Procedure and Computer Programs for the Structure Determination of Gaseous Molecules from Electron Diffraction Data

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The procedure and the different computer programs presently used in this laboratory for the structure determination of gaseous molecules from electron diffraction data are described in some detail.

Today, the determination of molecular structures from gas electron diffraction data rests heavily on computer calculations. As our papers on structure investigations normally do not include a detailed description of the applied methods, the procedures and programs presently used in this laboratory are given in the following article.

The programs available allow for several ways of carrying out a structure analysis; the chosen method depending upon the problem and the preferences of the investigator. Therefore, only the most commonly used procedures will be described.

THE EXPERIMENTAL DATA

The radial symmetric scattering pattern is recorded on photographic plates. To obtain data for a largest possible range of scattering angles, different nozzle-to-plate (camera) distances varying from 200 to 10 cm are applied. The apparatus described by Bastiansen *et. al.*[1] permits camera distances up to about 49 cm, while data from camera distances up to 200 cm may be obtained by a more recently constructed unit.[2] In practice, the data for a structure determination consist of results from two or three camera distances in the range 130 to 10 cm.

With data from the longest camera distance present, the lower s limit is about 0.6 Å^{-1}, while the data from the shortest camera distance have an upper limit of about s $= 65$ Å^{-1}. For each camera distance, the results from three to five plates are applied.

The microdensitometer traces of the photographic plates are read by an x,y reader (D-MAC line follower), and the x,y coordinates obtained, punched on paper tape, serve as input to the programs. A new photometer, giving numerical values for the x,y coordinates as output directly, has now been finished.

THEORY

The expression for the total scattered intensity is in the independent atom approximation given by:[2-4]

$$I^T_{tot}(s) = I_b^T(s) + I_m^T(s) =$$

$$\frac{I^0}{r^2} \left[\sum_{i=1}^{M} (|f_i|^2 + \frac{4}{a_0^2 s^4} S_i) + \sum_{i \neq j=1}^{M} \sum^{M} |f_i||f_j| \cos(\eta_i - \eta_j) \frac{\sin(R_{ij}s)}{R_{ij}s} \exp(-\tfrac{1}{2}u_{ij}^2 s^2) \right] \quad (1)$$

The superscript T stands for a theoretical function while the superscript E later will be used for an experimental curve. The different symbols mean:

$s = \dfrac{4\pi}{\lambda} \sin(\theta/2)$, where θ is the scattering angle, and λ is the electron wave length.

$f_j(s) = |f_j(s)| \exp[i\eta_j(s)]$ is the elastic scattering amplitude for atom j.

$S_j(s)$ is the inelastic scattering factor for atom j.

I_0 is the intensity of the incident electron beam.

a_0 is the Bohr radius.

r is the distance from the scattering centre.

R_{ij} is the distance between the atoms i and j.

u_{ij} is the root-mean-square amplitude of vibration of the distance R_{ij}.

M is the number of atoms in the molecule.

$I_b^T(s)$ is independent of the molecular structure in this approximation and is commonly referred to as the background.

$I_m^T(s)$ is the molecular intensity.

Data processing. Defining the intensity functions (theoretical and experimental)

$$J(s) = I_{tot}(s)(4a_0^2 r^2 s^4 / I^0) \quad (2)$$

and a modified scattering amplitude *

$$|f_j'(s)| = \frac{s^2 a_0}{2} |f_j(s)| \quad (3)$$

the following expressions are obtained:

$$J_{tot}^T(s) = J_b^T(s) + J_M^T(s) \quad (4)$$

with

$$J_b^T(s) = \sum_{i=1}^{M} [|f_i'|^2 + S_i] \quad (5)$$

and

$$J_m^T(s) = \sum_{i \neq j=1}^{M} \sum^{M} |f_i'||f_j'| \cos(\eta_i - \eta_j) \frac{\sin(R_{ij}s)}{R_{ij}s} \exp(-\tfrac{1}{2}u_{ij}^2 s^2) \quad (6)$$

A levelled intensity function is defined by [5]

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* In the first Born approximation, $f_j'(s) = Z - F(s)$, where Z is the atomic number and $F(s)$ is the X-ray atomic form factor.

$$J_{\text{lev}}(s) = J_{\text{tot}}(s)/J_{\text{b}}^{\text{T}}(s) \tag{7}$$

The theoretical levelled function then has the form

$$J_{\text{lev}}^{\text{T}}(s) = \frac{J_{\text{m}}^{\text{T}}(s)}{J_{\text{b}}^{\text{T}}(s)} + 1 \tag{8}$$

and the experimental one is

$$J_{\text{lev}}^{\text{E}}(s) = \frac{J_{\text{m}}^{\text{E}}(s)}{J_{\text{b}}^{\text{T}}(s)} + \frac{J_{\text{b}}^{\text{E}}(s)}{J_{\text{b}}^{\text{T}}(s)} = \frac{J_{\text{m}}^{\text{E}}(s)}{J_{\text{b}}^{\text{T}}(s)} + k(s) \tag{9}$$

If the agreement between theory and the experiment were perfect, $k(s)$ should be a constant due to the different scales of the two curves. In practice $k(s)$ is a smooth and slowly varying function of s.

A modified molecular intensity curve is defined by

$$I^{\text{E}}(s) = \left(J_{\text{lev}}^{\text{E}}(s) - k(s)\right)\left(J_{\text{b}}^{\text{T}}(s) \frac{s}{|f_{\text{k}}'||f_{\text{l}}'|}\right) = J_{\text{m}}^{\text{E}}(s) \frac{s}{|f_{\text{k}}'||f_{\text{l}}'|} \tag{10}$$

with a corresponding theoretical curve

$$I^{\text{T}}(s) = k \sum_{i \neq j=1}^{M} \sum^{M} g_{ij/kl}(s) \frac{\sin(R_{ij}s)}{R_{ij}s} \exp(-\tfrac{1}{2}u_{ij}^2 s^2) \tag{11}$$

where

$$g_{ij/kl}(s) = \frac{|f_i(s)||f_j(s)|}{|f_k(s)||f_l(s)|} \cos[\eta_i(s) - \eta_j(s))] \tag{12}$$

The atoms k and l in eqn. 12 are usually chosen to make $g_{ij/kl}(s) \approx 1$ for one or more of the distances in the molecule.

Radial distribution functions. A radial distribution (RD) function [2,3] is calculated by Fourier transformation of the modified molecular intensity curve according to:

$$\frac{\sigma(r)}{r} = \int_{s_{\text{min}}}^{s_{\text{max}}} I(s)e^{-ks^2}\sin(sr)\mathrm{d}s \tag{13}$$

where k is a damping constant, introduced to reduce the effect from the lack of data beyond s_{max}. k is usually in the range $0-0.004$ Å^2. The missing experimental data below s_{min} lead to an inconvenient zero line (envelope) for the experimental RD curve. This envelope may be subtracted, theoretical values may be applied between zero and s_{min}, or the experimental RD curve may simply be compared to a theoretical curve for the same s interval and damping. It should be mentioned that for $g_{ij/kl}(s) =$ constant, the peak corresponding to this distance on the RD function is Gaussian, which originally was the reason for defining the modified curves, eqns. 10 and 11.

Least-squares refinement.[6,7] Our least-squares refinements are usually carried out on the modified molecular intensities (10,11). $I^{\text{T}}(s)$ depends on m parameters with starting values x_1^0, x_2^0, $\cdots, x_m^0$, and the number of observations is n. Defining

$$\Delta_k(s_k, x_1, x_2, \cdots, x_m) = I_k^E(s_k) - I_k^T(s_k, x_1, x_2, \cdots x_m), \quad k = 1, 2, \cdots n \quad (14)$$

$$N_k = \Delta_k(s_k, x_1^0, x_2^0, \cdots, x_m^0), \text{ and } V_k = \Delta_k(s_k, x_1^0 + \delta x_1, x_2^0 + \delta x_2, \cdots, x_m^0 + \delta x_m^0) \quad (15)$$

the values of the parameters which minimize the weighted square error sum

$$S = \sum_{k=1}^{n} \sum_{l=1}^{n} P_{kl} V_k V_l \quad (16)$$

shall be determined. P_{kl} are the elements of the generally nondiagonal weight matrix (**P**). To obtain the shifts in the parameters, Δ is expanded and terms beyond the linear ones in the δx's are neglected. This leads to the normal equations

$$BX = Y \quad (17)$$

X is a column vector with the elements δx_i, and the elements of **B** and **Y** are given by:

$$B_{ij} = \sum_{k=1}^{n} \sum_{l=1}^{n} P_{kl} \left[\frac{\partial I_k^T}{\partial x_i}\right]^0 \left[\frac{\partial I_l^T}{\partial x_j}\right]^0 \quad (18)$$

and

$$Y_i = \sum_{k=1}^{n} \sum_{l=1}^{n} P_{kl} N_k \left[\frac{\partial I_l^T}{\partial x_i}\right]^0 \quad (19)$$

For a diagonal weight matrix, $P_{kl} = w_k \delta_{kl}$, the expressions (16, 18 and 19) reduce to single sums. The shifts are computed by solving (17) which in the linear case is the solution of the problem, in the nonlinear case however, an iteration should converge to the proper parameter values for sufficiently close starting values.

The standard deviations σ_i and the correlation coefficients $[(\varrho_x)_{i,j}]$ for the parameters may be expressed by the elements of the moment matrix for the parameters ($\mathbf{M}_x$) which is proportional to $\mathbf{B}^{-1}$ (Refs. 6 and 7).

The weight matrix **P** may be calculated from the observations if each intensity point is measured a sufficient number of times, say q. Let

$$\Delta_{ki}^E = I_{ki}^E - I_k^E \quad (20)$$

where I_{ki}^E is the k'th experimental intensity point on curve number i, and I_k^E is the corresponding mean value from the q measurements. The moment matrix for the observations (**M**) is then given by

$$M_{kl} = \sum_{i=1}^{q} \Delta_{ki}^E \Delta_{li}^E = \varrho_{kl} \sigma_k \sigma_l, \quad k,l = 1,2,\cdots,n \quad (21)$$

σ_k is the standard deviation for observation k, and ϱ_{kl} is the correlation coefficient between observation k and l. The weight matrix is then [6]
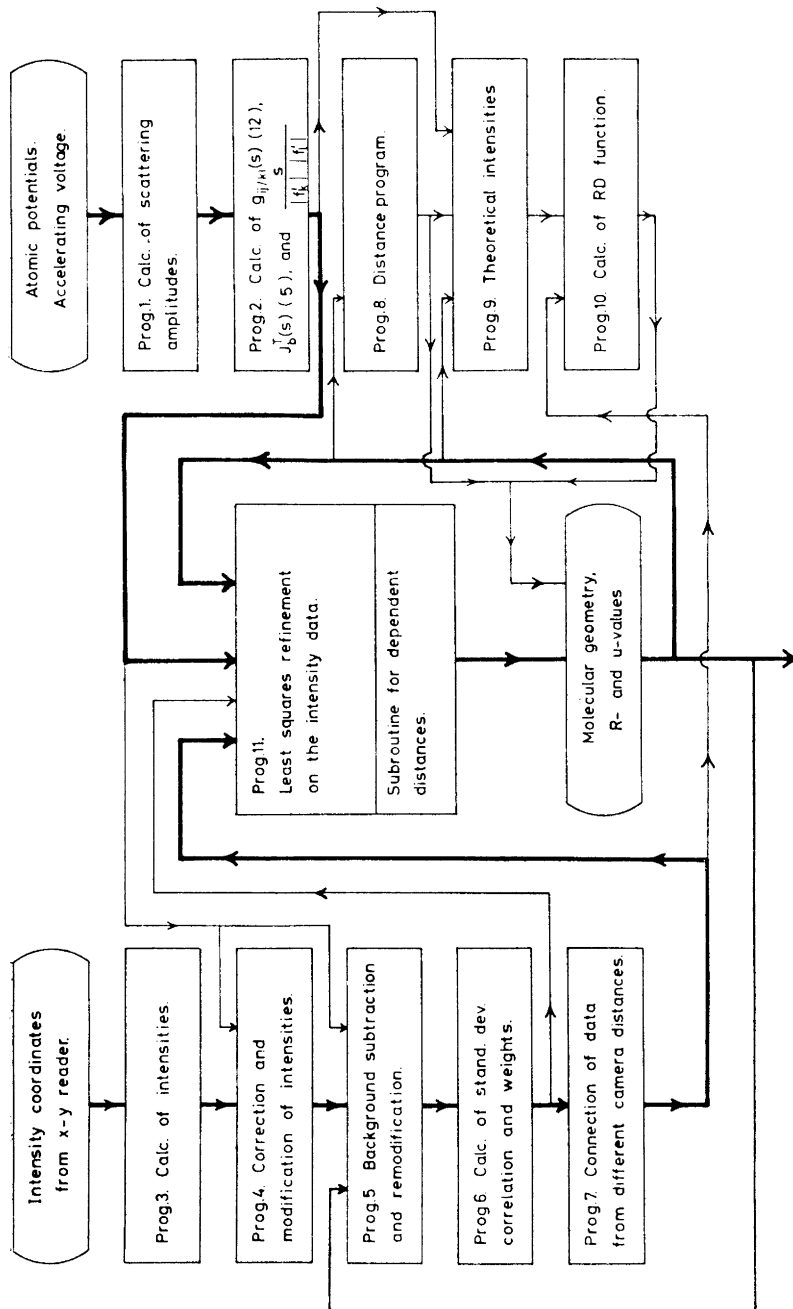
$$P \sim M^{-1} \quad (22)$$

Fig. 1. Flow of data and information in the described program system. Input to the system (the experimental intensity data, atomic potentials, and a first order model of the molecule) are symbolized. The programs are described in the text.

PROGRAM DESCRIPTIONS

The programs are written in FORTRAN for a CDC 3300 computer.

Program 1. This program computes elastic scattering amplitudes ($f(s)$) by the partial wave method.[4],[8] It is a modified version of the program originally written by J. Peacher, and it computes the scattering factors for atoms with a screening function of the type

$$\frac{Z_p(r)}{Z} = \sum_i {}^a\gamma_i\exp(-{}^a\lambda_i r) + r \sum_j {}^b\gamma_j\exp(-{}^b\lambda_j r) \tag{23}$$

where $Z_p(r)$ is the effective nuclear charge for the potential, Z is the atomic number, and the ${}^a\gamma_i, \cdots, {}^b\lambda_j$'s are parameters determined by the atomic fields. The number of the last type of terms may be zero. Accurate values of the parameters of (23) have been determined for TFD-atoms,[9] HF-atoms,[10],[11] and relativistic HFS-atoms.[11] A version of the program not dependent on analytical expressions of the screening function has recently been completed. As the accelerating voltage usually is kept close to 35kV, scattering amplitudes for the most common atoms for this voltage are stored on the magnetic disk.

Program 2. The functions $g_{ij/kl}(s)$ (12), $s/|f_k'||f_l'|$, and the theoretical background $J_b^T(s)$ (5) are computed. $g_{ii/kl}(s)$ is calculated for integer s-values, while the other two functions are calculated for s intervals of 0.125 and/or 0.25 Å^{-1} as specified by the user. The elastic scattering amplitudes are obtained from program 1, while published values are applied for the inelastic scattering factors.[12] The results are stored on the magnetic disk to be used by other programs.

Program 3. Uncorrected intensities at equidistant s values are computed from the coordinate values obtained by the x,y reader. About 300 points are used from each microdensitometer trace, and the intensities at equidistant s values are computed by Lagrange interpolation. The number of points in the interpolation scheme may be varied, but usually a four points interpolation is used. For camera distances larger than 35 cm, an s interval of 0.125 Å^{-1} is most often applied, while for shorter camera distances the use of an s interval of 0.25 Å^{-1} is common.

Program 4. The uncorrected intensities on the magnetic disk from program 3 are multiplied by the blackness correction, as the density on the photographic plate is proportional to the electron intensity only for low densities.

The data obtained from the left and right side of one plate are averaged. Previously all the intensity functions from one camera distance were averaged at this stage, but at the present this is usually not done. The intensities are corrected for effects from the rotating sector, for the use of planar photographic plates, and multiplied by s^4, giving a total correction of $s^4/(\alpha \cos^3\theta)$, where α is the aperture angle of the sector. To obtain the intensities on a form convenient for the background determination, the data are divided by the theoretical background, $J_b^T(s)$ (5). The results from this program are then the experimental levelled intensity curves (7,9), one for each photographic plate. The results are plotted and stored on the magnetic disk.

Program 5. The levelled intensity functions are plotted manually, and a smooth background is drawn. This background is fairly constant, usually in-

creasing somewhat at higher s-values and either increasing or decreasing at the lowest s-values. The coordinates for a necessary number of points on the background are punched and a smooth background function is computed by interpolation. This background is subtracted from the levelled intensity function, and the results are multiplied by

$$ J_b^T \; \frac{s}{|f_k'||f_l'|} $$

to obtain the modified molecular intensities, $I^E(s)$ (10). To check the results, the background and the function ($I^E(s)$ + background), are plotted by the computer, as the latter function is more illustrative than the levelled intensity function for short camera distances.

Plotting the background manually is somewhat laborious. Previously, it was of advantage to check the curves carefully for possible data processing errors at this stage. This is presently done by program 6, which was recently added to the system. Alternatively, the background determination may be carried out by a least-squares program. The refinement is based upon

$\dfrac{s}{|f_k'||f_l'|} \; J_{tot}^E(s)$, with $J_{tot}^T(s)$ given by eqn. 4. Using the approximation

$$ k(s) \approx \sum_{n=0}^{5} C_n s^n \tag{24} $$

N of eqn. 15 may be written

$$ N = \left[\{I^E(s) + J_b^T(s) \; \frac{s}{|f_k'||f_l'|} \; k(s)\} - (I^T(s,k,R_{ij},u_{ij}) + J_b^T(s) \; \frac{s}{|f_k'||f_l'|} \; k) \right] \approx $$

$$ \left[I^E(s) - I^T(s,k,R_{ij},u_{ij}) + J_b^T(s) \; \frac{s}{|f_k'||f_l'|} \; (\sum_{n=0}^{5} C_n s^n - k) \right] \tag{25} $$

where k, the R_{ij}'s, the u_{ij}'s, and the C_n's may be determined by the least-squares method. The program was originally written to analyze CO_2-data without drawing the background manually.* However, setting $k=0$ and varying only the C_n's, a smooth, slowly varying function going through the oscillations of the $I^E(s)$ part is obtained. According to our present, somewhat limited experience, this function is fairly close to the final background. If a theoretical intensity function is known, it may be used as input to the program, and k and the C_n's may be refined, giving a still better background. For simple molecules, also the R_{ij}'s and the u_{ij}'s may be refined. An average index of resolution according to

$$ R = k/\langle k(s) \rangle \tag{26} $$

is also computed. Input to the program is the levelled intensities for one plate from program 4, and $I^E(s)$ and the background may be filed for further calculations.

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* CO_2 has periodically been investigated for a camera distance of about 35 cm to check our wave length determination.

Program 6. The standard deviations of the intensity points $[\sigma_k(21)]$ and the correlation between the points $[\varrho_{kl}(21)]$ are computed from the different intensity curves for the same camera distance. Since these calculations are based upon only 3 to 5 curves, it is assumed that ϱ_{kl} is a function of $|s_k - s_l|$, and average correlation coefficients are computed. Nondiagonal weight matrices are calculated from the average correlation coefficients. This procedure and program will be discussed in a separate paper.[13]

Program 7. The intensity functions, one from each camera distance, are scaled to each other, either by least-squares refinement in the overlap region, or by applying scale factors given by the user. For a part of the overlap region a weighted average of the two curves is applied, the s-limits for this part, and the weights are specified by the user. The s-interval for the final curves are usually 0.125 Å^{-1} for s less than about 10 Å^{-1}, and 0.25 Å^{-1} for larger s-values. The resulting curves are stored on the magnetic disk.

Program 8. This program is a revised edition of the "Distance Program for Electron Diffraction" by G. Pauli and M. Druelinger, Indiana University. Temporary coordinate systems with origins at the center of each atom in the molecule are defined, and the input for each atom specifies the translation (bond length) and the two rotations (bond angle and dihedral angle) necessary to transfer this temporary system back to the temporary system of another specified atom. The temporary systems are transferred stepwise along the bonded network, and knowing all the transformations, the cartesian coordinates of each atom are computed in a reference system for the molecule. This reference system, being equivalent to the temporary system of atom 1, is defined by three atoms, atom 1 and 3 bonded to atom 2. Atom 1 is at the origin, atom 2 is at the negative x-axis, and the angle between the xz-plane and the plane of the three atoms is specified, formally as the dihedral angle of atom 3. The length and multiplicity of the distances are calculated from the cartesian coordinates of each atom in the reference system.

If it is difficult to give the exact value of some dihedral angle, this angle may be computed by a trial and error process. One of the cartesian coordinates of the atom in the reference system and the approximate dihedral angle (within $10° - 20°$ of the correct value) must then be given as input.

Program 9. A theoretical intensity function (11) is computed. The necessary input is $g_{ij/kl}(s)$, R_{ij}-, and u_{ij}-values. The computed intensity function may be plotted along with the experimental one and the difference to compare the two curves, and/or the curves may serve as input to the next program from the magnetic disk.

Program 10. Experimental and theoretical RD functions (13) are computed by this program. For the applied s-interval, the trapezoidal integration formula is sufficiently accurate. The necessary theoretical intensities are calculated by program 9 or program 11. The RD function may be plotted by the computer, and an additional function may be read from the disk and plotted in order to compare the curves. In the latter case, also the difference of the two curves is plotted. The results may be stored on the magnetic disk.

An enlarged version of this program also includes auto- and cross-correlation calculations. [14,15]

Program 11. This program refines the molecular parameters by the least-squares method.[6,7] A more general formula than (11) is used for the theoretical intensities.

$$I^T(s) = k \sum_{\text{distances}} \frac{A_{ij}}{R_{ij}} \, g_{ij/kl}(s)\alpha_{ij}\sin[(R_{ij}-\varkappa_{ij}s^2)s]\exp(-\tfrac{1}{2}u_{ij}^2s^2) = \sum_j F_j(s) \quad (27)$$

The sum is over all the different distances in the molecule. The distance R_{ij} repeats itself A_{ij} times due to molecular symmetry, and $\varkappa_{ij}$ is related to possible anharmonic vibrations between the atoms i and j;[16,17] it is often called the asymmetry constant.

The program allows refinement of α_{ij}, R_{ij}, u_{ij}, and $\varkappa_{ij}$. α_{ij} is usually kept equal to one for all the distances in the molecule, except when the molecule exists in more than one conformation. However, α_{ij} may also be refined to check the agreement between theory and the experiment.[18] Relations according to a certain molecular geometry are usually assumed between the distances, making some of the distances dependent parameters. If the model is determined by the k distances $R_1, R_2, \cdots, R_k$, the distances $R_{k+1}, R_{k+2}, \cdots, R_t$ are functions of the former distances, and it follows that

$$\frac{\partial I^T}{\partial R_i} = \frac{\partial F_i}{\partial R_i} + \sum_{j=k+1}^{t} \left[\frac{\partial F_j}{\partial R_j}\right]\left[\frac{\partial R_j}{\partial R_i}\right], \ (i \leqslant k) \quad (28)$$

It may be convenient to regard some angles as independent parameters. If R_i in eqn. 28 represents an angle, $(\partial F_i/\partial R_i)=0$, which is achieved by using the corresponding $A_i=0$ in eqn. 27.

A special subroutine must be programmed for each molecule. This subroutine computes the dependent distances and their derivatives from the independent distances and angles.* The partial derivatives $[\partial R_j/\partial R_i, (28)]$ are in many cases calculated numerically. If the shrinkage effect [19] is known, the values are included in the expressions for the dependent distances.

In principle, also relations between the mean amplitudes of vibration (u_{ij}) exist. The present version of the program allows a reduction of the number of u parameters only by giving some of them the same shift.

If the molecule exists in two conformations, α may be used to obtain the fraction of each conformation. The equal distances of the two forms are given $\alpha=1$. For the other distances $\alpha=\alpha'$ for one of the conformations, and $\alpha=1-\alpha'$ for the other, and α' is refined.

In the standard version, a diagonal weight matrix is applied. (Another version of the program using a nondiagonal weight matrix is discussed in a separate article.[13]) The weights are usually calculated from

$$\begin{aligned} w &= \exp[-w_1(s-s_1)^2] & & \text{for } s < s_1 \\ w &= 1 & & \text{»} \ \ s_1 \leqslant s \leqslant s_2 \quad (29) \\ w &= \exp[-w_2(s_2-s)^2] & & \text{»} \ \ s > s_2 \end{aligned}$$

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* The subroutine may also compute some dependent angles to obtain their values and standard deviations in the least-squares calculations.

where w_1, w_2, s_1, and s_2 are constants specified by the user. The standard deviations computed for the intensity points (program 6) are fairly constant for the data from one camera distance, though a slight increase with increasing s is usually observed. The standard deviations are increasing as the camera distances decrease. These results indicate that a smaller weight should be given to the data at higher s-values according to eqn. 29. However, most of the approximations in the theory [4] lead to uncertainty in the theoretical function at smaller s-values, and according to our experience, the agreement with the experimental data is somewhat poorer in this range. The weights are therefore usually reduced below $s \approx 5$ Å^{-1}. The weights for all the intensity points may also be given as data to the program.

Standard deviations and correlation coefficients for the parameters are calculated from the usual expressions.[6,7] For the dependent distances, the standard deviations are computed according to

$$\sigma_m{}^2 = \sum_{i=1}^{k} \left(\frac{\partial R_m}{\partial R_i}\right)^2 \sigma_i{}^2 + \sum_{i \neq j=1}^{k} \sum^{k} \left(\frac{\partial R_m}{\partial R_i}\right) \left(\frac{\partial R_m}{\partial R_j}\right) (\varrho_x)_{ij} \sigma_i \sigma_j =$$

$$= \sum_{i=1}^{k} \sum_{j=1}^{k} \left(\frac{\partial R_m}{\partial R_i}\right) \left(\frac{\partial R_m}{\partial R_j}\right) \left(M_x\right)_{ij} \tag{30}$$

As seen from eqn. 27, the least-squares problem is nonlinear in the R-, $\varkappa$-, and u-parameters and must be solved by an iteration from sufficiently close starting values of the parameters. Various modifications may be used to improve the convergency of the iteration. A special version of the program includes Hartley's modification.[20] An increase of the diagonal elements of the **B** matrix may be useful in the beginning of an iteration. Sometimes a special refinement program [21] is applied before starting the least-squares iteration. Usually, it is sufficient to apply "fudge-factors". Two constants c_1 and c_2 are specified by the user, and the applied shift in cycle i, $\delta x_i'$, is given by

$$\delta x_i' = c_1 \delta x_i + c_2 \delta x_{i-1} \tag{28}$$

where δx_i and δx_{i-1} are the shifts for cycle i and cycle i$-$1, respectively.

DISCUSSION

The programs are linked together as illustrated in the figure. Most of the input to one program is data filed in a previous calculation, and the different data files are standardized. Programs may therefore be added and changed without influence on other parts of the system, and this makes the system very flexible to use. The programs are stored on the magnetic disk, and several programs may be used in sequence during one computer job, storing intermediate results either permanently on the magnetic disk, or on scratch files which are automatically destroyed when the job is finished.

The necessary theoretical functions for the data processing are computed by programs 1 and 2, and the programs 3$-$7 carry out the data processing. A specific problem may be solved in different ways, but generally a first background must be obtained, and the resulting intensity curve is usually

applied for least-squares refinements by program 11. The results of the least-squares refinement are then used to improve the background by comparing theoretical and experimental intensities, and the experimental intensities from the improved background are again applied for the least squares refinement. If necessary, this process is repeated.

In many cases, the least-squares refinement may be started at once. For more complex problems the experimental RD function is compared to theoretical functions for different molecular models, and the least-squares refinement is started from a reasonable model obtained in this way (programs 8—10). For special problems, the auto- and cross-correlation techniques may be helpful.[14,15]

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