be as bad as 50% in extreme cases of systematic error. An individual set (excluding 11*a*, 11*b*, 12 and 13) differs from the set of mean F_h values, Table 4, by approximately 5–6%, *i.e.* the mean of $R_{i\mu}$ in Table 5 for these sets. We may interpret this evidence to mean that the absolute accuracy of any set is probably, at best, 5–6% (Mathieson, 1969). The change of curve shape in going from R_{ij} [Fig.1(*b*)] to wR_{ij} [Fig.1(*d*)] is, of course, related to the weights assigned in the two procedures – R_{ij} being a measure of mean deviation while wR_{ij} is a measure of percentage deviation. If the double peak is meaningful, then it indicates a sub-group at $wR \sim 0{\cdot}10$ and another sub-group at $\sim 0{\cdot}16$, contributions to the latter mainly relating to sets 4, 8 and 9; 4 and 8 being absorption-corrected Cu radiation sets and 9 involving Cu radiation for data for $l=0,1,2$.

We now consider the possible origins of the sources of error by analysis-of-variance of the data sets. As noted earlier, the experiments were each analyzed in terms of intensity I, d^*, h, k, l. The latter four variables are essentially angular functions, chosen to accord with the operational characteristics of 4-circle and equi-inclination diffractometers.

The results for the sets are again grouped in Fig.2 according to the basic design of instrument and radiation used.

Inspection of Fig.2 indicates a larger range among the slopes of the interaction effects for the angular variables d^*, h, k, and l for the equi-inclination than for the 4-circle diffractometers. For the 4-circle devices, there is no significant difference in the spread of slopes between the experiments using Cu or Mo radiation, except for the first point of the d^* curve of experiment 1 in which unfiltered radiation was used and for experiment 4 whose k dependence has no immediately obvious correlation or explanation. In the case of the equi-inclination diffractometers, the dependence on h,k,l is particularly obvious for experiments 12 and 13.

The intensity interaction curves for the Mo sub-group (1,5,7) of the 4-circle devices, using an $\omega,2\theta$ procedure and therefore homogeneous in this respect, are approximately parallel. The three experiments of the Cu sub-group (2,4,8) using an $\omega,2\theta$ traverse, show parallel trends but downward relative to 1,5,7. The, remaining Cu radiation member, 3, which involved the stationary-crystal stationary-counter technique does not lie parallel to 2,4,8. In the equi-inclination, group, the extreme trend with intensity as in 11*b*, 13 and 15 is, by contrast, upwards.

In the case of 13, an equi-inclination device with the crystal *c* axis mounted parallel to the ω axis, the extreme monotonic dependence on l [Fig.2(*a*)] suggests an instrumental malfunction that systematically increases with increasing equi-inclination angle. A similar but opposite trend is associated with experiment 12, in which the crystal *c* axis is again parallel to ω. It is of interest to note the interaction curves for sets 12 and 13. The trends are, in general, consistently opposite. The two experiments carried out on the same diffractometer (11*a* and 11*b*) do not appear to show any significant common systematic trend with any index. More-

Table 7. *Comparison of R for the different data sets*

wR				R			
Set No.	wR_i	$wR_{i\mu}$	$\lvert wR_{i\mu}^2 - wR_i^2\rvert^{1/2}$	Set No.	R_i	$R_{i\mu}$	$\lvert R_{i\mu}^2 - R_i^2\rvert^{1/2}$
6	0·068	0·059	0·034	6	0·055	0·040	0·038
11*b*	0·059	0·154	0·142	11*a*	0·044	0·093	0·084
11*a*	0·048	0·122	0·112	11*b*	0·040	0·092	0·083
10	0·045	0·064	0·045	10	0·024	0·049	0·043
8	0·028	0·131	0·128	13	0·019	0·478	0·478
13	0·026	0·460	0·459	8	0·014	0·071	0·070
5	0·024	0·070	0·066	7	0·012	0·049	0·048
12	0·021	0·204	0·203	9	0·011	0·039	0·037
9	0·021	0·138	0·136	5	0·010	0·046	0·045
15	0·019	0·082	0·078	12	0·010	0·141	0·141
7	0·018	0·080	0·078	3	0·010	0·031	0·029
1	0·018	0·092	0·090	15	0·009	0·058	0·057
3	0·013	0·080	0·079	1	0·008	0·053	0·052
16	0·012	0·089	0·088	2	0·008	0·084	0·084
2	0·011	0·087	0·086	16	0·007	0·048	0·048
4	–	0·163	–	4	–	0·091	–
14	–	0·096	–	14	–	0·062	–

over the sets appear to show marked differences of character as assessed on the basis of the interaction curves.

In summary, experiments showing deviations significant at the approximately 2σ level are indicated in Table 8.

Table 8. *Experimental deviations from average judged significant at the 2σ level*

Variable	Experiment
I	2, 4, 8, 11, 13, 15
A	1, 13
H	12, 13, 17
K	4, 12, 13, 14, 17
L	11, 12, 13

Because of the large trends for experiments 12 and 13 which could be consistent with the presence of appreciable systematic error in these data sets, the analysis-of-variance was repeated with these sets excluded. The F ratios of the thirteen-experiment subgroup were as follows:

	H_1	H_2	H_3	H_4	H_5
$F_{36,3038} =$	5·43	1·25	2·25	2·42	1·30

Significant values of F for these numbers of degrees of freedom are

$$F_{0\cdot005}=1\cdot72\ , \qquad F_{0\cdot05}=1\cdot42\ .$$

The systematic errors with l and also with d^* for the sets involved appear to be virtually removed. Those associated with the other variables remain. In the case of experiment 14, the small but significant trend with k may be related to the fact that the crystal b axis was parallel to the ω axis.

As has been noted in comparing the curves for R_{ij} and wR_{ij} [Fig. 1(*b*) and (*d*)], it is possible to select arbitrarily a group of sets in close common accord. Thus, based on wR_i (Table 7), we might choose group (1, 2, 3, 7, 15, 16) but for $wR_{i\mu}$, group (1, 2, 3, 5, 7, 15, 16). For $R_{i\mu}$, the group might be (1, 3, 5, 7, 9, 16), and from the interaction curves (see Fig. 2 and Table 8), 3, 5, 7, 9, 16. Taking into account these four criteria, a possible concordant group is group (1, 2, 3, 5, 7, 16). We can refer to this as a *modal* group and apply the analysis-of-variance to these experiments.† The values of the resultant F ratios were

	H_1	H_2	H_3	H_4	H_5
$F_{15,1300} =$	3·145	1·459	0·643	0·428	0·997

The significant value of $F_{15,1300,0.005}=2\cdot19$; hence H_1 may be rejected with confidence. However, for this group, any correlation with the angular variables, d^*, h, k, l, has disappeared. This result illustrates the difficulty of selecting a modal group based on subjective estimates of mutually good agreement. Part II of this report shows that important differences are still present among the concordant modal group as defined here. Thus the different and individual characteristics of each crystal in respect of its 'internal morphology' as differentiated for convenience into mosaicity, extinction and absorption might be identified as other appropriate variables. Although four sets were corrected for absorption (4, 8, 14 and 16), there is no indication from the data that these sets tend to form a more concordant sub-group differentiable from the other sets.

† The members of the modal group were mutually rescaled and the changes relative to the previous scale factors were small, being 1·000, 1·000, 0·990, 1·000, 0·995, and 0·999 for sets 1, 2, 3, 5, 7 and 16. The R_{ij} values were only marginally reduced (about 0·003 at most). Because the group of sets is chosen for concordance, the $R_{i\mu}$ values for the six sets were generally smaller: 0·033, 0·053, 0·037, 0·038, 0·020 and 0·026 as compared with the original 0·053, 0·084, 0·031, 0·046, 0·049 and 0·048 respectively.

The stability estimates given in the 2nd and 3rd columns of Table 1 tend to be rather large in some cases, indicating that improved stabilization or reference to a reliable monitor would be advantageous.

Concerning the material used, D(+)-tartaric acid, earlier tests had indicated its selection from a number of possibilities considered. It proved however to be less than ideal. Several participants found that the crystals, as received, had a rather large mosaic spread, resulting in extreme cases in crystals consisting of multiple individuals of nearly parallel orientation but rotated about [010]. For a large mosaic spread, it is possible that aperture dimensions become critical with $\omega,2\theta$ traverses (Burbank, 1964) and for intensity measurements to suffer systematic error with θ. There is no direct evidence from the data of this having occurred in the project. Interaction curves against d^* for the Cu/$\omega,2\theta$ group 2, 4, 8, which are likely to be most affected by such an error, show no marked deviations.

Summary

Magnitudes of error

No simple assessment of the accuracy of determination of structure factors can be given in this project. An average over all experiments could be misleading since this would include outliers, *i.e.* experiments which, although capable of yielding structural data when treated individually (see part II of the Report) are shown by inter-experimental comparison to differ significantly from the mean. For the group of sets remaining after elimination of outliers, it is possible to present several ways of assessing their accord.

Thus, (i) we may utilize the curve of R_{ij} for the sample, excluding sets 11*a*, 11*b*, 12 and 13 [Fig. 1(*b*)]. This provides a practical estimate of the agreement, measured as R_{ij}, likely to occur between two crystallographers measuring different crystals. The results of this project imply, for materials like D(+)-tartaric acid, a probable difference of 6% and outer limits of 3% and 10%.‡

‡ This assumes that the two crystallographers know that they have not made systematic errors of the magnitude which must be present in some of the experiments of this project. There is of course no way for an individual crystallographer to be sure of this in any one-shot experiment.

(ii) We may extract a group of sets which appear to be in best agreement according to specified tests. In this project, one such group is 1, 2, 3, 5, 7, 16 for which the mean measure of agreement for R_{ij} is 5·2%. This measure is, of course, highly selective and, in this sense, somewhat artificial.

(iii) We may consider the fit of an individual set with the set of mean values. With this criterion, the mean error in the group (1, 2, 3, 5, 7, 16) ranges from 0·020 to 0·053 in R and 0·053 to 0·071 in wR. With this criterion, the mean error is 5·8% and ranges from 3 to 9%.

Error-sources

Types of diffractometer and techniques

The present analysis indicates that the 4-circle group of diffractometers appear to yield results more mutually concordant than the equi-inclination group. For the latter, quite serious malfunctions can occur and may not be obvious to the experimenter (see Table 7 and also part II of the Report) without independent experimental evidence. It is particularly advisable to apply a careful check procedure in the use of such diffractometers.

The representation in the project of the various specific techniques is unfortunately uneven with only one example of measurement with stationary-crystal stationary-counter, set 3. Both sub-groups of the 4-circle devices using $\omega,2\theta$ scans, involving Cu and Mo radiation respectively, independently show a reasonable degree of internal concordancy. The ω scan method, used in all equi-inclination sets, appears to be associated with a lower degree of internal concordancy.

λ dependence

The trends with intensity of the Cu radiation groups 2, 4, 8 (of which 4 and 8 applied absorption corrections) relative to the Mo radiation groups 1, 5, 7, suggest the possibility of a wave-length dependence of systematic error.

Monochromatization

Apart from set 1 which used no filter (and this may account for the atypical first point in the d^* interaction curve) the procedures for monochromatization – β-filter, balanced filters, crystal monochromators – were all used, and there is no clear evidence that any one is better than others.

Count rates

Despite the fact that there must be counting losses in some experiments (Table 2), there is no direct evidence from the project data that high count rates are associated with the significant intensity trends noted for sets 2, 4, 8, (Cu) or 11*b*, 13, 15 (Mo).

Specimen dependence

The analysis appears to have shown that the variables I, d^*, h, k, l do not represent the complete range of error sources, nor necessarily the most important in a diffractometer experiment. The specific characteristics of each individual crystal may well contribute an important part of the total error. Independent experimental assessment of such specimen characteristics as mosaicity, extinction, absorption, *etc.* would be required to permit statistical allocation of the error magnitude to the specific property.

The representative nature of the project

It is advisable to remind readers that the number of participants in the project is, for statistical purposes, small. Although they are probably typical of the users of diffractometers, they cannot be regarded necessarily as completely representative. Secondly, the project involved measurement by each participant attempting to use his normal, routine procedure on an individual crystal, so that the project explored a wide range of variables likely to be encountered in practice. Thirdly the project data were measured in 1965–66. The assessments offered in this Report should therefore be considered within this framework.

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International Union of Crystallography
Commission on Crystallographic Apparatus
Single-Crystal Intensity Measurement Project Report
II. Least-Squares Refinements of Structural Parameters

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The structure factors measured by the participants in the single-crystal intensity project of the I.U.Cr. Commission on Crystallographic Apparatus on D(+)-tartaric acid have been used in least-squares refinement of the structural parameters. The structure factors submitted by each participant were subjected to two refinements – once with heavy atoms only and once with all atoms including hydrogen. The parameters resulting from these refinements differ by magnitudes which suggest that the estimated standard deviations of the positional parameters obtained in the least-squares refinements are not infrequently a factor of about two too small and about $\sqrt{2}$ too small on the average. The agreement for the thermal vibration parameters may be even worse – by an additional factor of about two. These results are consistent with the indications of serious systematic errors in some of the experiments revealed in part I of this report. A modal group of six experiments with good interexperimental agreement leads to least-squares refined position parameters that are also in fair agreement; the maximum value of the ratio of the externally estimated standard deviation to the internal estimate from the least-squares refinements is about 2·5. The finding that results of possible high precision but low accuracy are not uncommon in single crystal-structure investigations is confirmed.

Introduction

In part I of this report (Abrahams, Hamilton & Mathieson, 1969), later referred to as part I, the interexperimental agreement factors and classical analysis-of-variance techniques have revealed the presence of systematic errors in many diffractometer experiments. Such errors cause differences between relative structure factors, measured by different experimenters on different specimens of the same substance, to be much larger than the internal consistency of the individual experiments would suggest. The analysis-of-variance techniques used in part I are appropriate for revealing the nature of the systematic differences among experiments without recourse to a theoretical model. Nevertheless, it is of interest to examine the results of applying the usual least-squares refinement procedure to the structure factors to determine how the possible systematic errors are manifested in the refined positional and thermal parameters.*

Refinement procedure

Each set of structure factors was subjected to least-squares refinement, using the usual model for the oxygen and carbon atoms that

$$F(hkl) = K\sum_j f_j \exp\,[2\pi i(hx+ky+lz)]\exp\,[-\sum_{ik} h_i h_k \beta_{ik}]$$

* Since limited data sets consisting of no more than 332 independent reflections were used, none of the results below should be taken as definitive determinations of the average parameters in the D(+)-tartaric acid structure, especially since the reflections used extended only to sin $\theta/\lambda = 0{\cdot}5$ Å^{-1}.