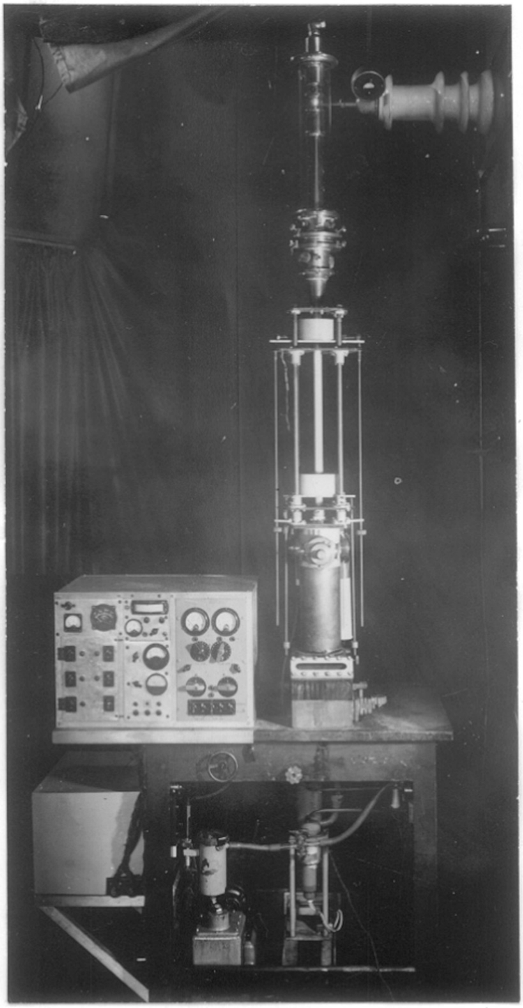
A New 200 KV. High Resolution Electron Diffraction Camera and its Uses in the Study of Surface Films.

By

H.C.LEWIS, B.Sc.

A Thesis Submitted for the Diploma of the Imperial College.

Applied Physical Chemistry Laboratories,
June, 1950.



General view of the Instrument.

CONTENTS

Page

GENERAL INTRODUCTION.

The early experiments of Thomson and Reid, and Davisson and Germer had shown that, in interacting with matter, electrons in motion were reflected as if they were guided by wave systems. There followed experiments by Thomson, Davisson and Germer, Kikuchi and Nishikawa which quantitatively established the validity of de Broglie's bold hypothesis. Since then, work in the field of electron diffraction has been mainly directed towards exploiting its possibilities as a means of studying the atomic structure of matter in surface layers in thin films. As a first result the slow speed electron technique originally used by Davisson and Germer is seldom employed, largely because it entails a laborious Faraday-cylinder scanning of the diffracted beams. On the other hand, Thomson's technique, in which electrons of about one third the velocity of light are used, thus enabling the directions of the diffractions to be recorded photographically, was soon established as being by far the most convenient technique. The research described in this thesis comprises the initial stage of an attempt to extend the applicability of this technique to surface structure problems by the use of higher-speed electrons and a higher degree of resolution than hitherto attained, and also to a further study of those electron diffraction phenomena of which a more exact knowledge is the pre-requisite of a closer understanding of fundamental principles. The instrument described is not therefore intended as a substitute for the Finch type camera in general use, but as an auxiliary to it.

The design of the various types of apparatus generally employed for electron diffraction before 1932 seems to have been influenced to a great extent by the older X-ray diffraction techniques. The considerable advantages that would be gained by devising methods of focussing and high-voltage regulation that were especially appropriate for electron diffraction were first noted by Finch (1933), who published details of a camera which was ingeniously designed to stabilise its own high-tension supply without the use of any electronic complications, and which was provided with a magnetic lens for focussing the beam and scanning the specimen. The extreme practicability of this instrument is evidenced by the fact that no substantial modifications in design have been found necessary in the course of the 17 years during which the several Finch cameras in use in this laboratory have been successfully applied to the study of a numerous variety of problems of surface structure, crystal growth, and electrodeposition.

Notwithstanding the striking simplicity of the apparatus, the quality of the photographs compares very favourably with that obtained with other types of electron diffraction camera, and there is therefore every reason to suppose that the design will be retained for all normal applications. On the other hand, the completion of some investigations has been deferred pending the modification of the apparatus to enable work to be carried out using electrons accelerated through 100-200 KV. as opposed to the 70 KV. which is found to be the practicable limit for the standard Finch camera. At the same time the development of the electron microscope suggested that use might be made of an electron optical system more elaborate than a single magnetic lens in order to improve the resolving power of the electron diffraction camera to a degree that would facilitate the detailed analysis of the various secondary effects in electron diffraction which are barely detectable with the simpler types of apparatus.

The present instrument is intended for use with any accelerating potential in the range 30,000 to 200,000 volts, and at the same time provision is made for an increase of resolution of an order of magnitude over that achieved by the standard equipment of this laboratory. The objective throughout has been to design an apparatus susceptible of stage-by-stage development followed by improvement in the light of experience gained in its use.

The decision to substitute a hot-cathode electron gun for the cold cathode discharge tube as a source of electrons drew attention to the scarcity of published work on the design of electron guns suitable for the illuminating system of the electron microscope and electron diffraction camera. The time therefore seemed opportune for a systematic study of this part of the subject, and a start has been made by the construction of an experimental gun equipped with mechanisms for making various internal adjustments during operation. It is not expected that this device will enable the multitude of design problems to be solved at once: the apparatus was built rather with a view to isolating at least some of the factors in order to determine where detailed attention to design was most necessary.

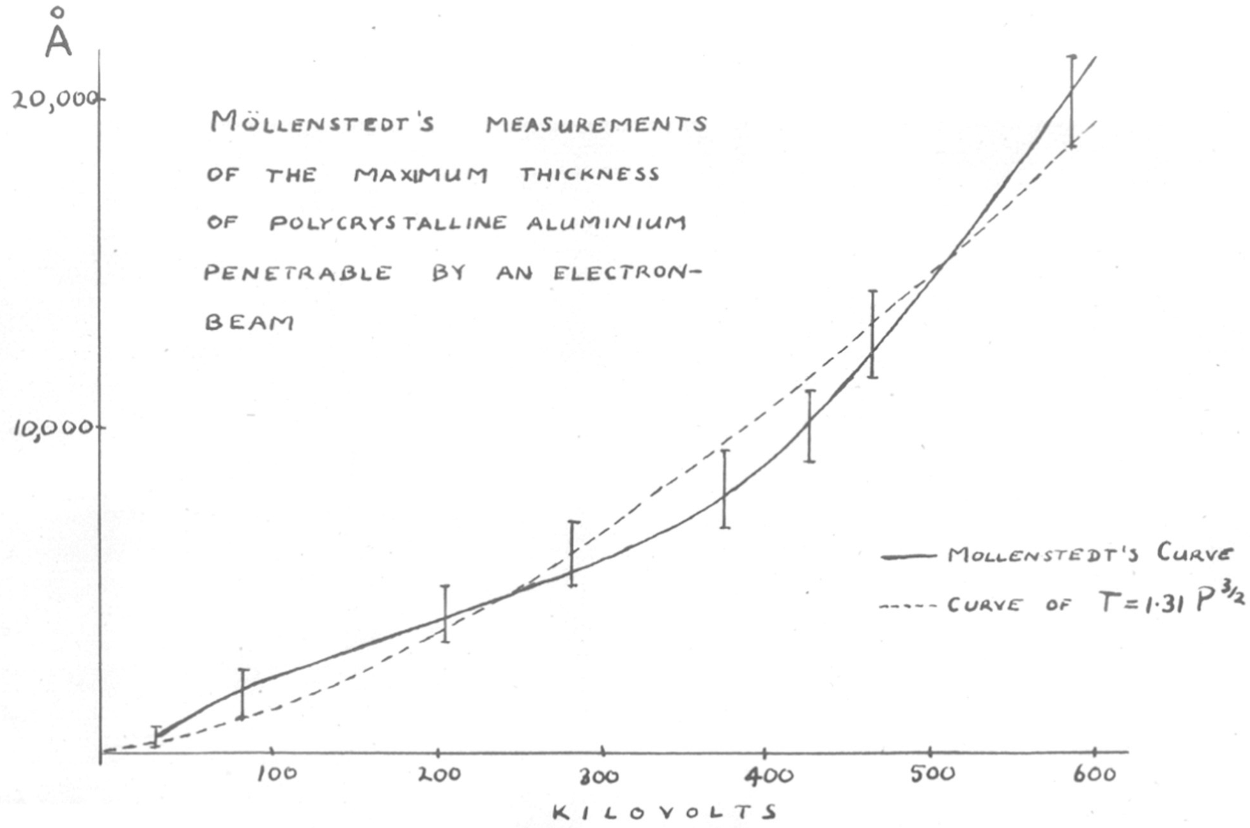

Å
20,000
10,000
MÖLLENSTEDT'S MEASUREMENTS
OF THE MAXIMUM THICKNESS
OF POLYCRYSTALLINE ALUMINIUM
PENETRABLE BY AN ELECTRON-
BEAM
MOLLENSTEDT'S CURVE
CURVE OF $T = 1{\cdot}31\, P^{3/2}$
100
200
300
400
500
600
KILOVOLTS

FIG. 1.

THE SPECIAL ADVANTAGES OF ACCELERATING POTENTIALS EXCEEDING 70 KILOVOLTS.

The special advantages which are gained by the use of accelerating potentials exceeding 70 KV. in electron diffraction arise for the most part in connection with diffraction by reflection from smooth surfaces: the transmission case has been investigated by Möllenstedt (1947) using electrons accelerated by fields approaching 600 KV., and his results indicate that the advantages here would be slight; we shall therefore deal only briefly with this case before passing on to a more complete discussion of the reflection of high speed electrons from smooth surfaces.

Möllenstedt's experiments were carried out with a view to determining the effect of higher accelerating potentials on the power of an electron beam to penetrate into solid matter. Fig. 1 is based on the results of these investigations; the ordinates represent the maximum thickness of a film of polycrystalline Al that would yield a discernible transmission pattern. The curve is not inconsistent with a 3/2 power law relating penetration to accelerating potential. It will be seen that an increase in transmission

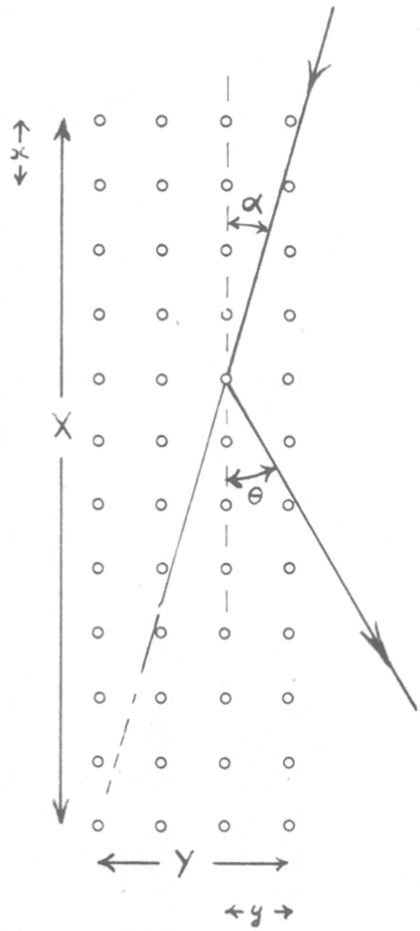


FIG. 2.

penetration of 50-100% can be expected when the accelerating potential is raised from 60 to 150-200 KV.

A further, and perhaps more important consequence of the higher electron speeds was that, for a given thickness of specimen, the ratio of the intensity of the diffraction pattern to that of the general background was appreciably increased. This advantage is liable to be offset at the very high voltages (~500 KV.) by the retraction of the pattern into the halation region of the central spot.

In the case of diffraction of electrons by reflection from a surface which is smooth to atomic dimensions, the depth of penetration of the beam below the surface is determined by several factors which we must now discuss in detail in order to analyse the effect of an increase in accelerating voltage.

Consider first the case of an electron beam incident on a portion of an atomically smooth crystal surface as depicted in cross-section in Fig.2. Here the atoms are spaced x apart along the direction nearly parallel to the incident beam, while X is the extent of the crystal effective for diffraction in this direction; y and Y have the corresponding significance for the direction in the plane of incidence normal to the surface. The beam is supposed incident at a small glancing angle α , and to emerge after diffraction at an angle θ to the crystal face. The intensity of the diffracted beam varies with direction according to an expression which contains the factors:

$$\frac{\sin^2 \frac{n\pi x}{\lambda}(\cos\alpha - \cos\theta)}{\sin^2 \frac{\pi x}{\lambda}(\cos\alpha - \cos\theta)} \cdot \frac{\sin^2 \frac{m\pi y}{\lambda}(\sin\alpha + \sin\theta)}{\sin^2 \frac{\pi y}{\lambda}(\sin\alpha + \sin\theta)} \qquad (1)$$

where n, m are the numbers of effective scattering centres in the X and Y directions respectively. The factor corresponding to the Z direction perpendicular to the diagram need not be considered here, since its effect is independent of angle of incidence and depth of penetration. The above expression is derived on the assumption that the contributions to the intensity from successive planes are equal, and although this assumption is not even approximately correct in respect of the planes parallel to the surface, the validity of the following discussion is not materially affected, since the discrepancy is in our favour.

The p^{th} order main maxima corresponding to these factors occur when

$$\cos\alpha - \cos\theta = \frac{p_x\lambda}{x}$$

and $$\sin\alpha + \sin\theta = \frac{p_y \lambda}{y}$$

and in view of the smallness of α and θ, these can be re-expressed as

$$\theta^2 - \alpha^2 = \frac{2 p_x \lambda}{x} \qquad \text{(circular ZONES)}$$

$$\alpha + \theta = \frac{p_y \lambda}{y} \qquad \text{(horizontal BANDS)}$$

Expressions for the angular width of these zones and bands can likewise be derived by considering the zeros of the factors in (1). Thus, taking $\sin\theta = \theta$, $\cos\theta = 1$, the angular <u>half</u>-widths are

$$\delta\theta_x = \left(\theta^2 + \frac{2\lambda}{nx}\right)^{1/2} - \theta \qquad \text{(ZONES)}$$

$$\delta\theta_y = \frac{\lambda}{my} \qquad \text{(BANDS)}$$

An exactly similar pair of equations determine the variation in α that can be tolerated at a given θ:

$$\delta\alpha_x = \left(\alpha^2 + \frac{2\lambda}{nx}\right)^{1/2} - \alpha$$

$$\delta\alpha_y = \frac{\lambda}{my}$$

Substitution of representative values shows that $\delta\alpha_x$ is several times greater than α for the first order diffraction, and $\delta\alpha_y$ is of the same order of magnitude for m = 1, 2, or 3 at accelerating potentials in the region of 70 KV., so that the Laue condition corresponding to the X-extension of the crystal is completely relaxed, and the second one is likewise relaxed in so far as it restricts α. If the latter effect is disregarded, and attention confined to the increased path length in matter associated with faster electrons, we arrive at the totally fallacious conclusion that a beam of fast electrons will penetrate further into the body of the crystal than a slow-electron beam. We shall now present the argument which suggests that the contrary is the case, and that the correct tool for examining essentially surface phenomena is a fast-electron beam.

Since the intensity of scattering by a single plane of atoms can be calculated in principle, in the case of irradiation by a normally incident beam, by a direct consideration of the Fresnel zones, the amplitude of a beam diffracted by glancing reflection at an angle θ increases like $1/\theta$. To see this we note that, for the obliquely reflected beam, the area of the first effective Fresnel zone, and therefore the number of atoms included in it, is increased by this factor. Now, the scattering power of each atom in the crystal is, <u>for a given order of diffraction</u>, independent of the de Broglie wave length and θ, since the relevant formula involves these quantities only as the quotient $\lambda/\sin\theta$ which remains constant for a given order. Hence the amplitude contributed by the first Fresnel zone is directly proportional to the number of atoms in the zone, and therefore increases as 1/θ. It follows that the intensity increases like $1/\theta^2$. It is clear that at the small values of θ (< 1°) with which we are dealing, the proportion of the incident beam which is diffracted by the first atomic plane increases very rapidly as θ is further reduced, so that the residue of the incident beam available for penetration to deeper atomic planes is correspondingly limited, while the depth to which the beam can penetrate at all can be controlled by reducing the angle of incidence.

Although the relaxation of the Laue conditions predicted above is realised in practice in respect of the broadening of the circular Laue zones, the tolerance in angle of incidence is less than would be expected: but it has been noticed that, as the accelerating potential is increased, smaller grazing angles can be used.

Increased electron speeds are thus associated with

1. A smaller possible grazing angle of incidence,
2. A smaller value of θ due to decreased λ
3. A greater proportionate contribution from the first atomic plane due to the smaller θ.

All these effects tend to limit penetration of the beam into the body of the crystal. An attempt at a theoretical estimate of the magnitudes involved based on a dynamical theory of diffraction would simply amount to an attempt to solve what is essentially a boundary-condition problem with unknown boundary conditions. On the other hand, there are good experimental reasons for anticipating that, with the smaller usable angles of incidence attainable with beams accelerated through 150 KV., definite evidence of monolayers of oxygen if present on single crystal surfaces will appear in the diffraction pattern. In support of this we note that Jones, Scott and Sillows (1949) succeeded in obtaining clear evidence of increased diffuseness introduced into a SiC single crystal pattern by a surface film of amorphous SiO_2 known to be only 3 A. thick. The experiments were carried out using an angle of incidence of 2°. If this angle were reduced to 1/4°, and the relative intensity of coherent scattering maintained by increasing the accelerating potential, the proportionate contribution from the surface layer would increase about 50-fold, while the penetration below the surface layer would decrease both in depth and in strength.

In measuring by electron diffraction the thickness of an amorphous layer deposited on a crystalline substrate, the general method is to obtain a reflection pattern and then rotate the specimen to decrease the angle of incidence so as to reduce α until the substrate pattern is obliterated. The effective thickness of the amorphous layer presented to the beam increases as $1/\alpha$ (θ is constant for a given order diffraction). A calibration experiment is then carried out using a layer of known thickness - e.g. a monolayer of stearic acid. The validity of this method obviously depends on the accelerating potential being sufficiently high to ensure that the value of α at which the diffraction pattern would be extinguished even in the absence of the deposited layer is substantially less than the value at which it is obliterated due to its presence. It is clear, therefore, that with faster electrons at our disposal, it should be possible not only to detect, but also to measure the thickness of thinner films.

The foregoing discussion indicates the considerations on which our expectation of the usefulness of higher accelerating voltages were mainly based. We mention in passing that no peculiar interest attaches to the reduction in wave-length from 0.045 Å. to 0.03 Å.: it is true that the resolving power, in so far as it depends on λ , increases, but the gain is so small, and the added difficulties are so great, that the advantage on this score can be regarded as trivial.

DESIGN CONSIDERATIONS AND THE CONSTRUCTION OF THE INSTRUMENT.

I. THE ELECTRON SOURCE.

The limitation of the working voltage of the Finch type electron diffraction camera arises from the use of a single acceleration cold cathode tube as electron source; the discharge becomes rough and uncontrollable at about 70 KV. This difficulty can be overcome in three ways: we can use a hot-cathode single-acceleration electron gun, or a multiple-acceleration hot cathode gun, or a multiple acceleration cold cathode discharge tube. The adoption of two-stage acceleration would necessitate the use of a rather elaborate voltage regulating device in conjunction with the high tension supply, and the additional complication did not seem justified for the particular range of voltage (30-200 KV.) in view. Nor was the use of a controlled air leak for regulating the first accelerating field considered feasible, as it introduced difficult insulation problems. It was decided to have recourse to single acceleration with a hot-cathode source.

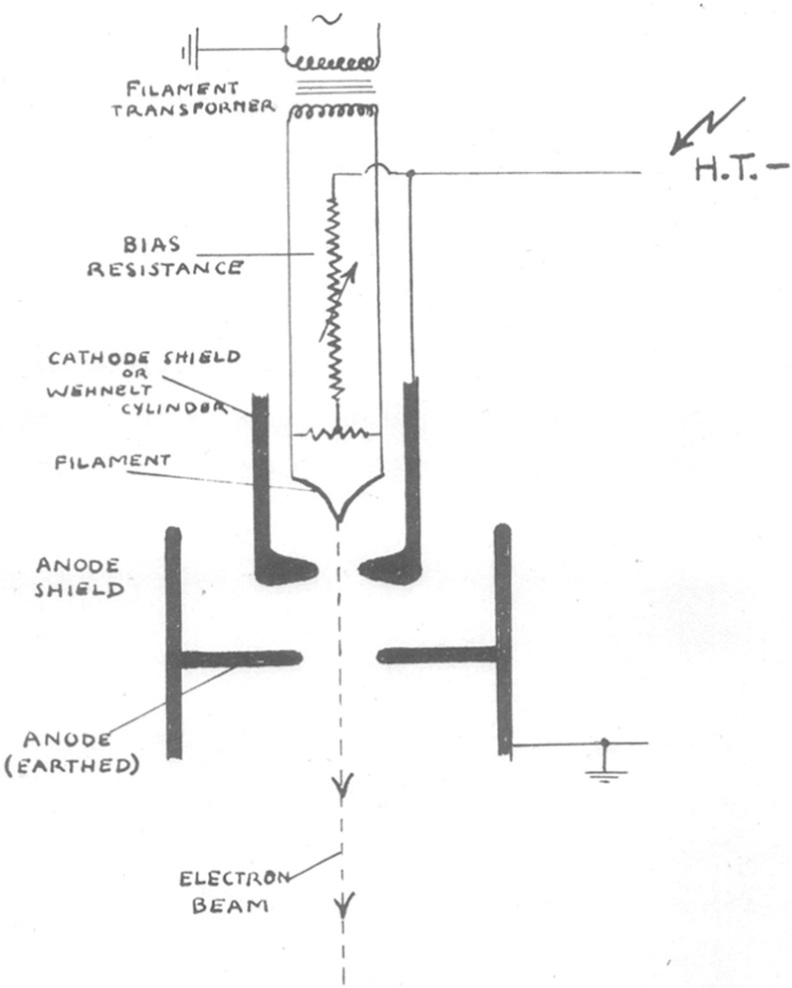


FIG. 3. ESSENTIALS OF SINGLE ACCELERATION ELECTRON GUN

1. Design Principles.

The essentials of the conventional three-electrode electron gun which has become familiar as the principal part of the illuminating system of the electron microscope are indicated in Fig.3. The cathode assembly comprises a filament mounted just behind the aperture of a Wehnelt cylinder. The anode is at earth potential and usually has a relatively large central orifice. The Wehnelt cylinder is maintained a few volts more negative than the filament. The three electrodes considered as a system can be regarded as an electrostatic electron lens, and an electron image of the source is formed by it at a position determined by the conditions under which the gun is operated. The actual shapes of the electrodes are at the disposal of the designer.

In view of the essentially experimental character of the design, the ceramic insulator into which the commercial type electron guns are usually built was discarded in favour of a more easily obtainable glass cylinder. Experience gained in this laboratory with an electron microscope tested up to 150 KV., and a preliminary "trial gun" (Fig.4) tested up to 180 KV. suggested that an 18-inch Pyrex cylinder would give adequate ~~operation~~ separation between the high-tension cap and the earthed base, provided the outside of the cylinder was kept

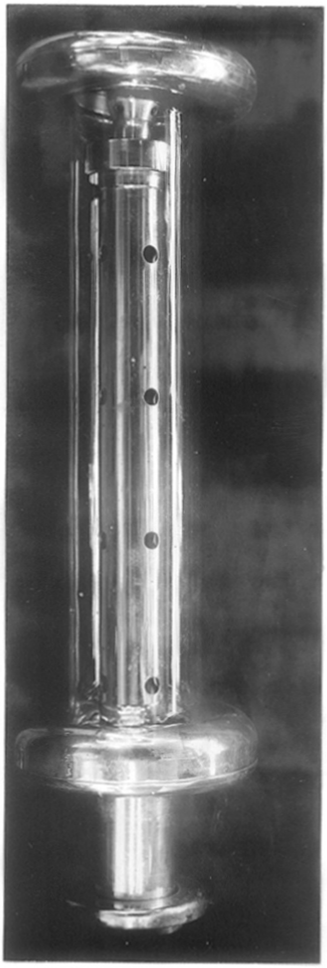


Fig.4. Preliminary Trial Gun.

quite clean.

The anode and cathode must be brought as close together as possible, otherwise the electron beam will tend to diverge unduly before it reaches the anode. The absolute minimum separation is set by the work-function of the metal used for plating the electrodes, but it is, of course, undesirable to approach this minimum, and in practice the separation usually adopted is about 1.5 cms. It is also usual to bring the sides of the anode above the level of the cathode in order to screen the glass as far as possible from contamination by evaporated tungsten. Haine (1947) mentions as an additional reason for this design factor the prevention of charging-up of the glass due to bombardment by secondary electrons from the anode, but this is only likely to occur when the anode-orifice is made extremely small, as is the case, for example, with the electron-gun of the type E.M.2 electron microscope, where the anode-aperture is intended to function as a beam-limiting diaphragm.

In principle, the anode-cathode gap can be positioned anywhere along the axis of the gun. The usual practice is to bring it close to the base, in order to put the earthed anode as far as possible out of reach of sparks which may

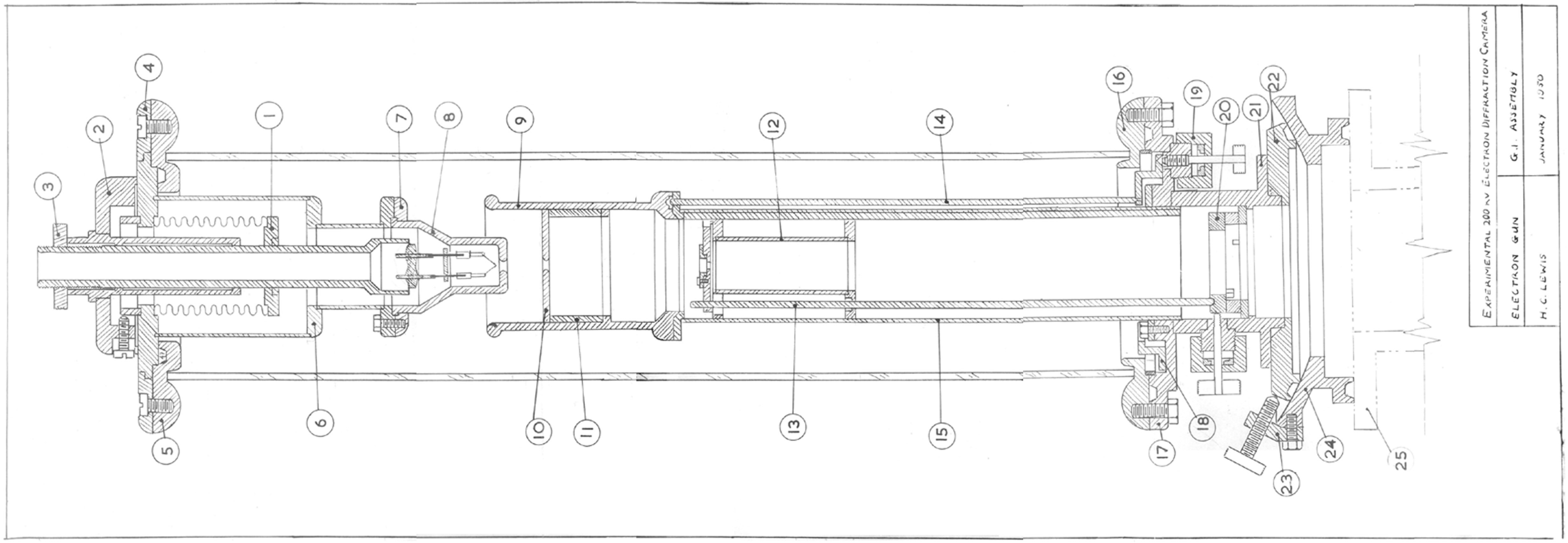


Fig.5.

Section through Electron Gun.

(The numbers refer to working drawings of components)

The vacuum glands were modified in the actual construction in accordance with the design shown in Fig.10.

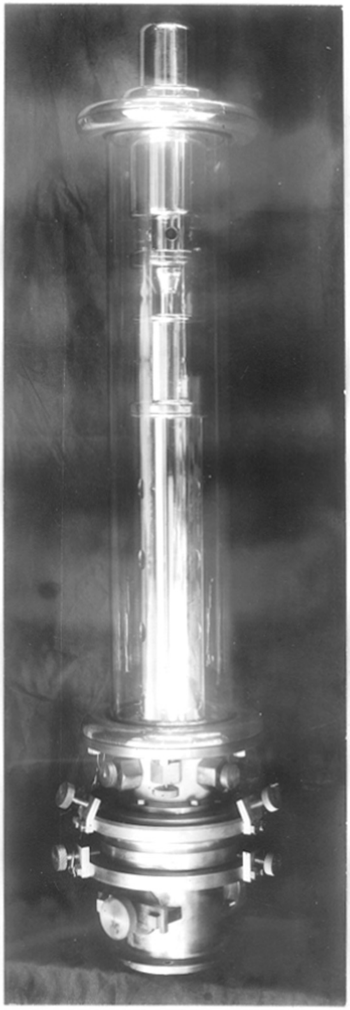


Fig.6. The Electron Gun.

TH-290

possibly track down the outside of the glass. The tests carried out with the preliminary gun in which the anode-cathode gap was very close to the top indicated that puncturing of the glass was only likely to occur as a result of very heavy sparking provoked by an accumulation of dust on the rim at the base; operated under these conditions, a puncture occurred at the level of the anode at 95 KV., but operated in a clean, dust-free condition, there was no tendency whatever to sparking even at 180 KV. Two advantages of bringing the anode-cathode gap as near as possible to the top of the gun are a gain in resolving power of the camera, and a small and compact cathode assembly that can be conveniently removed for filament renewal without the necessity of disturbing the whole gun.

ii. Design Details.

The twin-electrode glass-metal vacuum seal which carries the filament is mounted on a shaft which can be adjusted both laterally and vertically with respect to the Wehnelt cylinder. The former adjustment enables the sharply-bent tip of the 0.005 in-diameter tungsten wire filament to be centrally positioned behind the cathode aperture after it has been clamped in its supports. This obviates the

Fig.7. The Electron Gun.

Left. Anode assembly. Note adjustable top section of anode cylinder.

Right. The cathode assembly and glass envelope with corona cap removed to expose filament adjusting nut.

necessity of attempting to bend the filament wire itself into the correct position. The lowering or retraction of the filament within the Wehnelt cylinder, which is effected by means of a thumb-nut which can be operated while the gun is evacuated, affects the characteristics of the cathode lens in a way which will be discussed in a later section. The flexible vacuum joint necessary to achieve the adjustability is provided by a metal bellows. The end of the Wehnelt cylinder, which is the effective part of the cathode shield, is demountable to provide access to the filament, and is interchangeable with several variants. The whole cathode assembly is about 15 cms. long, so that the anode-cathode gap is about one-third of the way from the top of the gun. The main corona protection consists of the shaping of the external surfaces, no special corona shield being necessary apart from the small cap which covers the protruding end of the filament shaft and adjusting nut.

The anode is mounted in a 5-cm. diameter cylinder, the top section of which can be tilted slightly while the gun is in operation. The object of this adjustment is to enable the effect of slight assymetries in the accelerating field to be investigated. Alternatively, this tilting mechanism can be used to raise or lower the anode. The anode itself consists of one of five interchangeable variants; four of these are flat metal discs with variously sized central orifices, while the fifth is hemispherical (convex upwards) and can carry a diaphragm in the central bore. The anodes can be mounted at any predetermined height in the adjustable section of the anode-cylinder. There is also provision for a diaphragm to be mounted at any pre-set height in the fixed section of this cylinder. This diaphragm can be adjusted laterally for alignment purposes while the gun is in operation.

The adjustments of the anode and diaphragm are effected by mechanisms within the gun operated by six control rods brought out through specially designed vacuum glands (Fig.10). It was considered that these controls could later be adapted to operate other adjustable components that might usefully be introduced into the gun e.g. adjustable metal cylinders which would enable space-charge effects to be studied.

The unusual length of the gun and its intended use in conjunction with a narrow-bore focussing tube necessitates the provision of both lateral and tilt adjustments with respect to the camera. An interesting point arises in

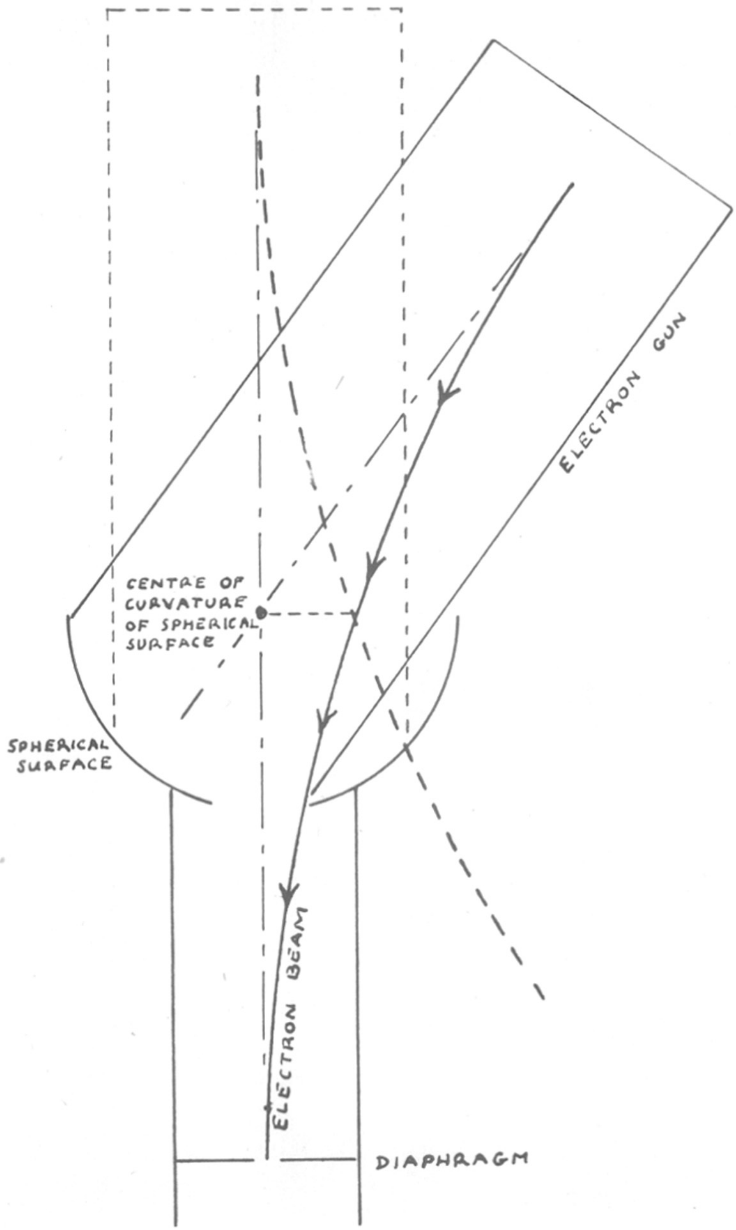


FIG. 8. LOCATION OF CENTRE OF CURVATURE OF SPHERICAL SURFACE

connection with the choice of centre of curvature of the spherical surface which must be incorporated. Preliminary experiments with the trial-gun indicated that deflection of the electron beam from the axis is due almost entirely to the earth's magnetic field, so that the path of the electrons is an arc of a circle. Reference to Fig.8 will make it clear that the beam can be made to pass vertically through any selected point (a diaphragm) on the axis of the camera by using the tilt adjustment only, provided the centre of rotation is mid-way between the diaphragm and the cathode. With this arrangement only slight lateral adjustment is necessary.

A departure is made from the practice of using metal bellows to provide the flexible vacuum joint in the aligning head: instead (Fig.9) an O-section neoprene gasket housed in a channel in each sliding member moves on a greased metal surface, the channel being not quite deep enough to allow metal-to-metal contact. We have found this extremely simple type of sliding vacuum joint completely trouble-free and very smooth and light in operation.

iii. Constructional Details.

The requirement that the components of the gun should

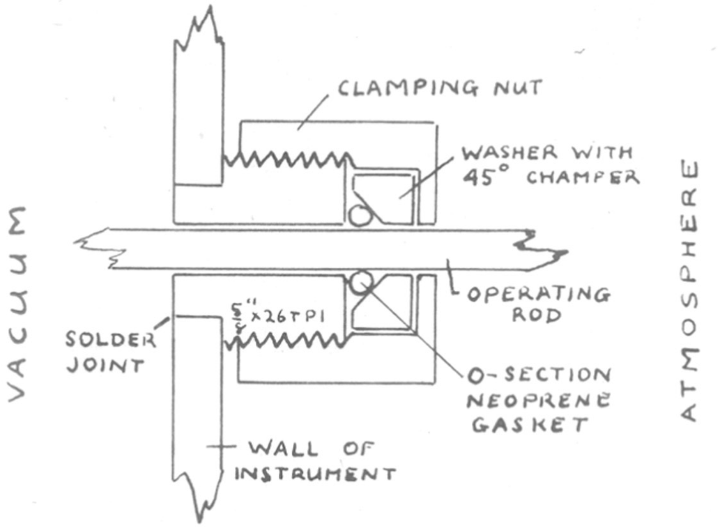


FIG. 10. DETAILS OF VACUUM GLANDS FOR OPERATING RODS
SCALE : 2 FULL SIZE

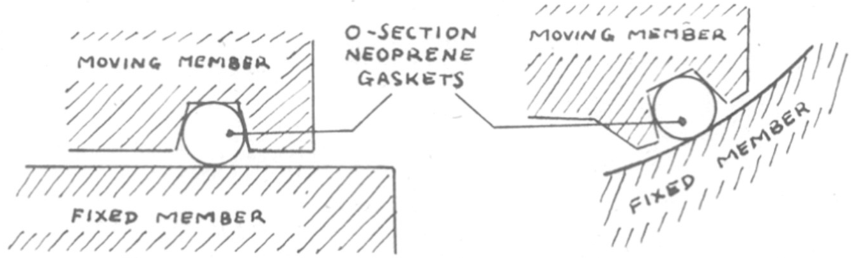


FIG. 9. SLIDING VACUUM JOINTS AS USED IN
ALIGNING HEAD

be entirely non-magnetic leads to the choice of brass as the most convenient material for the metal parts. The smooth working of the instrument depends to a large extent on a high polish being maintained on those metal surfaces which are exposed to electrostatic stress, and for this reason it is desirable to electroplate such surfaces. A suitable non-magnetic plating material is rhodium, and 0.00003 in. of this metal on an underlayer of 0.0005 in. of silver is very satisfactory: the cathode shield, its clamping ring, the top section of the anode cylinder, and the anodes were all heavily plated with platinum, 0.00025 in. being deposited on 0.0005 in. of silver. The reason for the use of platinum on these parts is that it has a work-function about three times as great as that of the usual plating metals.

Most of the joints were made with silver-solder which is stronger, cleaner, and generally easier to use than soft-solder; but where parts could not be machined after soldering, soft solder was used to avoid distortion of the brass through excessive heating. The ends of the Pyrex cylinder were lathe ground so as to be accurately parallel and perpendicular to the ~~main~~ mean axis; the metal flanges into which the anode and cathode assemblies are spigoted were then sealed into position with Apiezon W wax.

The demountable vacuum joints are made by O-section neoprene gaskets housed in channels cut to such dimensions as to ensure metal-to-metal contact of the mating members. The vacuum glands through which the operating rods pass are of original design as shown in Fig.10. We have found this type of seal completely reliable and much lighter in operation than the ordinary Wilson seal.

II. THE ELECTRON OPTICAL SYSTEM AND RESOLVING POWER.

The term resolving power, when used in connection with electron diffraction cannot be usefully defined so as to provide a direct measure of the performance of any particular diffraction apparatus. If the term is to signify the natural analogue of what is meant by resolving power in physical optics we must define it as, "the minimum detectable increment in lattice spacing expressed as a fraction of lattice spacing" or, what amounts to the same thing, "the minimum detectable increment in ring diameter expressed as a fraction of ring diameter", for

$$\frac{\delta d_{hkl}}{d_{hkl}} = -\frac{\delta D}{D} \qquad (1)$$

where D is the diameter of the ring corresponding to the lattice spacing d_{hkl}. By applying the usual criteria of resolvability to the case of a diffraction pattern, the R.H. member of the above relation transforms to the following formula for resolving power:

$$1{\cdot}73\,\frac{d_{hkl}}{\lambda}\cdot\frac{S_0}{2l}$$

where λ is the de Broglie wave length of the electron beam, S_0 is the diameter of the spot, and l the camera length, i.e. the distance from the specimen to the photographic plate.

The factor 1.73 arises as a consequence of assuming that the radial intensity distribution in a ring is such as would be obtained by uniformly distributing circular spots around its circumference. In order to consider the resolving power of the instrument in isolation from any particular application, the formula is derived with the additional assumptions that the natural ring width is zero, and that the beam convergence is very small. Of the three disposable quantities, λ , l, S_0, the first two have to be adjusted to the requirements of the experimenter, so that the problem of improving the resolving power becomes one of reducing the spot-diameter S_0.

The size and quality of the spot ultimately produced on the screen is obviously primarily dependent on the size and quality of the cathode image formed in the electron gun. This image acts as the source for the electron optical system. Since, however, it is the arrangement of diaphragms and lenses comprising this electron optical system that provides the means of studying the formation of the cathode image, we must discuss this arrangement first, assuming for our present purpose that a usable cathode image has been formed.

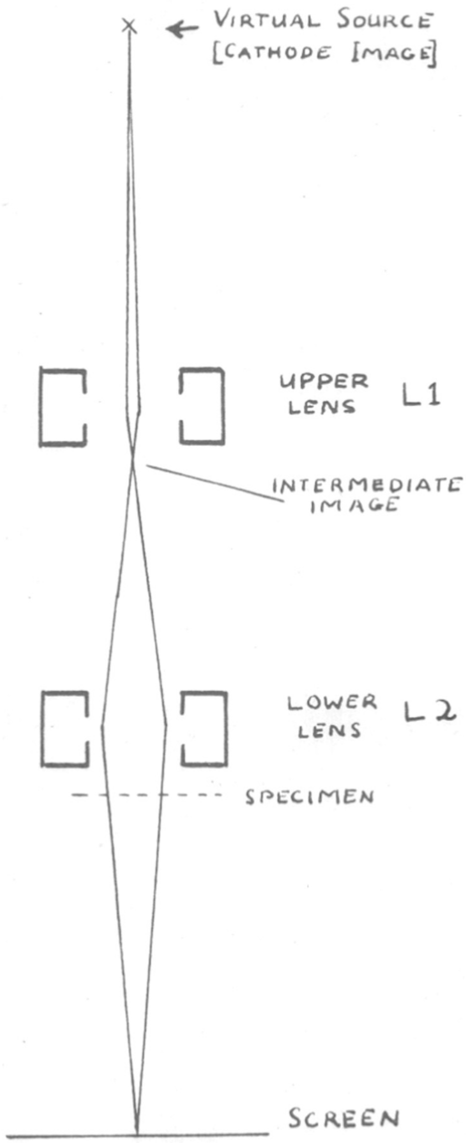


FIG. 11. PRINCIPLE OF TWO-LENS SYSTEM

The principle of the electron optical system by which a reduced image of the virtual source is focussed onto the screen is illustrated in Fig.11. A reduced intermediate image of the cathode-image is formed by the powerful magnetic lens L1, and the final image focussed onto the screen by the weak lens L2. It will be noticed that the placing of both lenses above the specimen preserves the main advantage of the Finch type focussing system in that the possibility of introducing distortion into the diffraction pattern is eliminated.

i. <u>The Design of the Electron Optical System.</u>

In the Finch type camera the focussing is brought about by means of a simple short solenoid in which no magnetic material is used to concentrate the field. On increasing the current in such a lens the focal length decreases to a minimum which is comparable with the dimensions of the coil itself, that is, to about 15 cms. A shorter focal length is never required, since the function of the single lens is simply to produce a real image of the effective source on the screen about 50 cms. away. But for the more powerful lens which is necessary in the two-stage focussing system, recourse must be had to the use of iron or steel shielding to concentrate the magnetic field: the focal length then

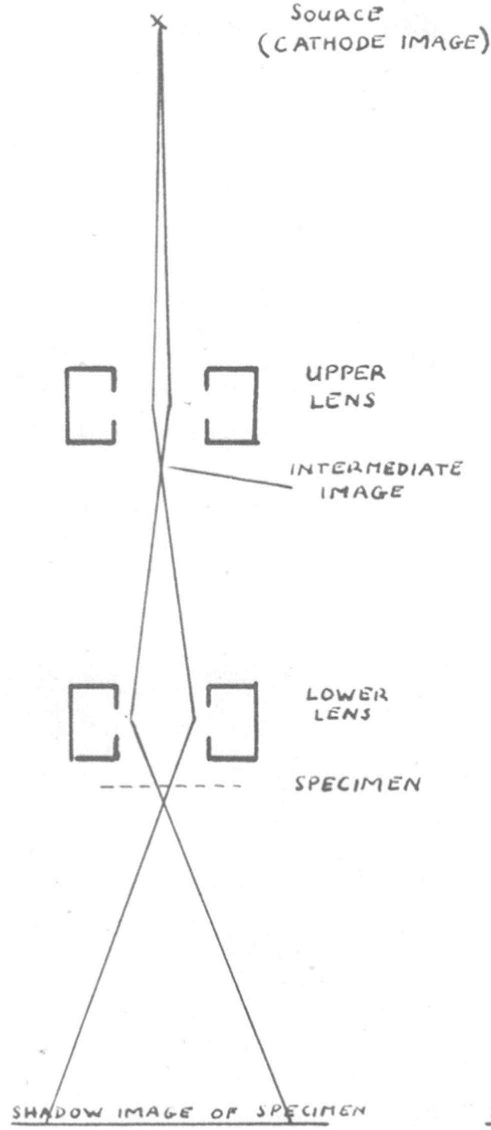


FIG. 12. APPLICATION OF LENS SYSTEM TO SHADOW MICROSCOPY.

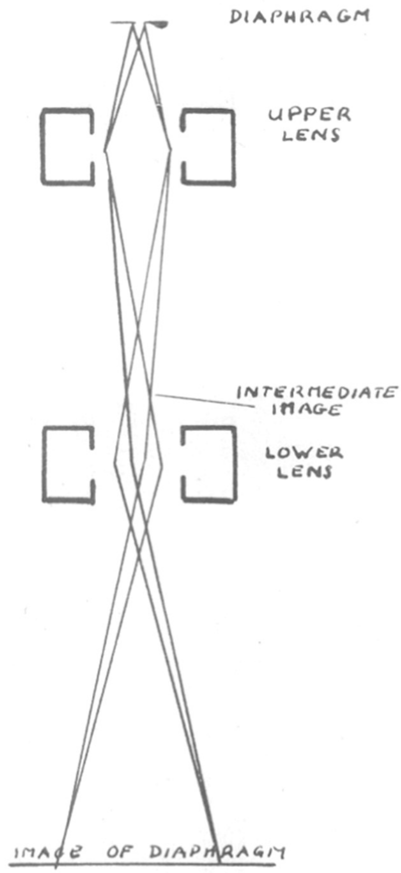


FIG. 13. USE OF LENS SYSTEM TO OBTAIN MICROGRAPH OF DIAPHRAGM.

becomes reducible to a value of the same order as the central bore of the lens. Hence it is immediately evident that a departure in design from the wide-bore focussing tube hitherto incorportated in the Finch type camera is an essential pre-requisite of an increase in resolving power by electron optical means. It is, of course, only the upper lens that must be a powerful one if the instrument is intended for use exclusively as a diffraction camera. On the other hand, if provision is made for increasing the power of the second lens until the beam is brought to focus very close to the specimen plane, there is a possibility of using the instrument as a shadow microscope. A still greater advantage of adopting the iron-shielded type of coil for the second lens is that the two-lens system then becomes a very flexible electron optical device for examining the quality of the cathode image, so that the determination of the optimum operating conditions for the electron gun is greatly facilitated. In addition, magnified electron images of the diaphragms in situ can be obtained, and this is a valuable asset when high resolution is being sought.

The considerations outlined above led to the decision to make the two lenses identical in design and with a minimum focal length of two cms., but to provide for the easy

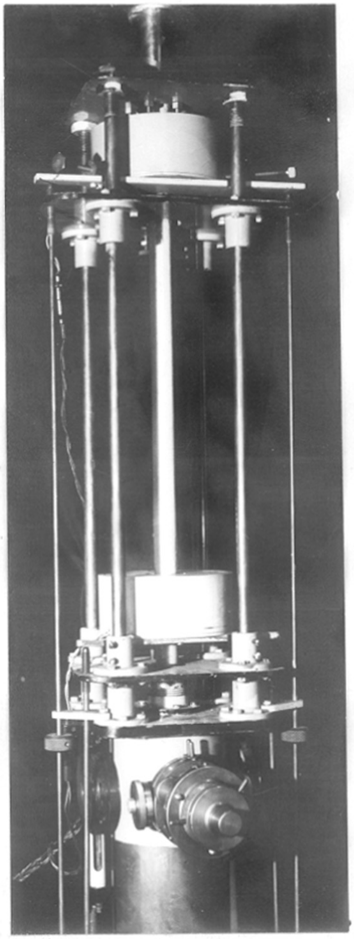


Fig.14. The Focussing Section.

substitution of a more powerful upper lens with a focal length of 4 mm. as soon as sufficient experience with the two-lens system had been gained. The former arrangement would give a reduction in spot diameter of about 1/20, and the latter of about 1/100.

The substitution of a narrow-bore focussing tube for the 2-1/4 in. diameter tube in current use entailed a radical alteration of the general design of the upper section of the instrument. Fig.14 shows the arrangement when the 2 cm. focal length upper lens is in use. The lenses are carried on 1/4 in. brass plates attached to the four main supporting columns by heavy bushes, thus ensuring the mechanical rigidity of the whole framework which serves to support the electron gun. The focussing tube itself has a 7/8 in. diameter bore and is 26 in. long. It is easily demountable to allow interchange with the alternative tube for use in conjunction with the 4 mm. upper lens.

ii. <u>The Electron Lenses.</u>

It is well known that the electron optical characteristics of an iron-shielded lens are practically independent of the shape of the pole-pieces, provided that the iron is not saturated. The focal length for a given

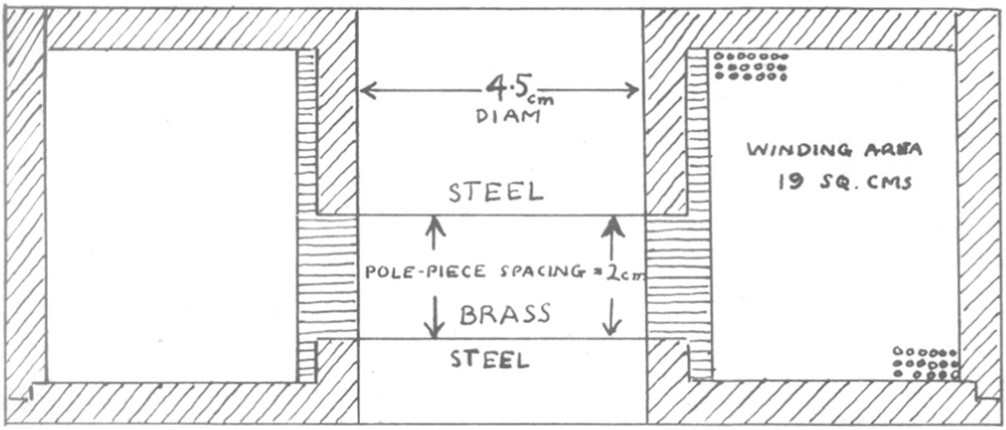


FIG. 15. SECTION THROUGH LENS

SCALE : FULL SIZE

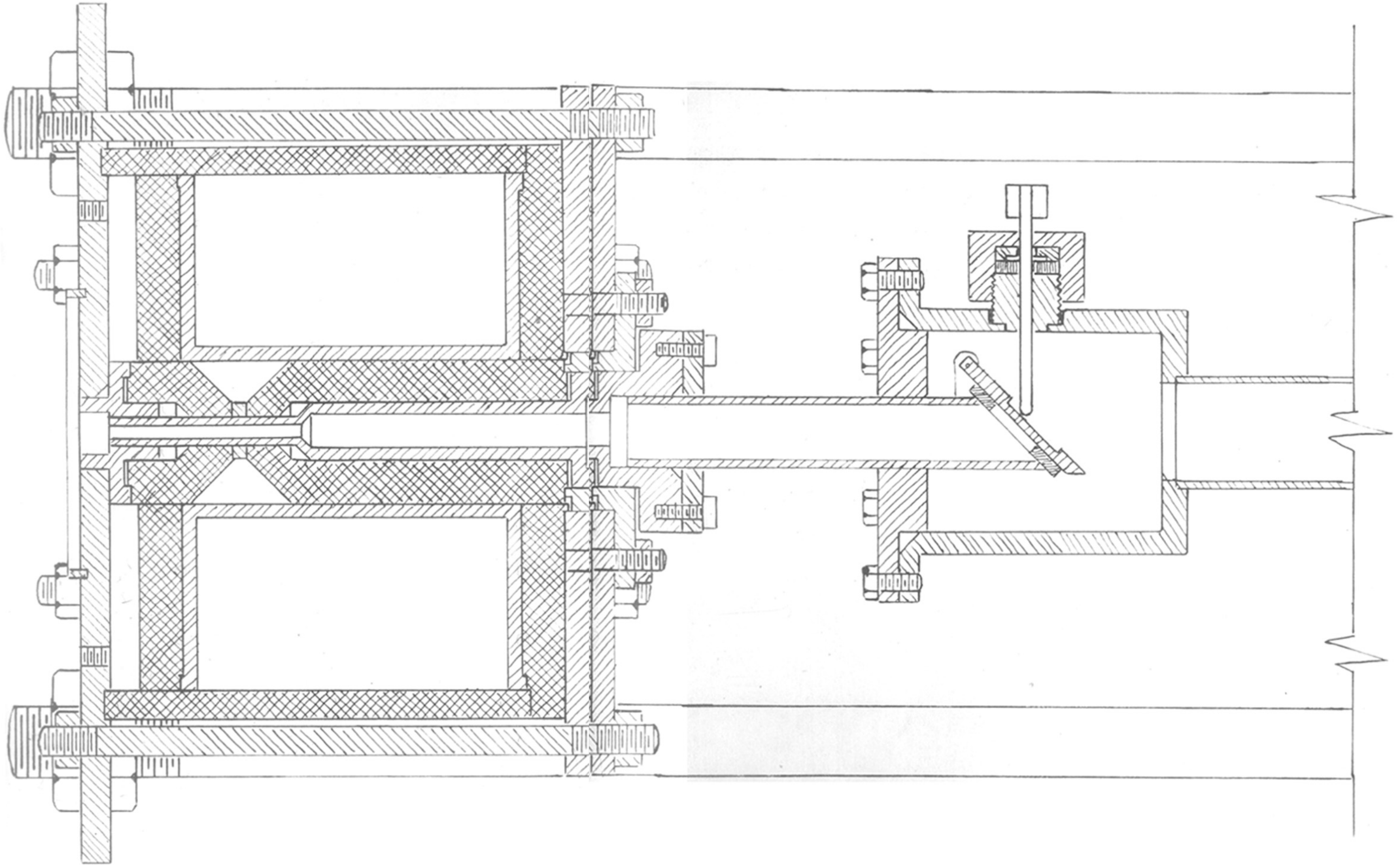


Fig.16.

Section through focussing system, showing short focus upper lens and flap valve. The soft-iron pole-pieces and mild-steel coil casing are shown in crossed shading; the remainder of the metal parts are of brass.

electron wave-length depends almost entirely on the pole-piece spacing b, the pole-piece diameter d, and the number of ampere-turns IN providing the exciting field. There is thus an infinite number of ways in which a lens can be made with a specified focal length, and it was by combining considerations of feasible dimensions, heat dissipation, achievable stabilised currents, convenience in construction, availability of material, and so on, in the light of general experience and the predictions of theory that the final design was produced. The whole process does not lend itself to brief explanation, but the following discussion represents the general line of reasoning employed in determining the design of the 2 cm. lens shown in Fig.15 and the 4 mm. lens in Fig.16. A more exact calculation of lens constants would be extremely involved and of no particular value.

Von Ments and le Poole (1947) found that the focal length of an iron-shielded electron lens can be estimated to within 10% by means of the following formula :

$$f = 3.6\,(b^2 + d^2)^{\frac{1}{2}}\,(10/K + 0.04)$$

where b is the spacing between the pole-pieces and d the diameter of the bore, both in cms., while K is the <u>excitation quantity</u> which is defined by

$$K = (IN)^2/V$$

where IN is the number of ampere-turns providing the exciting field and V is the accelerating voltage of the electron beam. Now the maximum current that could be conveniently stabilised with the resources at our disposal was 300 mA., and the maximum resistance load which could be tolerated was 800 ohms. The most convenient value of $(b^2 + d^2)^{\frac{1}{2}}$ from the point of view of mechanical design was about 5 cms. Substituting this in the formula for f, neglecting the 0.04 in the bracket, and setting f = 2 cms. gives K = 90. At 100 KV. this makes IN = 3,000, so that N = 10,000 turns. The available wire with the necessary current carrying capacity was 29 S.W.G. enamelled copper, which winds at just over 5,000 turns per square inch. Allowing 50% extra for insulation, a total winding area of 3 sq.ins. is required, and this could be most conveniently provided by the dimensions indicated in the figure. The wall thickness of 1/4 in. was expected, in view of general experience to be quite adequate to avoid saturation of the iron.

In the construction of each lens the top and bottom sections were machined out of solid mild steel blanks and soldered to the brass spacer to provide the coil former, which was then insulated with three thicknesses of Empire cloth before the 10,000 turns of wire were wound on by means of an improvised coil-winder attached to a lathe. Every third layer was separated by a strip of good quality tracing paper to add rigidity to the windings. The outer wall of the lens was made from a length of 6-in. steel seam tubing skimmed down to give a "push-fit" on the former.

In the case of the alternative (4mm.) upper lens, the main lens body is first clamped rigidly to the top plate of the focussing section. The pole-piece unit is then pushed up into the central bore and clamped, the clearances being arranged so that the vacuum sealing gasket is compressed by the correct amount when the pole-pieces are in position. The pole-piece unit itself is built onto a brass liner in such a way that no brass-iron soldered joint is involved: a point of advantage, since we have found that such joints are liable to develop leaks in course of time. This construction also enables the process of assembly to be carried out without re-heating or straining the pole-pieces after they have been heat-treated. The focussing tube is free to slide in the vacuum seal which joins it to the pole-piece sleeve, thus avoiding mechanical over-rigidity of the whole structure. The small inverted flap-valve which is incorporated enables the vacuum to be maintained in the gun while air is admitted to the camera body. The gun is continuously evacuated by its own oil-pump.

The calculations for design purposes are practically identical with those applied to the 2 cm. lens, except, of course, that $(b^2 + d^2)^{\frac{1}{2}}$ has a different value. The minimum focal length is actually about .35 cm. The coil has 10,000 turns. The pole-pieces are of pure soft iron, the rest of the magnetic circuit being of the softest mild steel available. The walls are made thicker than is strictly necessary so as to minimise flux leakage at joints. The iron and steel parts are turned from solid.

iii. Magnetic Screening Requirements.

The possibility of scattering of the electron beam from the walls of the instrument must be considered here. Owing to the small apertures in the beam-limiting diaphragms, such scattering cannot occur on account of the beam-divergence alone, but the curvature of the beam due to the effect of the earth's magnetic field is great enough to cause it to strike the walls of the focussing tube unless this is suitably screened. A calculation, based on classical theory, of the radius of curvature of the path followed by the electron beam gives

$$R = \frac{1}{H}\sqrt{\frac{2mV}{e}}$$

where H is the horizontal component of the earth's magnetic field, e/m is the specific charge of the electron, and V the accelerating potential. Taking H = 0.18 oersted, e/m = 1.77×10^7 e.m.u., V = 100 KV., and noting that 1 KV. = 10^{11} e.m.u., we find

$$R_{100KV.} = 1,700 \text{ cms.}$$

(neglecting the relativity correction which is small and in any case in our favour). After travelling a distance ℓ cms. the displacement of the beam from its initial path is therefore very nearly

$$\ell^2 / 3400 \text{ cms.}$$

The effective length of the focussing tube is 76 cms., and if we ~~supply~~ apply the above formula to this while making allowance for the screening effect of the lenses over two sections of the path, we find that the beam displacement at the lower end of the focussing tube is 1.53 cms., while the diameter of the bore is 2.22 cms., so that a 100 KV. beam could only traverse the tube if the gun were adjusted so that it entered obliquely. For this reason it was essential to screen the section of the focussing tube between the lenses

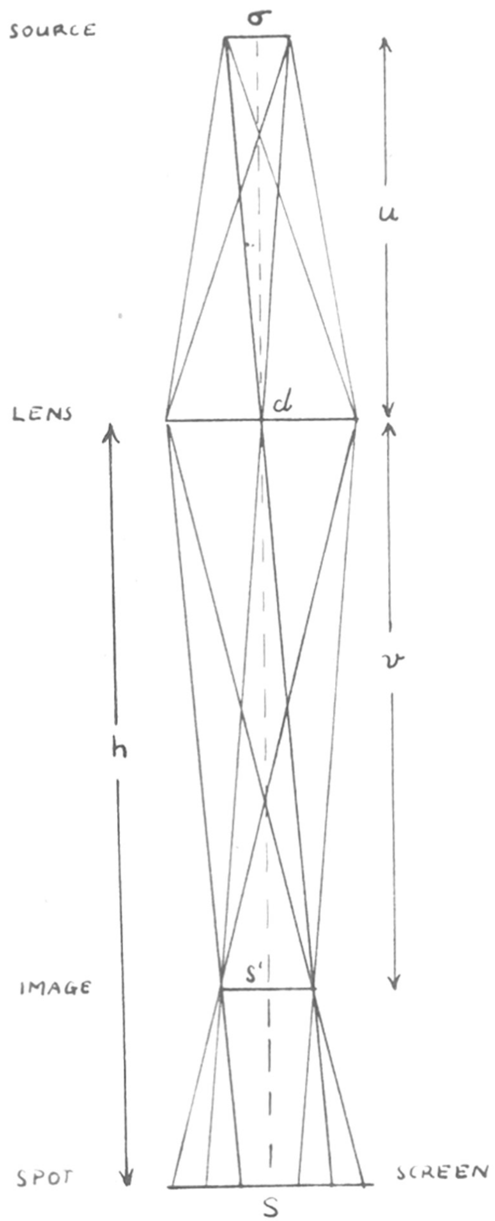


FIG. 17 EFFECT OF DEFOCUSSING

with Permalloy C. The deviation of the 100 KV. beam is reduced to 0.725 cms. by this screening.

iv. Stability Requirements.

In order to determine the degree of stabilisation necessary for the high tension supply and current supply for the lens coils, we must first investigate the way in which the spot size will be affected by fluctuations in focal length of the lenses, that is, we must derive a relation between the proportionate variation in spot size and proportionate variation in focal length. It will be sufficiently accurate for our present purpose if we apply the geometry of light-optics for the case of a thin lens.

Fig.17 represents the final lens slightly defocussed so that the image S' of the virtual source σ is formed at a distance v from the lens instead of on the plate at a distance h from the lens. If f is the focal length under the conditions depicted, f_o the correct focal length to bring the spot into focus on the plate, d the cross-sectional diameter of the beam at the lens, and u the distance of the virtual source from the lens, we seek an expression for $(S - S_o)/S_o$ in terms of $(f - f_o)/f_o$, where S, S_o are the diameters of the defocussed and focussed spots respectively.

From the geometry of the figure $$\frac{S'}{S - d\,\frac{h-v}{v}} = \frac{v}{h}$$

From optical considerations $$\frac{h}{v} = \frac{S_o}{S'}$$

Combining these we have $$S_o = S - d\left(\frac{S_o}{S'} - 1\right)$$

which gives $$\frac{S - S_o}{S_o} = d\left(\frac{1}{S_o} - \frac{1}{S'}\right) \quad \ldots (1)$$

But $$\frac{\sigma}{S_o} = \frac{u}{h} \text{ and } \frac{\sigma}{S'} = \frac{u}{v}$$

therefore $$\frac{1}{S_o} - \frac{1}{S'} = \frac{u}{\sigma}\left(\frac{1}{h} - \frac{1}{v}\right)$$

$$= \frac{u}{\sigma}\left(\frac{1}{f_o} - \frac{1}{f}\right)$$

so that (1) becomes $$\frac{S - S_o}{S_o} = \frac{f - f_o}{f_o} \cdot \frac{u}{f} \cdot \frac{d}{\sigma}$$

Now in our instrument, u is of the same order as f, and d/σ, while varying widely according to the operating conditions, would be unlikely to approach 1,000 even with the stronger upper lens in use, so that at the worst it would need a fluctuation of as much as 0.1% to double the spot diameter. There is, however, a second way in which the fluctuations in lens power can affect the resolving power, for owing to the non-coincidence of the geometric and electron optical axes of the lenses, when the system is aligned to give an anastigmatic spot, the spot may be deflected from its natural position by as much as a centimetre. This deflection, it is reasonable to assume, will be roughly proportional to the lens power so that focal length fluctuations may have a serious effect unless they are always less than one part in 10,000. Coil supply stability of about 1/20,000 should therefore be aimed at.

It will be seen from the foregoing that stable focussing is much more important than accurate focussing: a fortunate result, for the former is easy to achieve, whereas the latter is not. The lens power, of course, depends directly on the accelerating voltage as well as on the square of the coil current, and it would appear therefore, that the same order of stabilisation is necessary in each case: but we must also take into account that the high tension voltage enters as a factor determining the ring diameter of the diffraction pattern. The de Broglie wave length, and therefore the ring size, depends on the square root of the voltage. A ring radius of 5 cms. therefore justifies voltage stabilisation of 1/25,000 to reach the limit of

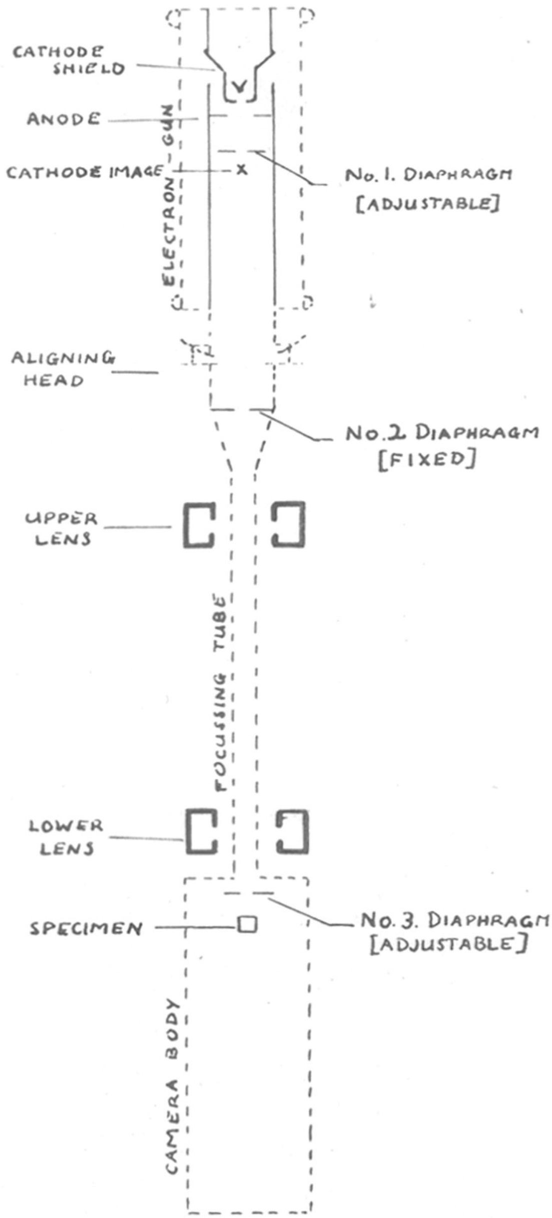


FIG. 18. POSITION OF DIAPHRAGMS

resolution associated with a spot of 0.0001 cm. diameter No direct determination of the stability of our high tension set has yet been carried out, but as this set has been used to supply an electron microscope ~~it~~ that would be sensitive to fluctuations of the above order, we believe that the degree of smoothing is quite adequate for our purposes.

V. The Formation of the Cathode Image.

The electron gun itself, as has already been pointed out, can be regarded as an electrostatic lens which will produce a virtual image of the emitting area of the filament at a point on the axis determined by the conditions under which the gun is operated. Some of the factors influencing the size, quality and position of this image have been studied by Hillier and Baker (1945). Other published comments are mostly by way of passing reference and frequently misleading.

In order to determine the exact position of the cathode image a diaphragm having a 0.015 cm. aperture is set at position 2 (Fig.18). This forms an ideal pinhole camera for the electron source, so that if the gun is moved laterally by a known amount the unfocussed spot on the screen moves in the opposite direction by a measurable distance.

The length from the diaphragm is known, and hence the height of the source above the diaphragm can be calculated. In practice, the positions of the unfocussed spot corresponding to ten successive translations of the gun through 0.87 mm. were recorded photographically for various operating conditions. It was found that the cathode image is always situated below the anode, the actual distance varying only slightly from 9 cms. Variations due to changing the filament height could not be isolated from experimental errors sufficiently to deduce any definite relation, but the tendency was for the image to move in the same direction as the filament.

Having ascertained the position of the cathode image, its quality can be investigated by interchanging the power supplies to the two focussing coils, so that the lower lens becomes the stronger one. This virtually converts the instrument into a low power two-stage electron microscope in which the cathode image itself takes the place of the object. The magnifications obtainable, ranging from 10x to 100x are very convenient for studying the behaviour of the cathode lens.

If no special precautions are taken the cathode image will

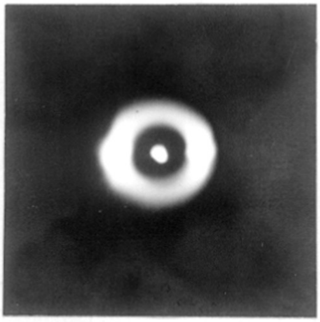


FIG. 19.

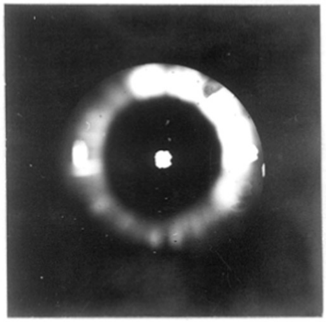


FIG. 20.

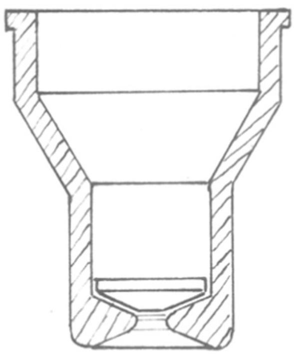


FIG. 21. SECTION THROUGH WEHNELT CYLINDER SHOWING INTERNAL MASK

usually be found to consist of a relatively small central spot accompanied by two or more equally bright, or even brighter satellite spots which may or may not be completely separated from it. The outer spots may be so numerous as to coalesce to form an almost uniform heavy halo. Fig.19 is a magnetic micrograph of this type of image. As the bias voltage is increased the halo tends to attach itself to the central spot, unless the filament is set very nearly level with the cathode aperture, when the bias ceases to have much effect on this type of image. Tilting the anode slightly sometimes causes the halo to break into two segments: a fact which perhaps lends justification to the description of the halo as being due to the extremely high spherical aberration of the cathode lens. The practice of simply "blocking off" the unwanted side spots by intercepting the part of the beam producing them by means of a diaphragm is unsatisfactory, since most of the intensity is thereby lost. Furthermore the aperture in a diaphragm used in this way has to be made to an exact size, and its position adjusted very accurately.

Further investigation of the nature of the cathode image can be carried out if the greatly enlarged pinhole image of it formed by a diaphragm placed within the gun itself is studied. Fig.20 was obtained by this method. The irregular shape of the central spot in this case corresponded to the irregular diaphragm which was purposely used in order to facilitate identification of the factors contributing to the formation of the image. An immediate inference from the sharpness of this pinhole image is that the central spot of the cathode image is small compared with the size of the aperture in the diaphragm, which in this case was 0.012 cm. It will be noticed that a brilliant reproduction of the shape of the central spot occurs at one ~~one~~ point of the halo, and that two other less bright reproductions occur partly superimposed at the diametrically opposite positions. The rest of the halo seems to be made up of more or less blurred and rather faint similar images. Viewed on the screen these images have a wavering intensity which increases during the 10 or 20 seconds after the filament has been switched on. The whole effect gives the impression of being due to an accumulating charge. This impression is supported by the fact that variation of the bias resistance becomes completely ineffective as soon as the halo has established itself. It would appear, therefore, that contamination on the cathode orifice results in the production of subsidiary point sources distributed round a circle enclosing the central spot. It is found that moving the filament off centre within the Wehnelt cylinder displaces the central spot relatively to the halo. In order to avoid the necessity of applying drastic cleaning processes to the highly polished electroplated cathode orifice, a mask was pressed out of 0.005 in. copper sheet and fitted inside the Wehnelt cylinder (see Fig.21). The orifice in this mask is slightly smaller than the cathode aperture so that the emergent electrons cannot strike the Wehnelt cylinder at all. The copper can easily be freed of contamination by dipping in nitric acid, or a new mask can be made in a few minutes by means of a specially shaped punch made for this purpose. The insertion of the mask entirely eliminated the halo.

vi. Beam Limiting Diaphragms.

If the beam is limited by a diaphragm having a small circular aperture, it would be expected that the unfocussed spot formed on the screen would likewise be circular. We find, however, that this is not always the case, the spot often being irregular in shape and larger than the geometry of the system should allow. On the other hand, if the lens system is used as a two-stage microscope by interchanging the coil supplies as explained above, an enlarged image of the aperture in the diaphragm can be obtained which is sharp and circular. The only satisfactory explanation of these facts seems to be that considerable scattering takes place at the diaphragm itself: when the lens system focusses an image of the diaphragm onto the screen, the scattered rays merely add to the intensity of the periphery of the image without detracting from its definition; but if the cathode image is used as the object for the lens system, the rays scattered at the diaphragm result in a sort of "penumbra effect". When using cold cathode diffraction apparatus it has been the practice to use the collapse of this "penumbra" into the central spot as the criterion for focussing, so that it is an image of the diaphragm and not of the true source that has been used for diffraction. It is clear that if by this method of operation is followed with a hot cathode source, the advantage of the extremely small virtual image source produced by the hot cathode gun is lost. The cleanliness of the diaphragm is therefore of paramount importance in the attainment of high resolution.

Although the cathode image is extremely small, the electron beam associated with it is sufficiently divergent to strike the inside of the anode cylinder and other parts of the gun. This sets up a general scattering of electrons some of which will find their way through the main beam-limiting diaphragm and give rise to various spurious haloes and background to the central spot. This can be eliminated by setting a diaphragm with a 0.01 cm. aperture in the adjustable carrier near the anode so that all but the central part of the beam which diverges from the cathode image is intercepted. Although this is the ideal method of dealing with the problem, the presence of this upper diaphragm renders the alignment of the gun extremely difficult, and unless exceptionally good definition is being sought, an almost equally effective way of eliminating background due to these scattered electrons is to introduce a diaphragm held just above the specimen by means of a second specimen carrier, this being the standard practice with the cold cathode cameras in this laboratory.

With a main beam-limiting diaphragm of 0.01 cm. aperture the beam divergence at the upper lens is 0.01/38 = 0.00026 radian. If the upper lens is used at a focal length of 2 cms., the divergence of the emergent beam will be approximately 0.00026 x 58/2 = 0.0075 radian or about 0.4 deg. On reaching the second lens the cross section of the beam is therefore about 0.0075 x 46 = 0.35 cm., which is excessive. It is impracticable to use a main diaphragm with an aperture of much less than 0.01 cm., and it is therefore necessary when using the upper lens to reduce the beam cross-section at the specimen by means of the final diaphragm referred to in the last paragraph even if the upper diaphragm is in use in the gun. It will be seen that unless the upper lens is used in conjunction with this diaphragm nothing is gained from its use, since the reduction in source size is accompanied by an increase in beam width with the associated spherical aberration effect at the second lens and excessive beam convergence at the specimen. The latter defect is particularly undesirable when measurements are being made which require an exact knowledge of the angle of incidence. This is the case, for example, when electron diffraction is used as a means of measuring the thickness of thin films. It is necessary for such experiments that the final beam convergence should be small compared with the angles of incidence used, and for this reason it is sometimes an advantage to use only the top lens. For other work in which extremely high resolution is not required the lower focussing coil can be used alone; for the very highest resolution, both lenses must be used and the final beam convergence controlled by the lower diaphragm.

vii. Control of the Beam.

The usual method of directing the electron beam onto the desired part of the specimen in the case of instruments which employ a simple unshielded focussing coil is to tilt the coil relatively to the axis of the instrument: this technique greatly facilitates the rapid scanning of the specimen, and it was originally intended that the lower lens in our instrument should be used in this way. Unfortunately it was found that, due perhaps to the different form of the field-curve, tilting an iron-shielded lens has a comparatively small effect on the direction of the beam, but a considerable adverse effect on the quality of the spot. It is necessary, therefore, for high resolution work, to leave the lower lens in alignment and to control the diffraction by moving the specimen. The practicability of an alternative arrangement whereby the lens is controlled by separate magnetic deflecting coils is under consideration.

viii. Elimination of Electrostatic Charge from Specimen.

It is well known that in many cases an electron diffraction pattern is seriously distorted by an accumulated electrostatic charge on the specimen. This effect is particularly marked in the case of diffraction by reflection from insulating materials, and with this type of specimen it is sometimes impossible to obtain a well defined diffraction pattern unless special precautions are taken to eliminate electrostatic charge from the diffracting surface. When high resolution is being sought, even slight "charging up" effects which can normally be ignored, must be eliminated.

The device generally employed to disperse the charge consists essentially of a source of very low velocity electrons in the neighbourhood of the specimen. A study of the conditions under which this arrangement takes effect was carried out by Brubaker and Fuller (1945) who concluded that the charge on the specimen accumulated as a consequence of disequilibrium between the rates of absorption of primary electrons and emission of secondary electrons, and that this charge, whether positive or negative, is conducted away when the residual gas is ionised by low-velocity electrons directed into the region near the specimen by the auxiliary electron gun or "decharger". The validity of this explanation is by no means certainly verified, and it is in any case difficult to make a quantitative study of the problem. We have, however, found that with a primary beam accelerated through fields exceeding about 100 KV. the effectiveness of the decharging becomes more noticeably dependent on the value of the small accelerating potential applied to the decharger. For this reason provision has been made for the control of the decharger voltage over a range of 300 volts.

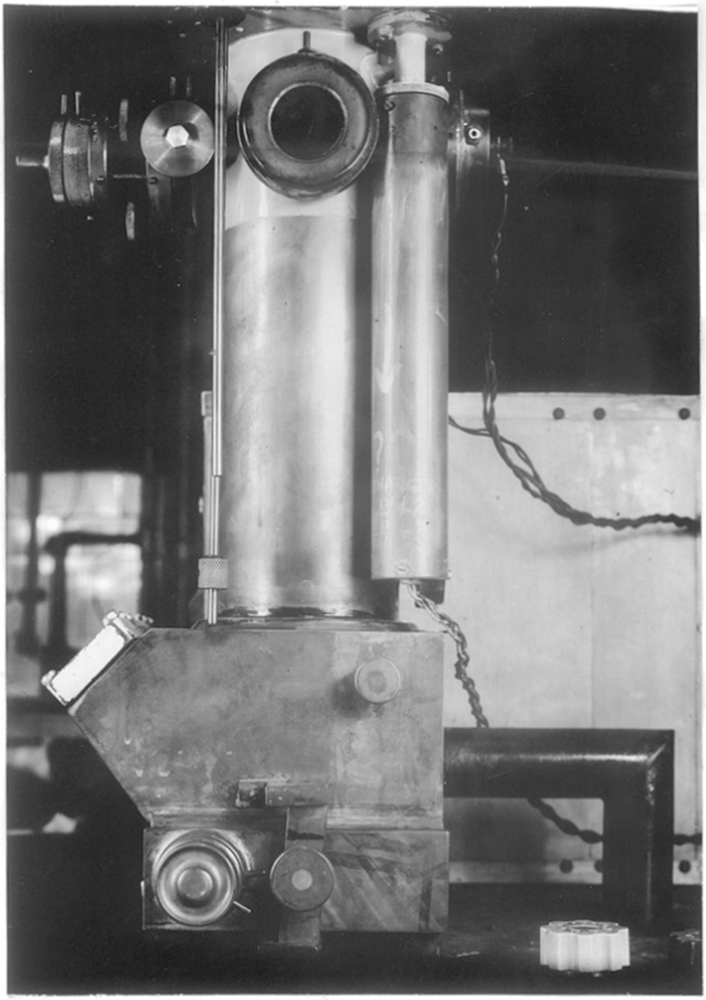


Fig.22. The camera body, with specimen holder (top left) and decharger (top right) in position; showing also the Pirani gauge head, triple plate holder, and vacuum pumping line.

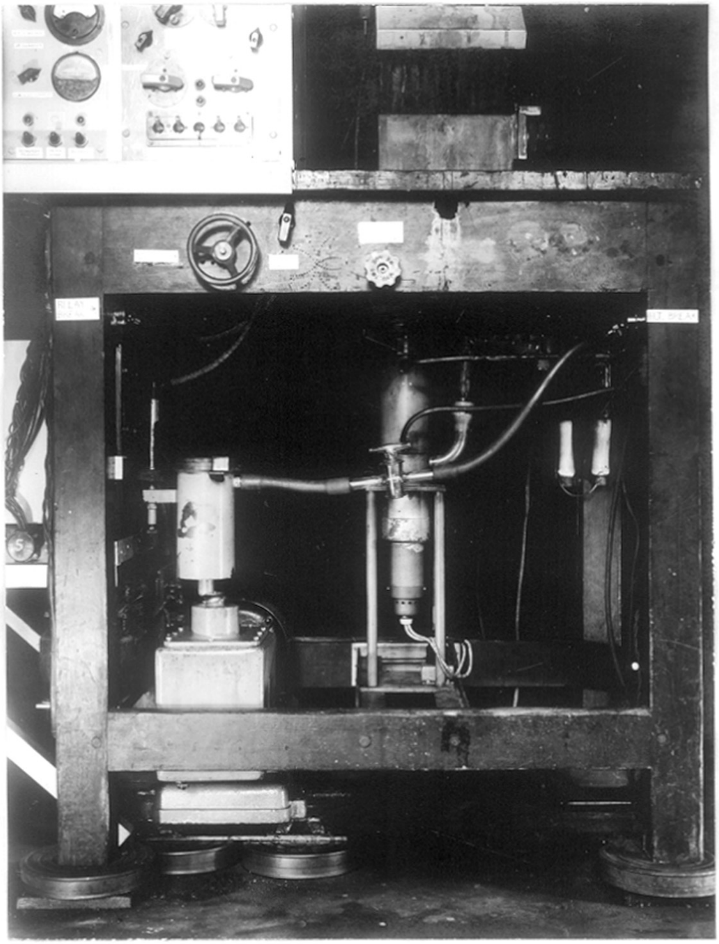


Fig.23. The arrangement of the vacuum pumps. Note the vibration absorbing units referred to on p. 60.

III. CAMERA BODY AND GENERAL CONSTRUCTION.

Apart from some minor refinements incorporated in the light of experience gained in this laboratory, the section of the apparatus below the focussing tube - namely the specimen chamber and its accessories, and the camera chamber and plate-holder - are identical with the corresponding section of the standard Finch apparatus as described in detail elsewhere (Finch and Quarrell, 1933).

As will be seen from Fig.22, the main pumping line is brazed into the back of the camera casting near to the base instead of being connected near to the top. This arrangement, which enables the diffusion pump to be placed conveniently beneath the bench (Fig.23) becomes possible with a hot cathode diffraction apparatus since the injector system used in conjunction with the cold cathode instrument is dispensed with.

The five ports in the specimen chamber which are provided for the insertion of the specimen holder and other accessories are, in accordance with well-tried practice, equipped with flanges which are ground flat and, when not in use, sealed off with a glass plate. The specimen holder itself, however, has been modified from the standard design by the cutting of a circular channel to hold an O-section neoprene gasket, thus eliminating the use of grease on the most frequently demounted joint. The use of greased ground surfaces for the semi-permanent vacuum joint at the viewing port has been eliminated by the substitution of a bolted joint sealed by a compressed neoprene gasket. The plate-holder, which holds three quarter-plates, and carries the fluorescent screen is of the standard Finch design, but is secured by a screw clamp as shown in Fig.22.

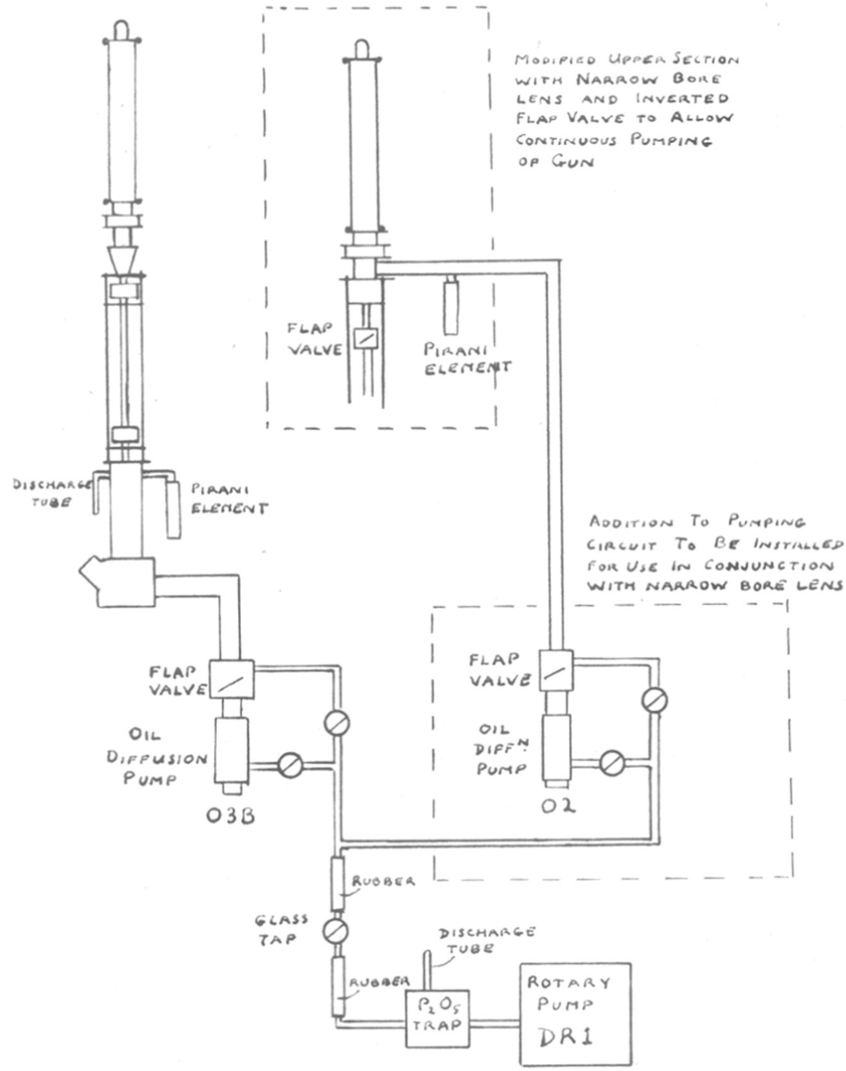


FIG. 24. SCHEMATIC DIAGRAM OF VACUUM CIRCUIT

IV. ANCILLARY EQUIPMENT.

i. Pumping Equipment.

The vacuum circuit is shown schematically in Fig.24. The forepump can be connected either directly to the apparatus for rough pumping from atmospheric pressure to about 5×10^{-2} cm. Hg, or via the diffusion pump for normal pumping to working pressure. The stop-cocks used to enable this change-over to be carried out are of the oil-sealed type. The diffusion pump can be isolated from the main apparatus by means of a flap-valve: this avoids the loss of time which would otherwise result if the pump had to be allowed to cool before opening the diffraction camera. A length of rubber pressure tube included in the backing circuit prevents direct transmission of vibration from the mechanical pump to the main apparatus. The remainder of the pipework is of metal, the demountable joints being rubber-sealed. No liquid air traps or similar precautions are necessary for this apparatus. Silicone oil (D.C.703) is used instead of the ordinary Apiezon B since it has the advantage of being stable with air at operating temperatures.

The pumping equipment itself consists of a Metropolitan-Vickers type O3B oil diffusion pump backed by a type DR.1 rotary pump. This set is rated to reach pressures below 10^{-6} mm. Hg, and we have verified by means of a McLeod gauge that, under test conditions, a pressure of 10^{-6} mm. is certainly reached. However, the virtue of this set lies in its great pumping speed below 10^{-3} mm. Hg rather than in the theoretical minimum attainable pressure. In practice the vacuum that can be reached in the apparatus is determined by the pressure at which a state of equilibrium exists between the rate of outgassing from the surfaces of the instrument walls and the pumping rate, and this pressure is obviously determined by the pumping speed.

A poor vacuum results in three distinct undesirable effects: inelastic scattering by gas molecules of sufficient electrons to cause an appreciable deterioration in contrast of the diffraction pattern; rapid disintegration of the filament, resulting in the necessity of frequent renewals; "striking" of the gun; the latter effect is liable to occur at pressures of about 5×10^{-3} mm., and, of course, no attempt should be made to operate the instrument when the vacuum has deteriorated to this extent. We are of the opinion that 2×10^{-5} mm. is the minimum pressure that should be tolerated, and once the vacuum characteristics of the apparatus ~~has~~ have been ascertained, there was little difficulty

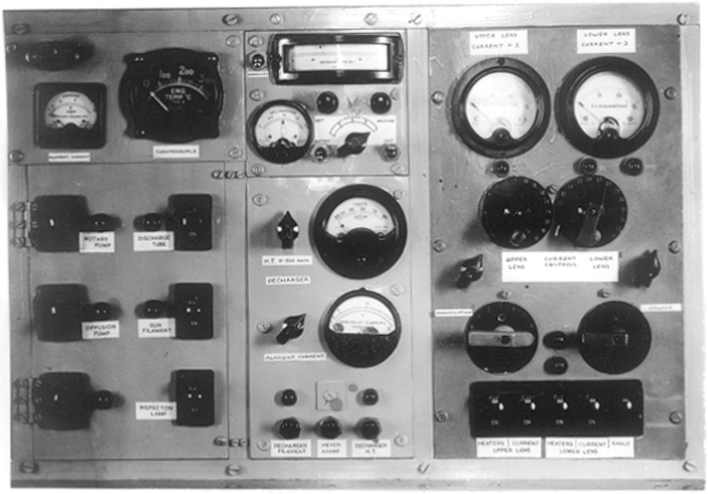


Fig. 25. The Control Panel.

in achieving this figure as a matter of routine.

The additional type 02 diffusion pump shown in Fig.24 for use in connection with continuous pumping of the electron gun has not yet been installed, but the necessary connection has been provided in the pumping manifold beneath the gun.

ii. Pressure Gauges.

In the case of the cold cathode diffraction cameras in general use in this laboratory, the state of the high tension discharge is a sufficient indication of the condition of the vacuum, but with the more stringent vacuum requirements associated with hot cathode work, the incorporation of separate vacuum gauges becomes a necessity. The position of these gauges is indicated in Fig.24. The Geissler tubes serve to indicate the stage in the evacuation at which the diffusion pump can be brought into operation and to give warning of very large air leaks. The supply to these tubes is provided by a 4,000 volt transformer in series with a 10-megohm current-limiting resistance.

The glass bulbs containing the Pirani and compensating elements are enclosed in a brass cylinder which

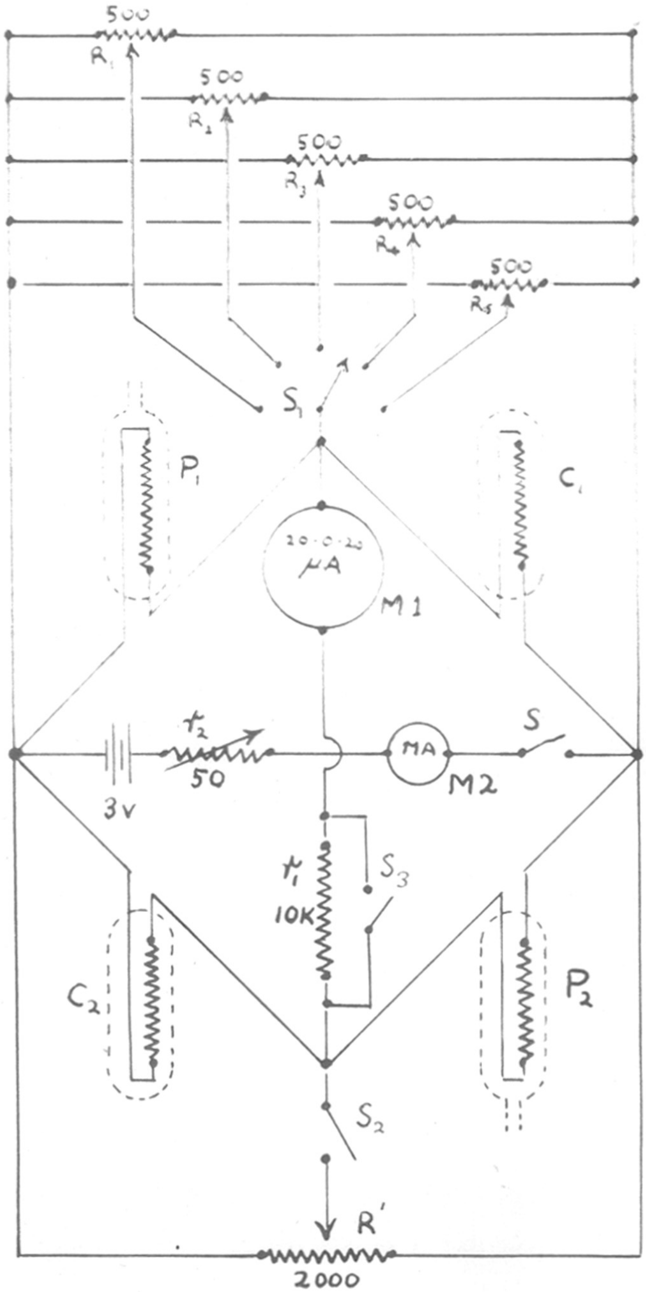


Fig. 26. Bridge Circuit for Pressure Gauge

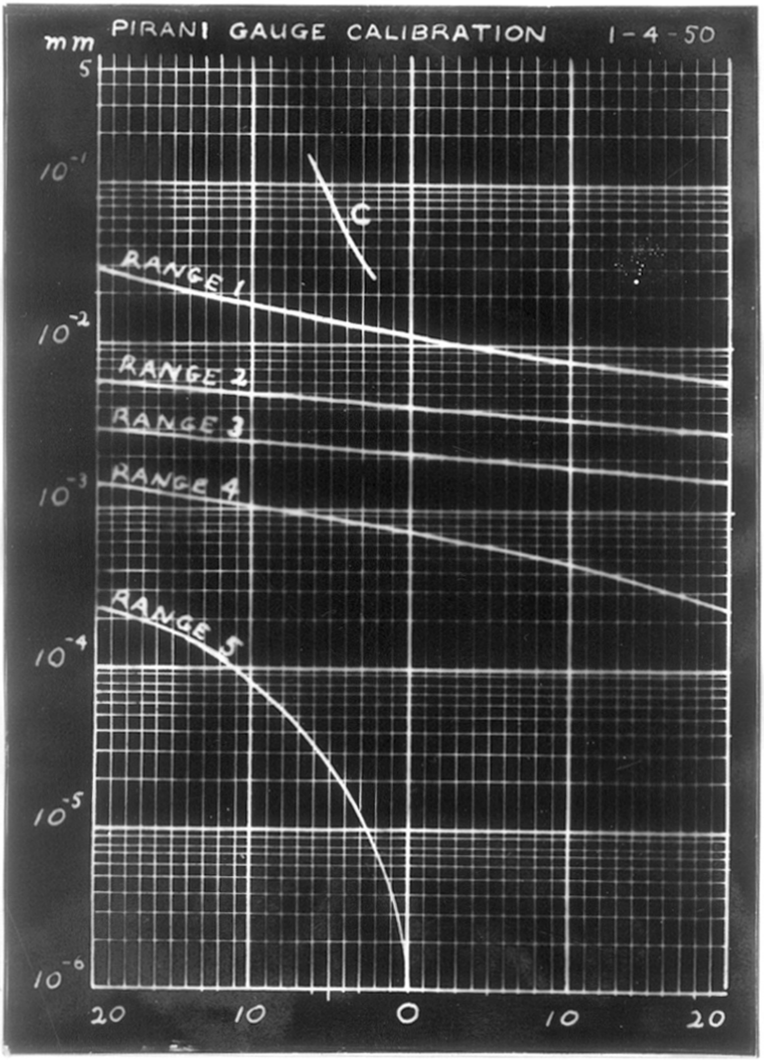

PIRANI GAUGE CALIBRATION
1-4-50
mm
5
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
C
RANGE 1
RANGE 2
RANGE 3
RANGE 4
RANGE 5
20
10
0
10
20

Fig.27.

is mounted as shown in Fig.22. The sensitivity of the associated bridge circuit shown schematically in Fig.26 can be inferred from the calibration curve reproduced in Fig.27. The two Pirani elements, P_1, P_2, and the two compensating elements C_1, C_2, are connected so as to form a Wheatstone bridge which is shunted by the five 500-ohm pre-set potentiometers R_1 .... R_5, whose centre taps are connected to the five-pole range selector S_1. These potentiometers are adjusted during calibration so that the bridge balances at 10^{-6}, 8×10^{-4}, 2×10^{-3}, 1.2×10^{-2} mm. according to the range selected: the meter reading then gives continuous coverage over the range of pressures 10^{-6} to 2×10^{-2} mm. A further potentiometer R' can be brought into use by the switch S_2 and enables the bridge to be balanced at any pressure in order to render it in the most sensitive condition for leak detecting. The pressure indicating meter M_1 is a 20-0-20 microammeter protected by the high resistance r_1 which is short circuited by S_3 after the correct range has been selected. The source of current is a 3-volt dry battery in series with the on-off switch S , an adjustable resistance r_2 and the set current meter M_2. The complete circuit is built into a small chassis which slides into the main control unit and carries the control panel which forms the top section of the complete panel shown in Fig.25.

iii. Filament Supply to the Electron Gun.

The unusually high voltage at which the electron gun was intended to operate gave rise to difficulties in connection with the heating of the filament, no suitable oil immersed transformer being available to supply the necessary low alternating current at high tension. This problem was solved by building a transformer into the ceramic bush which conveys the high tension lead through the partition separating the camera cubicle from the high tension set: the ceramic itself provided the insulation between the secondary winding which was placed inside the bush and the primary which was wound round the outside. For the secondary, use is made of an iron-cored solenoid which formed the primary of an old induction coil. The output leads from this winding are connected directly to the filament of the electron gun (see Fig.3). The ~~secondary~~ primary winding, which consists of 1,000 turns of No.24 gauge enamelled copper wire wound onto an earthed metal former built round the outside of the bush, is supplied, via ballast and control resistances from the A.C. mains. An unforeseen consequence of the open magnetic circuit of a transformer so constructed manifested itself in the unwelcome presence of an alternating magnetic field powerful enough to perceptibly disturb the electron beam. It was found, however, that this effect could be completely eliminated by suspending a compensating element at a position with respect to the camera symmetrically opposite to the high tension bush. When the current in this solenoid is correctly adjusted the varying magnetic field is reduced to a vertical one through the axis of the camera, and the electron beam is no longer affected.

iv. High Tension Supply.

The 220-kilovolt high tension set was designed in this laboratory and constructed by Messrs Ferranti. The set consists essentially of a 50-cycle 110 KV. transformer, a voltage doubling network, and a 0.015 μf smoothing condenser. A diode valve in series with the output serves as a current limiting device. The 50-cycle input to the primary of the high-tension transformer is tapped from the A.C. mains by a saturated-core variac which serves to control and regulate the voltage. A voltmeter across the primary side is calibrated to give direct indication of the D.C. output.

The bias resistance for the electron gun consists of a ½-megohm potentiometer secured within the hemispherical

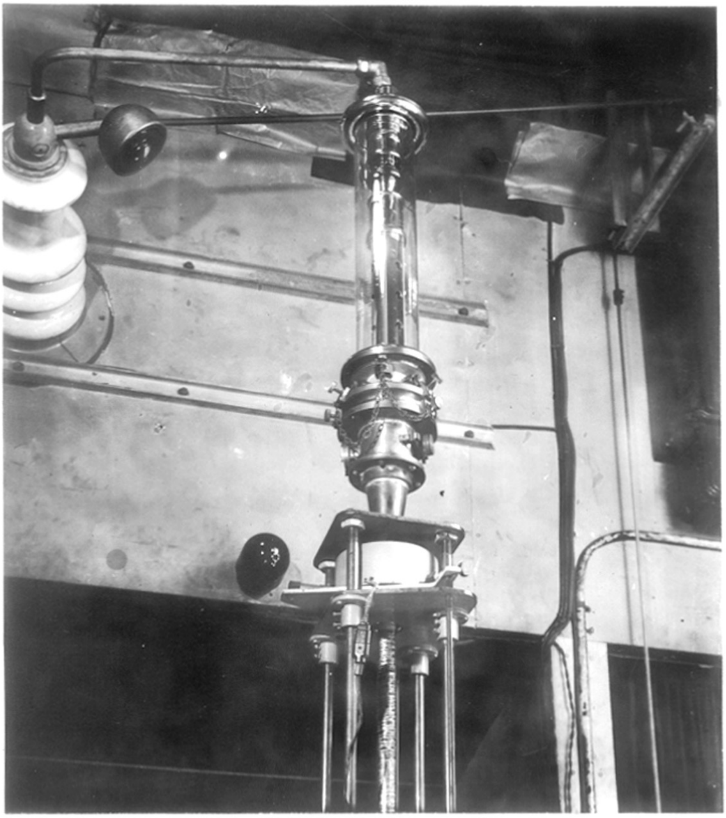


Fig.28. Arrangement of the high tension connections. The shell mounted on the ceramic bush holds a beam-current meter and contains the variable bias resistance: an insulating rod connecting the latter to the control transmission on the right can be seen behind the top of the electron gun. Note also the chain connections beneath the gun which ensure that the base sections are earthed. The top of the gun is 10' above floor level.

shell which carries the beam-current meter. This potentiometer is connected by a five-foot length of insulating rod to a system of gears and shafts which brings the bias control within convenient reach of the camera operator (see Fig.28).

v. Lens Coil Current Supplies.

We have seen in a previous section that the degree of current stability necessary for the lens coil supplies is one part in 20,000. For the upper and lower lenses, each having a resistance of 800 ohms, currents controllable in the respective ranges 100 to 300 and 10 to 150 mA. are required. The circuit adopted for the stabilisation of these power supplies is a modification of a design developed by Brown and Challice (1948) for use with an electron microscope, and the action of the circuit has been explained by Challice (Ph.D.Thesis, 1949). The greater maximum current and wider range of control required for our instrument necessitated a recalculation of the values of the circuit components, and a modification of the control resistance network. It was, however, decided to retain Mullard ML.37 series control valves in conjunction with a 6AC7 amplifying pentode. The circuit diagram is reproduced in Fig.30. The complete method by which the

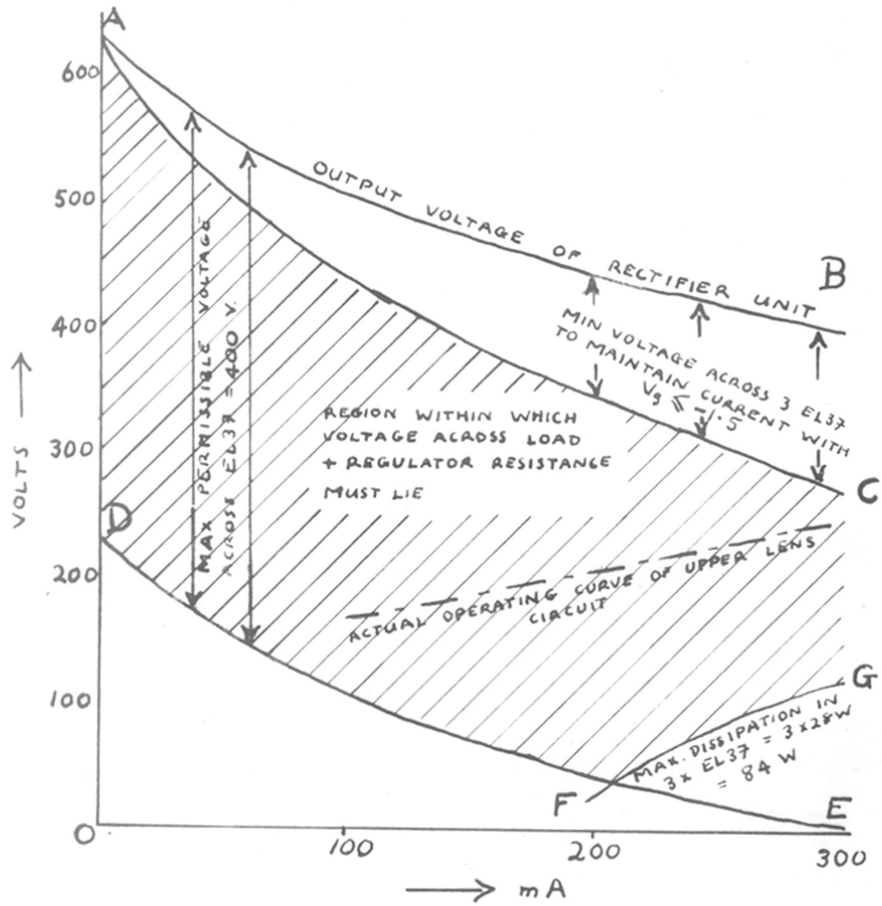


FIG. 29. DESIGN DIAGRAM FOR LENS STABILISERS

values for the circuit components are calculated is too lengthy to reproduce in detail, but the governing principles of the theoretical design process can be represented graphically as follows:

The input voltage to the stabiliser is derived from a conventional full wave rectifier unit, and it will therefore vary according to the current drawn, in the manner indicated by the curve AB (Fig.29). This curve represents the voltage available to be dropped across the series valve, the load, and the regulator resistance. Now for a given current the minimum share of this available voltage that must be allotted to the series valve is determined by the condition that the grid must not become more positive than the cathode, and for safety we set $V_{g\ max} = -1.5$ volts. Subtracting the ordinates of the $I_k V_a$ curve specified by $V_g = -1.5$ volts from the curve AB therefore gives the current curve AC representing the maximum voltage available for dropping across the load and regulator resistance. The minimum permissible voltage is reached when the anode-cathode voltage of the series valve is at its rated maximum, and the curve DE indicating this limit of the working range is derived by subtracting $V_{a\ max}$ from the ordinates of AB. So far we see that the voltage

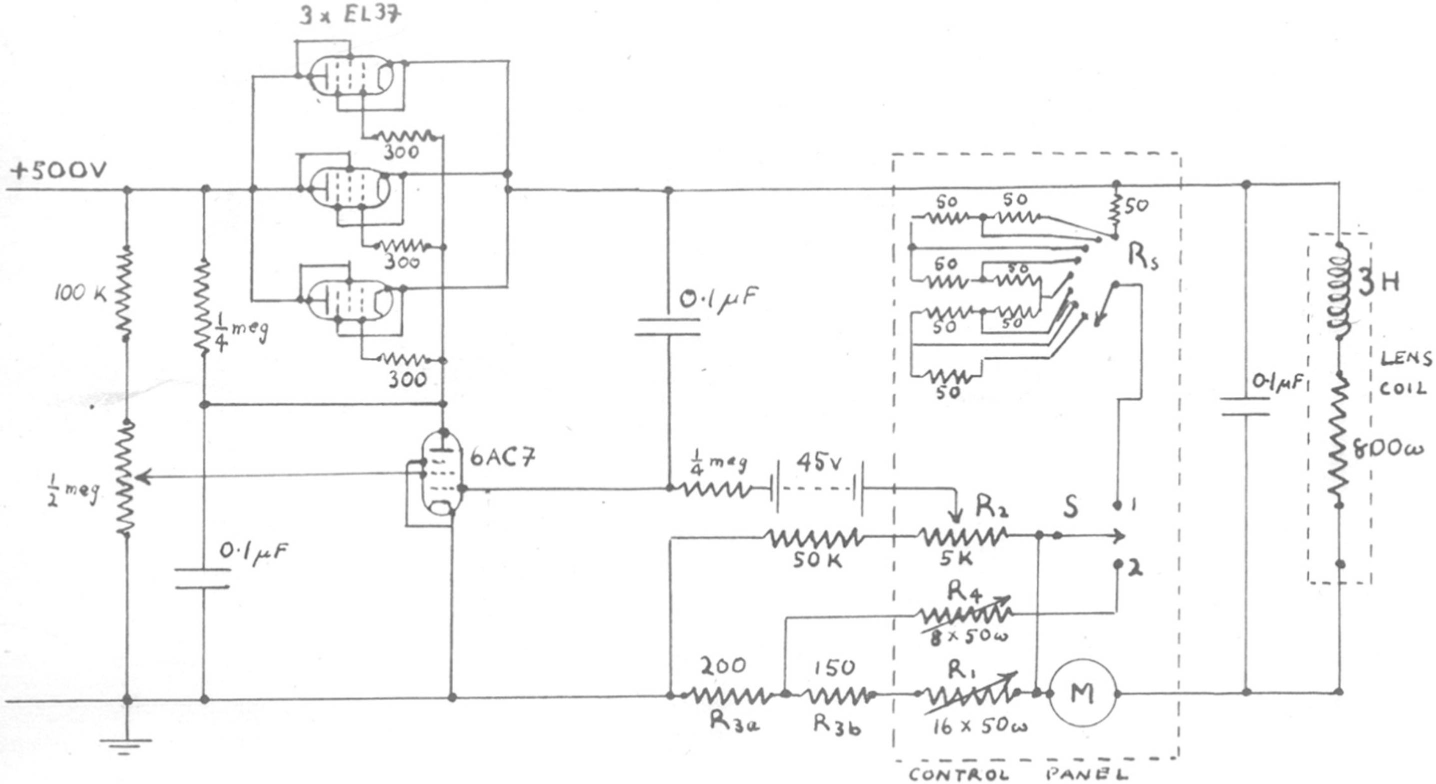


FIG. 30. CIRCUIT DIAGRAM OF LOWER LENS COIL CURRENT STABILISER

across the load and regulator network must lie between the curves AC and DE. A further limitation is set by the condition that the power dissipated in the valve must not exceed its rated maximum; the ordinates of the dissipation curve, when subtracted from AB give the curve FG which completes the boundary of the region within which the current-voltage curve of the regulator section must lie.

In the case of the circuit for the lower lens as depicted in Fig.30, the lower currents are obtained by means of the variable shunt R_s used in conjunction with the normal coarse regulating resistance R_1 and the fine control potentiometer R_2. The higher current range required to operate the camera as a shadow microscope is obtained by setting the switch S to position 2; this disconnects the shunt and by-passes the main regulator resistance R_1 and ballast resistance $R_{s\,b}$ through a variable step resistance R_4. The upper lens supply is identical with this circuit except that the switch S and the resistances $R_{s\,b}$, R_4 and R_s are omitted. The potentiometer from which the screen grid voltage of the amplifier valve is tapped is adjusted for minimum ripple as indicated on a C.R.O. connected across the load. The performance of the circuit is considerably better than is necessary for our purposes, the output being stable

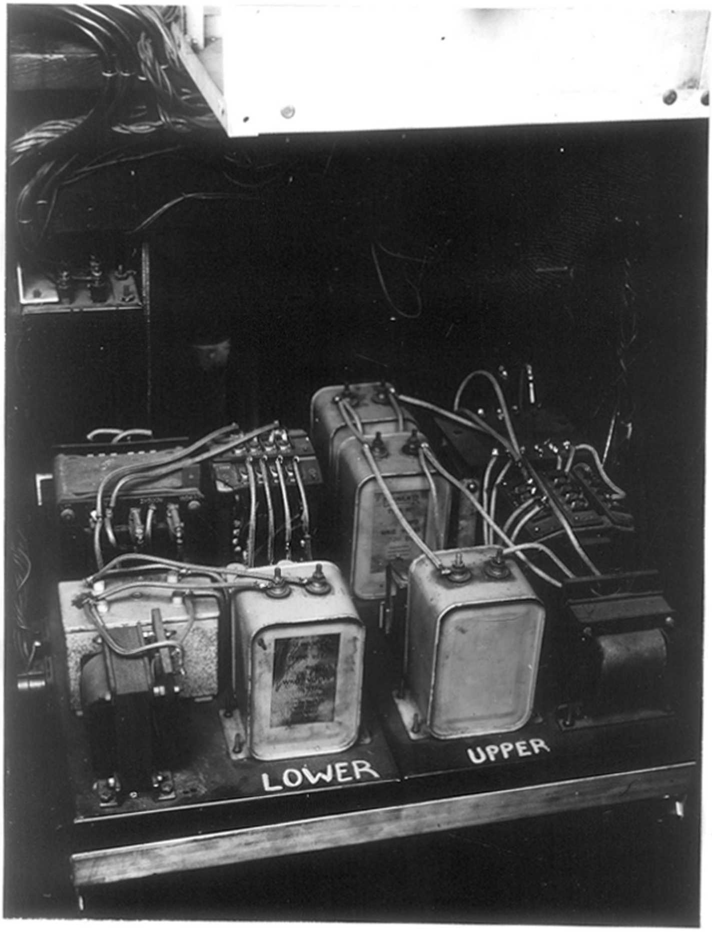


Fig.31. The Power Units for the Lens Coil Current Stabilisers.

to within one part in 200,000.

The two stabilising circuits are built into a single compact frame chassis which fits into the main control unit and carries the panel forming the right hand section of the complete control panel shown in Fig.25. The connections to the rectifying units and lenses are made by multipin plugs and jack-plugs to allow easy withdrawal of the chassis for maintenance. Separate switches are provided for the valve heaters so that the H.T.switches take effect immediately. Indicator lights are provided to facilitate operation under dark room conditions. The switching system is entirely relay-operated to allow compact and regular arrangement of the controls. In view of the considerable heat-dissipation of the closely packed valves, the circuit is air-cooled by a centrifugal fan which automatically comes into operation when high currents are being used in the upper lens.

The two power packs are mounted on a rack to one side of the table and are shown in Fig.31, but the rectifying valves are mounted separately behind the main switch panel to allow more ready access for inspection.

vi. The Decharger.

The decharger at present in use if of the very simple design that has always been found completely effective in this laboratory. It consists of a coiled tungsten filament set behind a large (ca. 1 cm.) orifice in a cylindrical anode. The circuit supplying the necessary H.T. and heating current employs a metal rectifier bridge fed from A.C. mains. The output from this bridge is smoothed and supplied to a potentiometer whose centre tap is connected to one side of the decharger filament: an electrostatic voltmeter gives direct indication of the decharger voltage. The filament current is drawn from a 10-volt transformer and controlled by a series resistor. The chassis into which this circuit is built fits into the control panel beneath the Pirani unit.

It is planned to build a more elaborate decharger employing a three-electrode principle similar to that used in the electron gun in order to facilitate a more exact study of the decharging effect which sometimes appears to operate under critical conditions when very high voltages are used to accelerate the primary beam. It is hoped, also, that the use of a more carefully designed decharger will minimise contamination of the specimen by tungsten, and reduce fogging of the plates by light emitted from the filament.

vii. Vibration Insulation.

The complete apparatus with all ancillary equipment with the exception of the mechanical forepump is built onto a robust wooden bench which is effectively insulated from every type of vibration by the specially designed shock absorbing units widely used in this laboratory for the mounting of optical microscopes as well as for heavier apparatus. In these units, the higher frequency vibrations are absorbed by a block of 300 dry filter papers, and the lower frequencies by a ½-inch thickness of medium-hard rubber; rubber alone is not effective, since it transmits higher frequencies. No trouble has been experienced from vibration even though building work has been in progress close at hand.

V. PREPARATION OF THE INSTRUMENT FOR USE.

The cycle of adjustment operations is best considered as commencing from filament renewal. When this becomes necessary, the cathode assembly is withdrawn from the gun and the Wehnelt cylinder detached to expose the filament clamps. The filaments with which we have had most success have been narrow hairpin shaped, rather than V-shaped. The newly mounted filament is centred with respect to the Wehnelt cylinder and retracted to a position about 5 mm. above the aperture. Before replacing the cathode assembly, the upper diaphragm holder is removed. The camera is now evacuated and the high tension adjusted to 120 KV. With the bias set to maximum, the filament current is increased from zero until an image appears on the screen: this image will usually be in the form of an annulus. On further increasing the current, a central spot appears and finally the annulus fades completely. The filament is now lowered until the spot becomes a single image of the diaphragm. The tilt and position of the gun is then adjusted for maximum intensity while keeping the spot in the position on the screen known to correspond to symmetrical passage of the beam through the focussing tube. The filament is now re-centred without disturbing its height, and the adjustable diaphragm replaced and moved until the spot reappears. The bias is finally adjusted for maximum intensity subject to the maintenance of a good quality spot.

If the upper lens is to be used, it is aligned by setting the current to a low value and adjusting the tilt for zero deflection of the beam, and then switching to a high current and adjusting laterally for zero deflection, returning to a low current and adjusting the tilt, and so continuing until the spot is undeflected by any alteration of current. The lower coil is adjusted to give an anastigmatic spot, and the lower diaphragm brought into alignment if required. The camera is then ready for use.

When it is intended to operate the instrument using accelerating potentials above 100 KV. it is advisable to raise the high tension above this value in 5-kilovolt steps with the gun filament supply switched off, until the gun ceases to "flicker". The cause of the flickering is perhaps connected with the presence of dust particles carried into the gun when air is admitted. If the gun "strikes" during the process and fluoresecence fails to black out within ten seconds, the voltage should be momentarily lowered. The high tension is raised in this way to about 10 KV. above the operating voltage and then adjusted to the final value before switching on the filament supply. With these precautions the gun will be found to operate quite smoothly. An outburst of violent sparking is usually an indication that dust particles or fibre strands have become attached to the outside of the gun, and the high tension supply should be switched off immediately.

APPLICATIONS OF THE INSTRUMENT.

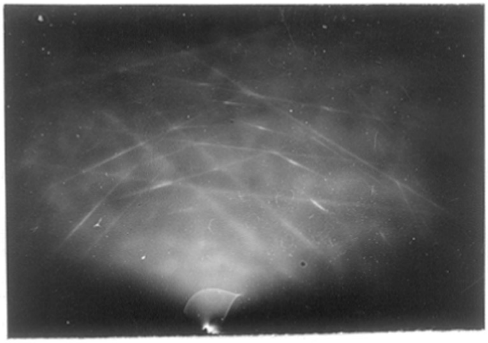
The work so far carried out with the instrument was undertaken mainly with a view to ascertaining the extent to which the expected advantages of diffraction with higher speed electrons would be realised in practice, and to determine what further advantages, if any, might be revealed. A variety of specimens was examined, and the diffraction patterns obtained at various electron speeds compared. Pending the incorporation of additional corona protection in the high tension set, we have found it necessary for the time being to restrict the accelerating potentials to 140 KV. or less during the exposure of plates.

Before considering in detail the more interesting patterns obtained, it can at once be stated that, quite apart from any specific advantages in particular cases, the use of higher speed electrons has in every case reduced the general difficulties of "finding" a pattern to a degree which would in itself justify the adoption of the high voltage technique in general practice. This greater facility in carrying out diffraction experiments at higher voltages is to some extent attributable to two factors: first, the "charging up" of reflection specimens is greatly reduced, and in some cases does not occur at all; or, to be more precise, it does not become important except when unusually small glancing angles are used. The use of the decharger can therefore frequently be dispensed with so that possible contamination of the specimen by tungsten oxide is avoided, and likewise fogging of the photographic plate by light from the decharger filament. A second notable advantage is that with the use of accelerating potentials of about 120 KV., the contrast response of the fluorescent screen more nearly matches that of the photographic plates; we can say generally that if there is no trace of a diffraction pattern on the willemite screen, none will appear in the photograph. This has never been true of lower voltage practice where it is frequently necessary to develop the plate before the quality of the pattern can be judged.

Diffraction from Quartz Single Crystal.

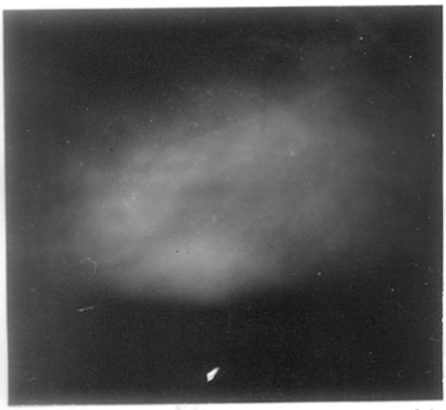
A quartz single crystal surface provides an ideal electron diffraction specimen for comparison purposes since it can be used repeatedly and accidental contamination removed without changing the surface layers.

The surface of a slightly convex single crystal quartz



HV 80

Fig.32. Quartz. 130 KV. Angle of incidence 3'.

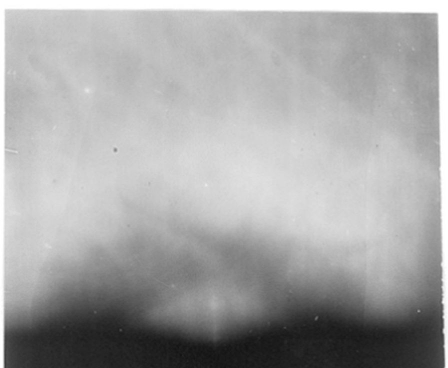


HV 131

Fig.33. HV 132

Fig.34.

Highly polished quartz surface. The amorphous polish layer which obscures the pattern in Fig.33. (60 KV) is penetrated by the 130 KV beam (Fig.34). On reducing the angle of incidence, the pattern is again obscured (Fig. 34a)



HV 133

Fig.34a.

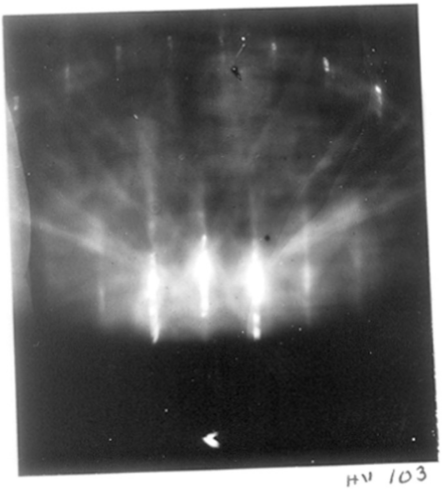
lens was first examined by reflection diffraction in order to determine the minimum angle of incidence at which a discernible diffraction pattern would be yielded. It was found that, at voltages above 110 KV., the distance between the shadow-edge and the undeflected beam spot could be reduced to zero, and that a clear diffraction pattern was obtained even when this distance was less than 0.05 cm., corresponding to a glancing angle of incidence of about 3' (See Fig.32). The adjustment of the decharger was extremely critical in this case.

The reverse side of the same quartz lens yielded further interesting results. It was not possible to obtain any diffraction pattern other than a diffuse scattering at the very low angle of incidence, and even when this angle was increased to 1° the pattern only became visible when accelerating potentials above about 100 KV. were used. Figs.33 and 34 show the patterns yielded at 60 and 130 KV. respectively, the angles of incidence and azimuth being the same in each case. It will be seen that with the lower voltage the barest trace of a Kikuchi line pattern can be detected, but with 130 KV. the pattern is quite clear. The high voltage pattern can again be obliterated by reducing the angle of incidence. These observations may be taken as clear evidence of the existence of an amorphous polish layer.

The presence of a Beilby layer on polished quartz has been established previously by the following argument independent of electron diffraction. It is found that if a scratch is made on the surface, all trace of it can be removed by a polishing process. That this obliteration is due to the flow of an amorphous layer and not to a wearing-down of the surface is evidenced by the fact that the scratch reappears after etching. The failure of previous workers to obtain electron diffraction evidence of this amorphous layer can now be attributed to the limitation of the range of electron speeds at their disposal, for since a relatively large angle of incidence must in any case be used to obtain a diffraction pattern from a very smooth insulator surface with 60 KV. electrons, the further increase in angle of incidence necessary to penetrate a Beilby layer was too small to give clear indication of its presence. The present sucess with a 130 KV. electron beam suggests that the usefulness of electron diffraction for the study of polish is now greatly extended.

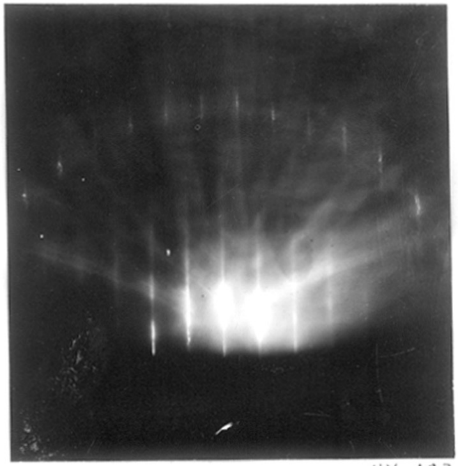
Mica Reflection.

Several specimens of mica were examined by reflection diffraction in the expectation that the increased penetration



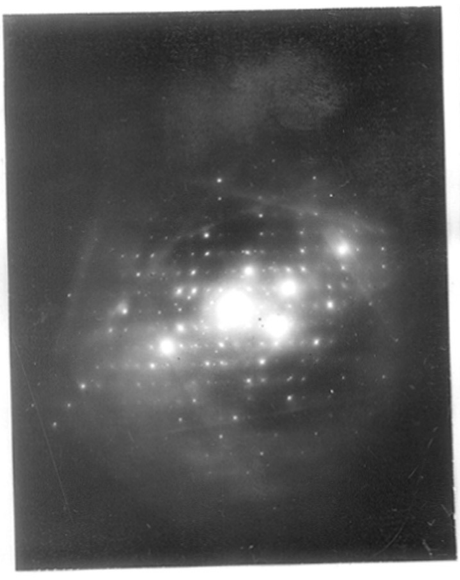
HV 103

Fig.35. Mica. 70 KV.



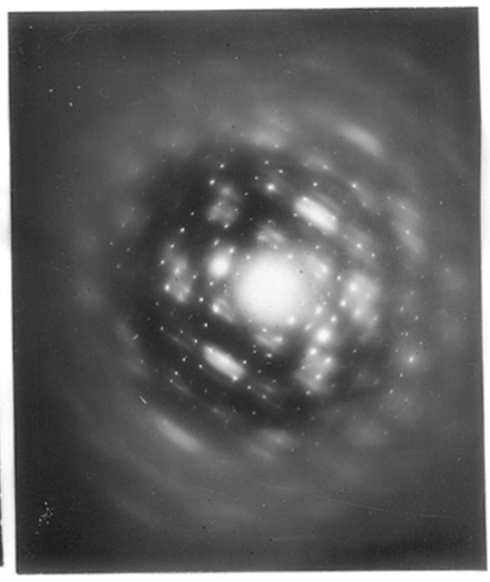
HV 102

Fig.36. Mica. 140 KV.



HV119

Fig.37. Anthracene. 110 KV.



HV124

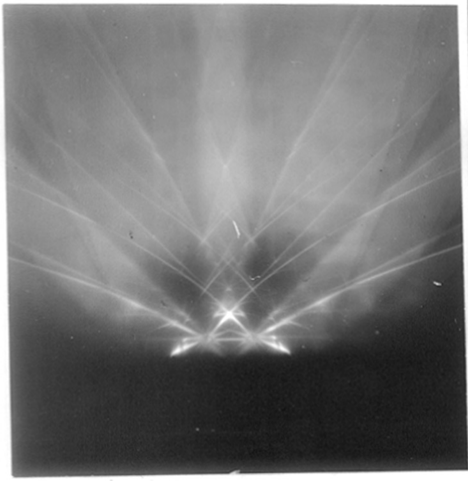
Fig.38. Anthracene. 120 KV.

possible with fast electron beams would give interesting results in view of the large lattice spacing normal to the cleavage face. No significant difference was apparent, however, between the patterns obtained at widely varying voltages. Figs. 35 and 36 show patterns yielded at 70 and 140 KV. respectively.

Anthracene.

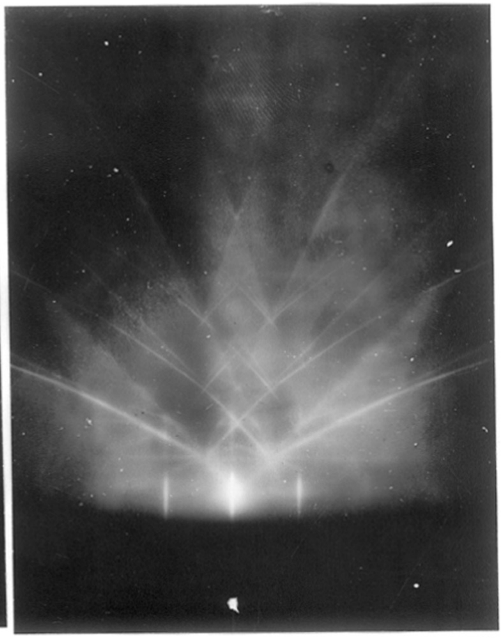
Single crystals of anthracene obtained by sublimation were examined by transmission diffraction. The ease and rapidity with which patterns can be obtained using a high voltage beam proves a great advantage in dealing with specimens such as this which evaporate rather rapidly in vacuum. Kikuchi lines seldom appear in anthracene patterns obtained with a 60 KV. electron beam, but they are readily obtainable at 100 KV. or above. Unfortunately, during the long exposure necessary to reveal these lines, the specimen often evaporates until it is too thin to yield them. However, Fig.37 shows clear Kikuchi lines in a pattern obtained at 110 KV.

It was found that with the fast electrons it was particularly easy to obtain the very interesting type of pattern shown in Fig.38 (120 KV.). The explanation of the



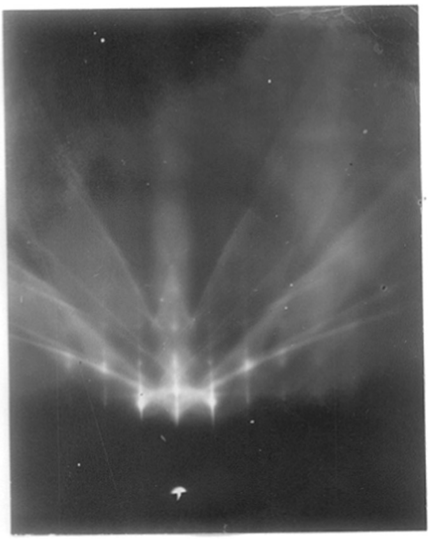
HV105

Fig.40. Periclase. 130 KV.



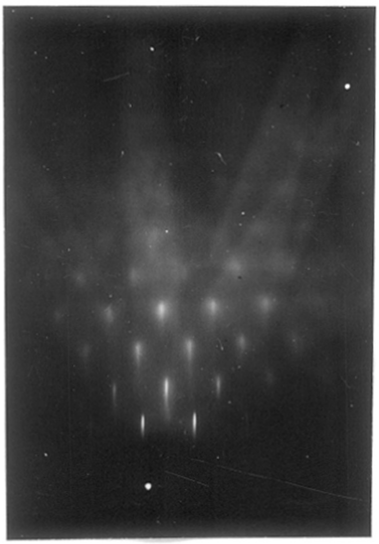
HV106

Fig.41. Periclase. 65 KV.



HV92

Fig.42. Rock Salt.



HV 137

Fig.43. Copper single crystal, 140 KV. Angle of incidence 3°.

diffuse regions and of the streakiness parallel to one lattice row has been discussed in detail by Charlesby, Finch and Wilman (1939), and the exceptionally clear patterns now obtained, and of which Fig.39 is an example, confirms their conclusion that the phenomenon is due to molecular diffraction and anisotropic thermal oscillation.

Periclase.

The pattern obtained by reflection from a periclase cleavage face at 130 KV. (Fig.40) compares very favourably with that obtained at 65 KV. (Fig.41) from the same crystal.

Rock Salt.

In the case of rock salt specimens the almost complete absence of any "charging up" effect with electron speeds above 90 KV. was particularly noticeable, and greatly facilitated the obtaining of patterns of which Fig.42 is an example.

SUMMARY.

The general inference from experience gained in the course of about 150 diffraction experiments carried out on various specimens and using high electron speeds is that the gain in ease of operation is so great as to more than justify the recommendation of 100-120 KV. as the most satisfactory

range of accelerating potentials for general electron diffraction purposes. The reduction in grazing angle of incidence necessary to carry out a complete study of amorphous layers due to polish, and which it was expected would be achieved at about 150 KV., has in fact been reached at 110-120 KV. The use of a decharger can generally be dispensed with when using fast-electron beams except when dealing with extremely small grazing angles, when its adjustment becomes critical.

Acknowledgements.

The author is deeply grateful to Professor G.I.Finch, M.B.E., D.Sc., F.R.S., for his constant interest and encouragement in the work and for his many helpful suggestions.

The author would like to thank Mr.D.P.D.Webb, B.Sc. for his generous assistance during the construction of the apparatus.

Thanks are also due to Messrs C.C.Wakefield and Co. and the Department of Scientific and Industrial Research for the grants which enabled this research to be carried out.

References.

Von Borries and Ruska, Zeits.f.techn.Physik., 20, 229, 1939.

Brubaker and Fuller, J.App.Phys., 16, 128, 1945.

Challice, Ph.D.Thesis, London, 1949.

Charlesby, Finch and Wilman, Proc.Phys.Soc., 51, 479, 1939.

Darbyshire, Zeits.Krist., A.86, 385, 1933.

Ellis, J.App.Phys., 18, 879, 1947.

Field, Rev.Mod.Phys., 18, 353.

Finch and Quarrell, Proc.Roy.Soc., A.141, 399, 1933.

Finch and Wilman, Erg.d.exakt.Naturwiss., 16, 353, 1937.

Haine, Journ.I.E.E., 94 (I) 451, 1947.

Harding, Phil.Mag., 23, 271, 1937.

Hillier and Baker, J.App.Phys., 16, 469, 1945.

Hiller and Baker, J.App.Phys., 17, 12, 1946.

Johannsen, Ann.d.Physik, 18, 385, 1933.

Jones, Scott and Sillows, Proc.Phys.Soc., 62, 333, 1949.

Morton and Hutter, Proc.I.R.E., 32, 546, 1944.

Von Ments and Le Poole, J.App.Sci.Res., B.1, 3, 1947.

Mollenstedt, Nachtricten.Gottingen, 1946, I, 83.

Morton, Rev.Mod.Phys., 18, 362.

Schlessinger, Proc.I.R.E., 32, 483, 1944.

Thomson and Cochrane, Theory and Practice of Electron Diffraction, Passim

Zworikin, Morton, Hillier, Ramberg and Vance, Electron Optics and the Electron Microscope, Passim.