# X-RAY EQUIPMENT FOR CRYSTALLOGRAPHY

ON November 25, 1944, a meeting of the X-Ray Analysis Group of the Institute of Physics was held in the Physics Department, University of Leeds. The meeting was opened by the chairman of the Group, Sir Lawrence Bragg. Sir Lawrence said that the necessary equipment and assistance had been found for scientific men to carry out emergency programmes of war-time research, and this must be extended in post-war planning. As the progress of crystallography is conditioned by the provision of equipment, it had been decided by the X-Ray Analysis Group to bring manufacturers and users together to discuss design and development, so that in the future suitable apparatus might readily be available.

The first two papers were on the development of the demountable X-ray tubes of Messrs. Metropolitan-Vickers Electrical Co., Ltd., and were presented by Dr. R. Witty and Mr. P. Leech. In the first paper, Dr. Witty spoke about the firm's standard demountable crystallographic X-ray tube, and emphasized the two points on which differences of opinion have been found among the users of the tubes, namely, the automatic gear of the instrument and the stability of the electron emission of the filament. He agreed that early types of automatic gear caused some trouble and loss of time, but considerable improvements have been made during recent years. Filament current will be stabilized either by galvanometer or electronic relays. Mr. P. Leech then spoke about the rotating anode tube with a seal of the Muller-Beck type which is being designed for post-war manufacture. A shielded filament tube is being contemplated to overcome the deposition of tungsten on the target. Ancillary spectroscopic equipment is being designed including low-angle, Weissenberg and high-temperature cameras.

Mr. R. A. Stephen, of Messrs. Philips Lamps, Ltd., drew up a series of specifications for a sealed-off X-ray tube. He said that multi-anode tubes cannot be manufactured at present because of mutual contamination of surfaces. Single-target tubes of copper, cobalt, iron, chromium, molybdenum and tungsten are made; but zinc and manganese targets cannot be used because of the high vapour pressure of these metals. The characteristic radiation output is given by the equation $P = K(V - V_c)^{1\cdot65}$, where $V$ is the applied and $V_c$ the critical radiation voltage. This holds for kilovoltages up to four to five times the excitation voltage of the characteristic radiation. In a good vacuum the insulation is estimated as about 10–20 kV. per mm. A word of warning was given about the use of unknown transformers with self-rectifying tubes, as this may put high electrical strain on the instrument due to the suppressed wave. X-ray windows are usually of 0·12 mm. Lindemann glass, which is capable of standing the required mechanical strain; but it must be carefully shielded from electron bombardment. Beryllium windows are in use, and a beryllium alloy (containing up to ½ per cent titanium) is being investigated as a window material. These metals have the advantage of not requiring to be screened. Figures of absorption of these different materials were compared. Traces of water vapour in the sealed-off tube cause the formation of tungsten oxide on the filament, and evaporation of the oxide and subsequent reformation of water vapour give a reversible reaction which produces deposits of tungsten on the target 10,000 times greater than would be expected. It is therefore essential to use a 'getter' during the life of the tube. The design of a shielded filament was discussed. The minimum safe diameter of a crystallographic X-ray tube with a Lindemann glass window was given as 60 mm. The maximum tolerance loading per unit area of a target is given by the equation

$$T_M = \frac{2W}{\pi k}\{a \log (b/a + \sqrt{1 + b^2/a^2}) + b \log (a/b + \sqrt{1 + a^2/b^2})\},$$

where $W$ is the watts per cm.$^2$ on a rectangular focus $2a \times 2b$, $T_M$ is maximum temperature permissible in the target, and $k$ the thermal conductivity of the anode.

Mr. Stephen then discussed the design of the line focus. The usual size is 12 mm. × 1·2 mm. and this gives an effective focal-spot size of 1·2 mm. × 1·2 mm. when the beam is taken off the anode at an angle of 6° to the line focus. If crystallographers were to standardize spectrometer design, then it might be possible to build a tube with a line focus 36 mm. × 1·2 mm. This would give an effective spot-size of 3·6 mm. × 1·2 mm., which would be admirable for use with a slit-type collimator. With a monochromator this beam could be concentrated further into an area of either 1·2 mm. × 1·2 mm. or 3·6 mm. × 0·4 mm., depending upon the direction of reflexion of the beam from the crystal monochromator.

Dr. D. P. Riley, of the Cavendish Laboratory, Cambridge, read a paper on monochromators. Slides were shown of X-ray pictures of liquids with spurious lines due to the characteristic absorption of reflexion of part of the white radiation. These false intensity peaks were shown to be absent when a beam of radiation from a monochromator is used. In designing a monochromator the crystal should be capable of rotation and also a lateral movement parallel to the axis of the tube. A crystal used as a monochromator should reflect a strong beam of sufficient breadth to bathe the sample under investigation. The crystal, which should not be mechanically deformable, should have a reflecting face of 3–5 mm.$^2$. It should also have a high $F$ value, be mosaic but not polycrystalline, and should be reasonably stable to heat, humidity and X-rays. It was said that planes with low Bragg angles are best, but the reflexion must be clear of the main beam of X-rays. Examples of monochromators were given. In using reflected radiation, a beam of wave-length $\lambda$ may also contain harmonics $\lambda/2$, $\lambda/3$, etc., which arise from the white radiation. The reflected radiation is seriously polarized, and this gives rise to difficulties in intensity calculations on reflexions. It was suggested that tube manufacturers should be asked to give some idea of the state of polarization of the X-ray beams from the tubes they supply. For low-angle work a perfect reflector such as calcite, giving a very sharp beam, is better than a mosaic crystal of the pentaerythritol type.

The discussion was opened by Sir Lawrence Bragg, who urged that if automatic gear is produced it should be really 'foolproof', and that some 'figure of merit' of an X-ray tube should be given by the manufacturers. He also asked that information as to the best form of monochromator should be made available. Dr. H. Lipson (Cavendish Laboratory, Cambridge) asked for greater co-operation between

designers and users; he stated that the filament life in a Metropolitan-Vickers tube is 300–500 hours. Dr. Witty replied that magnetic and electric automatic gear should be 'foolproof'; vacuum relays are being improved, but are more of a problem. Dr. W. T. Astbury (Textile Physics Laboratory, University of Leeds) said that the inventing scientific worker gets little return for his efforts and is often himself held up for lack of apparatus. He believed that for further progress in design, automatic gear in experimental tubes should be kept down to a reasonable minimum. Dr. E. Green (Unilevers, Ltd.) directed attention to a Russian X-ray tube in which a spirally-grooved rotating anode acts as its own molecular pump. Mr. H. P. Rooksby (General Electric Co., Ltd.) described briefly a demountable X-ray tube which has been in continuous use at his laboratory for ten years. Dr. I. MacArthur (Textile Physics Laboratory, University of Leeds) suggested that a monochromator might be placed inside the X-ray tube to produce a maximum intensity. Dr. A. Taylor (English Electric Co., Ltd.) said that from a sealed-off X-ray tube a beam has been obtained in which iron lines have been found present due to the evaporation of iron from a filament lead on to the target of the tube.

Dr. W. T. Astbury opened the afternoon session with a description of the high-output X-ray tubes which have been developed in his laboratories. The rotating anode tube has been evolved in three stages. It has a rotating copper target with a mercury seal and the body is stainless steel. Two difficulties were anticipated with this type of instrument. The first was the stability of the rotating mercury columns. An equation of the overall height of the mercury meniscus rotating with an angular velocity $\Omega$ and of internal and external radii $a$ and $b$ is given by

$$h = \frac{\Omega^2 a^4}{g(b^2 - a^2)^2}\left[\frac{b^4 - a^4}{2a^2} - 2b^2 \log_e b/a\right] \simeq \frac{\Omega^2 a \delta}{3g}(1 - \delta/2a), \text{ where } \delta = (b - a).$$

When $\Omega = 500$ r.p.m., then $h = 0{\cdot}3$ in. for the inner and $0{\cdot}45$ in. for the outer rotating system, which agrees near enough with observations. In the latest instrument $(b - a) = \frac{1}{8}$ in., but this can be reduced. Thus it can be said that the mercury column is stable. The second difficulty is evaporation from the mercury into the vacuum, but this has been eliminated by covering the meniscus with a layer of Apiezon oil, and no contamination of the vacuum chamber or anode then occurs. Dr. Astbury described how the first instrument was designed as a gas tube; this was then modified to contain a filament source. The third tube was a completely re-designed instrument which ran at 70 milliamperes and 30 kV. This is the limit of the high-tension equipment at the moment available, but there is no reason why much higher currents should not be used.

A much simpler and cheaper moving target tube has been designed which will be within the reach of any research worker requiring beams of reasonably high intensity. The instrument is continuously evacuated, and the anode is a flat hollow bar (through which runs the cooling water) which oscillates backwards and forwards at three complete oscillations per second. The vacuum seal is made by means of a tombac bellows at each end. A second and slower motion is given to the bar to vary the position of the instantaneous zero of the main oscillation and thus prevent the development of hot spots during running. Alternative methods of sealing the vacuum joint of the oscillating anode were suggested. Dr. Astbury visualizes a long oscillating tube incorporating a series of anodes all sealed by a single pair of bellows and operated from the same pumping system, and each having two windows. (At the time of the meeting the instrument had not been developed to the stage of giving an X-ray beam, but since that time the tube has run steadily at 45 milliamperes and 30 kV. and very good photographs of ramie have been taken in 2 minutes with a $\frac{1}{2}$ mm. collimator and a distance of 2 cm. (see *Nature*, **155**, 108; 1945).

Dr. I. MacArthur reviewed the development of moving-target tubes, and gave special prominence to features such as continuous cooling and vacuum seals in the instruments. The main point is not the overall power of the tube, but rather the maximum loading per unit area of the focal spot. This is determined by focusing, which might be improved by the electron gun method as used by Siegbahn, or by magnetic means. A clean target is also necessary; tantalum filaments were found to be less likely to sputter the anode than tungsten, although they have a shorter life. Dr. MacArthur referred to the possible use of the rotating anode as a molecular pump. He warned would-be makers of rotating anode tubes against porosity in vital metal parts.

Replying to questions by the chairman in the discussion, Dr. Astbury said that the rotating anode of his tube can be changed easily without draining away the mercury, and that at 70 milliamperes and 30 kV. the tube is 8–20 times as fast as a normal Philips tube. Dr. Kathleen Lonsdale described the 50 kW. and 5 kW. moving anode tubes at the Royal Institution. The output of a Shearer tube running at a nominal 5–10 milliamperes and 40–45 kV. was given as 400 watts, and a comparison of the speeds of photography of a normal Phillips tube, a Shearer tube, and the 5 kW. tube was said to be 1 : 8 : 24. The Shearer tube was used with a modified Wehnelt break and an induction coil, and a good beam was said to be maintained over a period of four hours.

Mr. Stephen said that the load on a Philips tube can be increased, but this will reduce the life of the tube. He asked that collimator sizes should be standardized, and then manufacturers would be able to design tubes for special purposes; at present the normal sealed-off tube has to be used on all occasions. Mr. T. S. Millen (Metropolitan-Vickers Electrical Co., Ltd.) outlined the optimum conditions for a rotating anode used as its own molecular pump. Replying to Mr. Rooksby, Dr. Astbury, Dr. MacArthur and Dr. Green said that the porosity of the metal in the rotating anode tube is in the outer casing and not in the anode itself. This leakage is closed by coating lightly the affected parts with shellac. Replying to a question of relative positions of focal spots in the Matchlett and Philips tube, Mr. Stephen said that with $9\frac{1}{2}$ cm. cameras the extra 1 cm. distance between focal spot and window in the Philips tube is immaterial, and is necessary because the windows of the Philips tube are of Lindemann glass. Dr. A. Taylor suggested that alloy targets might be used so that with suitable choice of filters different monochromatic beams might be obtained. Mr. F. A. Bannister (British Museum) told the meeting that minerals can be readily examined using beams from Shearer tubes, the purity of radiation of which is universally accepted.

F. HAPPEY.