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AN IMPROVED X-RAY DIFFRACTION CAMERA

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For obtaining high quality X-ray diffraction patterns with minimum exposure times, an improved Debye-Scherrer camera has been designed and is produced by North American Philips Co., Inc. Various factors entering the design of the camera and interrelated in their influence on the properties of the patterns are considered in this article. The necessary compromises between such properties as line intensity, line sharpness (resolution), contrast and line shape are favorably affected by several resources. Line intensity is improved without loss of resolution by using a rectangular collimating aperture for the X-ray beam. Contrast is enhanced by surrounding the primary beam on its path to and from the specimen by a collimator and exit tube, thus diminishing the film blackening resulting from radiation scattered in the air. It is shown that these "anti-air-scatter tubes" must have definite dimensions in order to insure the limitation of air scatter to a minimum without causing undesirable blind areas in the diffraction patterns and avoiding any scattering from the tubes themselves. The optimum forms of the tubes for different purposes have been computed, and the cameras can be provided with one of several systems designed in accordance with the results of these computations. The tubes are made easily interchangeable. Handling of the cameras in general is simplified to a considerable extent by the application of certain mechanical design principles outlined by Buerger.

In this article a description is given of an improved camera for the photographic recording of X-ray diffraction patterns of polycrystalline (e.g. powder) specimens according to the Debye-Scherrer method. There is no need to enumerate here the many purposes which these patterns may serve. It is sufficient to state that they are used extensively in research and industrial laboratories for the identification or chemical analysis of materials, for the examination of crystalline structure and grain size, and for the analysis of stresses, texture etc. [1]).

Fig. 1 illustrates the well-known principle of the Debye-Scherrer camera. This camera is a cylindrical enclosure along the axis of which the cylindrical specimen is mounted; the film strip is placed against the inner wall of the camera. The X-ray beam (made as monochromatic as practicable) enters the camera through an aperture in the cylindrical wall. Parts of this beam are diffracted at definite angles by the tiny crystals in the specimen and produce characteristic diffraction lines on the film, while the diffracted part of the (primary) beam passes on to a stop or receiver, or may leave the camera through an aperture in the opposite cylindrical wall.

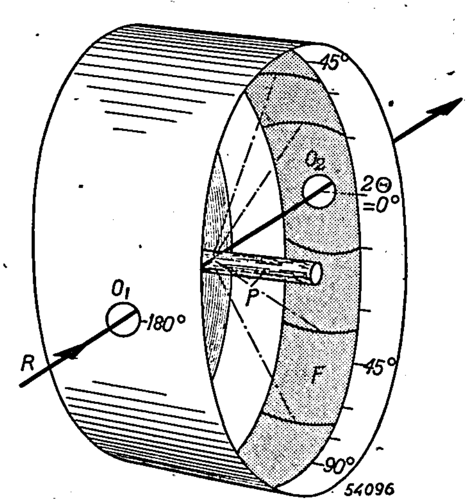

Fig. 1. Principle of the Debye-Scherrer camera. A monochromatic X-ray beam R enters the cylindrical camera through an aperture O_1 and is partly diffracted in the specimen P. The undiffracted part of the beam leaves the camera through the aperture O_2. The diffracted rays cause the lines (ring segments) of the diffraction pattern on the film F.

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[1]) A survey of the applications of the Debye-Scherrer method can be found, for instance, in Philips Techn. Rev. 5, 157-166, 1940. A few months ago an article in this Review was devoted to the "Geiger counter X-ray spectrometer", an apparatus also produced by North American Philips Co., Inc., and enabling the direct measurement of the intensity distribution of X-ray diffraction patterns (Philips Techn Rev. **10**, 1-12, 1948 (No.1)). The Geiger counter method and the photographic method are well suited to supplement each other due to their different performance characteristics.

A well designed Debye-Scherrer camera should meet the requirements that, on the one hand, the recording of a pattern be made as easy and quick as possible, while, on the other hand, the highest possible quality of the resulting patterns be secured.

As far as ease of handling is concerned, the cameras produced by North American Philips Co., Inc., have been designed in the main in accordance with the cameras described a few years ago by M. J. Buerger [2]). The principles outlined by Buerger were aimed, among other things, at facilitating the alignment of the camera with the X-ray tube, the recovering of the alignment after temporary removal of the camera from its position, the centering of the specimen in the camera, the rapid mounting of the film, etc. The successful realization of such principles depends mainly on precise mechanical construction. Those are not the subjects which are extensively treated in this article, although some details relating to the aforementioned principles are given in *figs 2* and *3* and explained in the accompanying legends. On the contrary, this article deals much more thoroughly with the speed of recording and the quality of the patterns obtained.

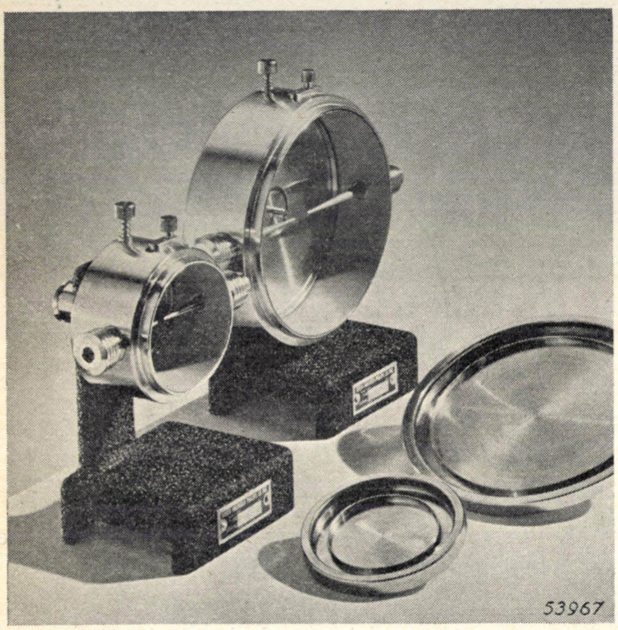


Fig. 2. The Debye-Scherrer cameras with diameters of 57.3 and 114.6 mm, as manufactured by North American Philips Co., Inc. Next to the cameras are the covers, fitting light-tight on the cameras, so that it is not necessary to make exposures in a dark room nor to cover the film with black paper. In each of these cameras the primary X-ray beam passes through two conical tubes between which the very thin rod-shaped specimen is mounted. After placing the specimen roughly on the axis of the camera it is accurately centered by means of a threaded "pusher" perpendicular to the axis. This is facilitated by slipping a small magnifying glass on the end of the exit tube. During the exposure the glass is replaced by a cap containing a small fluorescent screen, on which the primary beam and the shadow of the specimen may be observed. Thus the alignment of the camera and the centering of the specimen are checked. The fluorescent screen is covered at the inside with black paper, making the camera light-tight, and at the outside with lead glass, absorbing the remainder of the primary beam without obstructing the view of the fluorescent screen. The film is placed in the camera according to the Straumanis method: the film takes up nearly 360°, and two holes of normalized width of 9 mm are punched in it where the tubes are to be inserted. Careful design and machining of the tubes make it possible to remove them easily for mounting the film and to replace them in exactly the correct alignment. The film is expanded tightly against the inner wall of the camera by means of a movable finger which pushes one end of the film, the other end being fixed by a stop.

Properties involved in quality of pattern

Information about the nature of the X-rayed specimen can be deduced from the positions (i.e., the diffraction angles) of the diffraction lines, from their relative intensities and from the line shape. High quality patterns, therefore, will exhibit the following properties:

1) Recording of all lines that are of importance. This especially refers to very weak lines, which may often be important in quantitative chemical analysis by X-ray diffraction, and to lines at very small or very large diffraction angles (2Θ near zero or 180°, Θ = Bragg angle).
2) Great sharpness of lines. This will enable the diffraction angles to be determined very accurately and at the same time it will provide a good resolution of adjacent lines.
3) Good contrast of recorded picture, i.e., minimum and relatively uniform background density on the film in comparison to the density of the lines. A strong background would not only affect the observable recording of very weak lines mentioned in (1), but also impair the measurement of the correct relative intensities, for strong as well as for weak lines.
4) Good line shape, i.e., correct contour, uniform density, and constant intensity along the usable length of each line (insofar as the desired data do not actually depend on broadening, spottiness or specific intensity distribution of each line). This property is important for good definition of the position of the lines and for correct intensity measurements.

In order to meet the requirement of quick recording of patterns, especially in view of point (1), we must add to this list of desiderata:

5) High intensity of diffracted rays.

[2]) M. J. Buerger, The design of X-ray powder cameras, J. Appl. Phys. **16**, 501-510, 1945. Cf. also: M. J. Buerger, An X-ray powder camera, Amer. Mineralogist **21**, 11-17, 1936.

Camera design considerations relating to quality of pattern and speed of recording

When considering the design factors which influence the abovementioned properties it becomes apparent that in several instances the requirements are conflicting. For the sake of convenience let us start from the sharpness of lines.

Sharp definition of the lines mainly depends on the diameter of the camera, the width and divergence of the primary X-ray beam, and the thickness of the specimen.

Simple geometrical considerations show that the lines become sharper (i.e. narrower in relation to their mutual separation distances) with an increase

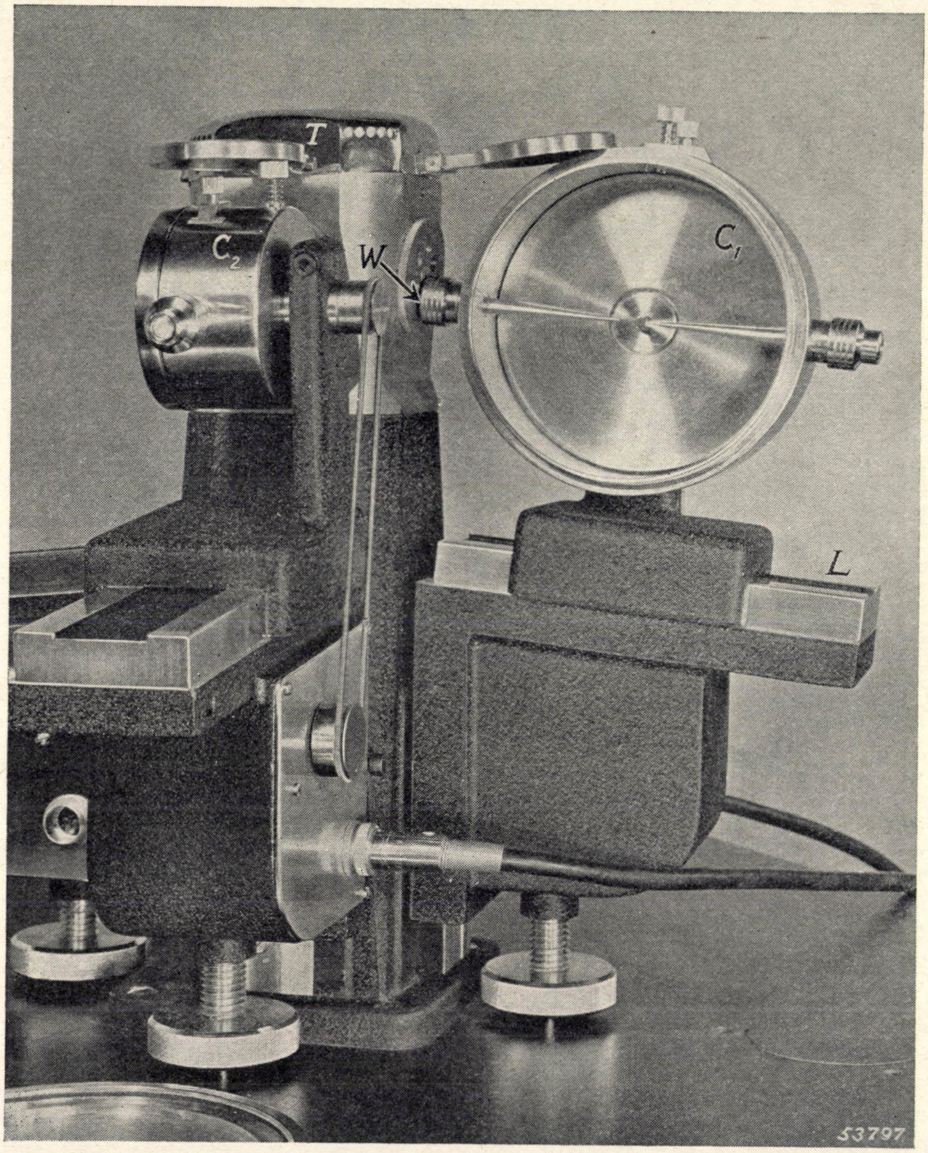


Fig. 3. The two cameras, C_1 and C_2, set up with an X-ray tube T on the desk of a Philips diffraction apparatus (the same tube can, as a rule, be used for from one to four cameras at the same time). Each camera is supported by a bracket and rests on a flat track L. The bracket being fixed, the camera may be rocked around and translated along the specimen axis; the track may be translated along and rocked about a vertical axis (parallel to the X-ray tube housing). This permits an easy aligment of the diffraction camera with the corresponding exit window (W) of the X-ray tube. Once aligned, the camera may be removed for film processing and reloading, and repositioned on the track without need for further alignment. Small and large cameras may be interchanged without altering the position of the track. The X-ray tube, too, is designed to allow changing tubes (e.g., for varying the target material) without affecting camera aligment. The tracks make an angle of 6° with the horizontal plane. Hence, if the camera is rocked around its axis so that the two tubes are parallel to the track, the X-ray beam used makes an angle of 6° with the horizontal face of the X-ray tube target. By slightly rocking the camera and raising the track this angle can be made smaller (e.g., 3°). While the exposure is being made the specimen is rotated continuously around its axis by a pulley attached to a small electric motor covered under the track.

in the diameter of the camera. Enlarging the camera, however, is soon limited by the rapid decrease in line intensity. In order to keep the exposure times within reasonable limits, one does not go any further, as a rule, with the camera type under consideration, than the frequently used 114.59 mm diameter. The Philips cameras possess diameters of 114.59 and 57.3 mm [3]).

Next, consider the divergence of the irradiating X-ray beam. Lines will be sharper for a smaller beam divergence, as may be seen from *fig. 4*. On the other hand, a larger area of the X-ray tube focus contributing to the irradiating energy corresponds with a greater divergence of the beam (the distance between focus and specimen, and the angle at which the anode surface is viewed being given). Hence, a compromise between line sharpness and line intensity again will be necessary.

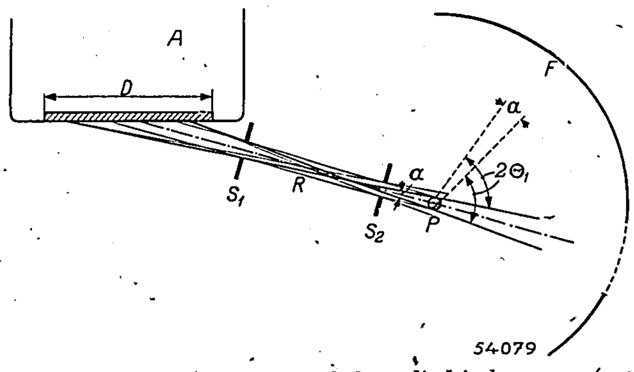


Fig. 4. Vertical cross-section of the cylindrical camera (not drawn to scale), showing film F, specimen P, anode A, focal spot D and beam collimating apertures S_1 and S_2. The divergence of the primary X-ray beam R (in this case limited by aperture S_1 and specimen diameter) is α. The same angular divergence appears between rays diffracted at the outermost points of the specimen cross-section and contributing to one diffraction line ($2\Theta_1$ = diffraction angle).

Finally the thickness of the specimen (which also determines the necessary width of the primary beam) should be considered. To obtain sufficiently sharp lines, the specimen must be extremely thin, in some instances 0.3 mm or even less. Evidently the X-ray energy diffracted by the specimen will, then, be very small. So we have once again the conflict between line sharpness and line intensity. In this case, however, other requirements mentioned above are involved in the conflict too. Diminishing the specimen diameter is apt to create a very unfavorable condition with respect to the contrast of the pattern and the line shape. This may be seen in the following way:

[3]) These seemingly odd diameter values have the advantage that the diffraction angles 2Θ are in a simple way numerically related to the line distances measured in mm on the film: 1° of 2Θ corresponds to 1 mm with the 114.59 mm camera, to $^1/_2$ mm with the 57.3 mm camera.

The main cause of the occurrence of a uniform background on the film, detrimental to contrast, is the scattering of the primary beam in the air through which it passes on its way through the camera [4]). Given a certain path length of the beam in air and assuming the beam width in all cases to be equal to the thickness of the specimen, the air volume contributing to the undesired scattering will decrease proportionately to this thickness. The volume of the specimen contributing to the desired diffraction, however, decreases according to the square of the specimen thickness, so that the air scatter will be relatively more pronounced the thinner the specimen.

Similarly, the unfavorable effect on the line shape may be understood. Given the average size of the crystals oriented at random in the specimen, the number of crystals irradiated decreases with the square of the specimen thickness, and so does the average number of spots from which each diffraction line is built up, while the line width on which the spots are distributed diminishes only proportionately to this thickness. Thus, the lines in the diffraction pattern will be less uniformly blackened and, in the extreme case, tend to get a spotty appearance on decreasing the specimen diameter. (This is quite noticeable in the examination of compounds of low crystal symmetry, for which a given type of lattice planes will attain an orientation suitable for reflecting only in a single or very few positions of the crystal.)

The various conflicts pointed out to exist between desired features and necessitating as many compromises in camera design, may have caused a somewhat gloomy impression as to camera performances which can be expected. Fortunately, there are several resources which favorably affect the necessary compromises and which have been applied in the Philips cameras with considerable success. One such resource, well-known for a long time, is the rotating of the specimen during the exposure (combined with a translation along its axis if desired); cf. fig. 3. This results in more crystals

[4]) We will not consider in this article causes such as possible fluorescence (X-rays or light) which is brought about by the primary beam in the specimen and which is non-directional, or the "white" (non-monochromatic) components of the X-ray beam which produce a diffraction spectrum for every wavelength that is present, all these spectra being superimposed to form a continuous background. These contributions to background (and hence indirectly also the contribution from air scatter) may be diminished by special measures for making the primary beam more monochromatic, e.g. by means of a "crystal monochromator". Such a device may also be used with the Philips cameras. The scattering of the beam at various apertures through which it passes will be discussed below.

coming temporarily in an appropriate position for contributing to a specific diffraction line and, hence, in diminishing the spottiness of lines. Other resources, also well-known but, in our case, applied in a novel manner, are concerned with the X-ray beam geometry and the reduction of air scatter. These will be treated to somewhat greater detail in the following parts of this article.

Geometry of beam collimating system

The desired beam shape is obtained by means of a collimator system, commonly consisting of two pinhole apertures (cf. fig. 4). When diminishing the specimen thickness to values of, say, 0.5 or 0.3 mm, the width of the X-ray beam had to be adapted to the specimen thickness by making the collimator apertures narrower. This was necessary, because all parts of the primary beam failing to strike the specimen do not add to the line intensity but only to the undesired air scatter. This necessity, however, applies only to the width of the beam in the "equatorial" cross-section of the camera and the specimen, represented in fig. 4. The width of the beam in a direction parallel to the specimen axis, need not be limited to the same content. This means that the beam need not be made rotation-symmetrical but that it can be given an oblong cross-section, by taking a rectangular slit rather than a circular pinhole as a first collimator aperture; cf. *fig. 5*. The obvious advantage of this is that a relatively long part of the rod-shaped specimen is irradiated, resulting in a higher intensity of the diffraction lines and, at the same time, in a more uniform blackening of the lines and truer relative intensities because a larger number of crystallites are irradiated [5]).

A limit to the slit length is set by the fact that with too great a divergence of the X-ray beam in a direction parallel to the specimen axis, the line sharpness and line shape will again begin to suffer. This is explained with reference to *fig. 6*. An ideal diffraction line is the intersection of the film with a cone of rays the apex of which lies in the specimen and the axis of which is the direction of

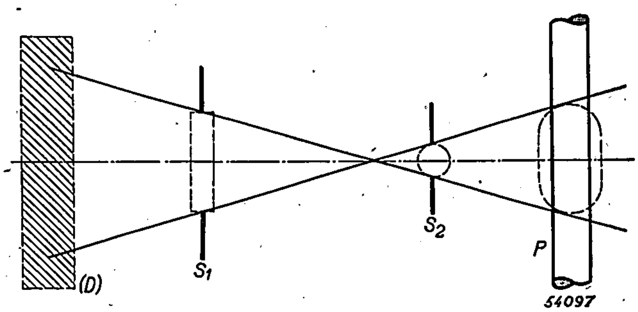


Fig. 5. Horizontal section of the primary X-ray beam. The first collimator aperture S_1 is made rectangular; S_2 is the circular second aperture; P, the specimen; D, projected focal spot. At P the X-ray beam has an oblong cross-section. (The cross-sections of D and of the beam at S_1, S_2 and P are turned in the plane of the drawing and indicated by dotted lines.)

the primary ray. With a diverging primary beam each ray will produce its separate diffraction cone. Each diffraction "line", then, is formed by the superposition of a continuous series of cones with different axes. This results in a broom-shaped broadening of each line on either side of the "equator" of the film, and in the case of a strongly diverging primary beam it also results in a line broadening at the equator itself. The effect is very noticeable at very small and large diffraction angles where the arcs form complete circles.

This well-known "slit effect" is not troublesome with the aperture dimensions chosen for our cameras. The first aperture is 0.5 mm wide and 2 mm long, the second one is circular, with a diameter of, say, 0.5 mm. The resolving power for adjacent lines proves to be hardly less than that of the usual systems with two circular pinhole apertures of 0.5 mm diameter, and it is certainly better than

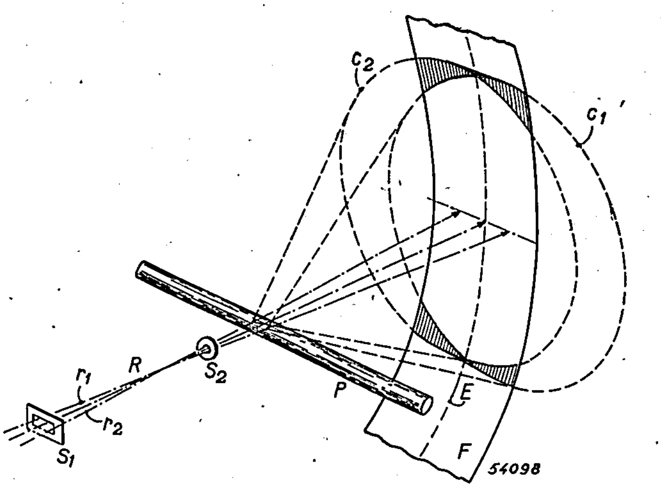


Fig. 6. Slit effect on shape of diffraction lines. Diverging rays of the primary X-ray beam R produce different cones of diffracted rays, each having its original ray as an axis. Diffraction lines on the film are obtained by the superposition of eccentric rings arising from the intersection of such cones with the film. c_1 and c_2 are the rings produced on the (virtually extended) film surface by the rays r_1 and r_2 respectively; E is the "equator" of the film F.

[5]) On the use of a rectangular first collimating aperture see e.g. F. Halla and H. Mark, Röntgenographische Untersuchung von Kristallen, Leipzig 1937, p. 156. The successful application of this device depends on extreme precision in the mechanical construction and particularly in the alignment of the slit with the focus and the specimen. Special precision techniques and tools have been developed for this purpose in the case of our cameras. The slit is punched out of a piece of lead by means of a special hob, which, when pressed further (without withdrawing), cuts a circle out of the lead. Thus the slit is automatically centered in a circular lead disc which may then be fixed at its proper place in the collimator tube.

that of a similar system with 0.75 mm apertures. Compared with the first system, an intensity factor of 3 is gained, when this collimator system is completely filled with radiation.

The value of 0.5 mm was chosen for the aperture widths because it was assumed that in general the specimen would be no thicker than 0.5 mm, and also because this value is well adapted to the focal spot dimensions of the Philips X-ray tubes supplied for use in the Philips diffraction apparatus. A few remarks should be made concerning this latter point.

The collimator system must, on the one hand, insure the beam width and divergence not to exceed the limits set by the desired resolution. On the other hand, it is desired that the radiation of the X-ray tube be used to good efficiency. This evidently will not be the case if the outermost points of the specimen circumference cannot "see" the full projected size of the focal spot because rays coming from the ends of the focal spot are cut off by apertures (cf. fig. 4). Therefore, we might say that the focal spot and the collimator system are well adapted one to the other if the first collimator aperture in fact could be omitted without affecting the beam shape [6]).

With the Philips X-ray tube mentioned above the focal spot is 9 mm long. At an angle of 3° from the face of the anode the projected width of the focal spot is 0.5 mm; in that case, rays from the entire focal spot pass through the apertures of size mentioned above to each point of the specimen cross-section, and the full radiation intensity is utilized. At an angle of 6°, however, which is more commonly used in diffraction apparatus, the projected width of the focal spot will be 0.9 mm, resulting in the ends of the spot not being used.

If the 3° angle is always used the only function of the first collimator aperture is to collimate the beam in case an X-ray tube having a larger focal spot than that of the Philips tubes is to be used.

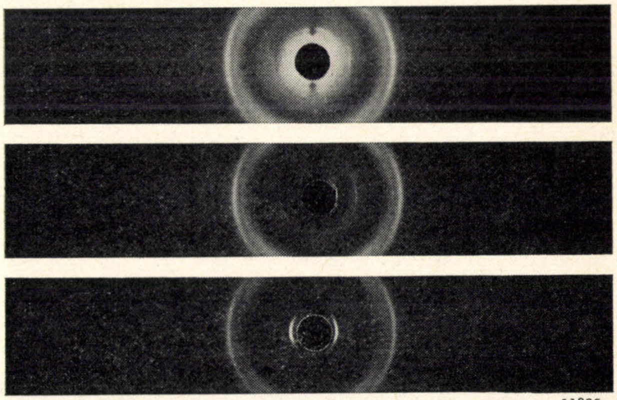
A similar consideration applies to the 2 mm length of the first aperture: the dimensions of the focal spot of Philips X-ray tubes in this direction being on an average only 1.2 mm, the collimating action of the first aperture in this direction likewise becomes apparent only if other tubes, having wider focal spots, are used. With the focus width of 1.2 mm mentioned above, the first aperture is not completely filled by the beam and the intensity gain as compared with two circular 0.5 mm pinholes, therefore, is somewhat smaller than indicated above (gain factor about $2\text{-}2^1/_2$).

Anti-air-scatter tubes

The deleterious effect of air scatter on the contrast of diffraction patterns can be eliminated by evacuating the camera. This technique, though applied in some cases, is not convenient in instruments for routine investigations. An alternative and much simpler resource consists in confining the primary beam on its path to and from the specimen within two metal tubes (see fig. 2). The entrance tube also contains the collimator system. Film background is reduced because the air path is now limited to the distance between the ends of the two tubes.

As is well known, incorporating the beam collimating system in a tube inside the camera has, moreover, the important advantage that the focus-to-specimen distance can be made smaller, entailing a considerable increase in intensity.

To reduce air scatter to a minimum one would prefer to bring the tubes as close as possible to the specimen. It is obvious, however, that sufficient space must be allowed around the specimen for the emission of the rays diffracted by the specimen. The diffraction lines at angles near 0° and 180° evidently are most liable to be intercepted by the rims of the tubes, and it was quite a common deficiency in cameras previously designed that "blind areas" were caused on the film at these diffraction angles, i.e., around the holes punched in the film at the entrance and exit region for the primary beam. Such a blind area is clearly visible in the pattern of a commercial soap reproduced in *fig. 7b*. On the other hand, fig. 7*a* exhibits the dense background which is produced, particularly near the exit film hole, if no exit tube or one of insufficient length is used.



a

b

c

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Fig. 7. Part of diffraction pattern of soap, containing the range of diffraction angles 2Φ from 0° to about 90°, taken with copper $K\alpha$ radiation and nickel filter.

a) No exit tube used. Considerable background occurs in the range of low diffraction angles, near 0°.

b) Badly designed exit tube. The tube, due to its contour, intercepts the diffraction lines which would otherwise be registered near the film hole (blind area effect).

c) Recorded with new 114.6 mm Philips camera. Background has been reduced to a minimum, without any blind area effect around the film hole, due to proper design of the collimator and exit tubes. Lines at small angles that could not be seen before are now reproduced very clearly.

Closer investigation shows that it is possible to compute specific dimensions of the anti-air-scatter tubes which give best results for different purposes. These computations have been made for our cameras and the tubes now employed have been designed in accordance with the results. For comparison, a pattern of the same specimen as in fig. 7*a* and *b* recorded with one of our improved cameras is

6) For a set-up without "first" collimator aperture, cf. A. Guinier, Radiocristallographie, Dunod Paris 1945, p. 80. The same principle is applied also in the Geiger counter spectrometer (cf. footnote [1])).

reproduced in fig. 7*c*. The occurrence of a "blind area" outside the film hole is completely avoided; some lines close to the hole, which could not be seen with previous systems on account of either background or blind area, can now be seen very clearly.

The line of thought for the computation of the tubes will be outlined below.

Computation of optimum form of anti-air-scatter tubes

Exit tube

Let us take the exit tube first; see *fig. 8*. The diameter of the camera, the diameter of the specimen and the diameter of the film hole are considered as being fixed. The primary X-ray beam — provisionally assumed to have rotational symmetry — is also considered to be given. The simplest form for the exit tube to take will be that of a truncated cone, with the base falling within the film hole while the width and the position of the tip still have to be chosen.

The exit port will, of course, have to receive the whole of the primary beam, whose edge is indicated by line I in fig. 8. The inner rim of the exit

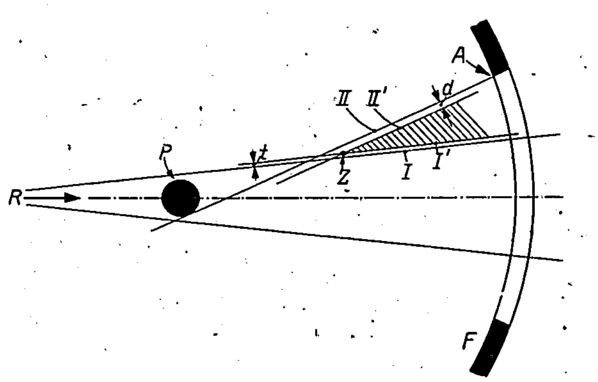


Fig. 8. Dimensioning of the exit tube. R, diverging X-ray beam; P, specimen; F, film; A, rim of the exit film hole. For the most favorable design the inner rim of the tip of the exit tube must lie at the point Z.

port will therefore have to be outside line I, measured from the axis, or rather outside line I', drawn parallel to I in order to provide a certain clearance t, so that the tip of the exit port will not be touched by the beam (otherwise there would be considerable scattering from this tip and the scattered rays would reach the film without hindrance).

On the other hand, we want to introduce the requirement that the abovementioned blind area effect should be completely eliminated, i.e., that the exit port should not obstruct any rays diffracted from the specimen, not even for the lowest usable diffraction angles. That means that the whole of the specimen must be visible from the rim A of the film hole. The outer rim of the exit port will therefore have to be within line II, and the inner rim within line II' which has been drawn parallel to line II at a distance d equal to the thickness of the wall of the tube.

The two conditions stated above limit the allowable position of the inner rim of the exit port to the hatched region between lines I' and II'. To envelop the primary beam over the greatest possible distance it is necessary to place the inner rim of the exit port in the extreme corner of the hatched area, i.e., at the point Z where the lines I' and II' intersect.

Collimator tube

Similar considerations lead to the required shape of the collimator tube, though here the problem is more intricate, as the collimating of the primary X-ray beam also is involved. We shall provisionally adhere to the supposition that the beam is made rotation-symmetrical, thus that both collimating apertures are circular, their widths, $2s_1$ and $2s_2$ (which need not be equal), being determined in advance on grounds in part explained above. Also it is assumed that the position of the first aperture can be considered fixed on the strength of certain geometrical and physical considerations; we have placed it at a distance $a = 20$ mm in front of the entrance hole in the film. We are still free to choose the position of the second collimating aperture, of course within certain limits.

We can start once again from the fact that the part of the collimator lying inside the camera may be shaped as a truncated cone, with the base falling within the hole punched in the film at the spot where the beam enters the camera. We shall now have to choose the most suitable width and position of the opening at the strip of the tube.

First of all we have to observe that this opening cannot itself be successfully utilized as a second collimating aperture. The primary X-rays are strongly scattered at the edges of the beam-limiting apertures. If the second beam-limiting aperture were to be located at the end of the collimator tube this scattered radiation would reach the film unhindered and that result would be worse than the air scatter it is desired to eliminate. The second collimating aperture must, therefore, be placed inside the collimator tube, and the tube tip (scatter cup), in addition to its function of diminishing air scatter, must keep the rays scattered by the aperture away from the film[7]). Thus we arrive at the configuration sketched qualitatively in *fig. 9*.

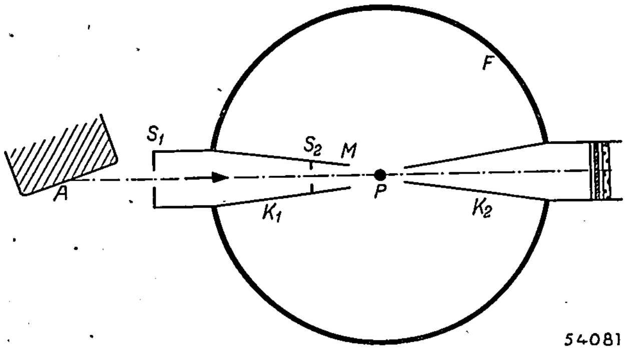


Fig. 9. Configuration of collimator and exit tubes K_1 and K_2 (qualitatively). The second beam-limiting aperture S_2 is inside the collimator tube, so that the rays scattered at this aperture are kept away from the film F by the tube port M (scatter cup). P, specimen, A, anode. (At the end of the exit tube, the three closures mentioned in the description of fig. 2 are indicated schematically.)

From *fig. 10* we can now deduce the conditions which have to be fulfilled by the collimator tube.

The tube tip must not cut off any part of the divergent beam formed by the apertures S_1 and S_2 (for if it did it would itself act as a second aperture). Hence the condition: 1) The inner rim of the scatter cup must be outside line I, measured from the axis, or rather outside line I' drawn parallel to I, again to allow for the necessary clearance t.

Furthermore, the scatter cup must intercept all scattered rays which, coming from the circumference of S_2, would fall outside the film hole on the opposite side of the camera (point A). Consequently: 2) The inner rim of the scatter cup must lie inside line III. The two conditions taken together indicate that the inner rim of the scatter cup must lie in the hatched region shown in fig. 10.

Finally, any blind area in the 180° region outside the entrance film hole must be avoided, which means that it must be possible to see the whole of the specimen unhindered from the rim C of the film hole. Hence it follows that the outer rim of the scatter cup must lie on the inside of line IV, or, if we draw a line IV' parallel to IV at a distance equal to the wall thickness of the tube: 3) The inner rim of the scatter cup must be on the inside of line IV'.

We see that all three conditions together still leave us free to place the rim of the scatter cup somewhere in the triangular part of the hatched field delineated by heavy lines. To envelop the primary beam over the greatest possible length, the rim of the scatter cup will have to be placed in the farthermost right-hand corner (W') of the triangle.

The position and the width of the collimator tube tip are thus fixed as soon as the position of the second collimating aperture S_2, as yet unspecified, has been established. When we vary this position we see that the more nearly horizontal line I (and I') becomes, and thus the smaller the divergence of the X-ray beam, the closer the point W' is brought to the specimen. It is therefore desirable to place the aperture S_2 as close to the specimen as possible. A limit to this is set because, as a closer examination will show, as S_2 is brought nearer to the specimen the apex (W) of the hatched area is shifted in the same direction. S_2 can only be shifted, therefore, until point W has reached the line IV', the delineated triangle in this case being reduced to that single point. The position of the second collimating aperture is then unambiguously established and the largest possible length of the collimator tube corresponds to this situation.

At the same time this position of the second aperture is also the most suitable for the design of the exit tube. In our discussion on this point with reference to fig. 8, which led to the tip of this tube being placed at point Z, we had assumed the primary beam to be limited by line I as given. It is apparent that point Z, too, now comes closer to the specimen the more nearly horizontal line I is made. The permissible lengths of both the collimator tube and the exit tube thus become a maximum when the position of S_2 is determined by the foregoing considerations[8]).

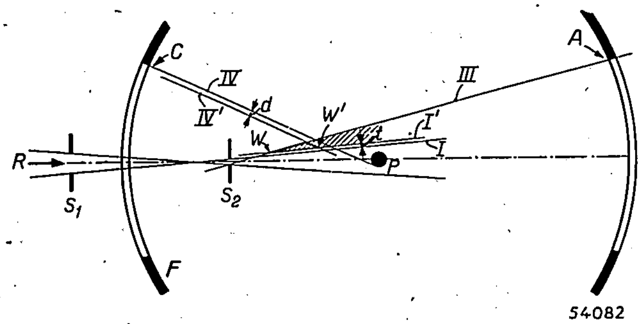


Fig. 10. Dimensioning of the collimator tube. R, X-ray beam; P, specimen; S_1 and S_2, apertures; F, film; C and A, rims of the entrance and exit film holes. With the position of S_2 being chosen arbitrarily as yet, for best results the inner rim of the collimator tube port must lie at the corner W' of the delineated triangle.

[7]) It is a common fault in many camera designs that this requirement is not met. The importance of the requirement was pointed out by M. J. Buerger, l.c. (1945), p. 505.

[8]) Incidentally, it should be pointed out that care must be taken to prevent rays scattered back from the wall inside the exit tube from returning through the tube tip and reaching the film. This requirement can easily be met by making this tube conical or by step-drilling it.

The optimum dimensions of the anti-air-scatter tubes can now be computed in an elementary, though not very simple manner, from the criteria established above. We introduce a system of coordinates ξ, η, having its origin O at the center of the entrance film hole as indicated in *fig. 11*. There are five unknowns: u, the ξ-coordinate defining the position of the second collimating aperture S_2; v and w, coordinates of point W, indicating the position and half the width of the collimator tube tip; x and y, coordinates of point Z, indicating the position and half the width of the exit tube tip.

According to elementary principles of analytic geometry we can write the equation for each of the straight lines I'-IV' in the form

$$\frac{\eta-\eta_1}{\xi-\xi_1}=\frac{\eta-\eta_2}{\xi-\xi_2},$$

where ξ_1, η_1 and ξ_2, η_2 are the coordinates of two fixed points through which the line is drawn. Since, according to the above, point W must be situated simultaneously on the lines I', III and IV', we get for the three unknowns u, v, w, the three equations:

$$I': \quad \frac{w+(s_1-t)}{v+a}=\frac{w-(s_2+t)}{v-u},$$

$$III: \quad \frac{w+s_2}{v-u}=\frac{w-f}{v-2R},$$

$$IV': \quad \frac{w+(r+d)}{v-R}=\frac{w-(f-d)}{v}.$$

$$II': \quad \frac{y+(r+d)}{x-R}=\frac{y-(f-d)}{x-2R}.$$

Elimination of u and v from the first three of these equations yields a rather cumbersome quadratic equation for w. After w is known, v and u may readily be computed, and likewise x and y with the help of u. We need not give here the detailed solutions and the computation of the numerical values.

Different types of anti-air-scatter tubes

In the above the rigorous requirement was set that nothing must be lost from the diffraction pattern, that is to say, the whole of the specimen must be visible from the rims of the film holes. In practice, however, this will never be required for the 0° section and the 180° section of the pattern at the same time. Organic compounds which often possess large interplanar spacings and, thereby, will produce very low angle diffraction lines, in general will not yield any "back-reflection" lines (near 180°). Conversely, inorganic compounds ordinarily give distinct back-reflection lines but rarely show lines corresponding to large spacings.

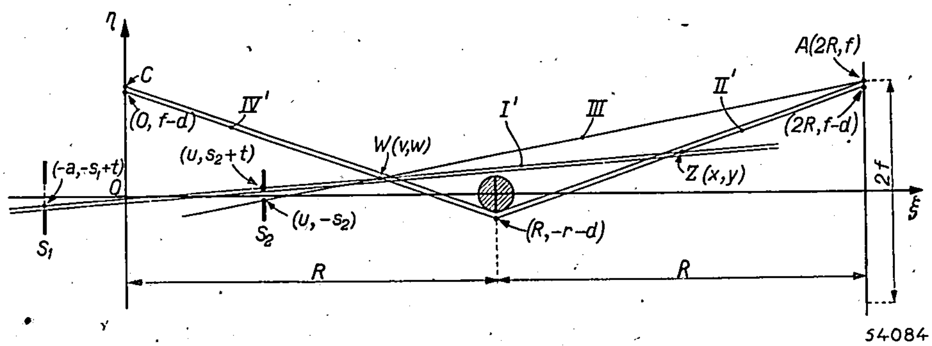


Fig. 11. The position of the second aperture S_2, and that of the points W and Z, indicating the best location and width of the tips of the two tubes, are given in the ξ, η-coordinate system by the coördinates u; v, w; x, y, respectively. The coordinates of all points used for the equations of lines I', II', III and IV' are indicated in the diagram.

Here R is the radius of the camera, r the radius of the specimen, d the wall thickness of the tube, t the desired clearance between the primary beam and the inner rim of the tube tip, f the radius of the entrance and exit hole punched in the film, s_2 half the width of the second collimating aperture; a and s_1 indicate the position and half the width of the first collimating aperture [9]). All these symbols stand for known numerical values.

Since point Z lies on the lines I' and II' we derive for the unknowns x and y the equations:

$$I': \quad \frac{y+(s_1-t)}{x+a}=\frac{y-(s_2+t)}{x-u},$$

[9]) These and the following equations contain two very obvious simplifications (cf. fig. 11): the circular cross-section of the specimen to which lines II and IV should be tangent was replaced by its vertical diameter; and the rim of the two holes in the curved film was assumed to have ξ-coordinates zero and $2R$, respectively. Actual computations showed that the latter simplification is permissible even in the case of the small camera ($2R$ = 57.3 mm) in which the film curvature is greatest.

Thus three different cases may be distinguished as regards the desired angle region of the diffraction pattern: the examination of forward diffraction only, of back-reflection only, and general work where neither very large nor very small angles are of interest but an optimal record of the entire "normal" angle region is desired. We have developed a number of different anti-air-scatter tube systems for these various purposes. Some of these are described below, the indicated figures corresponding to the 114.6 mm camera.

The first tube system (No. 77) is calculated for forward diffraction work. The rigorous blind area condition has been applied to the exit tube. Full line intensity is obtained at angles > 4.5° (the minimum angle permitted by the film hole). Lines at angles larger than 90° need not be recorded in this case. The collimator tube, therefore, has been extended so as nearly to touch the specimen. This has reduced the non-enveloped length of the primary X-ray beam to 19.9 mm.

An analogous tube system (No. 85) is made for back-reflection work. In this case only the collimator tube is subjected to the rigorous blind area condition, providing full line intensity up to angles of 175.5°. This tube system is used, e.g., for the precision determination of lattice constants. The non-enveloped beam length is 28.8 mm.

For the third case, i.e., all general work as in normal identification of compounds etc., the rigorous blind area condition has been replaced by a less rigorous one for both the collimator and the exit tube. This less rigorous condition consists in setting the requirement that, though not the entire specimen cross-section, at least part of it should be visible from the rim of the film holes. This means that a gradual intensity drop (a kind of "half-shadow") of the outermost lines towards the holes is allowed. *Fig. 12* illustrates the gain in tube length

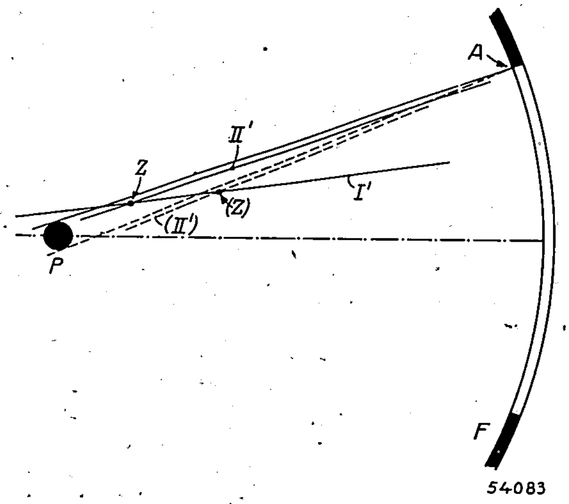


Fig. 12. For the strict requirement made above, viz., that the exit tube tip must not cause any obscuration of the specimen for points outside the film hole (rim A), in most practical cases the less strict requirement can be substituted that there should be no complete obscuration while a partial one could be tolerated. The former border line (II') dotted in this diagram may then be replaced by the full line II'. The intersection point (Z) will come closer to the specimen, at Z. The exit tube may, therefore, be longer. Similar considerations apply to the collimator tube.

obtainable by substituting this less rigorous condition for the rigorous one. Accordingly, the non-enveloped primary beam path with the third system of tubes (No. 55) has been reduced to 10.9 mm! The air scatter is extraordinarily weak with this tube system. The intensity drop of the diffraction lines as they approach the film holes is noticeable at angles smaller than 6° and greater than 172.5°.

For cases where high intensity is of prime importance, and a smaller resolution and stronger background are permissible, a similar tube system having a wider second collimating aperture has been designed (0.9 mm; No. 73). The non-enveloped beam length is 30.2 mm for these tubes.

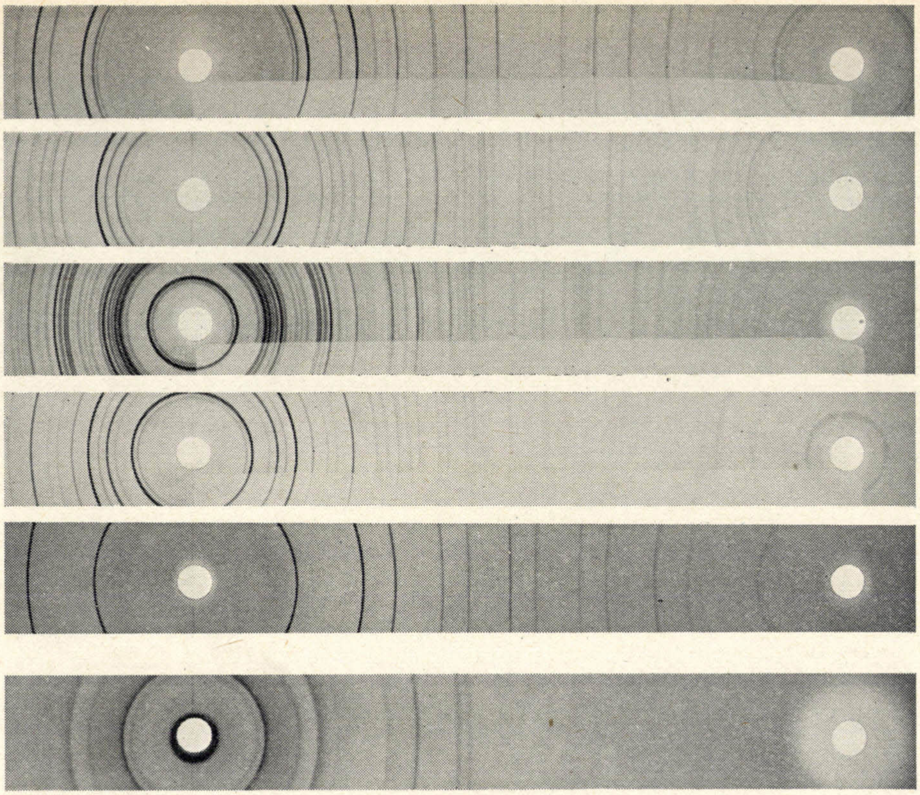
The same five equations as used above hold for the computation of the tubes with the less rigorous blind area condition, the only difference being that $+r$ has to be replaced by $-r$, as may easily be seen from fig. 12.

In the computation of collimator and exit tubes outlined above it was assumed that the X-ray beam has a circular cross-section. Its real shape, however, is oblong (fig. 5). In order to take in this oblong cross-section, a slit-shaped exit port evidently would be best suited. Nevertheless, for practical reasons the exit port, as well as the scatter cup, are made circular in all cases. This means that the exit port in the equatorial plane of the camera is about 3 times wider than is required for taking in the whole width of the primary beam. As a result, in order to prevent the occurrence of a blind region in the equatorial plane, the exit tube tip must be relatively a little farther away from the specimen, and the tube, therefore, has not the full theoretically feasible length. The difference is approximately compensated by the gain in length obtained by changing from the rigorous to the less rigorous blind area condition.

In *fig. 13* a number of patterns are reproduced which were recorded by the 114.6 mm camera, making use of the general purpose type No. 55 (films *a-e*) or of the forward-reflection type (film *f*) of anti-air-scatter tubes. The latter pattern has no blind area around the 0° film hole, the heavy broad diffraction ring in this region corresponding to a lattice plane spacing of ca. 15 Å, but has a large blind area around 180°. (The film of soap, fig. 7c, was also obtained with this type of collimator.) In the case of the former patterns (*a-e*), the very small partial blind area around both holes can be seen[10]). All the patterns are remarkable for their low background and their sharp and uniformly blackened lines. The exposure times for these patterns, taken with copper radiation at 45 kV_{peak}, 25 mA, were no longer than 1 to 2 hours, while a good pattern of quartz powder (*b*) could be obtained even with much shorter exposures (10 to 15 minutes).

Finally, it should be mentioned that for studies depending on differences in line shape, e.g., for the measurement of particle size by line broadening, or the examination of preferred orientations of crystallites by non-uniform line blackening, it is desirable to irradiate the specimen only along an axial length about equal to the diameter of the specimen. For this purpose collimators of a more conventional type having two circular pinholes are provided: with a first aperture of, e.g., 0.2 mm and a second one of 0.4 mm diameter, the beam

[10]) The blind area becomes visible by virtue of the faint residual background of the film which is caused mainly by white radiation diffracted by the specimen.



a Sodium chloride NaCl

b Quartz, SiO_2

c Dickite $Al_2O_3.2SiO_2.2H_2O$

d $(NH_4)H_2PO_4$

e Fluorite, CaF_2

f Montmorillonite, essentially: $Al_4(OH)_{12}Si_8O_{12}(OH)_8$

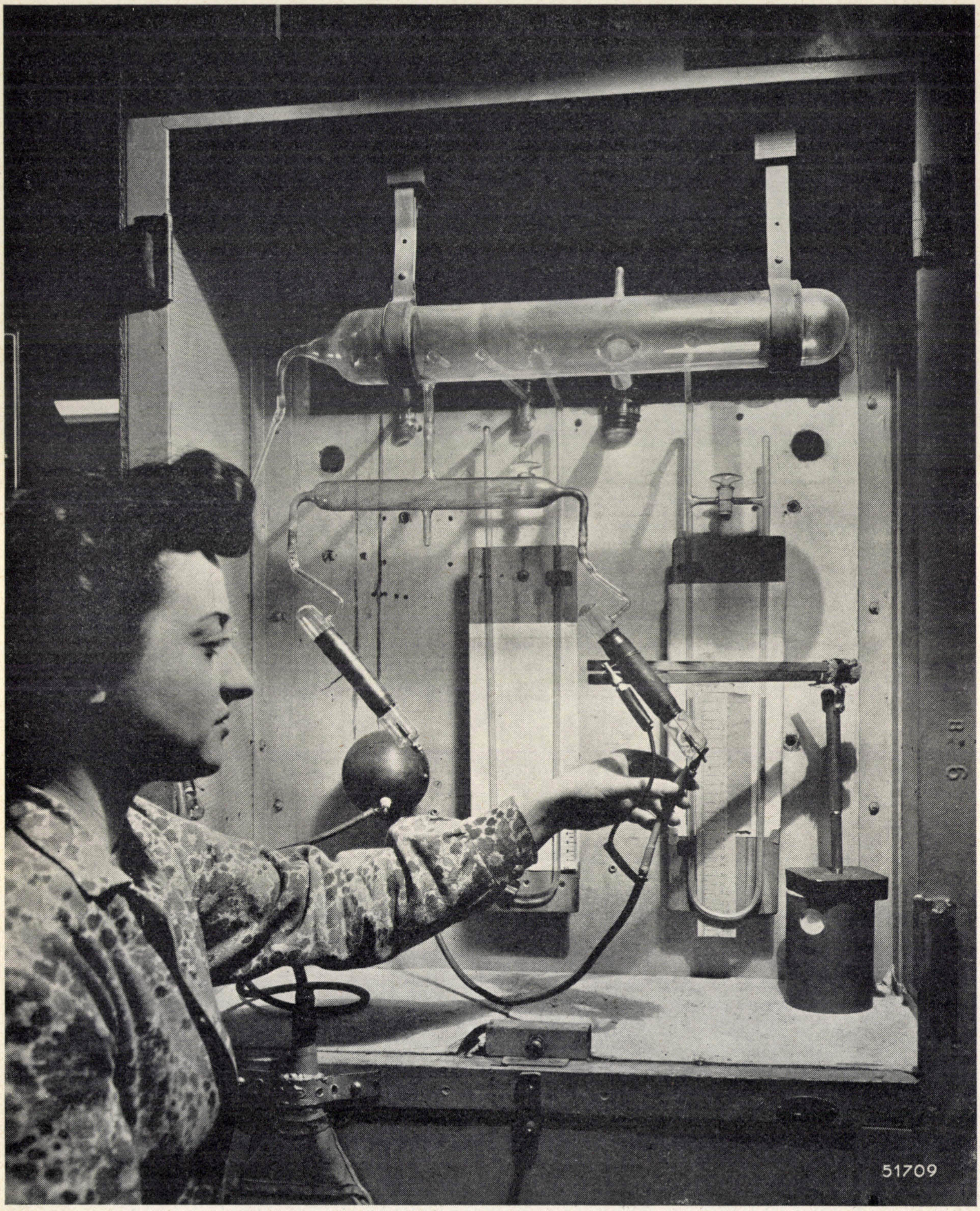
53826

Fig. 13. Six diffraction patterns showing performance of new Philips 114.6 mm camera. All taken with copper radiation at 45 kV_{peak}, 25 mA, with exposure times of 1 to 2 hours; *a-e* with general purpose type tube system No. 55 causing small blind areas around film holes, *f* with forward-reflection (only) type causing a large blind area around 180° film hole but no blind area near 0° hole. For films *e* and *f*, a 0.015 mm nickel filter was placed before the window of the X-ray tube, eliminating the undesired copper $K\beta$-radiation so that only the $K\alpha$-diffraction pattern is recorded. For *a*, *c* and *d*, a similar filter sheet covering the lower part of the film was used, so that the major portion of the film represents unfiltered radiation; this technique has the advantage of more rapid exposures combined with easy identification of the undesired $K\beta$-lines. Film *b* was taken with no filter at all. The pattern of fluorite shown in *e* was recorded with black paper being placed over the whole film to prevent the film from being blackened by visible fluorescence caused by the X-rays striking this compound (cf. footnote [4]). In some of the original patterns faint broad diffraction lines are discernible around the 0° film hole; these are caused by a slight devitrification of the Lindemann glass capillary used for mounting the specimen.

(of the 114.6 mm camera) has a width of 0.48 mm at the specimen. Of course intensity is low in this case; it is sacrificed to obtain optimum line shape.

High requirements are set for the precision of the machining in the manufacturing process of the collimator and exit tubes. It may be mentioned, for instance, that the inner diameter of the collimator tube tip must not deviate from the calculated dimensions by more than minus 0 and plus 5 microns. A similar precision is required for the tube fittings in order to insure the necessary exact alignment with the center of the camera. This requirement was made all the more difficult because it had to be combined with an easy interchangeability of the tubes of different types. A practicable solution was found in providing each tube with a precisely machined external surface fitting snugly into a receptacle in the camera wall, while the ultimate positioning on insertion results from matching of very accurately ground faces on the tube collar and the receptacle, these faces being normal to the beam axis.

EXHAUSTING GEIGER COUNTER TUBES



The photograph shows a phase in the manufacturing of Geiger counter tubes in one of Philips' American plants. These tubes, which are used for measuring X-ray or other radiation intensities, are filled with argon or other gases after the air has been exhausted. The gas filling process is checked by connecting the tube to a high voltage source and measuring the number of counts produced with a standard radio-active specimen. Gas ballast reservoirs and manometers for adjusting the pressure of the filling gas to the value desired are seen in the background.