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Research and Development Report

OPERATION OF AN X-RAY AND A
NEUTRON DIFFRACTOMETER
CONTROLLED BY A NON-DEDICATED
COMPUTER SYSTEM

by

MASTER

D. J. Dahm, J. E. Benson, D. M. Nimrod
D. R. Fitzwater, and R. A. Jacobson

October 1967

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IS-1701

OPERATION OF AN X-RAY AND A
NEUTRON DIFFRACTOMETER CONTROLLED BY A
NON-DEDICATED COMPUTER SYSTEM

D. J. Dahm, J. E. Benson, D. M. Nimrod,
D. R. Fitzwater, and R. A. Jacobson

ABSTRACT

The x-ray and neutron diffractometers under discussion are Hilger and Watts four-circle devices. Simultaneous control of these and other instruments is accomplished with a real-time computer system consisting of an SDS 910 computer linked to magnetic disks by an IBM 1401 computer. The operation of this system is discussed and compared with other automated systems currently in operation.

A complete set of programs for data collection including crystal alignment, generation of indices and angles, monitoring of standard reflections, and measurement of peaks and backgrounds have been written. Each program handles only a small portion of the task; additions or substitutions of programs are therefore easily implemented by the user to change the mode of operation. The same set of programs may also be recombined by writing one short calling program.

DESCRIPTION OF EQUIPMENT

The x-ray and neutron diffractometers currently operating under computer control at the Ames Laboratory Research Reactor (ALRR) are Hilger and Watts four-circle devices. Axis position is kept only by the computer. Each axis is equipped with a plastic disk concentric with its rotation. This disk contains a grating consisting of 3600 radial lines 0.1° apart. This grating passes before a lamp and a second grating to produce a Moiré fringe which is sensed by four photocells. The output from these photocells is used to interpolate axis position to 0.01° and to generate interrupts which allow the computer to keep track of axis position. The important thing is that this disk is located on the axis, not on a shaft connected to the axis by a gear, thereby eliminating errors due to backlash or gear wear.

Also contained on this disk is a silvered band which has a sharp discontinuity, the position of which is detected by another set of lamps and photocells. This discontinuity is the axis zero position and is the only external reference for absolute axis position. Consequently, the shafts must periodically be moved to the positions the computer believes to be zero to see if this is where the discontinuity occurs. This procedure is called a "datum check."

The computer system consists of an SDS-910 with 16K words of core storage. Auxillary direct access storage of five million words is provided by two magnetic disks. These disks are used for storage of source programs, object programs, and data. All communications with the computer are via teletype, either from the keyboard or punched paper tape.

Presently an x-ray and a neutron diffractometer are operating on-line. Two other neutron diffractometers and a continuous isotope separator are operating locally and are in the process of being put on-line. A bent crystal spectrometer and another neutron diffractometer will be put on-line in the near future.

AUTOMATIC COLLECTION OF DATA

We have just taken data on four compounds on our x-ray unit and one on our neutron unit. The remainder of this paper deals with our x-ray unit although much applies to both.

Our present data taking procedure uses balanced filters for scan data. Before data taking begins, the following information must be fed to the computer:

- (1) the Miller indices for three peaks and their approximate axis settings,
- (2) the wave length of the radiation being used,
- (3) the length of the scan in terms of a fraction of the $\sin \theta/\lambda$ of the reflection being measured,
- (4) the minimum and maximum θ of reflections to be measured,
- (5) the number of reflections between datum checks,
- (6) the number of reflections between position checks,
- (7) two intensity levels used in the position check,
- (8) the Miller indices for three standard reflections which will be scanned periodically if decomposition is being allowed for, and how often to scan them.

The rest is all automatic. If for some reason the computer is shut down during the data taking process, the next time the program is executed, it will take up where it left off.

As stated above, only approximate peak positions are necessary for input which means that the computer must find the exact peak positions. We call this process "tuning". The process of tuning a general reflection on a single crystal orienter so as to both maximize and center the diffracted beam is surprisingly difficult to rigorously define. One can easily get into an iterative process that can be carried on indefinitely without ever converging on the solution. With a computer intimately available during the tuning process it has been possible to define an algorithm which will converge for any reflection. By making some mathematical calculations during the tuning, the mechanical movements can be simplified.

When all axes are at their zero positions, the theta, omega, and phi axes are vertical, and the chi axis is horizontal and parallel with the undiffracted x-ray beam. The direction of positive rotation is counterclockwise, looking down in the case of theta, omega, and phi, and away from the x-ray source for chi, when all other axes are at their zero positions. The rotations expressed in degrees about the omega, chi, and phi axes are represented by ω , χ , and ϕ , respectively. The numerical value represented by θ is $\frac{1}{2}$ of the rotation about the theta axis expressed in degrees. There is complete mechanical independence of axis movement. The phi axis itself, however, is rotated

in a plane perpendicular to the chi axis by a chi-axis rotation; both the phi and chi axes are rotated by an omega-axis rotation. Physically, the crystal is mounted on the phi axis at the intersection of the four axes.

When a crystal is in position to diffract, the "reflecting" plane of the reflection is vertical and is therefore coincident with the instrument axis, omega. If the value of ω is near the Bragg angle, θ , the chi axis is also nearly parallel to the reflecting plane, and it is perpendicular to the omega axis. Hence, any reflecting plane may be positioned with chi and omega adjustments to give a maximum intensity in the horizontal plane within which the detector swings. This means that a general peak can be tuned by an iterative process of the following three steps:

- (1) Maximize the total intensity with respect to omega adjustments.
- (2) Center the detector in the diffracted beam with respect to left-right beam splitters on the detector face with adjustments in theta.
- (3) Center the diffracted beam with respect to top-bottom beam splitters on the detector face with adjustments in chi.

We have found that two or three iterations of these steps are sufficient for convergence.

Phi is left out of the tuning process because it in general is not parallel or perpendicular to the reflecting plane, and only two axes are necessary to orient the reciprocal lattice vector. The advantage of

using chi and omega is that, because they are perpendicular one may be tuned independently of the other with a minimum of interaction.

The chi-omega tuning process will in general leave ω at some value other than θ . However, at that point the computer may be used to calculate the chi and phi adjustments necessary to compensate for a rotation of omega to the Bragg angle. Essentially what is being done is a rotation of the reflecting plane about its normal.

The expressions derived¹ for the compensating chi and phi corrections for a given $\theta - \omega$ ($\Delta\omega$) are as follows:

$$\tan \Delta\phi = \frac{-\tan \Delta\omega}{\cos \chi}$$

$$\sin (\chi + \Delta\chi) = \sin \chi \cos \Delta\omega$$

$$\text{where } \Delta\omega = \theta - \omega$$

$$\Delta\chi = \chi_o - \chi_t$$

$$\chi_o \text{ is the } \theta = \omega \text{ solution.}$$

$$\chi_t \text{ is the tuned value}$$

$$\Delta\phi = \phi_o - \phi_t$$

It may be seen that the $\Delta\phi$ correction ranges from $-\Delta\omega$ at $\chi = 0^\circ$ to 90° at $\chi = 90^\circ$. The range of $\Delta\chi$ is 0° at $\chi = 0^\circ$ to $\Delta\omega$ at $\chi = 90^\circ$. The negative supplement of χ ($\chi - 180^\circ$) must be used in the $\Delta\chi$ expression for $|\chi| > 90^\circ$.

The general tuning algorithm is therefore followed by rotating omega so that $\omega = \theta$ and then calculating the required compensating $\Delta\phi$ and $\Delta\chi$ using the given expressions. The implementation of this algorithm on our system has experimentally verified the method and the expressions. We have the user feed to the data taking program the approximate angles for three independent reflections and let the system tune them and use the tuned settings for calculating the orientation matrix.

Let us now follow job flow after the program has just completed taking a reflection and control returns to the sequencing program. (Fig. 1). It should be kept in mind that each step is several programs, that there are additional processes and that many of the operations which are discussed are taking place at once, the language being flexible enough so that if properly programmed one is always waiting for the instrument and never the computer.

If the number of reflections recorded since the last datum check is equal to the input parameter, a datum check will be performed; otherwise, the sequencing program goes on to the next step. If the datum check passes, the Miller indices of the next reflection to be taken are recorded at a place on the magnetic disk reserved for what we call the restart reflection. (It is this reflection in this location that will be taken first if the program needs to be restarted at any time.) If the datum check fails, the sharp discontinuities on the axis are systematically searched for and when found, the core locations reserved for axis positions are reinitialized. Because some of the reflections taken since the

Fig. 1. Schematic of data collection.

last datum check may have been recorded with the axis in incorrect positions, the next reflection taken will be the one whose Miller indices were placed in the restart location at the last successful datum check.

Next, the sequencing program determines if a position check is warranted on the basis of the input parameter. A position check consists of recording the intensities of the three reflections which were used to calculate the orientation matrix by peak height measurements. Peak heights are used because they are much more sensitive to crystal position than scanned intensities. If the intensities are above a certain input level, it is assumed that the crystal has not changed its position and control returns to the sequencing program. If the intensity is below this level, there is a datum check to see if an error in shaft position could explain the intensity drop. If the datum check fails, a new position check is performed. If the datum check passes, the intensities are assumed to indicate an idea of the amount the crystal has shifted. If the intensities are below a second input level, it is assumed that the crystal's position has changed so much that manual intervention is required and the program terminates. If the intensities are between the two levels, the three reflections are tuned, and the orientation matrix is recalculated.

At this point the standards which are used to correct for crystal decomposition may be scanned. In many cases the user may not be allowing for decomposition and this step will never be executed.

After this it is time to return to routine data collection. The Miller indices of the next reflection are determined, the axis settings are calculated, and the intensity is recorded.

It should be obvious that there are operations in the above description which could not be included in normal operation with a small, dedicated computer. Also our system gives flexibilities in such ways as ability to change input parameters at any time during operation. Moreover, easily made changes of a few locations of the disk will alter the mode of operation such as changing method of output. In addition, if a method of data taking is desired which has not been provided for, a few changes or additions to the programs are easily made. If desired, a program being used may be changed and recompiled during the data taking process.

We believe this system leaves enough room for improvements in data taking techniques to satisfy the most ingenious experimenter.

REFERENCE

1. D. M. Nimrod, Ph.D. thesis, Iowa State University, Ames, Iowa (1967).