THE DESIGN OF A PROGRAMMED X-RAY DIFFRACTOMETER INSTALLATION

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THE installation consists of up to four four-circle X-ray diffractometers together with their measuring circuits, which are controlled on a time-sharing basis by a single punched-tape programmed control circuit. An order code allows the employment of a large number of different measuring sub-routines with moving or stationary crystals and detectors. The scanning range and speed, the insertion of filters, etc., can all be specified on the input tape.

Compared with moving-film cameras, which they are gradually replacing in X-ray diffraction studies on single crystals, automatic counter diffractometers are expensive instruments. In spite of the high initial cost, however, diffractometer methods are generally found to be economically preferable to photographic techniques when the cost of the additional equipment and of the labour required in the densitometry of X-ray photographs and in processing the results of the measurements is taken into account; in addition greater accuracy is possible with counter techniques.

The economic balance shifts even more in favour of a diffractometer which is in full and constant use: the instrument should thus be as versatile as possible to make it suitable for a variety of different measuring techniques and, moreover, for a number of crystallographic problems other than those of straightforward collection of data for three-dimensional structure factor determination.

The special facilities which may be required in different investigations have been examined in the companion paper.[1] It was shown there that amongst the many different geometrical arrangements possible for single-crystal X-ray and neutron diffractometers[2, 3] the most generally flexible is that of the four-circle diffractometer, illustrated in Figs. 1 and 2. For the automatic operation of such an instrument its crystal and detector shafts are set to pre-computed positions according to instructions supplied on an input medium such as punched paper tape; the versatility of the installation can be greatly extended by the provision of an order or command code which allows shaft-setting, measuring and other instructions to be combined in different ways in the input programme. It is the purpose of the present article to describe in greater detail the features of such a four-circle diffractometer installation which is about to go into production. This installation is the X-ray version of a basic instrument designed for either X-ray or neutron diffractometry. Some of the mechanical parts and most of the control circuitry are common for both applications, but there are important differences, especially in the way in which the equipment is used. The neutron version is described fully elsewhere.[4]

The Diffractometer

A photograph of a model of the diffractometer is shown in Fig. 2. The crystal, normally mounted on a standard goniometer head, can be rotated about the ϕ-, χ- and ω-axes, while the detector arm turns about the 2θ-axis.

The Four Circles Each of the four shafts or circles can be independently positioned by a moiré fringe method.[5] To this end, each shaft carries a radial transmission grating which generates fringes as it passes over a fixed grating incorporated in the pick-up head which also contains a lamp and photocell assembly. Pulses produced by the movements of the fringes are counted on solid-state counting circuits. The positioning accuracy is 0·01°. In addition, a reference mark is provided on each circular grating which is detected by a separate pick-up head, thus making possible the location of a datum point to a reproducibility which is also made 0·01°. Small adjustments of the χ, ω and 2θ reference pick-up heads allow the datum points to be made to correspond to exactly zero degrees; the ϕ datum point depends on the direction of the axes of the crystal mounted on the instrument: the ϕ grating can, therefore, be rotated with respect to the goniometer head.

Although the accuracy of the moiré fringe positioning method is independent of the gearing and the backlash in the drive, precision gears are used throughout and backlash is reduced by spring-loading the final gears; the absolute positions of the circles can thus be read to an accuracy of 0·01° by means of scales or revolution counters and micrometer heads.

Each shaft is provided with a D.C. motor with an integral tachogenerator: the nominal setting speed is 20°/sec for the 2θ shaft and 10°/sec for each of the others.

Rotation of the crystal through 360° is possible about the ϕ, χ and ω shafts, although the last of these may be limited in certain positions of the X-ray tube. The maximum 2θ angle of the detector arm is 155° on one side of the incident beam and 115° on the other. Limit switches are provided on all shafts: the rotations of the χ and ω circles are limited to just under and just over 360°, respectively, to prevent winding up of the connecting leads to the pick-up heads.

The final crystal shaft has an axial fine adjustment with a range of $\frac{3}{4}$ inch.

The X-ray Tube Bracket The front surface of the main casting carries a bracket supporting a horizontal plate on which the shockproof shield for the X-ray tube is mounted. This shield will accept any standard Philips X-ray-tube insert; it is provided with fine adjustments for small axial and vertical translations of the tube, for a small rotation about its horizontal axis to bring the line focus into the horizontal plane, and for a rotation about a vertical axis through the target face to allow a variation in take-off angle between $\frac{1}{2}$° and 5°. In this way the optimum beam from any X-ray tube insert can readily be made to pass through the collimator which is pre-set to point at the geometrical centre of the instrument. The tube housing with its adjustments is fitted on a plate in one of two ways: when it is in the fully backwards position the vertical circle assembly is capable of an unobstructed 360° rotation about ω; in the fully forward position, where there is a consequent gain in intensity, this rotation is limited to $\pm 45°$ from the zero setting ($\omega = 0$ when the plane of the χ-circle is normal to the incident beam). Alternative mounting plates will allow the X-ray tube to be fitted so as to permit the use of crystal monochromators.

Bolted direct to the tube shield is a solenoid-controlled assembly which contains a fail-safe shutter and two attenuating filters either or both of which can be inserted in the incident beam.

The entire X-ray-tube housing can, of course, be removed, allowing the diffractometer to be placed in front of a fixed rotating anode or other X-ray generator, provided that the latter has a minimum window height of 20 inches above the table top.

A variety of collimators, including total-internal-reflection lead-glass tubes, can be fitted. A beam stop is mounted on a clip fitting on the collimator: it can, of course, only be used when full 360° rotation about ω is not required.

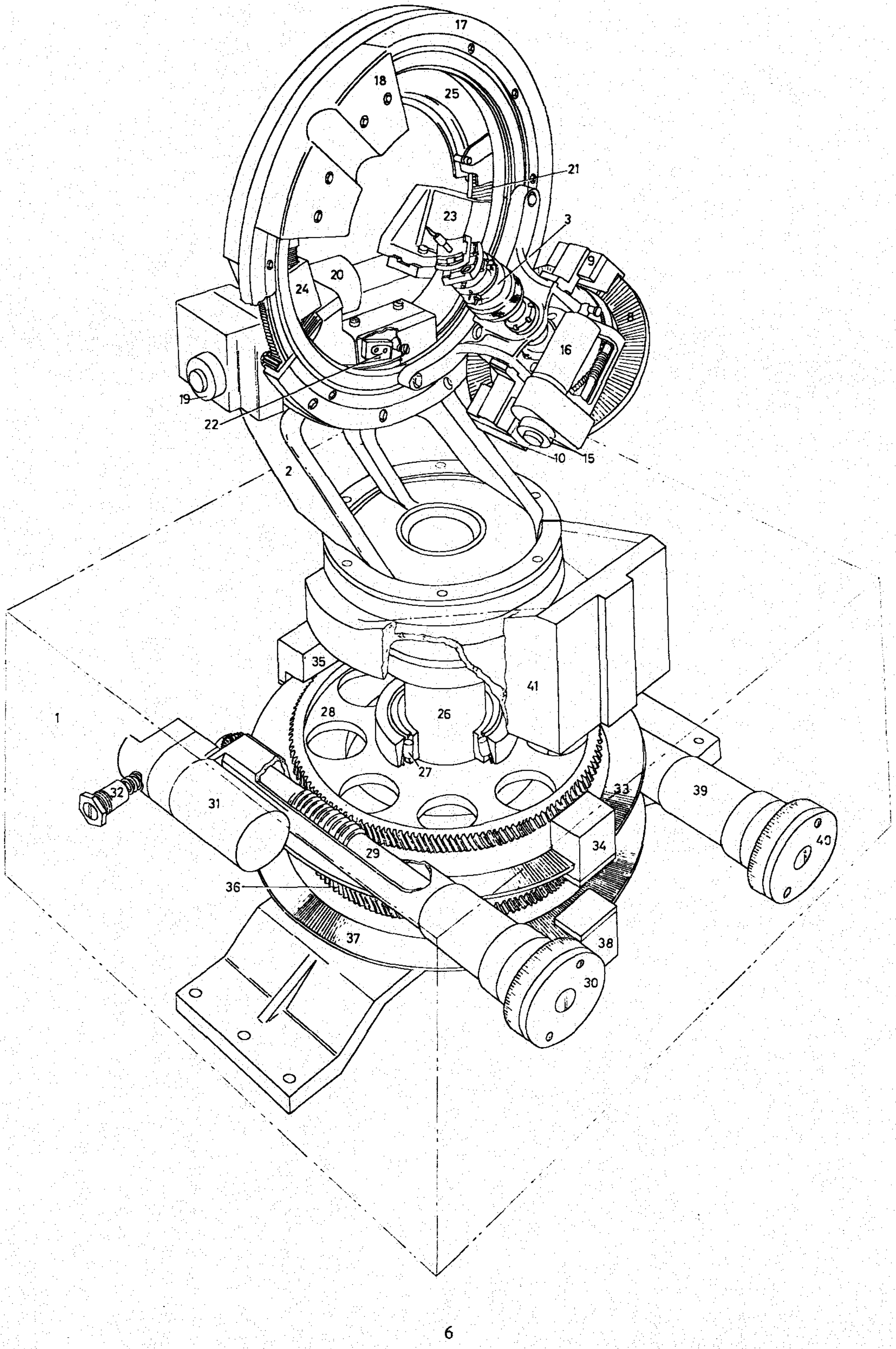
The Detector Arm The detector arm has provisions for a side-window xenon-filled proportional counter for the softer radiations or for an end-window scintillation counter for the detection of harder X-rays. Two further solenoid-operated filter holders, intended to be fitted with balanced filters, are sited in front of the detector. A diffracted-beam collimator guards against air-scattered radiation. The counter arm also carries two pairs of shutters by means of which the top or bottom or the right or left halves of the counter collimator can be obscured. This device, due to Furnas,[6] is employed during the alignment of the crystal.

As can be seen from Fig. 2 the rigidity of the main bearing incorporating tapered roller races is such that a much more massive detector arm than the standard one can be substituted: the basic instrument can thus be modified for high-resolution low-angle studies with a long detector arm or for neutron diffraction studies in which the detector must be surrounded by massive shielding.

The leads to the ϕ/χ assembly motors and pick-up heads are taken to a socket panel in the base of the instrument into which all leads from the instrument are plugged.

The Order Code

An incremental shaft-setting method has been adopted in the present installation so that a signed increment is specified for each shaft in turn and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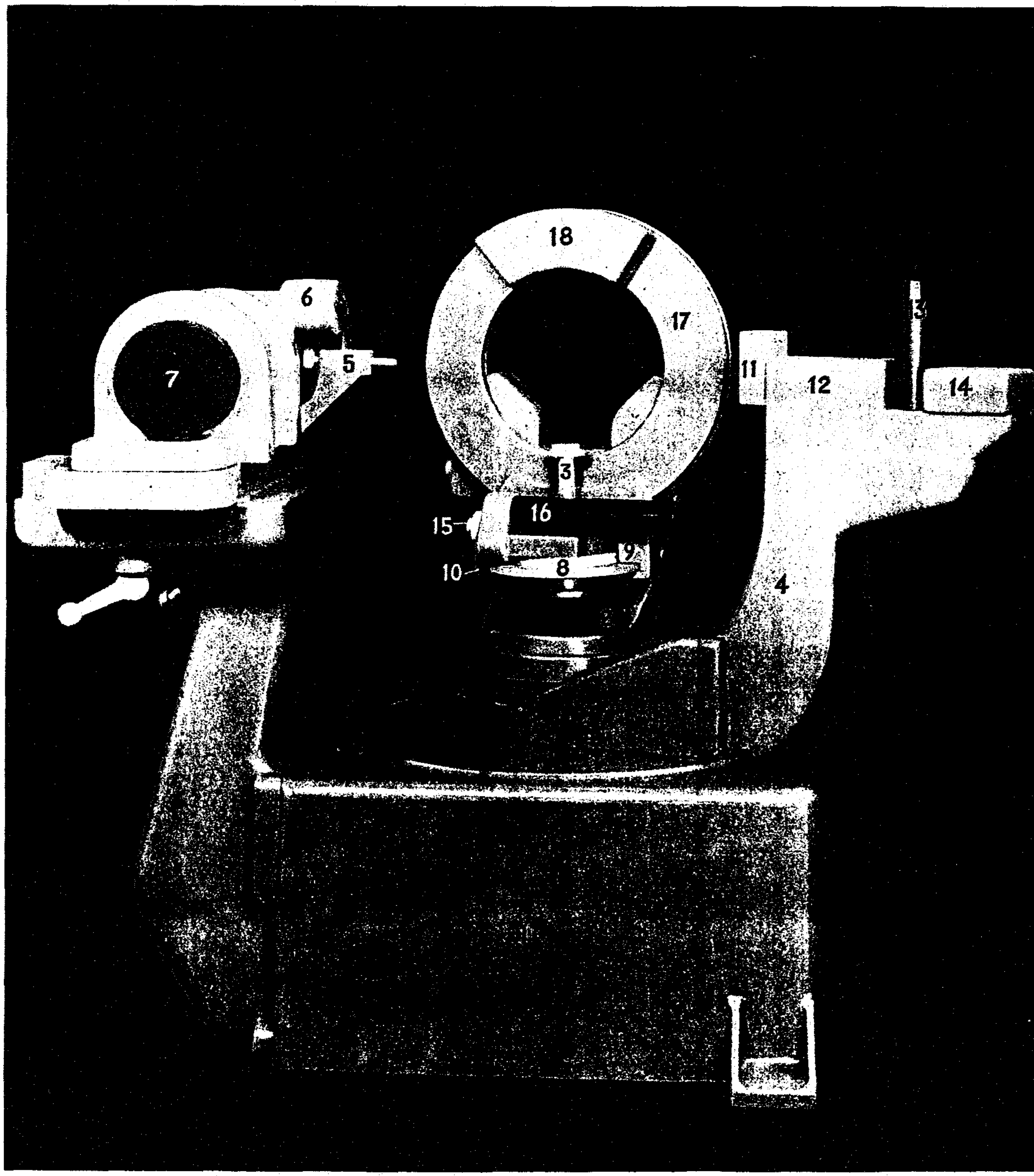

Fig. 1 (opposite) and Fig. 2 (above)—Diagram and wooden model of the diffractometer
1. Diffractometer Base. 2. ϕ/χ Circle Assembly. 3. Goniometer Head Fitting. 4. Detector Arm. 5. Collimator Bracket. 6. Filter and Shutter Assembly. 7. X-ray Tube Shield. 8. ϕ Grating. 9. ϕ Positioning Pick-up Head. 10. ϕ Reference Pick-up Head. 11. Top-Bottom and left/right Mask Assembly. 12. Balanced Filter Assembly. 13. Proportional Counter. 14. Pre-amplifier. 15. ϕ Micrometer Dial. 16. ϕ Motor and Tachogenerator. 17. χ Circle. 18. Counterweight. 19. χ Micrometer Dial. 20. χ Motor and Tachogenerator. 21. χ Grating. 22. χ Limit Switch. 23. χ Main Pick-up Head. 24. χ Reference Pick-up Head. 25. Protective Cover for χ Grating. 26. Hollow 2θ Shaft. 27. Tapered Roller Race. 28. 2θ Worm Wheel. 29. 2θ Worm. 30. 2θ Micrometer Head. 31. 2θ Motor and Tachogenerator. 32. Spring Loading for 2θ Worm. 33. 2θ Grating. 34. 2θ Main Pick-up Head. 35. 2θ Reference Pick-up Head. 36. ω Worm Wheel. 37. ω Grating. 38. ω Main Pick-up Head. 39. ω Worm Housing. 40. ω Micrometer Dial. 41. Bracket for Detector Arm

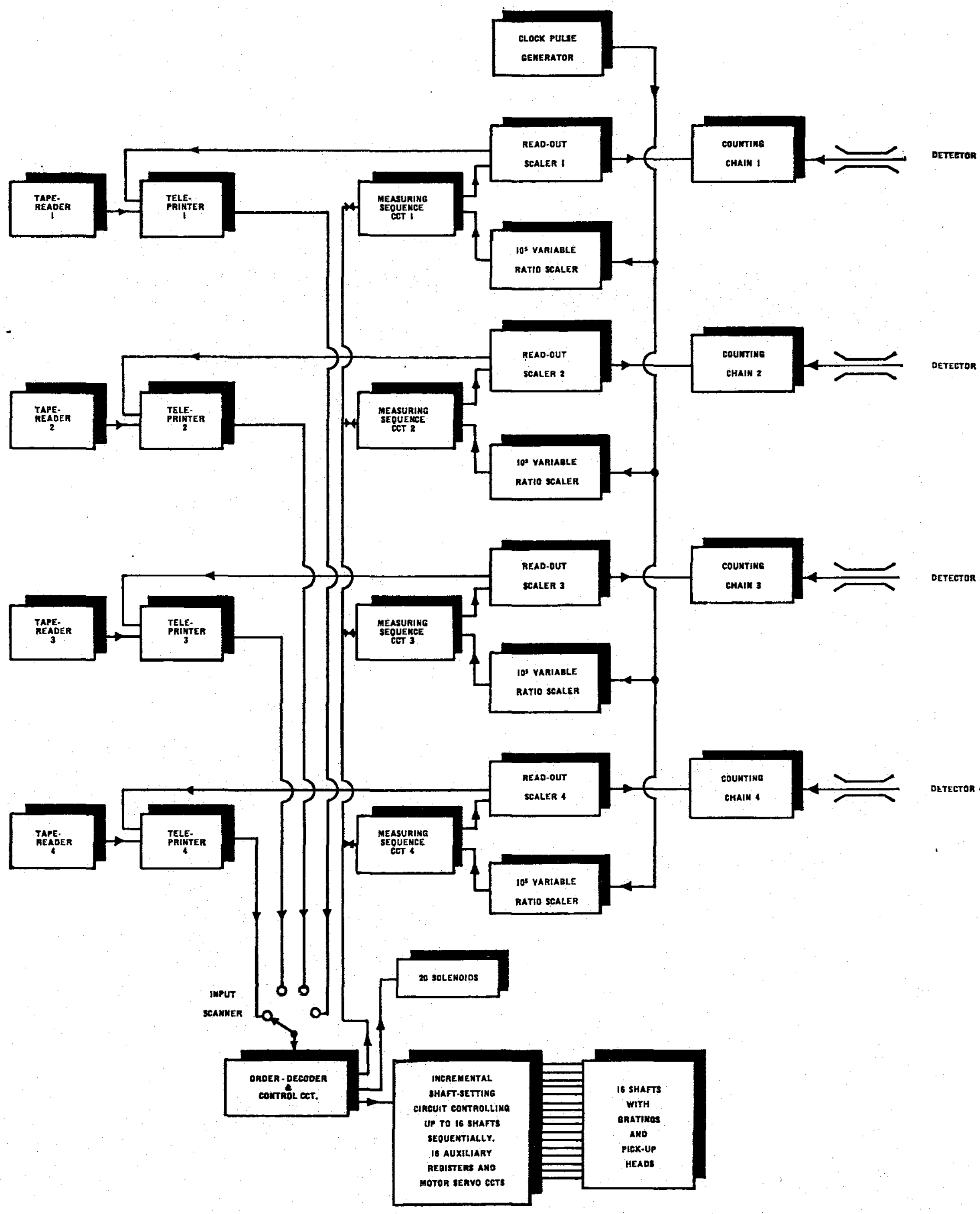

Fig. 3—Block schematic circuit diagram for a four-diffractometer installation

the arrival at the correct position is determined by a count-down to zero. In view of the high setting speeds obtainable with the moiré fringe method, little time is lost by setting the four diffractometer shafts in sequence so that only one incremental register is required.

Sequential setting requires some form of clamping of each circle after setting. This is achieved by providing auxiliary registers with a capacity of $\pm 0{\cdot}05^{\circ}$ which remain coupled to their setting motor servo-circuits during the disengagement of these circuits from the main incremental register. A small departure of a circle from its desired position through vibration or gravity is thus corrected by its own live servo-circuit.

Another consequence of the adoption of an incremental system of setting is the need for an absolute datum point for each circle. After a number of settings all shafts are returned to this datum point for checking purposes and to prevent the accumulation of small errors. Such a return to datum is accomplished by a special order (see x05, Table 1); arrival there is detected by means of a special mark on the grating and an additional pick-up head.

With this setting procedure it becomes logical to supply the setting instruction for a particular shaft in the form of an order having the following constituents:

1. An initiating symbol, indicating that the code groups immediately following must be decoded.
2. A code group, in practice consisting of two decimal digits, indicating that the instruction refers to the setting of a shaft.
3. Address of the shaft to be set.
4. Signed increment.
5. Terminating symbol.

The form of this instruction was extended into an order code to cover all the possible operations carried out during an experiment, including the insertion of balanced filters and attenuating foils and the instructions referring to the making of a measurement. This order code, together with a few special characters, is listed in Table 1.

One feature of the control of the installation by means of this order code is that the sequence of operations is flexible. The execution of one instruction leads inevitably to the reading-in of the next order which alone can initiate the next operation. This feature is essential if the installation is to be capable of being connected directly to a digital computer for on-line control (see page 12).

The other feature is that the installation lends itself to the control of a number of diffractometers. It will be noticed that the operations specified in Table 1 are of two kinds: those of the first kind are either instantaneous, such as the insertion or removal of a filter, or very rapid, such as the setting of a shaft; the other operations involving the making of measurements are necessarily slow to allow the accumulation of a statistically significant number of counts. While a measurement is being made, the circuits concerned with reading-in, decoding, and shaft-setting can thus be used to set a second diffractometer. In fact, the relative time scale of the setting and measuring operations is such that at least three other diffractometers can be set and made to initiate their own measuring operations before the measurement carried out by the first instrument is completed. In order to make such interleaved operation possible up to four input channels can be provided, each having allocated to it the four shafts and the five filter-operating solenoids of a different diffractometer. (By means of an internal patchboard, the allocation can be changed, thus making possible, for example, the control of a larger number of single-axis powder diffractometers.)

Fast and slow operation orders are dealt with in two different ways so as to permit maximum speed in a multi-channel installation. After a fast-operation order has been read-in on one channel no action is possible on any other channel until the execution of the order has been signalled back to the control circuit. On the other hand, as soon as a measurement has been initiated by an appropriate order on a given input channel, a scanning circuit examines the other input channels in turn to find one on which an ' interrupt relay ' has been set, signifying that this channel is free to read-in a new order.

Any channel can be disengaged by a special x06 order, by a fault in the channel, or by reaching the end of its input tape. After disengagement of an input channel it is omitted from the scan determining the provenance of the next order until re-engaged by the intervention of the operator.

All orders are terminated either by a ' comma ' or a ' full-stop '. After an order terminating in a ' comma ' the next order is always taken from the

TABLE I
THE ORDER CODE

Order	*Function*	*Address*	*Contents*
x01	Set shaft position	Any one of 16 shafts	±5 decimal-digit-number of 1/100° units
x02	Set ω-shaft and 2θ shaft simultaneously at twice the speed of ω	Any one of 4 ω-shafts	±5 decimal-digit-number of 1/100° units
x03	Set filter solenoid	Any one of 20 solenoids	—
x04	Reset filter solenoid	Any one of 20 solenoids	—
x05	Set shaft by specified amount to bring it back to datum; print error (±); set fault register if error exceeds ± 4/100°; disengage channel after two faults unless fault count override key is *on*	Any one of 16 shafts	5 decimal-digit-number of 1/100° units
x06	Disengage	Any one of 4 channels	—
x07	Return to starting point of scan specified in preceding x11, x12, x21 or x22 order	Any one of 16 shafts	—
x11	Carry out measuring scan with one shaft moving in steps, each step timed by output of variable-ratio scaler; at end of scan print-out and reset measuring scaler	Any one of 16 shafts	± or *; 2 digits (plus necessary number of significant zeros) specifying monitor counter ratio; 3 digits specifying number of steps; 1 digit p specifying the step size $\frac{2p}{100°}$, where p = 0 to 6. The direction of the scan is specified by the sign ±; if the sign digit is * (in 5-hole code) measurement is made without movement
x12	As for x11, but with 2θ moving through steps of twice the size of ω-steps	Any one of 4 ω-shafts	As for x11
x21	As for x11, but with print-out and scaler reset after each step	As for x11	As for x11
x22	As for x21, but with 2θ moving through steps of twice the size of ω-steps	Any one of 4 ω-shafts	As for x11

Letter shift: After this character is read all succeeding characters are ignored until the next ' figure-shift ' is received. If ' letter-shift ' occurs in the middle of an order the preceding codes are erased. (This is used in correcting orders set up on the keyboard.)

> character: If a limit-switch on any circle is operated during the programme, the tape is searched until the next > character is found. This character normally precedes x05, the order for returning to datum, and appears with x05 at regular intervals in the programme tape.

→ character: This provides a conditional disengage signal when the stop/run key is set at stop. It is used in programme testing.

TABLE II

MEASUREMENT OF A REFLEXION USING BALANCED FILTERS

x01 01 ± ------.	Set ϕ-shaft ↓
x01 02 ± ------.	Set χ-shaft ↓
x02 03 ± ------.	Set $\omega + 2\theta$-shafts ↓
x03 01 .	Insert β-filter ↓
x11 03 * ------.	Measure background on one side of Bragg peak (b_1) ↓
x12 03 + ------.	Measure intensity across the peak using $\omega/2\theta$ scan ↓
x11 03 * ------.	Measure background on other side of Bragg peak (b_2) ↓
x04 01 .	Remove β-filter ↓
x03 02 .	Insert balanced α-filter ↓
x11 03 * ------.	Measure background (b_2) ↓
x12 03 ------.	Measure intensity across peak using scan in reverse direction ↓
x11 03 * ------.	Measure background (b_1) ↓
x04 02 .	Remove α-filter ↓
	Proceed to next reflexion

TABLE III

SEQUENCE IN MULTI-CHANNEL OPERATION

Channel 1	2	3	4
x01 01	→x01 01	→x01 01	→x01 01
x01 02	x01 02	x01 02	x01 02
x02 03	x02 03	x02 03	x02 03
x03 01	x03 01	x03 01	x03 01
x11 03	x11 03	x11 03	x11 03
x12 03 →	x12 03 →	x12 03 →	x12 03
x11 03 →	x11 03 →	x11 03 →	x11 03
x04 01	→x04 01	→x04 01	→x04 01
x03 02	x03 02	x03 02	x03 02
x11 03	x11 03	x11 03	x11 03
x12 03 →	x12 03 →	x12 03 →	x12 03
x11 03 →	x11 03 →	x11 03 →	x11 03
x04 02			
x01 02			
etc.			

TABLE IV

VARIATION OF INTENSITY WITH AZIMUTH

x01 01	Set ϕ-component increment ↓
x01 02	Set χ-component increment ↓
x01 03	Set ω-component increment ↓
x11 03*	Measure intensity ↓
x01 01	Set ϕ-component increment
	etc.

same input channel, but after an order terminated in ' full-stop ' the other input channels are scanned in turn for an interrupt. In the absence of any special priority requirements fast duration orders are terminated by a comma and measurement orders by a full-stop.

Two Programming Examples

In the first illustration we shall assume that all four channels of the installation are occupied by identical four-circle diffractometers and that these are all making measurements according to the flow-sheet of Table 2. The probable order of execution of the orders will be as in Table 3.

In the second example it is assumed that there is only one diffractometer which is being used to determine the absorption correction for a particular reflexion by measuring the variation of diffracted intensity with rotation about the azimuthal angle. The increments specified in the section of the programme illustrated in Table 4 are the desired increments of the azimuthal angle resolved into its ϕ, χ and ω components (by means of a computer).

Circuits

The overall arrangement of the circuits of a four-channel installation is shown in Fig. 3. The main components of this block schematic diagram are briefly described in this section.

Input Circuit The standard input circuit for each channel consists of a tape-reader reading five-hole paper tape in any chosen code. Its output is read into a Creed Model 75 teleprinter arranged for five-wire parallel input and fitted with a reperforating attachment, keyboard and transmitting and synchronizing contacts. The output from the transmitting contacts is fed into a solid-state decoding circuit. All input information is thus monitored by the teleprinter; during initial setting up, and for testing purposes, its keyboard is available as an alternative input device.

The clock pulses for the solid-state shift register, which acts as a routing circuit and distributes the decoded signals to appropriate staticizers, are taken from the synchronizing contacts of the teleprinter. These circuits are capable of functioning at a much faster clock rate than the synchronous speed of 11 characters per second of the teleprinter, so that faster tape-readers can be substituted at will.

The Measuring Scaler The diffracted X-rays in each channel are detected by means of a scintillation or proportional counter whose output pulses are first amplified and then passed through a single-channel pulse analyser; from here they pass into a solid-state six-decade ring scaler. The count accumulated by the scaler is recorded, the most significant figure first, by the teleprinter appropriate to that channel; here, too, a faster output device may be substituted. The scaler is gated by signals from the main control unit; closing of the counting gate initiates a print-out followed by re-setting of the scaler.

Clock Pulse Generator Clock pulses for the timing of individual counts and of steps during the scan across the reflexion are derived from a 20 kc/s crystal oscillator via a scaler with a scaling ratio of up to 10^5. The scaling ratio is under programme control, its first two decimal digits, plus the number of significant zeros, being specified in the measuring order.

When an unstabilized source of X-rays is used a monitoring counter chain can be substituted for the crystal oscillator.

In certain problems, it is desirable to time a pre-set number of counts instead of counting for a pre-set time. For such cases the variable-ratio scaler is connected in the measuring chain and the read-out scaler in the timing chain.

The Positioning Circuits By means of appropriate circuitry, add and subtract pulses are derived from the photoelectric pick-up heads fitted to each circle, one such pulse corresponding to the movement of one-tenth of a moiré fringe, or, since the radial gratings have 3600 lines, to a clockwise or anti-clockwise rotation of the circle of 0·01 °. These pulses are fed into a five-decade reversible solid-state ring counter which is pre-set, decade by decade, from the input tape via the decoder. While the shafts are rotating, a separate decoder determines the distance still to go and this information controls the speed of the driving motor. Some twenty to forty counts from the end of travel the speed is reduced to a creep.

As has been mentioned above, the main register is shared between the various axes, but a low-capacity auxiliary register is provided for each shaft to effect quasi-clamping when the shaft is set.

Manual Control A manual control panel is provided near the diffractometer to enable the shafts to be moved non-automatically while a crystal is being aligned on the diffractometer. The initial setting and centring of the crystal is, of course, done optically and to this end a long-focus microscope is mounted on the X-ray tube bracket.

Mechanical Arrangement of Circuits The individual circuits are on 8 inch × 5 inch printed boards. The number of board types has been kept to a minimum to afford the greatest degree of interchangeability and ease of servicing. The circuits are almost 100 per cent solid-state and the use of electro-mechanical devices, such as relays and stepping switches, has been avoided wherever possible.

The boards are mounted in a console on the top surface of which the diffractometer stands.

On-line control by a Computer

The efficient use of a diffractometer installation such as that described necessarily requires some form of closed-loop system involving an external digital computer with punched paper tape as an intermediate step. A programme tape is first computed using unit cell dimensions which may themselves have been determined on the diffractometer. The sequence of measurements is then carried out under the control of this programme tape and the diffractometer records its output on a second output tape, which in turn is analysed by the computer. A new programme tape may be prepared as a by-product of this analysis which may, for example, specify re-measurements of faulty data or of weak reflexions with longer counting times.

Clearly, the complete process of producing a complete error-free set of results to a given minimum statistical accuracy can be carried out more rapidly if the output of the diffractometer measuring circuits is directly coupled to a digital computer which can make any necessary decisions as to remeasurement under different conditions or even as to re-aligning the crystal when this has become mis-set. Schemes involving the use of small general-purpose computers have been proposed by Pauling,[7] and by Katz.[8] Even with small computers, however, the actual computing time would be very short indeed compared with the total period during which the diffractometer is in operation; it thus seems desirable that the computer used for the control of the diffractometer be a time-sharing computer, only one of whose tasks would be the control of one particular instrument. A considerable amount of storage capacity would, however, be needed for storing the sub-routines for dealing with the various operational contingencies, thus limiting the usefulness of the time-shared computer for other problems on which it might be simultaneously engaged. It follows that only a large computer, which possesses the necessary organization of its different access-time stores becomes economical for such on-line control. These considerations have led to the design of a pilot installation at Manchester University which will use the MUSE computer, the prototype of the Ferranti ATLAS computer.[9] The diffractometer used in this work is identical with that described here. The Manchester installation promises to be both versatile and economical granted continuous access to a large time-sharing computer.

The present assembly is well-suited to optional on-line control, by almost any type of digital computer. Conversion from tape to computer control can be made very easily since only three signal lines are needed for communication between the computer and the diffractometer circuits:

1. Five-wire line from the computer output to the diffractometer input circuits, used for conveying orders to the diffractometer.
2. Five-wire line from the diffractometer output circuits to the input of the computer, used for transmitting numerical results to the computer.
3. Single-line lead, used for signalling the execution of an order and the consequent readiness to accept further instructions. This signal is used to set an interrupt in the controlling computer.

Conclusion

The installation which has been described here was planned as a coherent whole, but the control and measuring circuits and the form of the order code are such as to make it perfectly possible to substitute mechanically quite different instruments for the present diffractometer in one or all channels of the installation. It is intended to design compatible special or general purpose diffractometers as the need for them arises during the continuing development of the art of diffractometry.

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SPECTRUM COMPARATOR (*continued from page 3*)

was mounted horizontally at the bisector plane of the inner rear surface of the instrument at a distance of about 6 cm. from the stage. The lateral supports of the lamp were screwed to two right-angle arms fixed to the sides of the instrument by milled-head screws. The form of these arms provides the possibility of adjusting the lamp at different positions. The lamp is operated by the original switch and the 8W ballast is fastened by screws to the back of the instrument.

3. The inner surface of the instrument was covered with a thin polished aluminium sheet acting as a reflecting mirror. This sheet is kept in place by screws in suitable positions.

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