THE

ELECTRON MICROSCOPE

Its Development, Present Performance and Future Possibilities

by

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FOREWORD TO THE AMERICAN EDITION

THE progress in electron microscopy since the closing of the manuscript for the first British edition was both extensive and intensive. The several hundred papers which have appeared in the meantime on applications of electron microscopes hardly give an idea of the extension of their use, as an ever increasing proportion of the instruments is tied down in routine work in hospital and factory laboratories, and the times are long past when almost every successful micrograph was published. No attempt was made, or could be made, to give a full account of this extensive development in the limited space of this monograph. On the other hand I have tried to give a fair and up-to-date report on the basic progress, and I have revised the first edition in every point relating to the theory of image formation, wherever the views formerly expressed required alteration. Here, I must express my indebtedness to Dr. James Hillier of the RCA, for a correspondence which has greatly clarified my views, and led to the discovery of phase contrast as a new factor in image formation. I am also very thankful to Dr. J. Hillier and Dr. E. G. Ramberg for allowing me to see the manuscript of their outstanding paper on "The Magnetic Electron Microscope Objective: Contour Phenomena and the Attainment of High Resolving Power," which describes the ingenious methods by which these authors have now almost completely bridged the gap which at the time of the first edition still separated the practical performance of the electron microscope from the theoretical limit.

A short report on new commercial electron microscopes has been added, and I wish to thank Prof. Louis de Broglie, Paris, for allowing me to include a note on the work which is carried out under his general direction on that most intriguing new instrument, the proton microscope.

D.G.

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FOREWORD TO THE FIRST BRITISH EDITION

THIS monograph is an amplified version of a lecture, delivered on the 4th of March, 1943, before the Cambridge University Physics Society. In its present form it intends to be both an introduction to the electron microscope and a critical contribution to its theory. There is no doubt that in the near future this new instrument will be used by thousands of medical, biological, and chemical workers who have no theoretical acquaintance with electronics. It may be hoped that the fundamentals of electron optics which are briefly explained in Chapters 2 and 3 will enable them to understand the basic operation of the instruments described in Chapters 4 to 11. The last chapters, 12 to 14, are of a more advanced character, and are addressed in the first place to physicists and electronic engineers. An appendix on the diffraction theory of the microscope has been added for the benefit of those who are no optical specialists.

At the present stage of electron microscopy, it was not possible to include more than a cursory survey of the applications of the new instrument, and little more than a hint at laboratory technique which is growing so fast that any review of it would be obsolete by the time it is printed. However, the fundamental development of the instrument has now reached a stage beyond which progress is likely to be slow and difficult. At this point, it seemed appropriate to look into the future, and to try to explore in imagination the avenues of further development. As this is a proverbially risky undertaking, the author is quite prepared to join the ranks of other, more illustrious, prophets who have failed.

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CHAPTER 1

INTRODUCTION

SEVENTY years ago the great optician Ernst Abbé discovered the theoretical limit for the resolving power of the optical microscope. No objective, however perfect from the point of view of geometrical optics, can resolve details finer than about one half of the wavelength of light in the medium in which the object is embedded. Even by using ultraviolet rays and immersion the Abbé limit cannot be substantially reduced below 0.1 microns or 1000 Ångströms.*

It appears that Abbé was depressed rather than elated by his discovery, which meant that he could not go on endlessly perfecting the microscope to which he had devoted his life's work. This can be guessed from his remark ". . . it is poor comfort to hope that human ingenuity will find means and ways to overcome this limit."

Human ingenuity was indeed powerless at Abbé's time, and for some time after, until new physical discoveries opened the way for progress. The first hope emerged, in 1912, when the wave nature of X-rays was discovered. X-rays had indeed the short wavelength required for a supermicroscope, and, unlike the short ultraviolet rays, they had also sufficient penetrating power, but no X-ray microscope could be constructed for lack of X-ray lenses. By the very reason of their short wavelength, matter is not a refracting but a dispersing medium for X-rays. Therefore, geometrical X-ray optics could not develop on the lines of light optics, and although in the last ten years respectable progress has been made in the imaging of objects with X-rays

* 1 micron = $1\mu = 10^{-4}$ cm; 1 Ångström = 1Å = 10^{-8} cm

by means of reflecting elements, it can no longer be expected that the X-ray supermicroscope will ever materialize.

The way for the supermicroscope was opened in the years 1924–1926 by three discoveries. In 1924, de Broglie put forward his bold hypothesis of electronic waves. In 1925, Schrödinger discovered wave mechanics by combining de Broglie's ideas with Hamilton's analogy of dynamics and optics. Finally, in 1926, H. Busch discovered the lens property of axially symmetric electric and magnetic fields, and laid the foundations of geometrical electron optics.

It was a sheer coincidence that Busch's discovery followed so soon on the heels of the other two. When Busch worked out the electron trajectories in axially symmetric fields, he did not think either of wave mechanics, or of the Hamiltonian analogy. Only when he had finished his paper, and showed it to a theoretical physicist, did he hear the exclamation: "What a nice illustration of the Hamiltonian analogy!" It may be remembered that Sir William Hamilton wrote his celebrated Memoirs [1] almost exactly a hundred years before. It is amusing to think that during these hundred years many hundreds of able students studied the Hamiltonian theory of dynamics, and apparently there was not one to ask himself: "Well, if there is such a close analogy between dynamics and optics, what is the dynamical analog of a lens?" Presumably for most students, the Hamiltonian analogy was merely something to be acknowledged with a passing nod *en route* to the esoteric mysteries of Canonical Transformations and the Last Multiplier.

Lest experimental physicists might feel inclined to smile at the impractical attitude of mathematicians, it may be recalled that experimenters had in their hand an electron lens for more than 25 years (1899–1926), without noticing that it was one. This was the so-called *concentrating coil* of cathode-ray tubes, or Braun tubes as they were called at that time. The only thing that can be said in excuse of these experimenters is that their tubes were gas-discharge devices, and if anything queer happened in them, it was easy to blame it on the gas discharge. Only Busch was sufficiently impressed with the curious effects of the concentrating coil to ponder on the problem on and off for 15 years, and well he deserved to be the discoverer of electron optics!

When these discoveries had opened the way, the idea of an electron microscope emerged in many minds.* Work was started almost at once in two places—by Knoll and Ruska at the Technische Hochschule, Berlin, and by Brüche and his collaborators in the A.E.G. laboratory.† The first results, still very modest, comparable perhaps with the resolution of a good magnifying glass, were published in 1932. In another 4 years, the electron microscope had just about reached the resolving power of the ordinary microscope, and in 4 more years it had overtaken and beaten it by a factor of about a hundred. At this limit something like a brick wall was reached.

The greater half of this monograph will be devoted to a survey of the development which led the electron microscope to the present limit of about 8Å, where it has come to a stop. Toward the end, suggestions will be discussed by means of which the brick wall may perhaps be breached. Finally it will be shown that after overcoming this obstacle, there will be another, at about 0.5Å, made of much more impenetrable material than bricks. There is no detail in Nature finer than about 0.5Å, which could be resolved by any microscope, however perfect. In other words, resolution will have to stop here for lack of objects. But the interval between 0.5Å and 8Å probably contains objects of sufficient interest to make such an effort worth while.

* The author remembers discussions on the possibility of an electron microscope in Berlin physicist circles as far back as 1927.

† Allgemeine Elektrizitatsgesellschaft, Berlin.

CHAPTER 2

GEOMETRICAL ELECTRON OPTICS

ALTHOUGH the subject of electron optics has been exhaustively treated in several well-known textbooks,[2, 3, 4, 4a] it will be useful to recapitulate its principles to an extent sufficient to the understanding of the operation of electron lenses. This can be done much more simply now than at Busch's time, since we have been reminded of the Hamiltonian analogy.

Hamilton discovered that the trajectory of a material particle in a conservative field, that is to say, in a field in which the force can be derived from an ordinary, scalar, potential can be interpreted as the path of a light ray in a medium with suitable refractive properties. This follows at once from comparing the principle of least action of dynamics with the principle of least time in optics. According to the dynamical principle, the *action* connected with the trajectory along which a particle will travel from one given point to another will be less than for any other geometrically, but not dynamically, possible path.* The action W is defined as the time integral of the kinetic energy, that is,

$$W = \int \tfrac{1}{2} m v^2 dt$$

and as the velocity v is $\frac{ds}{dt}$, where ds is the length of a path element, this can be also written

$$W = \tfrac{1}{2} m \int v \, ds$$

Let us compare this with Fermat's principle of least time, the foundation of geometrical optics. In a medium of refractive index n, the velocity of light is $\frac{c}{n}$ if c is the velocity *in vacuo*.

* With the silent clause that the two given points must be sufficiently close together, so that the trajectories which can start from the first do not come to a focus before they reach the second. This does not restrict the analogy, as the requirements are precisely the same in optics.

Therefore, the time of transit between two points is

$$\frac{1}{c}\int n\,ds$$

and this must be a minimum. Comparing the two laws, we see that the dynamical trajectory and the light ray become identical *if the refractive index of the medium is assumed proportional to the electron velocity*. The proportionality constant is obviously of no importance.

At first sight it might appear that the analogy is very imperfect, as the velocity belongs to the electron whereas the refractive index is a property of the medium, that is to say, it must be a function of the position only. But this difference vanishes in the cases most important in electron optics, in which only *one* cathode is considered, which emits electrons with initial velocities negligible as compared to the velocities which they acquire in the field. In this case, the refractive index is proportional to the square root of the potential ϕ, measured from the cathode as zero level, and this is a function of the position only.

The analogy extends also to cases in which the initial velocities cannot be neglected. Let ϕ_0 be the initial energy of an electron emitted by the cathode, measured in potential units, *e.g.*, in volts. When this electron comes to a point of potential ϕ, its velocity measured in the same units will be $\sqrt{\phi + \phi_0}$. The refractive index will not be the same for all electrons, but this also has its optical counterpart in the *chromatic dispersion* of ordinary media. We talk, therefore, also in electron optics of *chromatic effects,* meaning by this the effects of initial velocities, or of varying cathode potentials.

In the case of a thermionic cathode with absolute temperature T, the average energy of an electron leaving the cathode will be 2kT ergs,* where k is Boltzmann's constant, 1.37×10^{-16}

* The average energy of electrons in an electron cloud of temperature T is $\frac{3}{2}$kT or $\frac{T}{7{,}730}$ volts. But more fast electrons will leave the cathode than slow ones, therefore, the average energy of the *emitted* electrons will be larger by a factor $\frac{4}{3}$.

ergs per degree. Expressed in electron-volt units, or shorter in "volts," this is $\frac{T}{5800}$ volts. For oxide-coated cathodes, this is about 0.17 volts, and for tungsten cathodes 0.43 volts. Calling this average initial value ϕ_0, it is convenient to define the refractive index as follows

$$n = \frac{\sqrt{\phi + \phi_0}}{\sqrt{\phi_0}} \tag{1}$$

In this expression, the arbitrary constant factor has been disposed of in such a way as to make the root mean square refractive index unity at the cathode. This is only slightly different from the average refractive index.

Equation (1) discloses an important quantitative difference between light and electron optics. Whereas the refractive index of optical media varies only in the limits 1–2, in an electronic device in which electrons emitted by a barium cathode are ultimately accelerated to 100,000 volts the refractive index varies in the limits 1–1,000. This is the main intrinsic advantage of geometrical electron optics, to which we shall return several times.

We can now immediately explain the lens effect of an axially symmetrical electric field, of which figure 1 shows an example. The equipotential surfaces are figures of revolution. They intersect the axis at right angles, and in the neighborhood of their vertex they are approximately spherical. If we imagine the space between these surfaces filled with a medium of a refractive index proportional to $\sqrt{\phi}$, we obtain a succession of lenses, somewhat distorted outside, but of the correct shape near the axis. As a combination of aligned lenses is again a lens, the imaging effect of the electric field is immediately evident. In the figure, increasing refractive index is indicated by shading of increasing density.

The explanation of the magnetic type of lens is somewhat more complicated. The force exerted by a magnetic field on a moving electron is at right angles both to the direction of the field and to the direction of motion. The effect is a twisting or

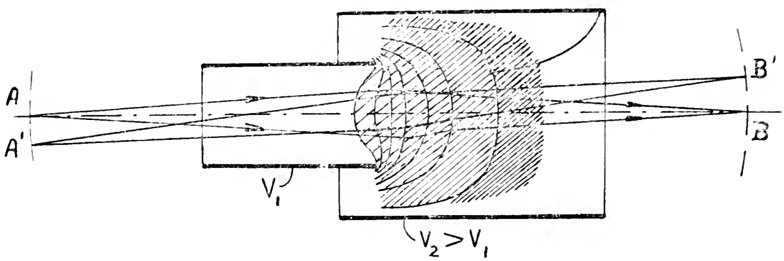

FIG. 1. Example of electrostatic lens

rotation of the electron trajectories around the axis, as shown in figure 2, an effect which has no counterpart in ordinary optics.

Forces of this kind have not been considered by Hamilton. However, starting from pioneer work by K. Schwarzschild, W. Glaser [5] could show that the motion of electrons in a magnetic field can be also derived from Fermat's principle by means of a suitable refractive index. This is of a very unfamiliar type, as it depends not only on the value of the velocity, but also on its direction, and little is gained by using it. Fortunately in the case of axial symmetry, it can be replaced by a much simpler law of a more familiar type.

This simplification is obtained by abstracting from the rotation around the axis and considering, instead of the trajectories, their projections by coaxial circles on a meridian plane, *i.e.*, on a plane passing through the axis. The rotation obeys a simple law. Let us call M the magnetic flux through a coaxial circle of radius r, and let v_t be the tangential velocity of an electron. If the electron proceeds from a position 1 to a position 2, its angular momentum mrv_t around the axis will suffer a change

$$(mrv_t)_1 - (mrv_t)_2 = \frac{e}{2\pi c}\,(M_1 - M_2) \qquad (2)$$

That is, *the angular momentum changes proportionally to the magnetic flux which passes between the two coaxial circles drawn through the initial and final positions respectively.*

This result can be put into an even simpler form if we introduce the vectorpotential **A**, from which the vector **H** of the magnetic field intensity can be derived by the operation **H** = curl **A**. In the case of axially symmetrical fields the vector **A** has always tangential direction, therefore, we can write it in the following as a scalar quantity, A. This is connected with the magnetic flux M by the simple relation

$$M = 2\pi r A \qquad (3)$$

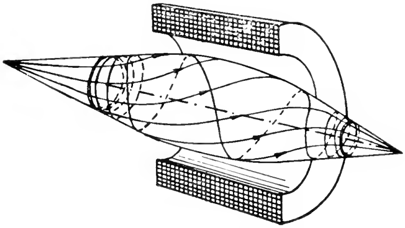

FIG. 2. Electron trajectories in magnetic lens. Curves orthogonal to the trajectories are drawn to show the variation of the pitch

The magnetic field lines, *i.e.*, the meridian curves of the tubes which carry a constant flux M have therefore equations $rA = \text{const.}$ Their shape is shown in an example in the upper part of figure 3.

The equation (2) becomes now

$$r\left(mv_t - \frac{e}{c}A\right) = \text{const.} \qquad (4)$$

The expression in brackets can be interpreted as the *total* momentum of the electron in the magnetic field. It consists of the mechanical momentum mv_t and a term of electromagnetic origin, $-\frac{e}{c}A$. (K. Schwarzschild, 1903.) Familiarity with this concept is a great help in understanding the somewhat complicated motion of electrons in magnetic fields.

If the electron has initially started from the axis, or if it has started anywhere *outside the magnetic field* with zero or negligible velocity, which is almost always the case, the constant at the right side of equation (4) becomes zero, and we have $mv_t = \frac{e}{c}A$, or

$$\tfrac{1}{2}mv_t^2 = \frac{e^2}{2mc^2}A^2 \tag{5}$$

This means that the kinetic energy corresponding to the tangential, *i.e.*, rotational motion of the electron has a potential of its own. Let us now combine this with the energy equation

$$\tfrac{1}{2}mv^2 = \tfrac{1}{2}m(v_m^2 + v_t^2) = e\phi \tag{6}$$

where v_m is the velocity in the meridian plane. Subtracting the two equations we obtain

$$\tfrac{1}{2}mv_m^2 = e(\phi - \frac{e}{2mc^2}A^2) \tag{7}$$

which shows that the motion in the meridian plane can be derived from a potential

$$U = \phi - \frac{e}{2mc^2}A^2 \tag{8}$$

This shall be called the *equivalent electrostatic potential* of the magnetic lens. It is also valid for any combination of electric and magnetic fields.*

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* On the problem of replacing Glaser's general refractive index by simpler expressions if a second integral of the dynamical equations is known, *cf.* Opatowski.[6] The equivalent potential appears to have been first used by C. Stoermer, Ann. Physik, **16**, 685 (1933), and also by L. Brillouin,[7] in 1941, in a less general form.

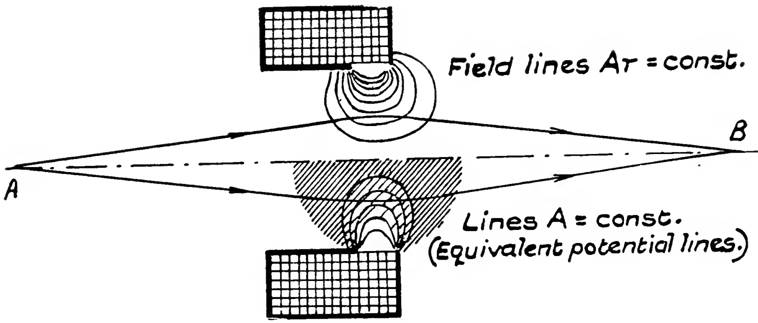

FIG. 3. Magnetic lens

This at once explains the lens effect of axially arranged coils. By its definition (3) the vectorpotential **A** is zero at the axis and increases approximately linearly with the radius r. This means that the equivalent potential decreases outside the axis and that the magnetic field has a repellent effect, *i.e.*, it drives the electrons back, toward the axis. Therefore, a magnetic lens is always a condensing lens. An example is shown in the lower half of figure 3. Increasing equivalent potential is indicated by increasing density of shading.

Having shown that axially symmetrical fields act as lenses, it remains to investigate whether they are *good* lenses. Of the numerous defects which lenses can have, only three are of interest in microscopy: the spherical aberration, the chromatic aberration, and the coma.

Spherical aberration, the most important of the lens defects in electron microscopy, is illustrated in figure 4. An axial point is imaged as a point strictly speaking only while the imaging rays are infinitesimally close to the axis. These are called *paraxial* rays, and their intersection with the axis is the paraxial or Gaussian image. At larger angles α, the intersection moves away from the Gaussian point by a distance which is called the longitudinal spherical aberration, and which in first approximation is proportional to α^2. If a screen p is placed at the Gaussian point, the bundle will intersect it instead of in a point in a disk of a diameter proportional to α^3. This diameter is called the transversal spherical aberration. In fact, the bundle has its smallest cross section not at p but at m, which is called the disc of minimum confusion. Its radius is one quarter of the spherical aberration. When an electron lens is focused, it is always m, not p, which is made to coincide with the screen or plate.

Scherzer[8] has proved the important theorem that in electron lenses, whether electrostatic, magnetic, or combined, the spherical

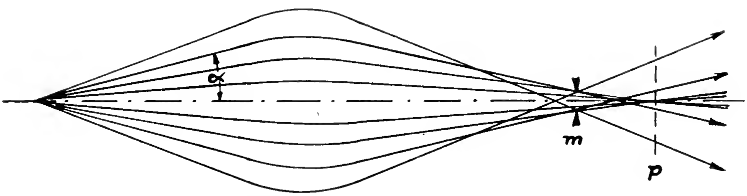

FIG. 4. Spherical aberration

aberration can never be eliminated as long as there are no space charges or currents in the space traversed by electrons. All lenses used to date in electron optics fall into this class. The spherical aberration has always the sign as shown in figure 4, *i.e.*, the strength of the lens increases with the angle α. In optics this is called an *undercorrected* lens. Corrected electron lenses are impossible.

Scherzer has proved also that the same fundamental difficulty exists in the case of chromatic aberration which is illustrated in figure 5. The faster electron (dotted line) will intersect the axis always *beyond* the slower electron. This defect cannot be corrected, because dispersing electron lenses cannot be realized with the means employed in present-day electron optics. We have proved this in the case of magnetic lenses. In electrostatic lenses, it is possible to realize a slight dispersing effect in a part of the field, but this is always more than compensated by the condensing effect. Quantitative examples will be given later.

The spherical and the chromatic aberrations are the only lens errors which exist at axial and at extra-axial points of the image field. The other defects—coma, astigmatism, image curvature, and distortion—are zero at the axis, and manifest themselves increasingly with the distance from the axis. Of these, only the coma increases with the first power of extra-axial distance, therefore, this is the only one of importance in microscopes in which the field is seldom more than a few degrees of arc. The coma is illustrated in figure 6. A lens, corrected for spherical aberration but not corrected for coma, images extra-axial points in the shape of an arrowhead pointing toward the axis. The point of the arrow corresponds to the ray which passes through the center of the lens; all other rays strike the screen at a greater distance from the axis. Rays passing through coaxial circles of the lens strike the screen again in circles; these are, however, not concentric, but fit between two straight lines which pass through the point A' and form an angle of 60°. This is the characteristic coma figure which increases linearly with the distance from the axis.

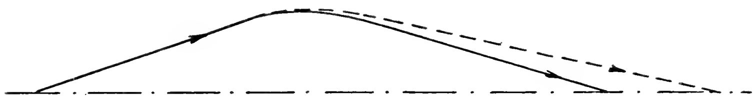

FIG. 5. Chromatic aberration

It is possible to correct electron lenses for coma alone, but this is of little interest as long as the spherical aberration cannot

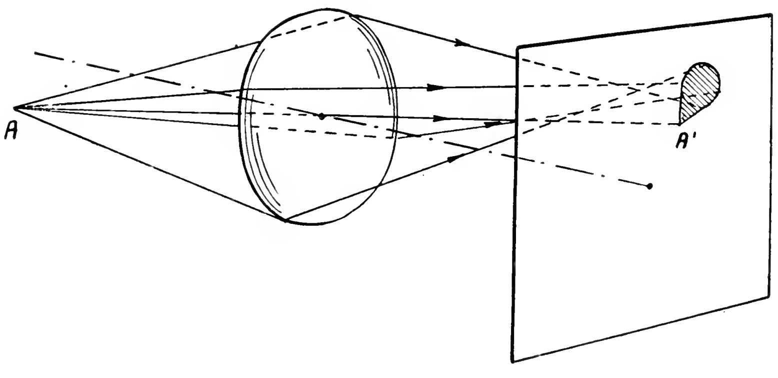

FIG. 6. Coma

be eliminated. Optical microscope objectives which are corrected for spherical aberration and coma are called aplanats. The most perfect specimens of the optical art, the apochromats, are aplanatic and chromatically corrected for three different spectral colors. Electron microscope objectives, however, are *entirely uncorrected.* Yet, these primitive lenses, which as regards optical perfection can be perhaps compared to a dewdrop, have beaten the marvels of the optical art by a factor of almost a hundred. How has this been achieved?

It is known that even the worst lens approaches perfection if its aperture is reduced to almost nothing. The high quality of present-day electron microscopes has been achieved by this simple artifice of using objectives with pinhole apertures or their equivalents. This is a line which light optics could never follow, as two phenomena would soon set a limit to it: diffraction at the aperture and insufficient light intensity. In electron optics, however, it was highly successful for two reasons. The first is the smallness of the diffraction effect, due to the extreme shortness of the de Broglie wavelength of the electrons, which will be discussed in the next chapter. The second reason is that the intensities which can be realized in electron optics are enormously greater than those available in light optics.

Intensity is defined in optics as the light flux per unit area at right angles to the stream and unit solid angle, *i.e.*, per cm^2 and *steradian.* It is known that the intensity divided by the square of the refractive index of the medium is an invariant along a beam of light. This means that if the light originates in a medium of refractive index n_0 and the original intensity is I_0, the intensity I which can be produced in this beam in a medium n cannot be more than

$$I = I_0 \left(\frac{n}{n_0} \right)^2 \tag{9}$$

though it can be less if some light has been lost by absorption or reflection. With the media available in light optics this means that the intensity can be increased at the best three to four times.

By the Hamiltonian analogy, the electron optical equivalent of intensity is the electron current per unit cross section and unit solid angle, and equation (9) is immediately applicable if we replace the square of the refractive index by the potential ϕ, *i.e.*,

$$I = I_0 \left(\frac{{}^0\phi}{\phi} \right) \tag{10}$$

We have already seen that the factor of I_0 can approach a million. In order to obtain an exact expression for the intensities obtainable with thermionic cathodes, it is necessary to apply equation (10) to every one of the electron groups with homogeneous velocity which are emitted by the cathode, and form the average of the quantity $\frac{I_0}{\phi_0}$. This has been first carried out by D. B. Langmuir,[9] and the result is

$$I = 3{,}700 i_0 \frac{\mathrm{V}}{\mathrm{T}} \tag{11}$$

where i_0 is the current density at the cathode, *i.e.*, the current per unit area (not per unit area *and* unit solid angle), T is the temperature of the cathode in degrees Kelvin, and the numerical factor is $\frac{11{,}600}{\pi}$. V is the accelerating potential, measured in volts. For example, with a barium oxide cathode with $\mathrm{T} = 1000°\mathrm{K}$ and $i_0 = 0.2$ amp/cm^2 we obtain, if $\mathrm{V} = 60{,}000$ volts, $I = 44{,}400$ amp/cm^2 steradian, which is 6.9×10^5 times more than the maximum intensity $\frac{0.2}{\pi} = 0.064$ amp/cm^2 steradian at the cathode.

In light optics, the intensity is a measure of the energy concentration in the beam, as the energy of a light beam does not change during its propagation. In electron optics, however, the energy of the electrons increases continually with acceleration, therefore, in order to obtain a fair comparison, we must calculate not the intensity I, but the power I.V per unit cross section and solid angle. This is $\frac{3{,}700 i_0 \mathrm{V}^2}{\mathrm{T}}$ watts/cm^2 steradian.

The highest intensity available in light optics is that of sunlight. The *solar constant, i.e.*, the solar radiation which falls on a square cm outside the terrestrial atmosphere is 1.94 cal/min or 0.135 watts. This corresponds to a black body temperature of 5770°K, and to an energy flow of 2,000 watts/cm^2 steradian. On the other hand a barium cathode, as assumed above, again with an accelerating voltage of 60,000 volts, can produce 2.6×10^9 watts/cm^2 steradian, that is, *more than a million times more.* With a tungsten cathode it is possible to realize a ten times higher figure, and with a point discharge from a cold cathode at least ten thousand times more again. The factor of a million, which can be realized with barium cathodes under ordinary circumstances, sufficiently explains why electron optics could score such successes in spite of the most primitive lenses. We shall see later that by now electron microscopy has about exhausted its enormous initial advantage; further progress is likely to be achieved along lines similar to those of light optics.

The high intensity of electron beams explains why it was possible in electron microscopy to cut down the numerical apertures to one hundredth or even one thousandth of the values which are used in light microscopy. The spherical aberration increases with the cube of the aperture angle, therefore, it could be very effectively reduced by this simple expedient. Reducing the aperture is far less effective in the case of the second important lens error, the chromatic aberration, as this is only in direct proportion to the aperture angle. Everybody who has looked through cheap pocket microscopes will have noticed that though their aperture may be so small as to make satisfactory illumination very difficult, the chromatic fringes still remain rather disturbing. To solve this difficulty, another simple artifice has been adopted in electron microscopy: strictly *monochromatic* illumination by electrons of very homogeneous velocity. But before discussing this further, it will be necessary to consider the other side of the analogy between mechanics and optics.

CHAPTER 3

ELECTRON WAVE OPTICS

UP to here, we have considered electrons as charged particles and, following Hamilton, established the analogy of their trajectories with the light rays of geometrical optics. It is well known that the concept of a *ray of light* is of limited validity, applicable only as long as the physical apertures in the optical system are very large in comparison with the wavelength. It is one of the greatest discoveries of modern physics that the idea of *electron trajectory* is subject to quite similar restrictions.

The bold idea of associating a wave with the motion of a particle emerged first in the head of Louis de Broglie, in 1924. By using the theory of relativity, this immensely powerful tool for physical discoveries, de Broglie could show that from the data of an electron in motion it is possible to define the data of a wave in such a way that their association is relativistically invariant, which in modern physics is equivalent to saying that it *might* have physical sense. He took a further step by combining this result with the other great source of modern physics, quantum theory. Classical physics is continuity physics, one physical datum in it is as good as any other. The only distinguished datum, "*constant of nature*," which occurs in it, is the velocity of light; but no combination of c with the dynamical data of the particle, *i.e.*, its mass m, and its velocity v, will give a length, therefore, as far as classical theory goes, the wavelength of these hypothetical waves could be just anything. Quantum theory, however, gives a new universal constant, the natural unit of action $h = 6.54 \times 10^{-27}$ erg/sec. The dimension of action can be also written momentum $\times$ length. A *wavelength* associated with the motion could be therefore defined as follows:

$$\lambda = \frac{h}{mv} \tag{12}$$

and this *de Broglie wavelength* fits also well into the relativistic scheme.

De Broglie started very far off, in a very general way; it is therefore not surprising that he failed to point out the concrete physical significance of these waves which Einstein was the first to recognize. Stimulated by Einstein's hints, E. Schrödinger, in 1925, approached the subject afresh from a new angle. Starting from the Hamiltonian analogy, he established the equation for the propagation of de Broglie's waves in electro-magnetic fields by that combination of deduction and guessing which is indispensable for every great physical discovery.[10] He showed that by means of his *wave equation* the older quantum theory, which up to then was a collection of somewhat arbitrary looking rules, not always in agreement with experiments, could be put on a solid and general basis which explained even those features of atomic and molecular spectra where the older theory failed. The direct experimental evidence, however, for the de Broglie waves was still outstanding. As early as 1925, W. Elsasser suggested that certain curious features in the reflection pattern of electrons on nickel crystals may be due to de Broglie waves. Fully convincing proof was obtained, in 1927, by Davisson and Germer, and almost simultaneously by G. P. Thomson.[11]

It was known since von Laue's discovery, in 1912, that crystalline bodies (for instance metals), in which the elementary constituents, ions, atoms, or molecules, are arranged in lattices of high regularity, can serve as a sort of diffraction grating for X-rays, as the spacing in these lattices is usually of the same order as the wavelength of X-rays which is equal to or larger than

$$\lambda_x = \frac{12.340}{V} \text{Å}$$

if V is the potential in volts which was used to accelerate the electrons which produced the X-rays.

The de Broglie wavelength of electrons, however, accelerated by V volts is, if equation (12) is expressed numerically,

$$\lambda = \frac{12.2}{\sqrt{V}} \text{Å} \tag{13}$$

For electrons of 60,000 electron-volts (ev) velocity this is about 0.05Å, which is about the same as the shortest wavelength of 250,000 ev X-rays. Therefore, we should expect diffraction patterns of much the same general character. This was indeed found by the experimenters mentioned, who verified also the value of the de Broglie wavelength given by equation (13). Since that time, electron diffraction has become a close rival of X-ray diffraction in the exploration of the structure of materials.

In electron optics, we work in general with apertures which, though very small as compared with those used in light optics, are enormously greater than λ. The smallest apertures used in electron microscopes have a diameter of about 0.001 in. $=$ 0.0025 cm $= 2.5 \times 10^5$ Å, which is five million times larger than the wavelength of 60,000 ev electrons. Neverthless, the diffraction effect is not only noticeable, but constitutes the effective limitation of the performance of electron microscopes.

If a wave starting from a point, a *homocentric* or *stigmatic* wave, passes through a lens, however perfect in the sense of geometrical optics, it can never again be brought together in a point. The reason is that the unavoidable aperture has cut out a part of the spherical wave front, and, to use Schrödinger's expressive words, ". . . the wound torn by the aperture will not heal." Instead of in a point, the source will be imaged in a diffraction figure, calculated, in 1834, by Sir George Airy. This consists of a central disk, *Airy disk*, surrounded by a succession of dark and bright interference fringes of rapidly decreasing luminosity (*cf.* appendix). It is convenient to describe this figure instead of in terms of its actual dimensions by the dimensions of the object which it appears to represent. The radius of the Airy disk, which extends to the first dark fringe, corresponds to an object of a radius

$$r_A = 0.61 \frac{\lambda_0}{a} \tag{14}$$

where λ_0 is the wavelength at the object, *i.e.*, $\lambda_0 = \frac{\lambda}{n_0}$ if n_0 is the refractive index of the medium in which the object is immersed, and λ is the wavelength in a medium $n = 1$. a is the aperture angle, that is, the angle between the marginal rays which pass through the lens aperture, and the optical axis.

This formula was applied by Airy to the apparent diameter of stars and to the resolving power of telescopes. It can be also applied immediately to microscopes, with a restriction: We must assume that the objects are *self-luminous*. Let us assume that we observe two minute particles under the microscope which fulfill this condition. It is evident that they will appear as separate particles only as long as their Airy disks do not overlap completely, so that a minimum of light intensity remains visible between the two maxima. How much is required for visibility, is to a certain degree a matter of convention, but for all practical purposes, it is sufficient to assume that the minimum spacing, which is called the *resolution limit* of the microscope is

$$d_A = 0.5 \frac{\lambda_0}{a} \tag{15}$$

To obtain this formula, we had to assume that the resulting figure for the two particles can be obtained by simple superposition of the intensities of the two Airy disks. This is a legitimate assumption in the case of self-luminous particles, as these emit light independently of one another, therefore, resulting intensities can be obtained by addition. This condition is satisfactorily fulfilled only in the *ultramicroscope* in which small objects are made visible by *dark field illumination, i.e.,* by illuminating them sideways, and in all rigor only in the fluorescence microscope in which the particles are illuminated by ultraviolet light. In all other cases, there will be definite and *small* phase differences between the light waves which appear to originate from the two particles and which are caused by refraction, dispersion or absorption of the illuminating wave. This means that these secondary waves can destroy one another in certain directions, and that the resulting intensity is no longer a simple sum of the component intensities.

This problem of *coherent* light emission was first investigated by Ernst Abbé who published his solution, in 1873. Abbé considered only the somewhat over-simplified special case of parallel illumination and showed that in this case the microscopic image is produced by an optical mechanism essentially different from the one which operates in the case of self-luminous objects. Abbé's theory of the microscope had the bad luck of finding ardent protagonists who claimed for it validity far beyond the limits in which it was proved, and by this did much toward starting a violent controversy which went on for almost forty years.* But for the present we can leave aside the mechanism of image formation, as Abbé's theory leads to the resolution limit

$$d_A = 0.5\frac{\lambda_0}{\sin \alpha} \tag{16}$$

which for the small apertures used in electron microscopy gives much the same result as Airy's equation (15). Airy's original formula, which was valid for small angles only, was also extended to large apertures by Rayleigh and other authors. It is rather fortunate that the suffix "A" can mean equally well "Airy" or "Abbé."

The equations (15) or (16) explain at once the second great intrinsic advantage of electron optics, which enabled the construction of highly successful microscopes with entirely uncorrected lenses. In light microscopy it was possible to make $\sin \alpha$ as large as 0.9, and, by immersing the object into a medium with a refractive index of about 1.6, the resolution limit could be brought down to about one third of the vacuum wavelength. The wavelength of yellow light, however, is about 0.6 μ or 6,000 Å, and for ultraviolet light as used in microscopy about 0.28 μ or 2,800 Å. Therefore, the visual resolution limit remained about 2,000 Å, and even photographically it could not be made better than about 1,000 Å.

* *Cf.* Appendix on Diffraction Theory of the Microscope.

The de Broglie wavelength of 60 kev electrons, however, is, as we have seen, about 0.05 Å, *i.e.*, roughly one hundred thousand times shorter than for visible light. It is therefore possible to make the aperture angle one thousand times smaller than in light microscopy, and still retain a roughly one hundred times better resolution. This, in short, is what electron microscopy has done up to the present.

At the end of the previous chapter, it was mentioned that the reduction of the aperture would have been insufficient by itself to eliminate the *chromatic* error, and that the great progress of electron microscopy was achieved by using *monochromatic* illumination. We can give now a more precise meaning to this concept which we introduced in geometrical optics on the basis of the different refrangibility of electrons of different velocities. In a beam of average energy V ev, in which electrons of energies differing by ΔV ev are present, the degree of inhomogeneity of wavelength can be expressed by the fraction

$$\frac{\Delta\lambda}{\lambda} = -\frac{\Delta V}{2V} \qquad (17)$$

This follows from equation (13). The same applies also if the voltage fluctuates by ΔV. In modern electron microscopes this has been reduced to less than 1 part in 25,000 or even 50,000, so that the de Broglie wavelength varies only by 0.004–0.002 per cent. To appreciate this, we may consider that in optics the sodium lamp is considered as a good monochromatic source. The two D-lines of the sodium spectrum differ by about 0.1 per cent in wavelength, which is fifty to one hundred times more than the corresponding value in a good electron microscope.

It is perhaps more appropriate to base the comparison on the variation of refractive indices in the two cases. A good measure of this is *the relative dispersion*

$$\frac{\lambda}{n}\frac{dn}{d\lambda}$$

In electron optics λ is inversely proportional to $\sqrt{V}$, and thus to

n, therefore, this quantity is always —1. This means that if the wavelength increases by 1 per cent, the refractive index drops by 1 per cent. The corresponding quantity for ordinary glass is about —0.02. But it is more correct to consider

$$\frac{\lambda}{n-1}\frac{dn}{d\lambda}$$

as the corresponding quantity in light optics, as practically all lenses utilize the refraction glass-against-air. This is about —0.07 for ordinary glass. Therefore, even on this basis of comparison, a modern electron microscope has a fifteen- to thirtyfold advantage over an optical microscope with a sodium lamp as light source. Unlike the high intensity and the shortness of wavelength, however, this is not to be considered as an intrinsic advantage of electron optics, but rather as a result of brilliant development work.

So far we have tacitly assumed that because electrons have a wavelength and can be diffracted, they are subject to the same laws as light waves in physical optics. This can indeed be justified to a certain extent, especially if we consider that the curious contradiction between waves and particles exists also in optics. Light behaves as a wave in refraction and diffraction, but when it exchanges energy with matter as in photoelectricity, Raman effect, and Compton effect it behaves like a particle, a *photon* with energy and momentum. The differences in the interaction with matter between electrons and photons can be mostly explained by the fact that at a given energy the latter have a very much smaller momentum. Nevertheless, there is a very great difference between light waves and electron waves, which must be pointed out, though it happens to be of small importance in electron microscopy.

Light waves are electro-magnetic waves *in space.* When Schrödinger laid the foundations of wave-mechanics, it appeared almost self-evident to assume the same of waves of electrons and of matter in general. However, during the further development of the theory, this turned out to be impossible. One electron, it is true, required only a three-dimensional space for its waves, and this could be identified with the space of geometry and electro-magnetism, but two electrons required six dimensions, three electrons nine, and so on. These *hyperspaces* are obviously only convenient fictions, quasi-geometrical interpretations of the somewhat abstract mathematics of quantum theory. If we admit this, it would be illogical to claim an exceptional position for *one electron,* to assume that the waves of the first electron occupy space, but that as soon as a second electron appears, one of the two has to go *outside* with its waves. The reason why in electron optics we can use this strictly speaking inadmissible picture of *electron waves in space* is the slight degree of interaction of electrons in electron beams. The greatest concentration which is realized near the object is usually less than 10^{12} electrons/cm^3 and in all the rest of the beam it is very much less.* What small interaction there is, can be usually considered as a *space charge effect.* This means that only *one* electron is considered as a particle at a time, the others are accounted for by imagining their charges spread out continuously over the space occupied by the beam. In this sense, the problems of electron optics are always *one electron problems* and Schrödinger's original picture can be used with impunity.† Moreover, in calculating the diffraction effects at apertures, we can apply the well-known methods and results of optics without any modification. Though the momentum of an electron is very much larger than the momentum of a photon of the same energy, it is not sufficient to shake a physical aperture appreciably. Important differences arise only in the case of scattering by single atoms or molecules, to which we shall return later.

To round off the picture, we must give a somewhat more precise meaning to the *intensity* of electron waves. If we calculate, *e.g.,* the Airy diffraction figure and the densities, which we must expect in it, this does not mean of course that an elec-

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* Electron concentration in metals, in which the quantum mechanical interaction becomes very marked, are of the order 10^{24}/cm^3.

† The improvements in the quantum mechanics of electrons due to Dirac can be ignored in electron optics, as the *spin* of free electrons cannot be observed.

tron will distribute itself over the photographic plate according to this law. *One* electron can appear only in one spot and can react only with *one* photographic grain or at most with a few, by secondary effects. If we expose the plate long enough, the density will gradually approach the calculated distribution. This is the essence of the statistical interpretation of wave mechanics, according to M. Born. The intensity distribution is a statistical law, which, if the numbers involved are large enough, will be very closely satisfied by the electrons, in the same way as gas molecules fulfill the gas laws, or human beings—often rather unwillingly—fulfill the laws of mortality statistics.

CHAPTER 4

ELECTRON MICROSCOPES WITHOUT LENSES

THESE devices, also called *simple microscopes,* utilize in a very direct and ingenious way the fundamental advantage of electron optics, the possibility of realizing enormous refractive indices. Everybody knows that the mercury column in a thermometer appears somewhat enlarged by the refraction of the glass. If we had a glass with a refractive index of say a hundred, the thermometer would look very nearly like a solid rod of mercury. In electron optics such refractivities and even larger ones can be realized, and this fact is the basis of the two types of *simple microscopes* which are without counterpart in light optics.

In the cylindrical microscope, invented by R. P. Johnson and W. Shockley,[12] a thin emitting filament is stretched in the axis of a cylindrical glass envelope which is coated with a fluorescent powder. This is shown in figure 7. The anode can be a wire spiral in contact with the tube wall, but this is not essential. It is known from the theory of cathode ray tubes that the usual fluorescent powders are good secondary electron emitters, and will maintain themselves very nearly at anode potential, provided that the anode is not too far off or shielded. The electrons move outwards radially in very nearly straight lines and produce a strongly magnified projection of the emitting patches of the filament surface on the tube wall.

The spherical type, shown in figure 8, was first used by E. W. Müller.[13] The fluorescent envelope is spherical, and the cathode is a wire which ends in a hemisphere with a very small radius, approximately in the center of the bulb. The main difference between the two devices is that in the spherical type relatively small voltages (of a few thousand volts) are sufficient to produce

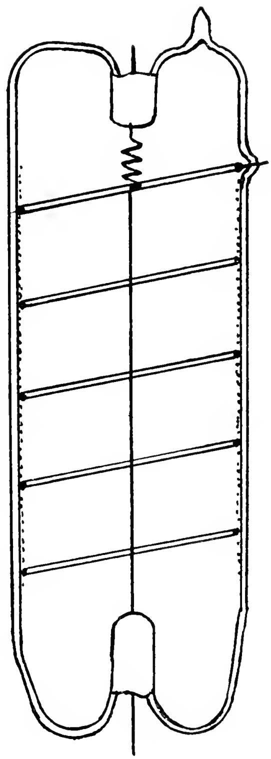

Fig. 7

cold or *auto-electric* emission from the sharp point. The magnifications obtainable with these simple devices are of the order of a few hundred thousand, and their resolving power is remarkably high, of the same order as of the supermicroscope, but their field of application is rather limited. They have been used by M. Benjamin and R. O. Jenkins [14] in a very thorough study of the auto-electric emission of various clean and contaminated metals. Their cathode was a thin wire of the metal to be investigated, etched to a fine point, approximately hemispherical, with a curvature radius of about 2.5×10^{-5} cm. As the bulb radius was 5 cm, the magnification was about 200,000. The pointed wire was welded to a filament, as shown in figure 8, which enabled the sample to be heated during observation. Figure 9 is an example of the many beautiful patterns obtained in this investigation. It shows the emitting patches of a minute single crystal of nickel and the changes which are produced in them by heat treatment.

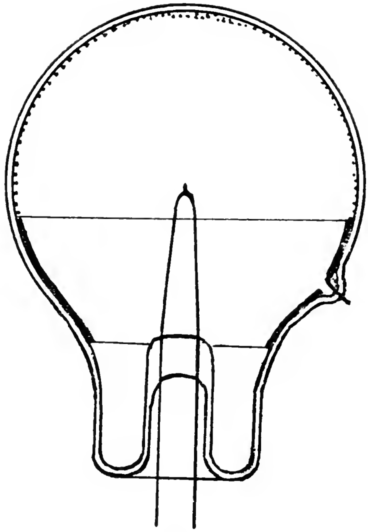

FIG. 8

On watching these microscopes in action, one can often see very small bright points moving hesitatingly about on the surface. Some of these are probably caused by single ions, crawling about on the crystal and producing increased local emission. To make a single ion visible is an achievement unmatched up to now by any of the more complicated microscopes.

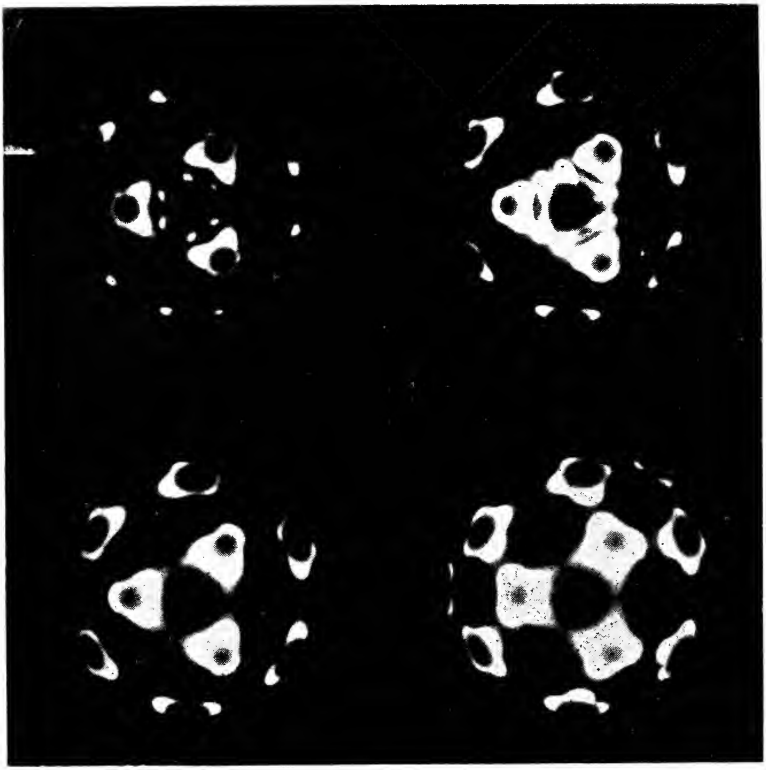

Fig 9. Photographs taken with the spherical microscope of M. Benjamin and R. O. Jenkins (G.E.C., Wembley), showing progressive activation of nickel crystal point by heat treatment. The original magnification on bulb with 100 mm diameter was about 200,000

CHAPTER 5

ELECTRON MICROSCOPES OF THE TRANSMISSION TYPE (SUPERMICROSCOPES)

THE electron microscopes in which the image is formed by means of electron lenses can be conveniently divided into two classes: for self-luminous and for non-self-luminous objects. A self-luminous object in electron optics is a cathode; it may be thermionic, photo-electric, radio-active, or a secondary emitter. The choice of objects is therefore rather restricted, but there are also other factors which restrict the importance of this type of instrument. In the other type of microscope, the image is formed by electrons emitted by a separate cathode, which strike the object after having been accelerated to a rather high velocity. Only transmitted electrons can be used, as true elastic reflection is a very rare process. Electrons which reemerge by diffusion with reduced velocity cannot be distinguished from secondary electrons, and microscopes which utilize these belong strictly speaking to the first class. Such *reflecting* microscopes, the electronic counterparts of the metallurgical microscope, were up to now rather unsuccessful, and they are not likely to have a great future, the more so, as we possess now in the *scanning microscope* (to be discussed in a later chapter), a fully satisfactory tool for surface investigations.

The first *electrostatic* microscope for self-luminous objects was constructed by E. Brüche and H. Johannson,[15] and started operating in 1932, almost simultaneously with the first *supermicroscope*. In the following, we shall deal only with the second type, not only because of its wider scope and greater general interest, but also because the microscope for self-luminous objects appears to have completed its development, about the year 1936.

References to comprehensive accounts may be found in the bibliography.[16] The supermicroscope, however, has become a success only in the last years, and there are reasons to believe in its considerable further development.

The transmission type or *supermicroscope* is called *magnetic* or *electrostatic* according to the type of its objective. The magnetic microscope is the older, and for the time being the more successful type. Its official birth year is 1932, as in this year M. Knoll and E. Ruska published their first paper [17] on it. The first successful electrostatic microscope was constructed, in 1939, by H. Mahl.[18]

In the ten years which followed the first publication by Knoll and Ruska the magnetic microscope has changed little in principle, but every detail was perfected by long patient work, with the result that the performance of the instrument was improved more than a thousand times. It is almost impossible to do full justice to work of this kind. The enumeration of principles, means, and devices is likely to leave out just those difficulties which it took the experimenters all their energy and patience to overcome. Only those who have worked with large and complicated experimental apparatus know what a challenge they are to the "cussedness of objects," and how often those difficulties which it took most time to overcome appear afterwards too trivial even to be mentioned in the publications.

Among the workers to whom the present development of the magnetic supermicroscope is mostly due may be mentioned: M. Knoll, E. Ruska, B. von Borries and M. von Ardenne, in Germany; E. F. Burton, J. Hillier, W. H. Kohl, A. Prebus, A. W. Vance and V. K. Zworykin, in the United States and Canada; L. Marton, in Brussels and later in the United States; and L. C. Martin, in Britain.* The development of the magnetic microscope to the commercial stage owes most to two great electrical firms: Siemens and Halske, of Berlin, and the Radio Corporation of America. In 1935, Siemens established a special

* The first British electron microscope was constructed by Metropolitan Vickers Electrical Co., Ltd. in 1936.[28]

electron-optical laboratory for E. Ruska and his numerous collaborators.† The R.C.A. took an interest in the supermicroscope, in 1938, when L. Marton (until then in E. Picard's laboratory in Brussels) continued his researches in the Camden Laboratories of the R.C.A. under V. K. Zworykin.

J. Hillier and A. Prebus built, in 1938, a highly successful supermicroscope at Toronto University, under the supervision of E. F. Burton and W. H. Kohl. In 1940, J. Hillier joined the staff of the R.C.A., and with A. W. Vance developed the first commercial supermicroscope, details of which were first published in 1941.[19-29]

The principle of the magnetic microscope is explained in figure 10 which shows the electron-optical arrangement side by side with its optical counterpart, the projection microscope.

The electron source in modern magnetic microscopes is a thermionic cathode, almost always a tungsten wire, bent to a V, pointing toward the object. This produces, in combination with an electron gun arrangement which will be discussed later, a divergent electron beam which is made approximately parallel by a first magnetic lens, the *condenser lens.* The beam next strikes the object which in some cases can be suspended in the meshes of a fine gauze, but in most cases is resting on an extremely thin supporting membrane. This is followed by a second magnetic lens, the *objective,* which forms a fifty to one hundred times magnified intermediate image. This in turn is magnified in about the same ratio by a second lens, the *projector lens,* and forms the final image, with a magnification of the order of several thousand, on the photographic plate.

In the early instruments, of which Ruska's magnetic microscope of 1934, shown in figure 11, is an example, the electrons were produced by a gas discharge from a cold aluminum cathode, in the same way as in high speed cathode ray oscillographs. In

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† Reinhold Rüdenberg, at that time Chief Electrician of the Siemens-Schuckert A.G., patented, already in 1931, several fundamental principles of the electron microscope, but experimental work in the Siemens concern did not start until 1935.[85]

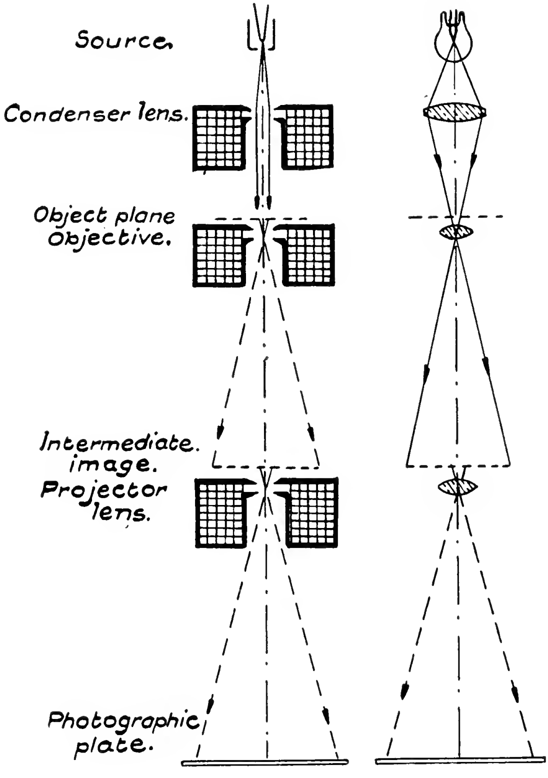


Fig. 10

fact, most of the technique of these early microscopes originated in high voltage cathode ray oscillography, and the first instrument of Knoll and Ruska was a slightly modified oscillograph. (For the history of this development, cf. reference 2.) In a gas discharge tube, it is almost impossible to keep the voltage variation below about 1 per cent; it was therefore a very essential step when this was replaced by a high vacuum—hot cathode discharge.

The magnetic lenses very soon assumed a shape from which later designs have departed only in minor points. Figure 12 shows details of the design of the objective in Ruska's microscope of 1934. The whole coil is enclosed in the vacuum envelope. As outgasing of the insulation and of the long and narrow interstices

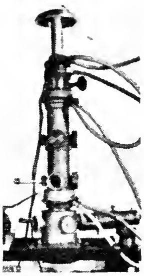


FIG. 11. E. Ruska's magnetic electron microscope of 1934

between the windings would be a very slow process, the whole coil is encased in metal. The gap in the iron casing is covered with non-magnetic material, such as brass. The pole pieces are interchangeable. In addition to these features, which recur in practically all modern microscopes, Ruska's design provides water cooling, a complication which was later found unnecessary.

Comparing the optical systems of an electron microscope and an optical microscope of the projection type, which are shown side by side in figure 10, it can be seen that they are qualitatively identical. There is, however, a striking quantitative difference in the illuminating beams. Optical microscopes which aim at high resolution are fitted with wide-angle illuminator systems. The advantage of wide-angle illumination for a good resolution was a result of Abbé's theory of the microscope, and though later investigations proved that this conclusion was really a rather daring extrapolation from the original theory, the result remained true, though it had to be buttressed by another theory. The best result is obtained when about two thirds of the aperture is filled by the illuminating beam, but this condition is not critical.

In electron microscopy, however, it appears from figure 10 as well as from practically all the literature of the subject, as if a radically different method had been chosen: parallel illumination. The difference is only apparent, as can be seen from a simple consideration.

Figure 13 shows a strongly magnified picture of the electron gun and a few characteristic electron trajectories. In order to make the drawing clearer, the cathode is assumed flat as in cathode ray tubes instead of hairpin-shaped. This makes practically little difference, as the active area of the cathode is usually only a few tenths of a mm in diameter. Electrons will start from every point of this area with different initial velocities and in different directions. Three of these trajectories are shown in the ilustration for three selected points, one starting at right angles to the cathode, the other two tangentially.

The accelerating field penetrates into the aperture of the shield and produces in it a very powerful lens. This bends the trajectories which have started at right angles to the cathode strongly toward the axis which they cross at a distance of usually 1 mm or less from the cathode. This point would be the image of the cathode, if the electrons all started at right angles to it, which would be the case if they had no initial velocities. Because of the tangential velocities the *cross-over* will be a disk. Strictly speaking, this disk has no well-defined limits. The intensity distribution in it is a reproduction of the Maxwellian velocity distribution of the electrons, and falls off with the radius r according to a law exponent $\left[-\left(\frac{r}{a}\right)^2\right]$. The constant, a, can be taken to represent the equivalent diameter of the disk. The cross-over was first discovered and thoroughly discussed by Maloff and Epstein.[30]

Outside the electron gun the trajectories pass through the condenser lens, and are bent parallel to the axis. (Collimated.) Only the principal trajectories, which have started at right angles to the cathode, can be exactly collimated. There remains a certain divergence in the beam which is illustrated by the angle δ. The root mean square value of it can be calculated as follows: The mean square tangential velocity components at the cathode are $\frac{1}{2}mv_x^2 = \frac{1}{2}mv_y^2 = \frac{1}{2}\mathrm{kT}$, therefore, the mean square tangential velocity is $\frac{1}{2}mv_t^2 = \frac{1}{2}m(v_x^2 + v_y^2) = \mathrm{kT}$, or in volts

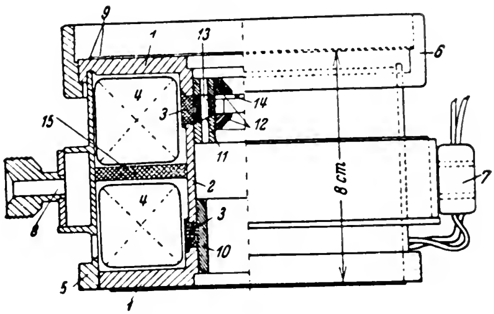


FIG. 12. Design of magnetic objective
(*E. Ruska*, 1934)

$\frac{T}{11,600}$. If these electrons are ultimately accelerated by v volts the root mean square divergence is

$$\delta^2 = \left(\frac{T}{11,600}V\right)^{\frac{1}{2}}$$

In the case of a tungsten cathode with $T = 2,600°K$ and $V = 60,000$ volts this is very nearly 2.10^{-3}.

Whenever the cathode is imaged on the specimen, the divergence cannot be less than this value, though it can be more, as a consequence of lens errors and space charge effects. But this divergence, small as it appears, is of the order of the apertures which can be allowed in uncorrected objectives. Numerous measurements, in particular those by R. F. Baker, R.C.A., demonstrate that the resolving power and contrast improve very appreciably if the illuminating angle is reduced considerably below this limit. This, however, is possible only if the current density at the specimen is reduced by defocusing below the maximum value which can be obtained by focusing the cathode or the cross-over directly on it, or if part of the beam is cut out by an aperture in the center of the condenser lens. Such arrangements have proved very successful, and the modern tendency in electron microscopy is to operate with almost parallel illuminating beams, with a divergence of less than 10^{-4}, down to a few 10^{-5}. In the absence of the specimen these beams fill only a small fraction of the objective aperture.*

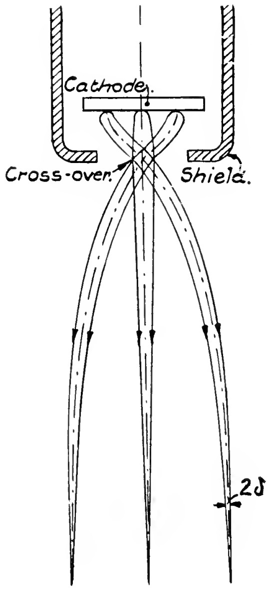


Fig. 13. Trajectories in electron gun

The limitation of the aperture by a physical diaphragm in the objective is a comparatively recent improvement in electron microscopy. It was introduced, in 1939, almost simultaneously by E. Ruska and B. von Borries [31] and by M. von Ardenne. Its introduction resulted in about doubling the obtainable resolution. The extremely fine bores are rather difficult to make, and certain disadvantages result from it. After prolonged operation a semiconducting substance of unknown origin forms a deposit on the diaphragm.[24] This acts like a very poor electrostatic lens and

* Cf. J. Hillier and R. F. Baker, A Discussion of the Illuminating System of the Electron Microscope, Jour. Appl. Phys. **16,** 469–483, August 1945.

reduces the obtainable resolution by a factor of three or more, that is, below the value obtainable without a physical aperture. Therefore, the diaphragm has been again discarded in most electron microscopes operating at the present time.

It appears at first sight that such microscopes without a physical aperture present an entirely different problem. Most investigators have indeed assumed that as the divergence of the electron beam scattered by the object is several times the divergence of the original beam, these microscopes could be considered as operating with parallel illumination, and they explained image formation by Abbé's theory, applicable to such cases.[31, 32] If this were so, it would be hard to understand why the physical aperture improved the definition only by a factor of about two. Even if the electrons were scattered only to twice the original divergence, the spherical aberration which grows with the cube of the angle ought to reduce the resolution by a factor 8. This paradox will be explained in the next chapter. For the present, it will be sufficient to note without proof that there is no very essential difference between the limitation of the aperture by a physical diaphragm and limitation by the illuminating beam. The effect of an illuminating beam, collimated as well as possible, is much the same as using a physical objective aperture corresponding to a little more than the root mean square divergence angle of thermal origin, calculated above.

It was mentioned in chapter 2 that even with apertures of the order of 10^{-3} radians, very steady driving voltages are required in order to reduce the *chromatic aberration* to the same level as the diffraction error and the spherical aberration. The power of a magnetic lens can be written

$$\frac{1}{f} = \text{const.}\frac{J^2}{V} \qquad (18)$$

where f is the focal length and J the current through the coil. A variation of f will have two effects. One is the *chromatic change of focus* which will cause the image of a point on the axis to appear as a disk. The other is the *chromatic change of magnifica-*

tion. If f changes but the position of the lens does not change appreciably, this effect is very nearly the same as if f had been kept constant and the lens shifted along the axis, and the result will be a radial shift of extra-axial points. If it were possible to make the coil current J strictly proportional to $\sqrt{V}$, both effects could be avoided. But this is practically impossible, as lens coils have very high inductivities and J cannot be made to follow $\sqrt{V}$ instantaneously. Therefore, the only possibility is to keep both J and V constant, and if the variation of f is to be kept within given limits, it is convenient to prescribe for J tolerances half as wide as for V. In the best modern magnetic microscopes these tolerances are about 0.002 per cent for V and 0.001 per cent for J.

It can be said that the development of the magnetic electron microscope, from about 1934 onward, went exactly parallel to the improvement in stabilization of the driving circuits. By 1940, this development had reached a stage at which further improvements in the stabilization, very difficult anyhow, would have brought no corresponding improvements in the performance of the microscopes. This progress was obtained, however, at the price of very elaborate and expensive auxiliary equipment of the magnetic microscope.

Electron microscopes, operating with electrostatic lenses only, are free from this necessity. The power of an electrostatic lens can be written approximately

$$\frac{1}{f} = \text{const.}\left(\frac{U}{V}\right)^2 \qquad (19)$$

where U is the potential difference between the electrodes of the lens. It has been assumed that there are in it only two different electrode potentials. By making $U = V$ this becomes a constant. This means that one group of the lens electrodes must be connected with the cathode, the other with the accelerating potential V. Such *fixed focus* or *unipotential* lenses were first used by Brüche and his collaborators in the A.E.G. laboratory, from 1932 onward. H. Mahl [18] utilized them, in 1939, for the first time in a high voltage electron microscope. C. H. Bachman and S. Ramo,[33, 34] in the Schenectady laboratory of the General Electric Company, constructed, in 1942, the first commercial electrostatic microscope on this basis. The great advantage of microscopes of this type is that they can entirely dispense with the elaborate voltage and current stabilizers which are indispensable in the case of the magnetic type.

In spite of this advantage, the electrostatic microscope has not been able to catch up with the achievements of the other type. The position appears to be similar to that of the cathode ray tube in television; the magnetic cathode ray tube threw almost the whole burden on the circuit designer, but the circuit engineer was equal to the task. The electrostatic cathode ray tube, however, in which circuit complications were avoided at the cost of complications in the electron-optical system, was less successful. Some of the reasons are discussed in a survey of electron optics by the author.[35]

CHAPTER 6

THE ORIGIN OF CONTRAST IN ELECTRON MICROSCOPIC IMAGES

Contrast by Scattering; A Result of Lens Defects

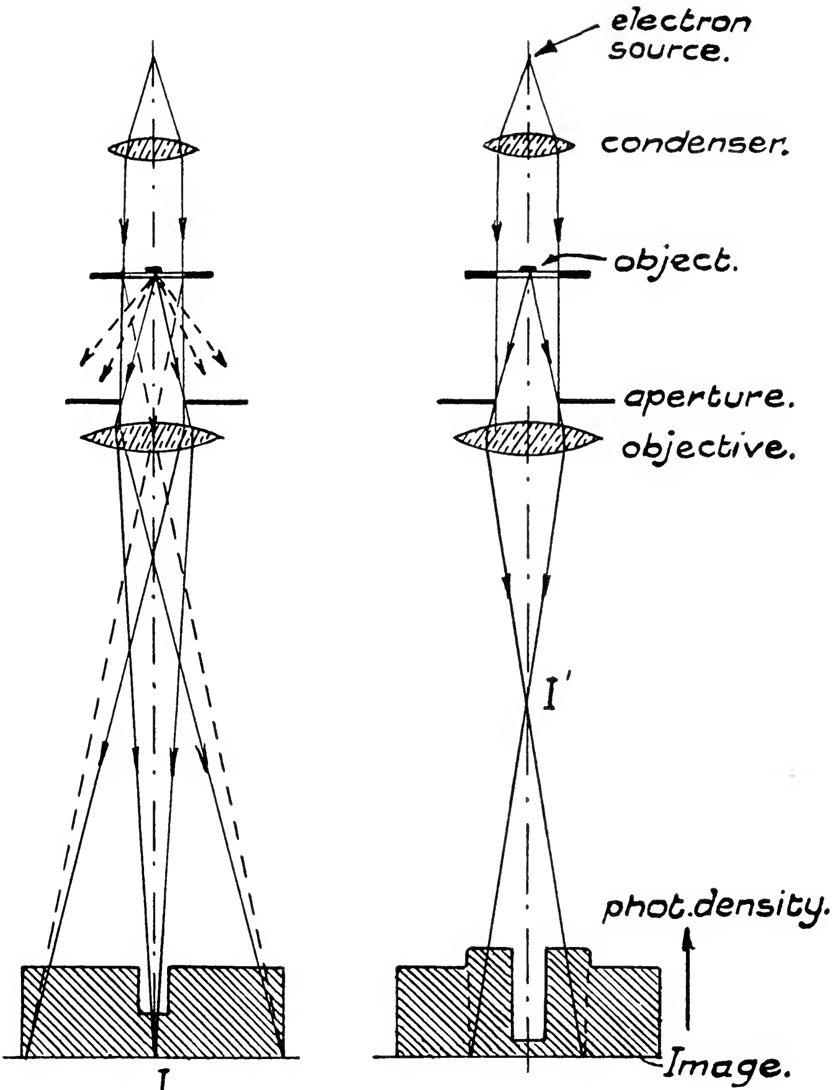
IN ordinary light microscopy the contrast arises mostly from different absorption in different parts of the specimen. Differences in refracting power and in fluorescence can be also utilized. The first electron micrographs were photographs of thin wires, metal foils with holes, and the like, that is, objects which showed great differences in electron absorbing power. This was thought a necessary condition for obtaining pictures, and Marton,[26] who was the first to apply the electron microscope to the investigation of organic cells, impregnated them with osmium salts. It appears that F. Krause (ref. 16, p. 55–60) was the first to discover that this was not necessary. In 1936, Marton [36] carried out the first theoretical investigation of the origin of contrast. He showed that *scattering* of electrons rather than absorption is the basis of electromicroscopic image formation. This general result is now well established. Marton's theory gave a satisfactory account only of the imaging of thick objects, or of objects resting on thick supporting membranes. It appears that most writers on the subject have applied it rather uncritically also to small objects and thin membranes, that is to say to the sort of objects usually employed to test the definition of electron microscopes. An important step was made, in 1939, by J. Hillier who pointed out certain effects arising from the chromatic error of electron lenses. It will be shown in the following that without these errors which most authors consider as regrettable imperfections, the high performance of the uncorrected electron microscope could not have been achieved.

When fast electrons collide with atoms, it happens very rarely that they lose their whole energy in a single collision. Most of the energy is lost gradually, in steps of 10–30 volts at a time. Some of the electrons are scattered elastically, *i.e.,* without appreciable loss of energy. Most of our knowledge of these processes is derived from experiments in gases and from theoretical investigations of the collision of electrons with single atoms and molecules.[39, 40] Our knowledge of the scattering of fast electrons in solids, however, is still rather rudimentary. The theoretical investigations have not yet gone very far, and the older experimental material, up to the year 1938, is of entirely inadequate accuracy. It will be shown that in electron microscopy a loss of about 10 ev by an electron of 60,000 ev velocity has observable effects, whereas, until 1938, no energy loss less than about 200 ev could be observed with certainty. The reason is that until the advent of the electron microscope, experimenters had no high voltage source at their disposal with a voltage fluctuation less than about 0.5 per cent. High voltage accumulator batteries were available in very few laboratories only. The development of negative feedback stabilizers, which was undertaken for the purposes of electron microscopy, at once enabled physicists to improve the accuracy of their measurements by a factor of about a hundred. G. Ruthemann,[41] in 1941, was the first to measure the velocity losses of fast electrons in thin metal foils with an error limit of less than a volt. He demonstrated that in solid bodies, as in gases, fast electrons lose their energy in well-defined discrete steps. This result is of considerable importance, as it justifies treating metals and other solids at least approximately as highly compressed gases, and using for them the very large amount of knowledge collected in the investigation of electrons in gases. Another important result is that, similarly as in gases, the number of elastically scattered fast electrons is rather small in comparison with the number of those which have suffered energy losses.

These results are sufficient for a discussion of the origin of contrast. Figure 14 shows the simplified ray-diagrams of an electron microscope of the transmission type, for elastically and inelastically scattered electrons. We assume a small object which either hangs unsupported in space or is supported by a membrane of negligible thickness. It will be shown later that the very thin organic membranes used for this purpose in electron microscopy fulfill the condition that most of the electrons of the original beam pass through them without collisions, and therefore can be considered as of *negligible* thickness.

For simplicity, we consider the problem as one of *geometrical* electron optics, *i.e.*, we consider the electrons as particles, neglecting diffraction effects. We can also for a start neglect the divergence of the illuminating beam, and consider it as parallel. If the object were not present, the illuminating beam would produce uniform blackening of the photographic plate. The object becomes visible by its scattering effect. In the case of elastically scattered electrons, obviously only those will make a difference in the image which are scattered *outside* the objective aperture. Those which are deflected by less than the aperture angle will produce the same density as if they had not been deflected at all, because, if the lens is assumed to be perfect, it will focus them in the image point "I" on the photographic plate.

Recapitulating the argument, the object is made visible by those electrons which are missing from the beam, because they have been absorbed by the physical aperture of the objective. This is unimpeachable *if we assume an objective free from chromatic and spherical aberration.* It is not admissible to consider this as the basis of a general theory of contrast, as most authors have assumed. We have seen that if the physical aperture is made sufficiently small, the spherical aberration can be practically eliminated. This is not sufficient to eliminate the chromatic error. However homogeneous the primary beam, the scattered beam will contain electrons of different velocities, unless we assume against the evidence of Ruthemann's experiments, and against the evidence of the theory of collisions that most of the electrons will be scattered elastically. If this theory were complete, it would be useless to smooth out the illuminating beam within limits of about 1 volt, if scattering produces differences of the order of 10–30 volts.



Elastic scattering a.) Inelastic scattering b.)

FIG. 14. The origin of contrast

There is also another objection. If the physical aperture plays such an essential part in producing contrast, how was it possible to obtain micrographs, prior to 1939, before the physical objective aperture was introduced? It could be said, and this seems to have been the opinion of the authors on this subject, that there always was an aperture, though in the older microscopes it was of the order 0.1–0.5 radians rather than 0.002. Good contrast could be produced in such instruments therefore only if a great part of the electrons is deflected by more than 0.1–0.5 radians. There is strong evidence, to be discussed in a later chapter, that in a single collision only a very small fraction of the electrons is deflected by more than 0.1 radians, therefore, contrast could be produced only by thick objects. Marton and von Ardenne were aware of this and built up their theories on the basis of multiple collisions. Multiple collisions mean very large energy losses, in fact, it is easy to see that the mean energy loss will increase with the square of the average deflection angle produced by a great number of successive collisions. This presents the dilemma in an even sharper form: If an electron suffers say ten collisions in the object, it will lose energy of the order of 100–300 volts. How was it, then, that the resolution of the electron microscopes was very noticeably improved when the fluctuation of the driving voltage was reduced from 10 volts to about 1 volt, although no steps were taken, nor could be taken, to reduce the *chromatic error* arising from energy losses of the order of several hundred electron volts?

The explanation is very simple, but rather surprising: There are two sorts of *chromatic error,* one arising from fluctuations in the driving voltage, the other from energy losses in the object. Although the first is harmful, the second is beneficial. This does not affect the resolution, but increases the contrast to such an extent that it can be said without exaggeration that microscopes without small physical apertures produce good pictures mainly *by* the chromatic defect of their lenses. It may be added that the spherical aberration has also a partly beneficial effect.

The explanation is contained in the right half of figure 14 which illustrates inelastic scattering, *i.e.,* assumes that the electrons have suffered energy losses in the object. These electrons will be *missing* from the beam which in the absence of the object would produce a uniform background on the photographic plate. If the object had absorbed them, or if it had scattered them all outside the physical aperture, there would be an area of zero photographic density corresponding to the image of the object. Some, or most of these electrons may be scattered inside the physical aperture. Because of the chromatic defect of the lenses they will be focused not on the photographic plate, but at I′, nearer to the lens, and they will fall on the plate inside a disk, with a radius corresponding to the chromatic error. An elementary consideration shows that as the focal length of a magnetic lens is inversely proportional to the electron energy V, the diameter of this disk will be, expressed in terms of equivalent object size

$$d_c = 2af\frac{\Delta V}{V} \tag{20}$$

where a is the aperture angle and ΔV the energy loss suffered by the electrons. In the illustration it is assumed for simplicity that their distribution is uniform over this disk. If a is large this assumption is not admissible, as the electrons may be scattered in a much narrower cone. In this case, their distribution will approach a shape similar to the probability law, without a sharp outer edge. As an example, let us assume $a = 1°$ as mean angle of scattering, $V = 60{,}000$ volts, $\Delta V = 20$ volts, and a focal length of 0.5 cm. This gives $d_c = 560$ Å, which is more than ten times the resolution limit obtained with microscopes without physical objective aperture. Figure 14 shows immediately that this large *aberration* does not interfere at all with the definition of the object. It merely produces a slightly darker border around the image of the object, the density difference at the edge of the object has remained unchanged, that is, it has the same value as if all the electrons which have been scattered by the object had been absorbed, irrespective of the size of the aperture. In photography contrast is measured as the ratio of the densities or the difference of their logarithms, therefore strictly speaking the contrast will reach this ideal value only if d_c is very large in comparison with the object diameter. Therefore, the contrast will even improve slightly with increasing value of this sort of *chromatic defect.*

In microscopes with a small physical aperture this effect is much smaller, but still noticeable. Electrons which have suffered a loss of ΔV ev will produce a border—dark in the original, white in prints or reproductions—of a width $af\frac{\Delta V}{V}$, measured in the plane of the object. Such *chromatic* borders have been observed in many micrographs, especially by E. Ruska and his collaborators. Sometimes the borders do not show one step only, as in figure 14b, but two, of about equal width. These *chromatic fringes* can be explained by the discovery of G. Ruthemann [41] that fast electrons lose their energy in solids mainly in discrete steps, in carbon for instance in steps of 24 ev. It appears that in most solids one type of collision is dominant, with a characteristic energy loss so sharply defined that Hillier [76] could base a method of microanalysis on it.

Similar borders appear also in numerous micrographs of objects supported by thin organic membranes. This is an indirect proof that a large proportion of the electrons in the illuminating beam traverses the foil without collisions. More direct evidence is contained in Ruthemann's experiments. He found that a large proportion of the electrons suffer no energy losses, and this proportion is larger in very thin foils than could be explained by electrons which have suffered elastic collisions only. A theoretical estimate, based on considering the solid as a highly compressed gas, gives 80–90 per cent as the proportion of electrons which will traverse a collodion foil of 100 Å thickness without collisions. The theory which has been sketched out is therefore applicable to conditions under which most electron micrographs are obtained. The essence of this theory is in short that *almost all electrons which have suffered inelastic collisions in the object will produce contrast as if they had been absorbed.*

But the theory can be extended to a certain measure also to elastically scattered electrons, assuming that they are scattered at angles of the order 3.10^{-3} radians or more. Such electrons will be absorbed by physical objective apertures of the usual dimensions. But if there is no physical aperture, the *spherical aberration* will play much the same part as the chromatic aberration did in the case of inelastically scattered electrons. Figure 14b can be again used as an illustration. As the spherical aberration increases with the third power of the deflection, electrons which have been scattered at appreciable angles by the object will be distributed over a relatively wide area of the photographic plate. Let us assume for instance that for an angle of 2.10^{-3} radians, that is about 7 minutes of arc, the spherical aberration is 10 Å. This means that electrons deflected by 30′ will be distributed over a radius of about 800 Å, those deflected by 1° over about 6,400 Å, which is practically equivalent to saying that they will not have any perceptible effect in the picture. The effect is enhanced by the fact that elastically scattered electrons will be in general deflected by wider angles than those which have suffered losses. However, the number of elastically scattered electrons is in general considerably less than that of the other group.

We can now understand the fact, mentioned without proof at the end of the previous chapter, that a well collimated illuminating beam is, in effect, equivalent to a physical diaphragm.

To sum up, electron microscopes without physical objective aperture produce good pictures not so much in spite of, as rather by virtue of, the lens defects. It can be said that micrographs, especially of small and thin objects were produced prior to the introduction of the physical objective aperture by an extraordinary combination of lucky circumstances of which the workers do not seem to have been aware. How these escaped attention for such a long time, can be perhaps explained by the general observation that unexpected difficulties are more stimulating to efforts of understanding than unexpected successes. There is also the reason, that the pioneers of the electron microscope were guided mostly by the analogy of the optical microscope, and the effects discussed are without a counterpart in optical microscopy. A microscope with parallel illumination and uncorrected spheri-

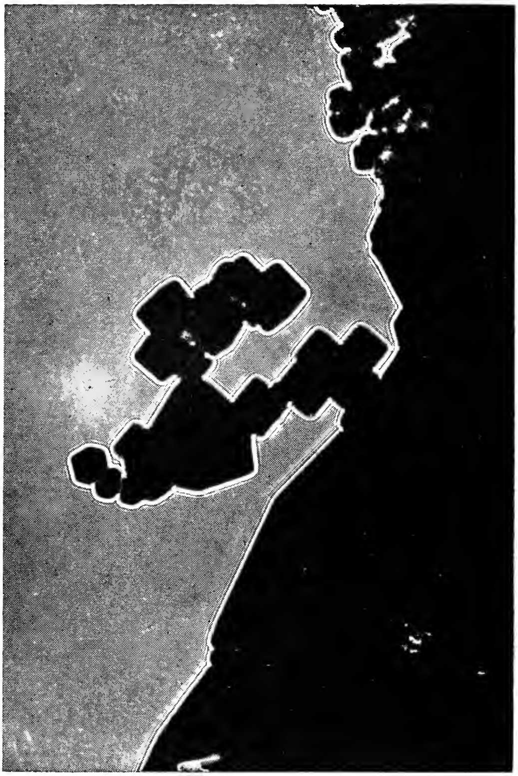


Fig. 15a

cal aberration would have very poor resolution. The effect corresponding to inelastic collisions is absent in ordinary microscopy, as light in general does not change its wavelength in the object. An exception is fluorescence microscopy. In this case, it would be possible to imitate the working of the electron microscope by reversing the usual process. Instead of observing the visible fluorescence through chromatically corrected objectives, one could use lenses corrected for spherical aberration, but deliberately uncorrected for chromatic aberration, and record the *negative* of the fluorescence, by taking photographs, in which the fluorescent particles would appear absorbing. But it would be, of course, even better to take the photographs through a filter which absorbs the fluorescence and transmits only the primary ultra-violet light.

It is evident that the beneficial effects of the chromatic and spherical aberration exist only so long as the illumination of the object can be considered as homogeneous and undiffused. A thick supporting membrane would destroy the high resolution altogether. The same is true of the resolution of details in thick objects. For instance, density differences in the membrane of the bacillus will remain invisible, which differences could be easily resolved if it were possible to spread out the membrane without the bacillus. This fact is of importance in the interpretation of micrographs.

It may be added, that the beneficial effects of lens defects turn into the reverse also in electron microscopes with *dark field illumination.* In these instruments, objects are shown up not in the negative by the electrons which have dropped out of the background illumination *(missing electrons)*, but in the positive by the scattered electrons themselves. This is at least one of the reasons why such instruments were so far rather unsuccessful.[21]

Contour Phenomena; Fresnel Diffraction

The *chromatic* fringes mentioned in the last section are very often masked by a much more striking contour phenomenon: the Fresnel diffraction fringes. These were discovered, in 1940, independently by J. Hillier [42] and H. Boersch.[42a]

In light optics, Fresnel diffraction arises if a part of a parallel or almost parallel light beam is cut off by the edge of some dark object. Instead of the more or less sharply defined shadow, one finds at some distance behind the object a sort of oscillatory transition between light and darkness, with several maxima and minima. This phenomenon can be very easily observed in light microscopes of only moderate magnification. If the microscope is sharply focused on the edge of the object, the contour appears sharp, but as soon as the objective is focused a little ahead or behind the object, the edge appears surrounded by fine fringes of uneven spacing, at both sides of the true contour. Precisely the same phenomenon is observed in electron microscopes under suitable conditions.

According to the elementary theory of Fresnel diffraction,* the fringes are caused by the interference of the primary, parallel wave, and an *edge wave*, that is to say, a cylindrical wave, starting from the illuminated edge of the dark object, without loss of phase. In a plane at a distance y from the plane of the object, dark fringes of the order n appear in the bright field at a distance x from the "true shadow" according to the equation

$$x^2 = ny\lambda$$

where λ is the wavelength.

The distance between two consecutive fringes is

$$\Delta x = \sqrt{y\lambda}(\sqrt{n} - \sqrt{n-1})$$

The fringes of order $n - 1$ and n will be separately visible only if the divergence of the illuminating beam is less than $\Delta \frac{x}{y}$. If it is more, the fringes will merge into one another.

Figure 15a shows exceptionally clear Fresnel diffraction patterns, recently obtained in the R.C.A. laboratory by J. Hillier and E. G. Ramberg. The distance of the 1st order dark fringe from the edge is about 80 Å. As the wavelength of 60 kev electrons is about 0.05 Å, the photograph must have been taken with a defocusing of about 1·3.10⁵ † Å or 13 μ. In the original, fringes are visible up to the 7th or even 8th order, with a spacing of only about 14 Å corresponding to an angle $\Delta \frac{x}{y}$ of only about 1·1.10⁻⁴. This agrees very well with the half-illuminating angle

* Cf. e.g. R. W. Wood, Physical Optics, Macmillan, New York, 2d ed., p. 220–223.

† To avoid misunderstanding, please note that the raised period between numbers stands for a decimal point and the lower period, for a multiplication sign, i.e., 1·3.10⁵ is 1.3×10^5.

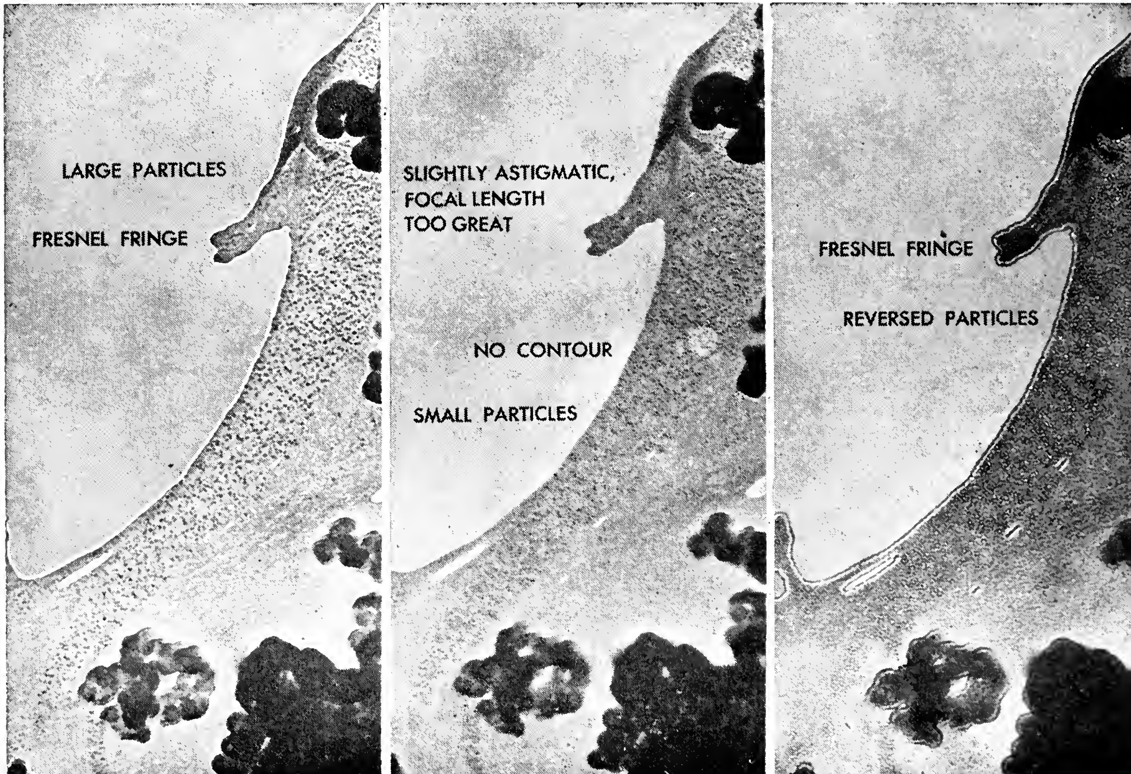

LARGE PARTICLES
FRESNEL FRINGE
SLIGHTLY ASTIGMATIC,
FOCAL LENGTH
TOO GREAT
NO CONTOUR
SMALL PARTICLES
FRESNEL FRINGE
REVERSED PARTICLES

Fig. 15b

given by the authors as 5.10^{-5}, *i.e.*, a total beam divergence of 10^{-4}. It can be seen that the observation of Fresnel fringes gives a useful method for measuring the very small illuminating angles used in modern electron microscopes.

Fresnel fringes are invaluable for ascertaining that the objective was really focused on the object. It will be shown in the next section that focusing to maximum contrast will almost invariably result in focusing a little off the object. But Fresnel fringes disappear entirely at exact focus, and by this fact give a very sensitive and reliable criterion. This is particularly clearly shown by the series of photographs, reproduced in figure 15b which were kindly sent to the author by Dr. James Hillier. All three photographs show the edge of a collodion film with colloidal gold particles sputtered on it. The photograph at the left was taken with an objective which was too weak, the central one with correct focus, the last one with too strong a lens current. It is remarkable that the contrast at the edge appears worst at best focus. An operator who had only the general appearance of the picture to guide him would have almost certainly chosen a somewhat defocused position.

Another interesting feature of this series is the way in which the appearance of the colloidal gold particles varies with varying focus. Similar phenomena are well known to every user of light microscopes. In the absence of any knowledge of the object, it would have been difficult to decide which of the three pictures is the closest representation of the original.

In the hands of Hillier and Ramberg the observation of Fresnel fringes has proved an invaluable tool for the detection and measurement of the most tenacious of lens errors: astigmatism in the center, due to ellipticity of the magnetic field. The central figure shows that it was not possible to focus a magnetic objective, which would have been considered as a very good one by any other test, simultaneously on a vertical and on a horizontal edge. It will be described in a later chapter how these authors used these observation methods for improving the objective, and finally achieved the best performance of which any electron miscroscope of the present type is capable.

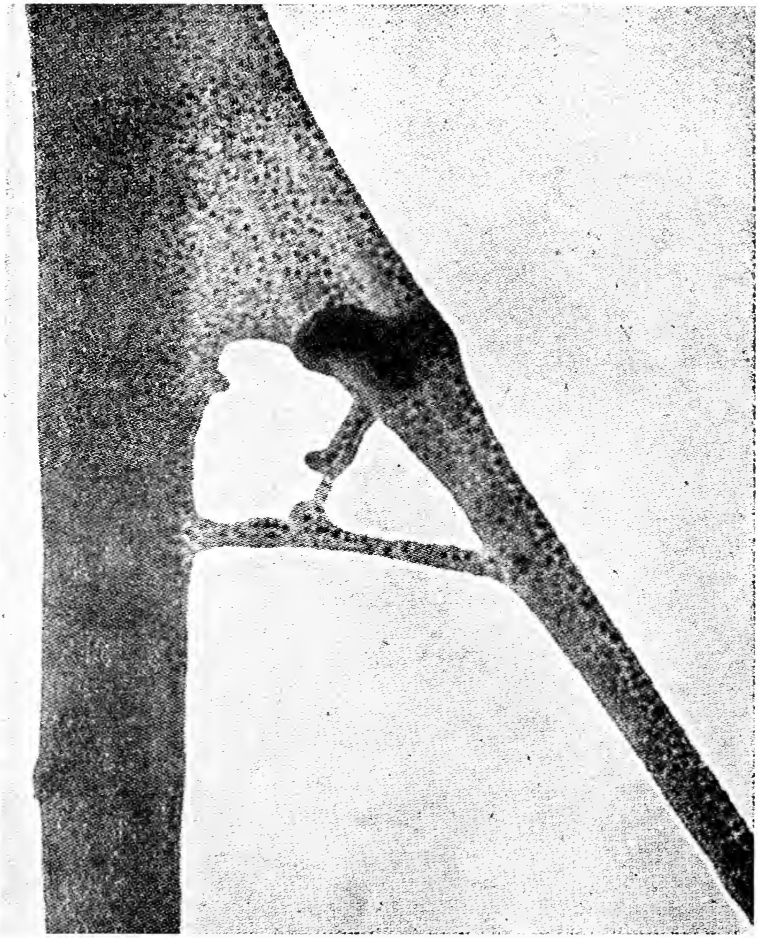


FIG. 15c. Extremely thin fibers of a synthetic rubber "Koroseal," photographed with R.C.A. electron microscope. Magnification about 100,000. Defocused photograph, showing Fresnel fringes. The dark centers are presumably *phase contrast* images of the giant molecules. *(By courtesy of B. F. Goodrich Co. and Dr. V. K. Zworykin, Associate Director of R.C.A.)*

Phase Contrast

One of the first photographs in which Fresnel fringes have been observed is reproduced in figure 15c. In 1943, the author received an original copy of it from Dr. V. K. Zworykin. On closer investigation, the Fresnel-fringe interpretation appeared somewhat doubtful. Only two fringes were visible, which may have been just as well steps in the light distribution, as in the case of *chromatic* fringes, with which they happened to coincide, if it was assumed that the characteristic loss was that in carbon, 24 volts, and multiples of it. But the main reason to question Hillier's and Boersch's interpretation was that the fine fibers showed detail which appeared incompatible with a defocusing of the order of 10 μ, which had to be assumed if the contour phenomena were to be explained as Fresnel fringes.

This dilemma was solved only quite recently. It appears that in addition to contrast due to absorption and to scattering there exists in electron microscopy a third: *phase contrast.* This is a wave-optical phenomenon, well known in light microscopy. Transparent objects, with a refractive index different from that of the medium (for instance: thin glass fibers) are almost invisible if the objective is sharply focused on them; but they produce very strong contrast on defocusing, if the focal plane of the objective happens to coincide with the focal points or lines of the object. In other words, what the microscope will be able to see in such a case, is not the object itself, but the disturbance caused by the object in the original, more or less plane wave-front.

It is known from electron diffraction experiments that solids refract electron waves with a refractive index which is

$$n = \sqrt{1 + \frac{V_i}{V}} \approx 1 + \tfrac{1}{2}\frac{V_i}{V}$$

where V is the volt energy of the electron and V_i the *inner potential* of the solid, about 12 volts for carbon. In electron microscopy it was generally assumed, against the evidence of diffraction experiments, that even in thinnest objects the effect of refraction would be entirely masked by scattering, *i.e.*, objects would behave like frosted glass, not like polished glass. But the above mentioned dilemma can hardly be solved otherwise than by assuming that very thin objects can in fact act as fairly regular refractors. This assumption is supported by the theoretical estimate, mentioned on p. 35, that 80–90 per cent of the electrons pass without collisions of any kind through organic foils of the order of 100 Å thickness, and by the evidence collected in electron diffraction experiments. Further evidence has been found recently by Hillier and Ramberg who could explain the fine details of the Fresnel fringes at the edge of thin collodion membranes only by assuming that an appreciable fraction of the primary wave traverses the membrane as a coherent wave, but with a phase retardation roughly as calculated from the above mentioned refractive index.

The discovery of phase contrast is too recent to say much about at the present time. It appears that it will have to be taken into careful consideration in the interpretation of almost all electron micrographs of very thin objects.

Some very interesting possibilities arise from the fact that very thin objects can act, at least partly, as regular refractors and retard electron waves by one quarter, or even one half wave without destroying their coherence. This means that in addition to the macroscopic, man-made electromagnetic fields, we may be able to use the microscopic inner fields of solids for electron-optical purposes, thus adding a new chapter to electron optics. The field of application is of course restricted by the limit of about half a wave maximum retardation; but there are some interesting optical devices which operate with retardations of a quarter wave only. Chief of these is the ingenious *phase-contrast microscope* of Prof. Zernicke * which allows to see the true shape of non-absorbing objects with refractive indices only slightly different from those of the surrounding media in their true shape. Experiments to translate this instrument into electron-optical terms are in progress.

* Prof. Zernicke, Physica **1**, 689, 1934; C. R. Burch and J. P. P. Stock, Journ. Sci. Instr. **19**, 71, 1942.

CHAPTER 7

THE RESOLUTION LIMIT OF THE UNCORRECTED ELECTRON MICROSCOPE

THE resolution limit of an electron microscope is defined as the smallest distance at which two small particles of sufficient scattering or absorbing power will appear as two, before merging into one. Figure 16 is a reproduction of a brilliant test photograph obtained by M. von Ardenne,[43] in 1940. It shows particles of colloidal gold, deposited from an extremely dilute solution on a thin membrane of collodion. It can be seen that two particles at a distance of 30 Å could just be separated; this, therefore, is the resolution limit. At the same time, the photograph gives also a fair estimate of the *detection limit,* as the diameter of the gold particles was about 10 Å, and this was just sufficient for good visibility.

Von Ardenne's resolution limit of 30 Å was the best achievement, in 1940, equaled in the same year by the R.C.A. In 1939, the best resolution stood at 50 Å, obtained by H. O. Müller, a collaborator of Ruska,[31] and by L. Marton and collaborators.[27] By the end of 1943, the limit was improved to 24 Å by V. K. Zworykin and J. Hillier,[44] while E. Ruska and his collaborators achieved about 18–20 Å. No further progress was made until 1946, when J. Hillier and E. G. Ramberg in a brilliant investigation eliminated the lens astigmatism which until then stood between the practical and the theoretical limit, and brought the resolution down to 8–10 Å.

We have seen that there are three kinds of limitation for the resolving power, due to: spherical aberration, diffraction and fluctuations of the driving voltage. The last is often called *chromatic error* but it is better to avoid this expression, as we have seen that a part of the chromatic error is beneficial. All

Individual particles

FIG. 16. Resolution test photograph taken by M. von Ardenne, showing gold particles of about 10 Å diameter on a very fine membrane

errors will be discussed in terms of the dimensions of an equivalent object.

The spherical aberration turns a point into a disk with a diameter

$$d_s = Cf\alpha^3 \tag{21}$$

where f is the focal length of the objective, α the aperture angle, and C is a dimensionless constant, which depends only on the shape of the electromagnetic field of the objective, not on its dimensions.*

The diffraction of the de Broglie waves at the aperture, whether limited by a diaphragm or by the divergence of the illuminating beam only, produces an Airy disk with a diameter $1.22\frac{\lambda}{\alpha}$, equation (14). As the intensity in the Airy disk falls off gradually toward the edge, the distance below which two Airy

* Some authors reserve the name *spherical aberration coefficient* and the symbol C for the length Cf, a regrettable practice, as coefficients should be always dimensionless.

disks would appear to merge into one is somewhat smaller. If we assume this factor to be 0.67 and use equation (13) for the de Broglie wavelength, we can write the resolution limit due to diffraction alone in the simple form

$$d_A = \frac{10}{a\sqrt{V}} \text{ Å} \tag{22}$$

Fluctuations ΔV of the driving voltage V produce in the magnetic microscope, but not in the electrostatic microscope with unipotential lenses, as we have already seen, a disk of diameter

$$d_c = 2af \frac{\Delta V}{V} \tag{20}$$

If two, or all three of these errors are of approximately equal size, it is a matter of some difficulty to estimate the *resulting error*. In optics, Max Born has solved this difficult problem [45] by treating geometrical and diffraction errors with a unified wave-theoretical method. It turns out that it is legitimate in first approximation to superimpose the intensities calculated separately from geometrical optics and from diffraction, though combination terms will also arise. To simplify matters, we follow a suggestion made by von Ardenne [37] to consider the three disks as bell-shaped *probability distributions* and to calculate the resulting error as the *geometrical* sum of the three diameters, *i.e.*, as the square root of the sum of their squares. If we accept this, it is evident that the three errors will have to be of the same order of magnitude in the optimum, or most economical design.

It can be seen from equations (21) and (22) that the spherical aberration and the diffraction error vary in opposite directions with variations of the aperture a. The sum of their squares will be a minimum at an aperture which makes the diffraction error $\sqrt{3} = 1.73$ times the spherical aberration. This gives for the optimum aperture an equation

$$a^4 = \frac{5.8 \times 10^{-8}}{Cf\sqrt{V}} \tag{23}$$

and for the optimum resolution limit, as produced by the combination of diffraction and spherical aberration by the rule of geometrical addition

$$d_{sA} = 750(Cf)^{\frac{1}{4}}V^{-\frac{3}{8}}\text{Å} \qquad (24)$$

where f has to be substituted in cm. This would be the best resolution obtainable if the variation of the driving voltage could be altogether suppressed. But obviously little would be gained by this. If instead we make $d_c = d_s$ the numerical factor of (24) is increased only by $\sqrt{\frac{5}{4}} = 1.12$, *i.e.*, to about 850 instead of 750.

Equation (24) shows that only three parameters of the electron microscope enter into the resolving power, provided that the fluctuations of the driving voltage can be kept sufficiently small. It appears from (24) that in order to obtain a high resolution the most effective method is to raise the potential V. This is really less effective than it appears, for two reasons. The first is that in a given magnetic lens field the focal length f is proportional to V. However, saturation of iron sets a limit to the field intensities which can be realized. The usual focal length for 60 kev electrons is about 0.5 cm, and the shortest which can be realized is about 0.3 cm. Assuming such a lens of maximum power we obtain

$$d'_{sA} = 35C^{\frac{1}{4}}V^{-\frac{1}{8}}\text{Å} \qquad (25)$$

As the resolving power increases only with the $\frac{1}{8}$ power of V, the gain is very small. In reality, it will be even less profitable to increase V beyond about 60 kv, as the penetrating power of fast electrons rises so rapidly that very small particles can no longer be detected for lack of contrast. High voltage electron microscopes have been constructed by M. von Ardenne [46] (200 kv), H. O. Müller and E. Ruska [47] (220 kv), and by V. K. Zworykin, J. Hillier and A. W. Vance [25] (300 kv), not with a view to improving the resolution but in order to investigate thicker objects. At 60 kv, which can be roughly considered as the optimum, the resolution becomes (with the strongest objective that can be realized)

$$d''_{sA} = 9C^{\frac{1}{4}}\text{Å} \qquad (26)$$

Thus ultimately, the resolution depends on the numerical coefficient C, which characterizes the spherical aberration. On the basis of Scherzer's theory of the spherical aberration, R. Rebsch [48] obtains a minimum value of $C = 0.1$ for the optimum shape of the lens field. Rebsch's value is based partly on a somewhat too optimistic estimate of the minimum distance at which two aberration figures merge into one, and 0.2 appears to be a better founded theoretical value. With this equation (29) gives a value of 6 Å for the resolution limit as determined by spherical aberration and diffraction.

Until very recently, there was a rather large discrepancy between this theoretical limit and the practical achievements. Several authors tried to explain this by values of C much larger than the theoretical optimum, and this seemed to be confirmed by attempts to measure the spherical aberration directly, which never gave values of less than about two. But J. Hillier and E. G. Ramberg, in a most important paper, still unpublished at the time of writing, proved conclusively that the discrepancy between theory and practice was in fact not due to large departures of the magnetic field in the axis from the theoretically recommended law, but to minute yet important departures from axial symmetry. The *ellipticity* of the field need be only of the order of $\frac{1}{10{,}000}$, that is to say, the departure from circular symmetry need not be more than one caused by a deformation of the round bore in the pole pieces of this magnitude, in order to prevent the achievement of resolving powers better than two to three times below the theoretical limit. The astigmatisms encountered within good magnetic lenses were in fact so small as to escape direct observation by any other method than that described in the previous chapter in connection with figure 15b. Once this method was found, correction became possible by screwing eight small iron screws into the gap between the pole pieces, controlling the results all the time by means of Fresnel fringes. This very laborious method led to brilliant results. The resolution limit of the R.C.A. electron microscope was at once reduced from about 20 to 10–8 Å, which is only very little more than the above calculated theoretical limit of 6 Å.

It may be mentioned that the above theoretical estimate with geometrical addition of errors cannot lay claim to great accuracy. More recent calculations by W. Glaser, using Born's method of unified treatment of geometrical and diffraction errors, seem to indicate that the estimate errs on the safe side, and that the theoretical limit may be 3.5–5 Å only.

We must now check the assumption that the required constancy of voltage can be realized. With $C = 2$, $f = 0.3$ cm and $V = 60$ kv, equation (24) gives 4.5×10^{-3} for the optimum aperture. This is very near to the value 5×10^{-3} used by von Ardenne in taking the micrograph shown in figure 16. The diameter of the objective aperture is 0.0027 cm, and the spherical aberration d_s is 5.35 Å. The relative fluctuation $\frac{\Delta V}{V}$ which according to equation (20) produces the same error is 0.002 per cent. This degree of stability and an even better one has been indeed realized in modern electron microscope circuits.[50, 24]

The formulas (25) and (26) were derived under the assumption that no lens stronger than a certain lens of maximum power can be realized because of saturation of the magnetic pole pieces. This limitation does not exist in the case of the electrostatic microscope, but it so happens that electrostatic lenses have very nearly the same limit, though for an entirely different reason. The power of an electrostatic lens is proportional to the intensity of its electric field, and beyond a certain intensity flash-overs occur through autoelectric emission. For the present the resolution of the best electrostatic microscopes is still considerably less than that of the magnetic type (about 50 Å).

CHAPTER 8

THE DETECTION LIMIT OF THE UNCORRECTED ELECTRON MICROSCOPE

THE size of the smallest particle which can be detected in an electron microscope of the bright-field, transmission type is closely connected with its resolution. Let us imagine a small disk-shaped object of diameter d which absorbs all electrons falling on it. The absorbed electrons will be missing in the picture of the uniform background, and the missing intensity (deficit), will be distributed according to a certain law, compounded of spherical aberration, variation of the driving voltage, and diffraction. In first approximation, let us assume the deficit to be uniformly distributed over a disk of a diameter equal to the resolution limit d_r. The particle will be visible if the contrast, defined as the ratio of deficit to background intensity is more than a certain minimum, say 10 per cent. This means that the small object will be visible if

$$d_{min} = \sqrt{.1}d_r = 0.316d_r \qquad (27a)$$

For a resolution limit of 30 Å, as in von Ardenne's photograph shown in figure 16, this is 9.5 Å, in good agreement with the experimental value which was about 10 Å.

In second approximation, we must take into account the peaked character of the distribution of the intensity in the error figure. This need not take us into a detailed investigation of the actual distribution, as the correction is small and rather insensitive to the actual shape. If we assume the distribution in the shape of a probability distribution, the resolution limit would be approximately the diameter on which the intensity has dropped to half the peak value. The minimum between two overlapping distributions would just about vanish if the centers were approached to this distance. Calculation gives a value of 1.10 for the peak factor, *i.e.*, to the ratio of the peak intensity to the intensity obtained by distributing it uniformly over the half-value diameter. If, however, we assume the distribution to have the same law as in the Airy diffraction figure, and imagine it replaced by a uniform distribution over 0.67 of the diameter of the central disk, which is the same assumption as we have made in the previous chapter, the peak factor becomes 1.13. Therefore, either of these two assumptions gives almost exactly

$$d_{min} = 0.3 d_r \tag{27b}$$

for the diameter of the smallest object which can be detected in a bright-field electron microscope of resolution d_r.

So far, we have assumed the cross section $\pi \frac{d^2}{4}$ to be absorbing. But as we have seen in chapter 6, inelastic scattering has practically the same effect as absorption, whether the electrons which have suffered energy losses ultimately reach the photographic plate or not. The case of elastic scattering is somewhat more complicated, but at least in the case of small physical apertures most of these electrons will also contribute to the contrast. We can, therefore, with good approximation replace *absorbing cross section* by *scattering cross section.*

J. Hillier,[38] who was the first to attempt an evaluation of the detection limit of the electron microscope, followed an argument similar to the above, but was led to the paradoxial conclusion that the uncorrected microscope with small aperture should be able to detect a single atom of atomic number exceeding 25, *i.e.*, heavier than manganese, if the atom would not move during the exposure. He found that taking account of the atomic electrons did not materially alter the lower limit to the atomic number of an atom that could be detected with the electron microscope. The difference in the laws of scattering will be explained in more detail in chapter 13. Though the uncorrected microscope cannot see single atoms, its detection limit is rather impressive. Von Ardenne's gold particles contained only about thirty atoms each. He was also able to obtain pictures of single large organic molecules, such as edestine.

It may be noted that the simple argument which led to equation (27) requires more careful elaboration when applied to microscopes in which the resolution is mainly limited by diffraction, *i.e.,* in cases in which the diffraction error considerably exceeds the two *geometrical errors.* In this case, the argument which operates with electrons, or superposition of positive and negative intensities may lead to appreciable errors. As a next approximation, it may be replaced by the following: We consider the inelastic scattering of an electron as absorption, followed by reemission. The absorption can be also replaced by reflection, because a black body cannot be exactly defined for the purposes of wave theory. In fact, all theories of the microscope and other optical instruments in which absorbing objects or diaphragms occur are only approximations, for reasons explained in the appendix. As regards re-emission, it is entirely justified to consider a particle which scatters an electron with energy loss as a self-luminous object. Moreover, as is shown in the appendix, even in elastic scattering, electrons suffer sufficient energy losses to make their waves *incoherent* in the sense of being unable to interfere with the primary electron waves. In this approximation, we must therefore proceed as follows: Consider the object first as absorbing or reflecting and calculate the diffraction figure in the image plane of the microscope. It is well known that this figure can have a wide variety of shapes. In many cases a bright region will appear in the center of the image of a small dark object. On this diffraction figure we must superimpose the Airy figure, produced by diffraction of the reemitted electrons at the objective aperture. Obviously, if two diffraction figures have widely different characters, the minimum detectable diameter might be appreciably different from the value given by equation (27). But the problem is more of theoretical than practical interest, as in present-day microscopes the geometrical errors are of the same order as the diffraction error.

CHAPTER 9

DESCRIPTION OF COMMERCIAL ELECTRON MICROSCOPES

ELECTRON microscopy reached a new stage, in 1941, when the Radio Corporation of America put on the market the first commercial electron microscope [24] of the magnetic type, after a year's experience in a number of selected research institutes. An instrument for research institutes [51] was also developed, in 1939, by Siemens and Halske in Germany, and commercialized at about the same time as the R.C.A. microscope. By the end of 1943, the large experience accumulated with the first model enabled the R.C.A. to complete a very much simplified magnetic microscope, a *desk model* of somewhat lesser performance. The first commercial electrostatic microscope, also a desk model, was completed by the General Electric Company, Schenectady, at the end of 1942. The first British magnetic electron microscope was put on the market by the Metropolitan Vickers Electrical Co., Ltd., in 1944.

1. R.C.A. Magnetic Miscroscope, Floor Model, Type B

Figure 17 is a photograph of two magnetic microscopes of the R.C.A., side by side. The floor model, "Type B," stands almost 7 ft high. Large as it is, it is considerably smaller than several of the experimental models which preceded it, and occupies very little floor space. This has been achieved by packing all the electrical supplies, control gear, and exhaust system into the comparatively small space of the pillar behind the microscope proper. The only exceptions are the mechanical pumps which are mounted separately to avoid vibration.

In the design of this instrument, no simplifications were attempted which would have entailed even the smallest sacrifice of performance. In spite of this an instrument was created, after an astonishingly short time of development, which can be successfully operated even by semi-skilled laboratory assistants; a fact which reflects the highest credit on the skill of the designers.

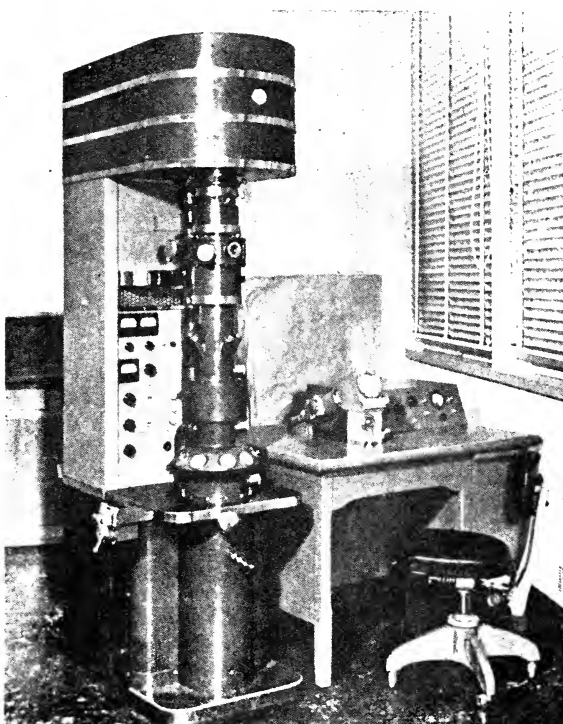


FIG. 17. Type B floor model and experimental desk model of R.C.A. magnetic microscopes

Figure 18 is a schematic section of the microscope. From top to bottom, it contains the discharge tube, a flexible joint with a double set of adjusting screws, the condenser lens, a second flexible joint, the object chamber, the objective lens, a third flexible joint, connecting the objective with the first *throw,* the

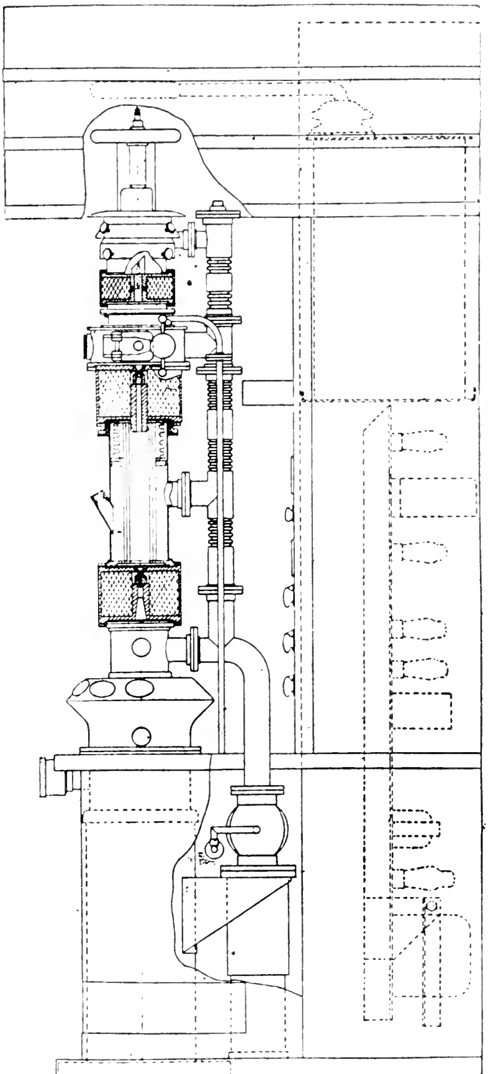


FIG. 18. Full section of R.C.A. Type B electron microscope

projector lens, and the second throw, followed by the photographic chamber. Every lens, the throw, the viewing chamber, the object chamber, and the photographic chamber is a unit by itself, accurately machined, and fitted together by means of large surface flanges, lagged with gaskets of duprene, a synthetic rubber. This sectionalized method of construction enables new developments to be embodied in existing instruments. It entails a very great number of vacuum joints; in addition to those mentioned, there are three rubber-lagged glass windows in the first throw, six in the viewing chamber, and one in the object chamber. Yet a vacuum of the order of 10^{-5} mm mercury can be realized without difficulty, and leaks are a very rare occurrence. The exhaust system consists of a three-stage oil diffusion pump, backed by a mechanical pump. No liquid air is necessary. Very quick evacuation is insured by two airlocks, one for the specimen, and one for the photographic plate, which can be changed without breaking the vacuum in other parts of the microscope. This is particularly important in the case of the discharge tube, as tungsten filaments are rather sensitive to atmospheric moisture. The air locks are preevacuated by a separate mechanical pump.

The discharge tube contains an electron gun, consisting of a V-shaped tungsten filament and a cathode shield, the operation of which has been explained in chapter 5. It is fitted with a corona shield, and these are the only parts of the microscope at —60 kv against earth, all accessible parts are earthed. The two sets of screws which can be seen in figure 18 below the discharge tube allow two independent adjustments, so that the tube axis can be exactly aligned with the condenser lens. The adjustment can be watched on an oblique fluorescent screen with a small central bore, visible in the figure just above the condenser lens. This can be observed through a special port. A set of screws just below the condenser lens (not visible in the figure) enable the condenser and the discharge tube to be aligned with the objective aperture as one unit. This adjustment can be watched on a fluorescent screen with a small hole on top of the projector lens. Finally, the objective lens, together with all the preceding

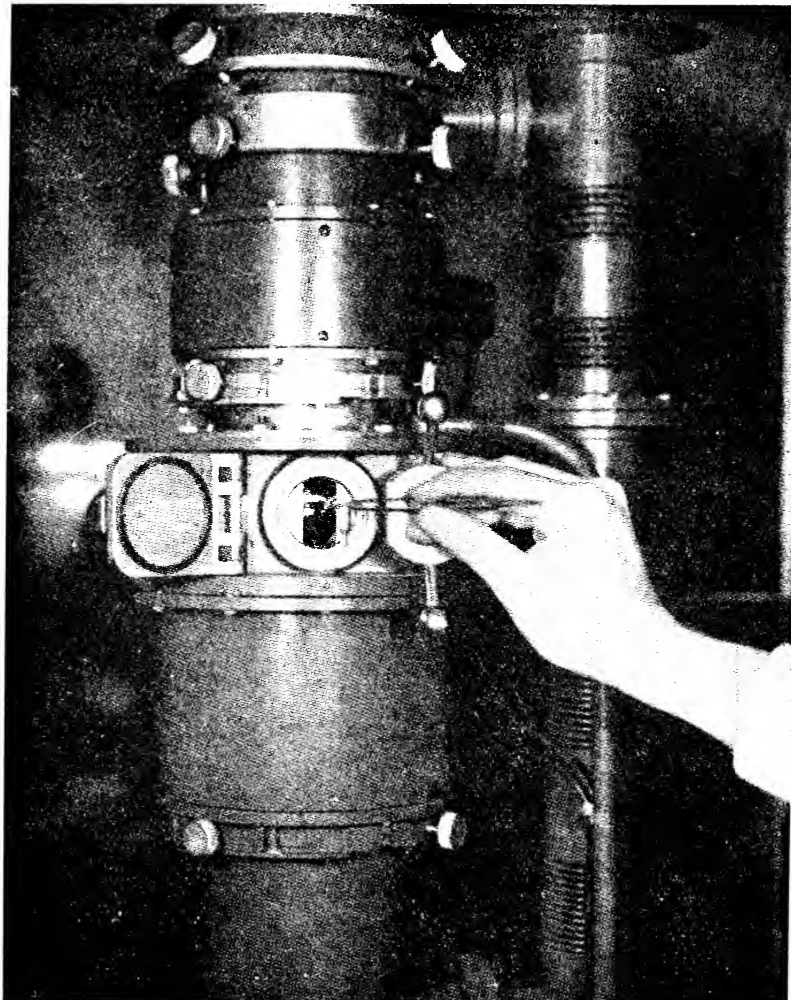


FIG. 19. Two electron lenses and air lock of the R.C.A. electron microscope

sections, can be tilted to align it with the rest. The final image can be observed on a fluorescent screen just above the photographic plate. Exposure is effected by tilting this screen out of the way. Once aligned the instrument will hold its adjustment for a long time, in many repeated exposures.

The object chamber can be seen open in figure 19. It contains a moveable object stage and the object changing mechanism. The stage allows moving the object in two directions at right angles to one another. Models have been made also for *stereoscopic* photography which enable tilting the object around a horizontal axis. The specimen holder is a small thimble which during operation rests about 5 mm above the center of the objective. It is attached to a lever system which is operated by

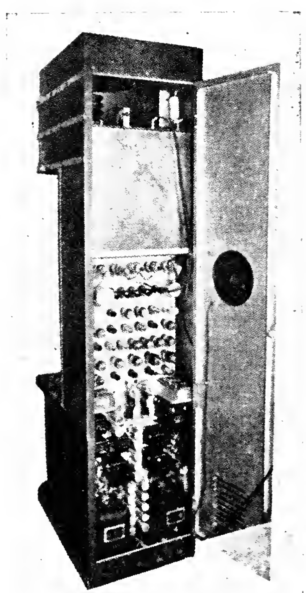


FIG. 20. Photograph of rear of Type B electron microscope, showing electrical supplies. The resonant circuit which produces the high voltage at 32 kc is contained in the oil tank, near the top

a handle. The manipulation of this handle lifts up the specimen holder and at the same time operates the airlock, isolating the object chamber from the rest. Air is admitted automatically on unlatching the door which can be seen in the figure, and the specimen holder can be removed. On loading, the same operations are performed automatically in opposite order.

The photographic plates used in the R.C.A. microscope measure 10 in. by 2 in. and can be used for a series of exposures, usually 2 in. by 2 in. each. Like the specimen, the plate is introduced through an air lock, without breaking the vacuum in the main chamber. Reevacuation takes only 2–3 minutes. Fine grain photographic plates are used with preference. The maximum obtainable magnification is about 20,000, which means that the finest observable detail of about 30 Å will be magnified to about 0.06 mm. In order to avoid any disturbance by photographic grain this ought to be about ten times the grain size, *i.e.*, the plate should have a resolution of about one hundred fifty lines (black and white) per mm. It would not pay to use plates of higher sensitivity, as the grain size goes up roughly proportionally to the sensitivity, and apart from the disadvantage that the microscope would have to be made longer or more complicated by adding a third stage, the available light intensity would decrease with the square of the magnification. Therefore, it is much preferable to magnify the photographs by optical enlargement to a size suitable for studying with the unaided eye. A final magnification of 30,000–45,000 is fully sufficient if the enlargements can be studied in the original, in half-tone reproductions as much as 200,000 may be required to bring out every detail clearly.

The pillar behind the microscope proper contains the high-voltage supply, the circuits to energize the coils, and the stabilizing and controlling equipment. Figure 20 gives an idea of the complication of the auxiliary gear which comprises 53 valves. Only a very cursory description can be attempted here.

Fluctuations of the driving voltage can be divided into two classes: a-c ripple and variations of the mean value. The latter are mostly *slow* variations, due to inconstancy of the line voltage and of the *constants* of the circuit components. Spontaneous high frequency *noise* is of relatively very small importance; it is mostly suppressed by the capacity of the coaxial cable which connects the high voltage supply with the microscope.

Ripple voltage in the R.C.A. microscope is enormously reduced by supplying the rectifiers with 32,000 cycles, instead of with the line frequency. This artifice was known in high speed cathode ray oscillography (Schering), but in electron microscopy, it has additional advantages. Not only can the ripple suppressing condenser be made much smaller, but it is much easier to shield the microscope from stray fields. In addition, the voltage regulator will work very much faster.

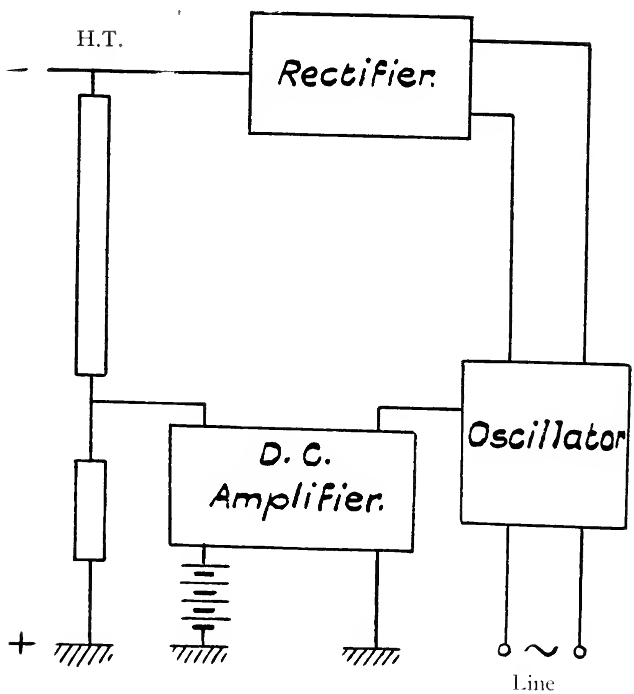


FIG. 21

Slow fluctuations, that is, slow in comparison with 32 kilocycles, are suppressed by an inverse feedback regulator.[50] The principle is indicated in figure 21. The high voltage output is divided by a potentiometer, which, it may be mentioned, takes a

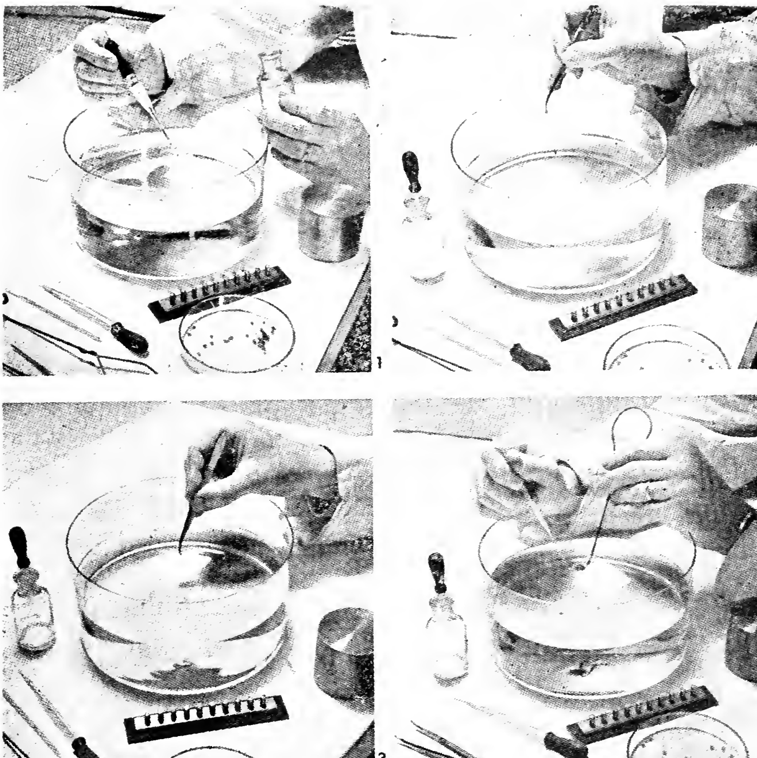


Fig. 22. Preparation of specimens for the R.C.A. electron microscope

current considerably larger than the maximum 0.5 millamperes of the discharge tube, so that the rectifier works under rather steady current conditions. The divided voltage is balanced against a dry battery. (Ordinary anode batteries fluctuate by less than three parts in a million during 30 second intervals.) The voltage difference is amplified by a d-c amplifier, and the amplified voltage is impressed on the screen grids of the output stage of the oscillator which supplies the rectifier with 32 kilocycles power. The sign of this voltage is so chosen that it will reduce the high tension if it is too high, and *vice versa. (Negative feedback.)* The smoothing effect of this circuit is so strong that what fluctuations remain must be ascribed to other causes. As A. W. Vance has shown, they are mostly due to variations in the potentiometer, caused in some way, not yet properly understood, by the circulation of the oil in which it is immersed. This unit maintains the output voltage constant within 0.004 per cent or better over 30 seconds, which is shown by experience to be the longest exposure required.

As mentioned in chapter 5, this microscope has a physical objective diaphragm of about 0.001 in. diameter. This extremely fine hole is drilled into a gold leaf with a finely pointed needle. Under optimum conditions, it improves the resolution limit of the R.C.A. microscope to about 24 Å, which is half of what is obtainable without the diaphragm. As the performance of the microscope with a diaphragm tends to fall off, and at the end of two months of operation is not better than 100 Å, it is not surprising that many users of the R.C.A. microscope prefer to discard the aperture and are satisfied with a steady resolution of about 50 Å.

The preparation of specimens for investigation is simpler than in ordinary microscopy, as no dyeing is necessary. Figure 22 shows four stages of the ordinary routine. First a drop of collodion (nitrocellulose) solution is placed on a water surface. This spreads out rapidly, and after the solvent has evaporated a solid film of about 100 Å thickness will float on the water.* Next, a small circular disk of very fine wire mesh is placed on top of the collodion film, and pressed down somewhat, so that it adheres to the film. The wire mesh is fished out (last figure) and its edges are freed from the collodion. The mesh which is now covered with a film is dried, and a drop of the suspension or

* Experiments of the author indicate that the artificial resin, "Petrex 5," manufactured by the Hercules Powder Company, Inc., lends itself even better for the production of extremely thin films than nitrocellulose.

solution which is to be studied is applied to it. Finally, the mesh with the preparate is placed over the opening of one of the small cartridges which can be seen in the foreground, and the cartridge is loaded into the object chamber of the microscope. This is the routine which can be followed, *e.g.*, in the investigation of bacteria and viruses. Other objects require special techniques of which a great number are in existence, and which are growing almost daily.

The R.C.A. Type B electron microscope has been fitted lately with a number of accessories for special investigations which are embodied in the new "Universal" E.M.U. model, to be described later.

2. R.C.A. Magnetic Microscope, Desk Model

Several years of experience in the application of the R.C.A. microscope, Type B, enabled the R.C.A. to compile interesting statistics of the most frequent applications.[51] It was found that the greatest interest lies in a range of sizes and details for which even a resolution limit of 20 Å is barely sufficient. But the statistics revealed also considerable interest in objects exceeding about 800 Å in size where a resolution of about 80–100 Å would be all that is required.

Statistics were made also of the magnifications preferred by research workers in various fields and of the number of exposures usually taken of one specimen. It turned out that relatively few workers availed themselves of most of the steps which are provided in the large R.C.A. model between the minimum of 1,000 and the maximum of 20,000, and that in the great majority of cases only one photograph was taken of each specimen. These and other considerations convincingly proved the need for a simplified instrument. It was decided, therefore, to develop a small electron microscope with only two magnifications, 500 and 5,000, and with only one exposure per plate. An experimental model embodying these and many other distinctive features has been described by Zworykin and Hillier.[51] Though some of its constructional details have been redesigned, most of the functional features have probably remained unaltered in the commercial model which has just appeared on the market at the time of writing.

A full view of the small microscope has been already shown in figure 17. Figure 23 is a slightly simplified section of it. The main differences from the large model will become evident from a comparison with figure 18. Starting from the cathode end, we note first of all the absence of the condenser lens. This has been replaced by electrostatic focusing, by impressing different potentials on the cathode shield which in figure 23 is called the *grid*. If the grid is at cathode potential, the cross-over will very nearly coincide with the object which is now illuminated by a bundle of fairly wide divergence. This bundle is not cut off, as the new microscope contains no physical objective aperture. It contains only a small diaphragm between the electron source

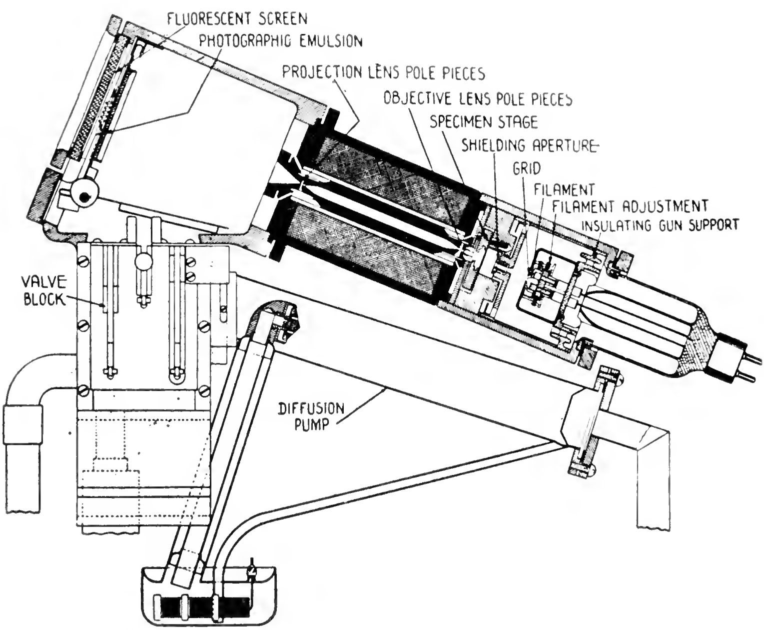


FIG. 23. Section of R.C.A. electron microscope, desk model

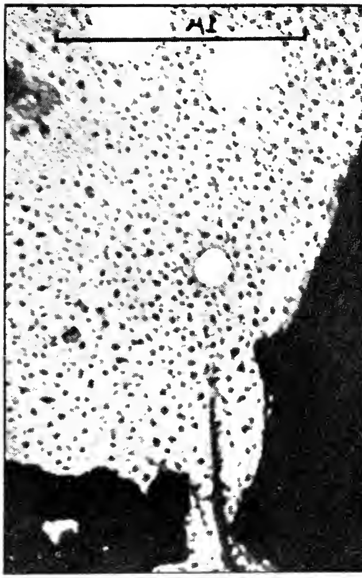


Fig 24. Test photograph of simplified R.C.A. microscope. Thin membrane spattered with heavy metal. Electronic magnification ×3,100

and the specimen, very near to the latter, in order to reduce the cross section of the beam and the heat development in the specimen. Such wide bundles produce large spherical aberration, and cannot be used for photographs with high resolution, but they are very useful for focusing the specimen. Apart from the higher intensity, a wide bundle has the advantage of smaller depth of focus and enables more accurate focusing. When the picture is taken a potential of —75 volts is impressed on the grid, which not only cuts down the current, but brings the cross-over very close to the cathode and makes the illuminating beam at the specimen very nearly parallel, with a divergence near to the irreducible minimum, discussed in chapter 5.

The specimen is no longer contained in a special object chamber with an air lock. The whole volume of the microscope is so small that a vacuum of the order of 5.10^{-5} mm mercury can be established in $1\frac{1}{2}$–3 minutes, according to the length of

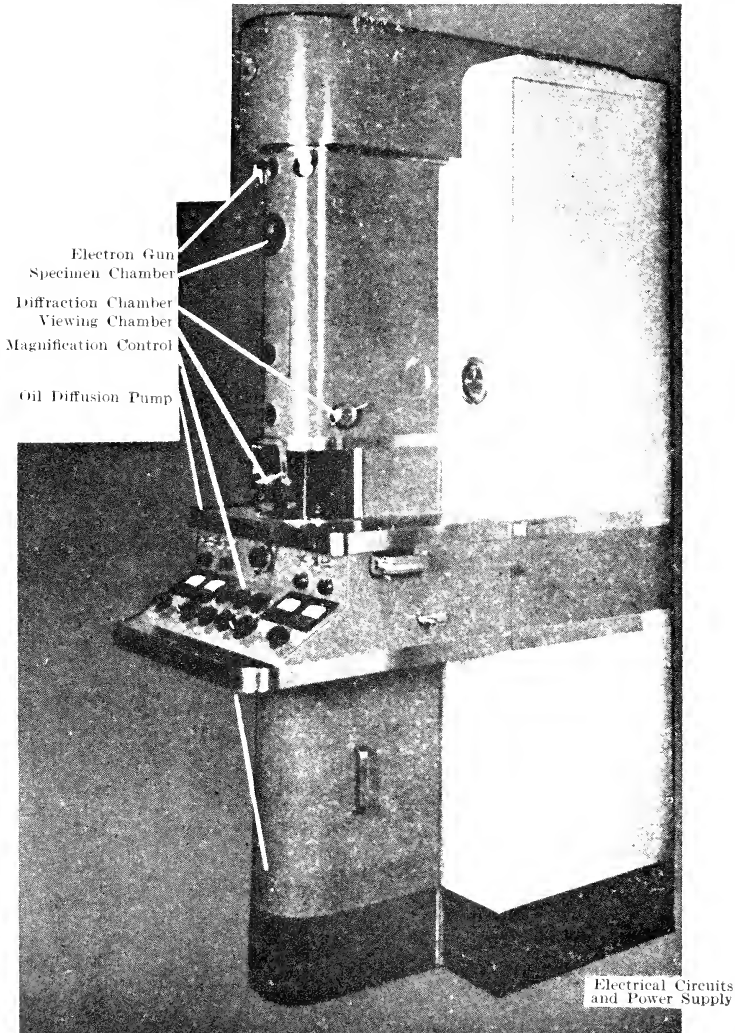


FIG 25. R.C.A. Universal Electron Microscope Model 1944. In addition to the features of Type B electron microscope, it contains a stereoscopic object stage and a diffraction adapter as permanent fixtures.

time it has been left open. The object is removed together with its stage, which allows scanning a circular area of the specimen 2.5 mm in diameter.

A striking new feature of the small microscope is the fact that the objective lens and the projection lens form one unit, energized by the same coil. The iron pole piece unit is machined in one, which enables superior accuracy to be obtained and obviates special alignment mechanisms. The figure shows the pole piece adjusted for a magnification of 5,000. If the lower magnification of 500 is desired, a separate cartridge has to be removed from the projection end of the pole piece. This can be done through the window of the fluorescent screen which can be easily removed. No steps are provided in the supply of the energizing coil, only a fine adjustment for the focusing, which changes the magnification only slightly.

The viewing chamber and the method of photography have also undergone radical changes. The fluorescent screen is now viewed in transmission. It will be noted with interest that the R.C.A. has developed new fluorescent materials which give a three- to four-fold increase of image brightness as compared with the willemite commonly used in American television, which was hardly, if at all, inferior to the best sulfide screens, preferred in Britain. At the same time the grain size of the fluorescent powder is fine enough to resolve about one hundred lines per mm.

Only a single photographic plate is used. An ingenious mechanism enables one to tilt this in front of the fluorescent screen, exposing it by opening the lid, to close the lid, and to tilt it again into a horizontal position, all by the movements of a single lever. The plate can be removed through a door, visible just below the fluorescent screen.

The exhaust mechanism has been also greatly simplified, and its operation is made nearly automatic. Three valves are worked by a lever in such a way that a complete pumping cycle corresponds to one to-and-fro motion of the lever. Only the timing is left to the operator. Evacuation is effected by a single-stage high-speed oil diffusion pump which is an integral part of the microscope unit and a mechanical pump which stands underneath the stenographer's desk on which the unit is mounted, but on a separate base.

The power supplies have been also very considerably simplified. Whereas the principle of high frequency rectification and inverse feedback regulation has been retained, the number of valves has been reduced to eleven. The accelerating potential is 30 kv in the small model against 60 kv in the large microscope, and its fluctuation is about 0.015 per cent against 0.004 per cent or less.

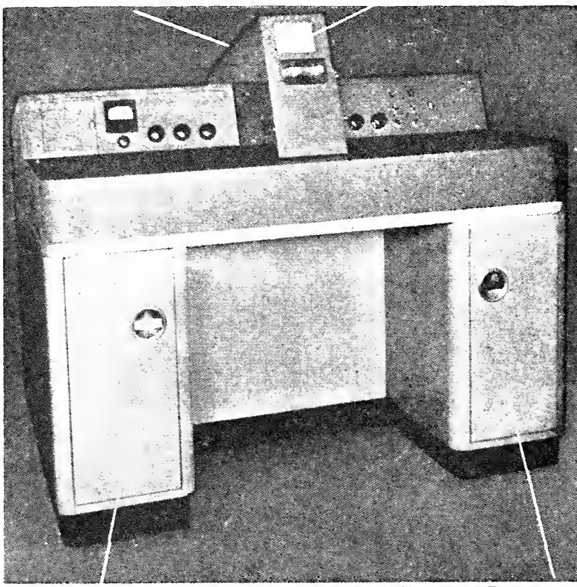


FIG. 26. R.C.A. Type EMC Electron Microscope (Console Model) Essentially as described in connection with fig. 23

The resolution of the small experimental model is not stated by its authors, but it can be judged from figure 24 which shows a membrane spattered with heavy metal particles. The instrument was just able to separate two particles which can be seen below the line indicating a length of 1 μ, near one end of it. From this it appears that under favorable conditions a resolution limit of 100 Å can be expected, sufficient for a wide range of applications.

In 1944, the Radio Corporation of America completed the development of two new commercial models. These are the Type EMU Universal Model, shown in figure 25 and the Type EMC Console Model, shown in figure 26. Apart from the striking elegance and convenience of its design the EMU model represents a progress in comparison with the Type B one, by embodying an almost continuous range of magnifications between 80× and 20,000×, an object stage for stereoscopic photographs, and the diffraction adapter described in chapter 14. The main features of the EMC model are substantially the same as described in connection with the experimental desk model.

3. Other Commercial Electron Microscopes

The Siemens & Halske A.G. in Berlin were the first to appear on the market, in 1939, with a commercial electron microscope, developed by E. Ruska and B. v. Borries. This very successful instrument was in many respects simpler than the American electron microscopes. It could be operated up to 100 kv, but the high voltage supply was of the kind used in commercial X-ray equipments. The lens currents were supplied by accumulators. In spite of the less perfect smoothing, this instrument achieved resolutions down to 18–20 Å, a performance which has been surpassed only quite recently by the R.C.A. microscope. There appear to be two reasons for this. One is that the electron gun of the Siemens instrument produced at the specimen a current density perhaps a hundred times better than the R.C.A. instrument prior to the redesigning of its electron gun, in 1945. The other is that the Siemens instrument was always used with an objective aperture which, unlike the one in the R.C.A. instrument, seldom gave trouble probably because the Siemens microscope was always evacuated with mercury pumps, not with oil pumps. The manufacture of the Siemens instrument was discontinued, in 1945, at the time of the occupation of Berlin.

In the second half of 1945, the first British commercial electron microscope was put on the market by the Metropolitan Vickers Co., Trafford Park, Manchester, England. It was developed by M. E. Haine. It is a magnetic instrument which operates with a carefully smoothed voltage of 50 kv, derived from the 50 cycle mains but regulated by a negative feedback controller. It has no objective diaphragm and is evacuated by an oil pump. Several instruments have given excellent service in practice, though they were mostly used for purposes where 50 Å resolution was amply sufficient. A new model with many novel features is in preparation.

The first French commercial electron microscope appeared on the market, early in 1946. It was developed by H. Bruck and P. Grivet and is manufactured by Compagnie Générale de Telegraphie sans Fil, Paris. This is an electrostatic instrument, the only one on the market at the present time. It was already mentioned that electrostatic instruments operating with *unipotential lenses*, that is to say with electrodes which are connected either to the cathode or to the anode are free from the necessity of very careful regulation of the operating voltage. One cannot entirely dispense with regulation, as E. G. Ramberg [52] has shown that at electron energies of 50–100 kev the relativistic corrections of electron dynamics are no longer negligible, and equation (19) on p. 28 ceases to apply rigorously, but smoothing to 2–3 per cent is sufficient, and this can be achieved by very simple means. The C.S.F. microscope is operated at 100 kv and contains two unipotential *single lenses*. These consist of two outer apertured electrodes, dished inwards, connected with the anode, and a central electrode, with a somewhat larger aperture, connected with the cathode. As unipotential lenses have a fixed focus, the magnification is fixed once for all at the convenient figure of 6500. The focusing of the object is done mechanically, by longitudinal displacement. The best resolution achieved to date is 80 Å. The C.S.F. microscope is very simple compared with any instrument of the magnetic type, as may be seen from figure 27. It is very easy to use, and has already given good service in several scientific institutes.

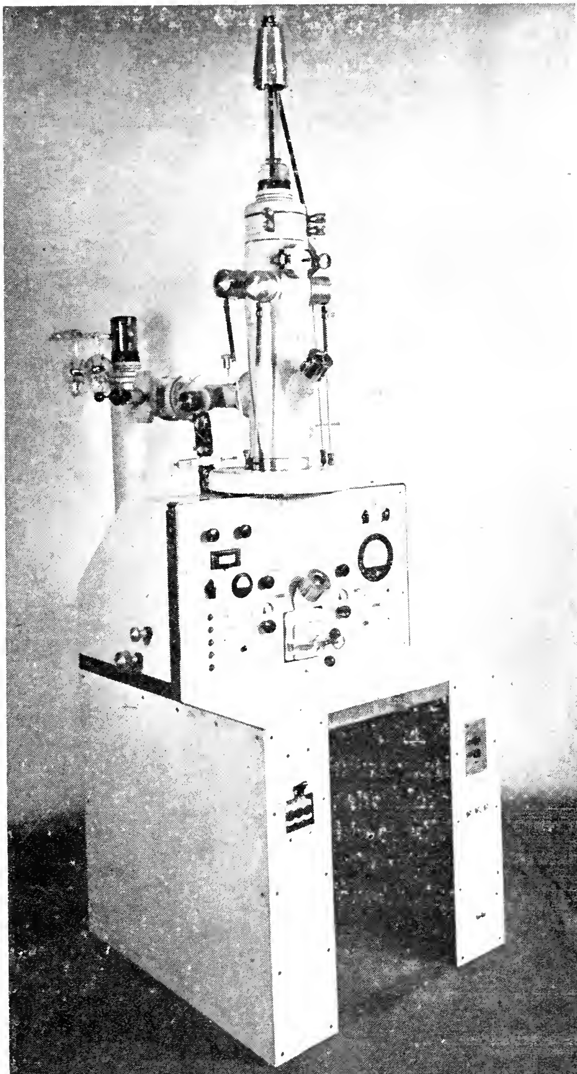


FIG. 27

The Philips Incandescent Lamp Factories, Eindhoven, Holland, have recently completed the prototype of a magnetic electron microscope with many novel and interesting features on the basis of research extending over many years by Dr. le Poole of the University of Delft. It is a desk model instrument with less than three feet total length, but operated with voltages up to 100 kv and with electronic magnifications not inferior to most other, much longer instruments. The saving in length is achieved by a third lens, *i.e.,* a second projector lens. Another interesting feature is the very large fluorescent screen, of 8 in. diameter, at the end of the tube, as in the R.C.A. desk model. This allows very accurate visual focusing. The film on which the photographs are taken is arranged very much nearer to the second projector lens, so that the photographs can be accommodated on 35 mm film. It is believed that with the fine grain of modern photographic material this size is entirely sufficient. Without the five times larger screen for visual focusing, only a small proportion of the photographs taken would do justice to the capacity of the film.

The objective is of radically different design from that of all other microscopes. Here the suggestions, discussed later, on p. 101, have been put into practice for the first time: A large lens, up to a certain limit, allows a shorter focal length and smaller spherical aberration than a smaller one. The bore of the pole pieces is 11 mm, about four times larger than in almost all other microscopes. The object is fixed in the center of the magnetic field. This allows realizing a focal length of only 3 mm at 100 kv, and by their large dimensions the pole pieces can be manufactured much more precisely than smaller ones. They allow also to accommodate an important new organ, an adjustable objective aperture which can be displaced axially and radially.

Another interesting novel feature is an auxiliary lens, between the objective and the first projector, which allows taking electron diffraction diagrams, without moving the object from the position in which photographs are taken.

CHAPTER 10

ACHIEVEMENTS OF ELECTRON MICROSCOPY

IT can be estimated that at the time of writing more than two hundred electron microscopes are in operation in research institutes all over the world, and new results are being published almost daily in medical, biological, and physio-chemical journals, the greater part of which is not accessible to the author. Therefore, the following short selection cannot claim to be fully representative or up to date.

As soon as the preliminary tests had proved the usefulness and reliability of the new instrument, the R.C.A. established a Fellowship for Biological Research with the Electron Microscope, under the auspices of the American Research Council. Figures 28, 31, and 32 are taken from a survey of the first year of this work, by G. A. Morton.[54] The photographs are all taken with the commercial Type B model of the Radio Corporation of America.

Figure 28 shows photographs of the typhoid germ and of *Bacillus Subtilis* side by side with pictures obtained with an optical microscope. It may be mentioned that the latter are not fully representative of what the best microscopes can achieve, especially those working with ultraviolet. In fact the flagellae and the membrane of the typhoid bacillus were first discovered with the ultraviolet microscope, in a magnification of about 1,500, but both are far better visible in the electron micrographs. The *Subtilis* photograph shows also minute crystals adhering to the bacillus body. Both electron micrographs are direct records of the scattering power of the bacillus body which has not been dyed in any manner. They are also approximate records of the density. L. Marton and L. I. Schiff have devoted an interesting study[55] to the problem of the translation of the photographic density in electron micrographs into data of object thickness and

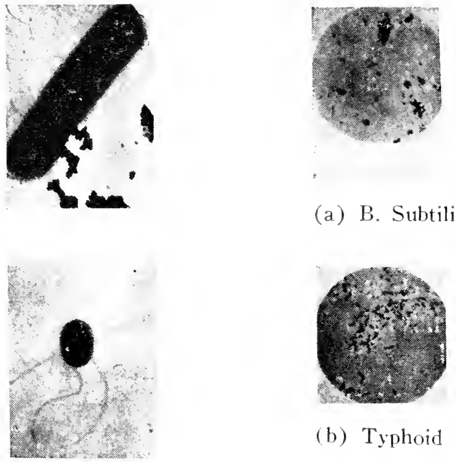


FIG. 28. Typhoid germ and *Bacillus Subtilis* electron and light micrographs

density. This for the time being is still a difficult and rather uncertain process, and in the case of small objects their theory requires correction for the reasons described in chapter 6. The biologist will obtain in most cases sufficient information from a few micrographs which show the bacilli in different positions or directly from stereoscopic electron micrographs. By these means it is possible to overcome the disadvantage of electron micrographs which like X-ray diagrams have very great depth of focus and make it difficult to locate details in depth. This

The bacteriophages, or bacterial viruses, of which three strains have been identified under the electron microscope, are the smallest known living organisms. For some time it was doubtful whether they could be classed as such. The strain here shown, Bacteriophage anti-coli PC, is the largest of the three. The "head" has a diameter of about 800 Å (0.08 microns), the "tail" has a length of about 1,300 Å. One particle of phage is sufficient to originate the lysis of a bacterial cell, and during the lysis an average of about a hundred new phage particles are generated. This photograph shows the phage particles swimming toward the bacteria. They were immobilized by drying five minutes after a suspension of the bacteria was injected with a drop of diluted phage solution.

(By courtesy of Dr. V. K. Zworykin, R.C.A.)

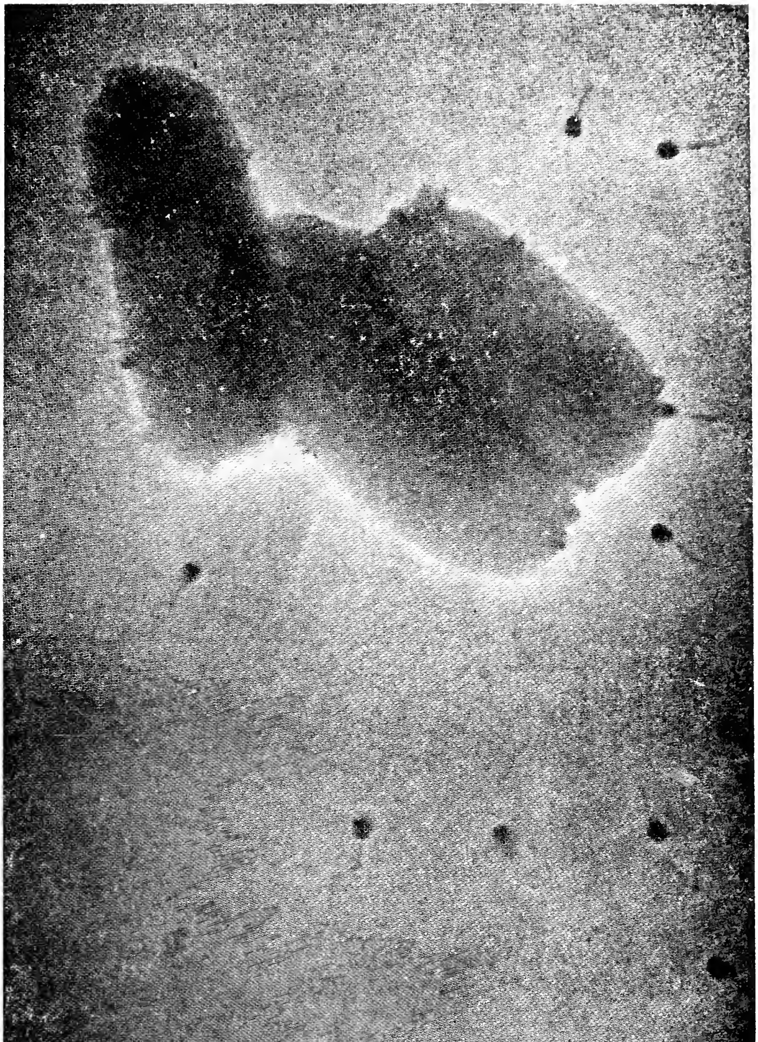


FIG. 29. Bacteriophage anti-coli and *Escherichia coli* × 25,000
(5 minutes in suspension)

slight disadvantage is however a very great advantage in the taking of electron photographs, as no extreme accuracy is required in focusing.

Whereas in bacteriology proper the electron microscope could add relatively little to the vast body of knowledge accumulated with the optical microscope, viruses and other submicroscopic organisms presented it with a magnificent field for discovery. One of its most important successes was the identification of the influenza virus.[56] But as the influenza virus is, even in electron micrographs, a rather unimpressive-looking particle, it appears preferable to select as an example a discovery which is spectacular as well as important.

Bacterial cultures are sometimes attacked by mysterious epidemics. F. W. Twort, in 1915, and F. d'Hérelle, in 1918, were the first to investigate them. D'Hérelle gave the name *bacteriophage* to the cause of the epidemics, and maintained that the phage is an ultramicroscopic living organism, parasitic in bacteria and reproducing itself. Direct proof was lacking until, in 1940–42, Pfankuch and Kausche, Ruska, and Luria and Anderson discovered and identified them with the electron microscope.[57] Figure 29 and figure 30 are reproductions of two brilliant photographs obtained by Luria and Anderson with the R.C.A., Type B instrument. Figure 29 was taken in a culture of *Escherichia coli* which was dried on to a thin membrane 5 minutes after it was injected with a drop of dilute suspension of the *Bacteriophage anti-coli PC*. The drying process has caught the phages which look remarkably like tadpoles vigorously swimming toward the bacteria. A few have already managed to bury their heads in them. The result of their activity is shown in the second picture, which was taken after the coli bacillus was exposed to the phage for 30 minutes. It shows destruction so complete that the phages themselves cannot be detected. There is ample evidence that, far from being destroyed, they have multiplied their numbers by a factor of about a hundred while feeding on the bacteria, which corresponds to a *generation* lasting only 4 minutes. It is very striking that the phage which seems to have so many of the attributes of animals contains

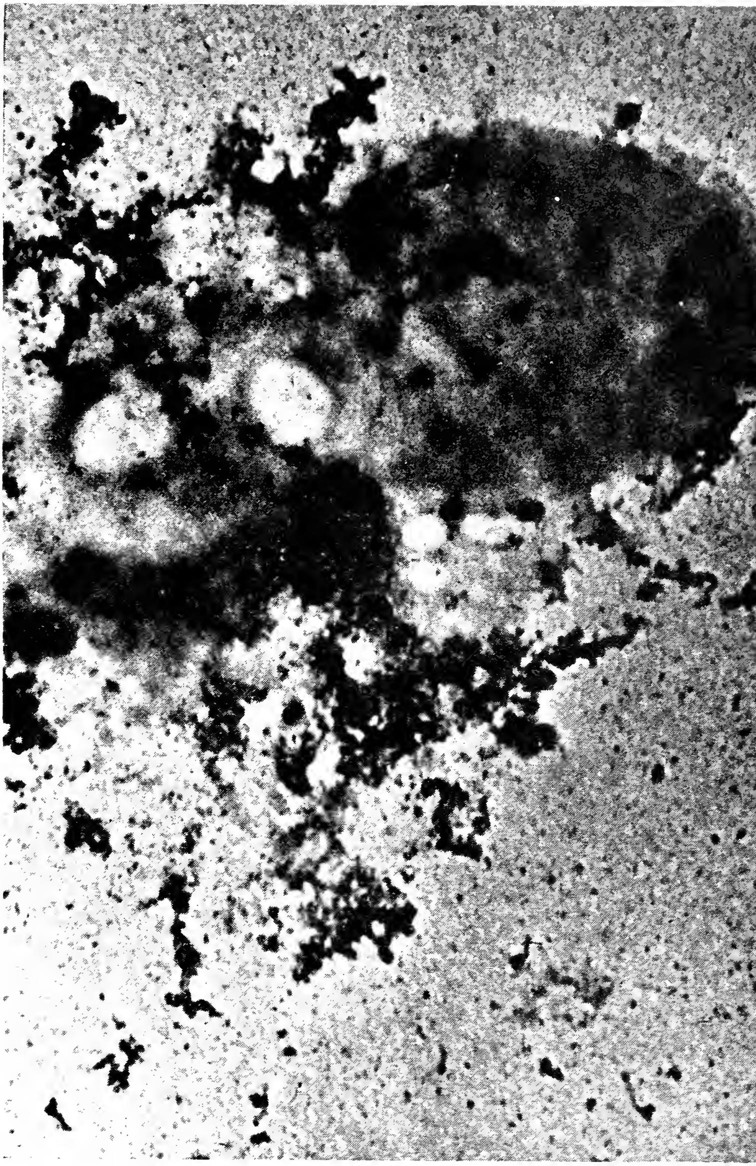


FIG. 30. Bacteriophage anti-coli and *Escherichia coli* × 35,000
The picture shows complete lysis of the bacterial cell after 30 minutes
(By courtesy of Dr. V. K. Zworykin, R.C.A.)

altogether only about a million atoms, and some investigators consider it as a giant protein molecule.

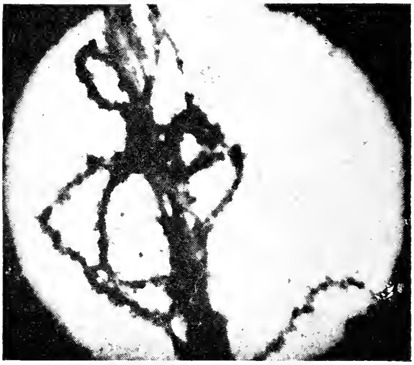


FIG. 31. Lampbrush chromosome. Magnification × 11,000

Figure 31 is a sample from another field of biological research. It shows a giant *lampbrush* chromosome. Genetics may become one of the most important fields for the electron microscope in the future. From experiments with electron bombardment of the eggs of *Drosophila Melanogaster,* C. P. Haskins has derived evidence that a single gene is confined to a space about the size of a protein molecule. To *see* single protein molecules is not beyond the range of the electron microscope as it is at present. When we have learned to see at least some of their details, important progress may be expected in the study of heredity.

Figure 32 is a photograph of that fascinating borderland subject of living and dead matter, the tobacco mosaic virus. So far the action of different chemical reagents and anti-sera on this virus has been tested under the electron microscope; in time we may also learn the secret of its procreative power.

One of the finest scientific achievements of electron microscopical research is the elucidation of the development process in photography.[59] As the electron micrograph in figure 33 shows, every silver grain appears as a tangled skein of a relatively very long and thin silver filament. There is now enough evidence to

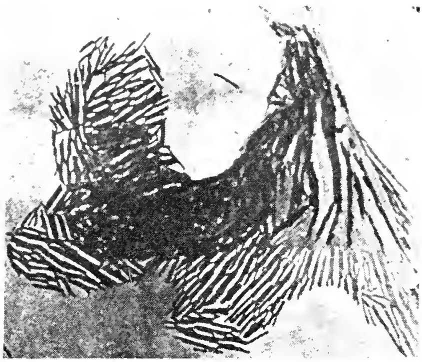


FIG. 32. Tobacco mosaic virus. Magnification × 21,000

make it practically certain that this filament was formed by extrusion during the development process. The extrusion starts from a spot of the silver bromide grain when the *latent image,*

FIG. 33. Photographic grains. Magnification × 125,000
(By courtesy of the Eastman Kodak Research Lab.)

formed by a few hundred silver ions was produced by the exposure. This spot is determined by a minute speck of sulfur in the gelatin of the emulsion. The micrograph at the left is especially remarkable, as the silver threads in the Lippmann emulsion are only about five atoms thick. They are still appreciably above the contrast limit of the R.C.A. microscope.

In metallography and other surface investigations, the transmission type electron microscope cannot be used directly. A great number of ingenious methods have been worked out to make film replicas faithfully reproducing the ups and downs of the surface, and thin enough to be used in transmission (500–

5,000 Å). One of the most interesting is the polystyrene-silica method of Heidenreich and Peck,[60] which was used in reproducing the calcite surface in figure 34a. The specimen is first molded into polystyrene, and this is coated by evaporated quartz. The most surprising effect was discovered, namely, that as long as the layer is thin enough, the silica molecules spread on the polystyrene like a liquid and form a perfectly smooth outer surface. After dissolving the polystyrene mold, the silica film can be introduced into the electron microscope.

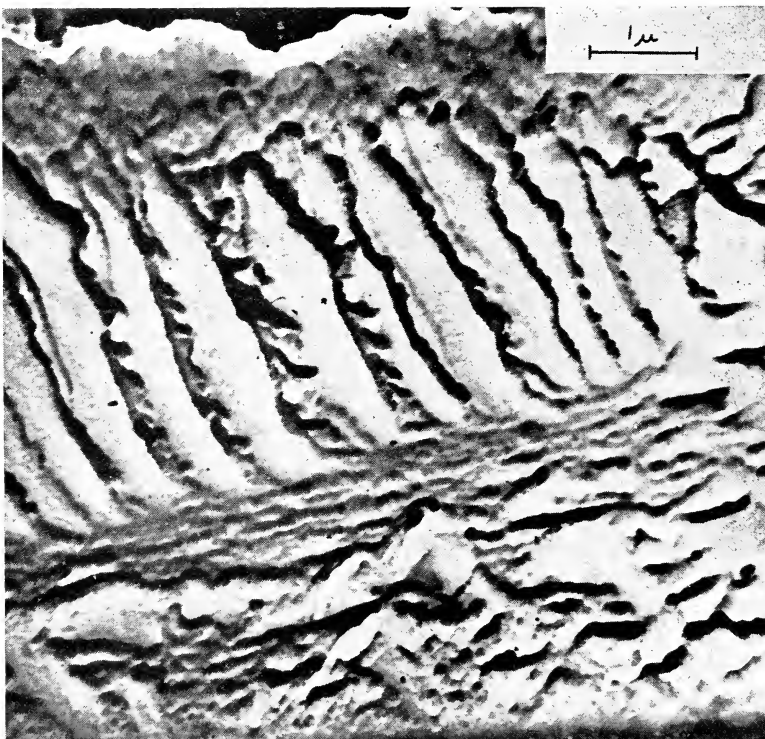


FIG. 34a. Electron micrograph of polystyrene-silica replica of a freshly cleaved calcite surface, obtained by R. D. Heidenreich, Dow Chemical Company.

(By courtesy of Dr. V. K. Zworykin, Associate Director of R.C.A.)

The most important recent addition to electron microscope techniques is the *shadowcast* method, chiefly connected with the names of Williams and Wyckoff.[84] It appears that it occurred to Müller first to show up fine details of surfaces by coating them partially with a thin layer of silver, condensed in high vacuum from a collimated atomic beam, at almost grazing incidence to the surface. Every minute projection on the surface, facing toward the beam will be coated, whereas parts facing the opposite way remain clear. But silver has a strong tendency to aggregate in specks, therefore Müller's attempts were only moderately successful. Williams and Wyckoff in a careful study

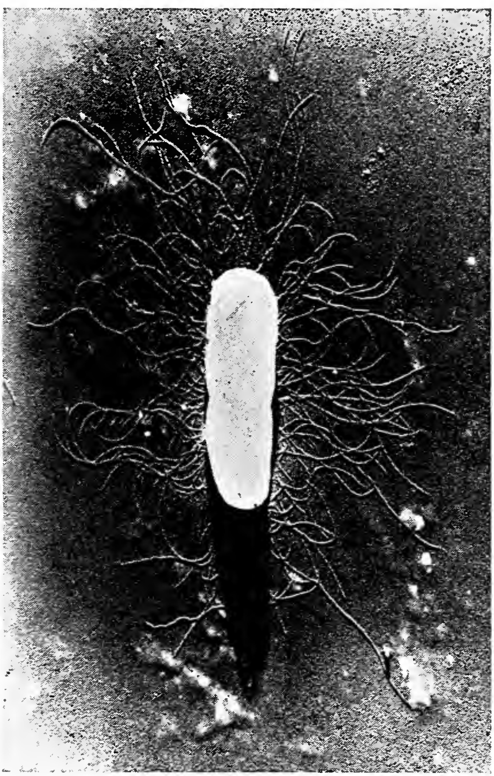


FIG. 34b. "Caryophanon"

drew attention to the excellent coating properties of chromium and uranium, which form fine, even, structureless layers. A thickness of 5–10 Å of uranium is sufficient to give well observable contrast. But the most often used metal is gold. Figure 34b is a brilliant example of a gold shadowcast, taken by Crowe and Robinow in the Cavendish Laboratory, Cambridge, England.

CHAPTER 11

THE SCANNING MICROSCOPE

IT has been mentioned in chapter 5 that attempts to produce an electronic counterpart of the metallurgical microscope were so far not very successful, nor are they likely ever to achieve the perfection of the transmission type of instrument. Transparent replicas will give information on the shape of the surface, but none on its constitution. It is therefore fortunate that we possess in the scanning microscope an instrument which gives information based on secondary emission, which is a highly sensitive function of the chemical constitution and physical state of the surface constituents.

The scanning microscope is entirely a by-product of television. Max Knoll, who by that time had left the Technische Hochschule, Berlin, for the Television Department of Telefunken, was the first to conceive the idea of producing an image of a surface by scanning it with an electron beam, collecting the secondary electrons, and applying the impulses to the modulating electrode of a television tube which is scanned in synchronism with the first beam.[61] A similar tube, called *Monoscope,* was later designed by C. E. Burnett,[62] of the R.C.A., for the purpose of testing television tubes. The same principle was utilized by M. von Ardenne for the first scanning microscope.[63] Finally, a highly perfected instrument with many novel features was developed by V. K. Zworykin, J. Hillier and R. L. Snyder,[64] in the R.C.A. Laboratory in Camden, a short description of which follows.

Figure 35 is a diagram of the electron optical arrangement. The problem here is to produce an extremely fine scanning spot of less than 500 Å diameter, which yet carries sufficient current to produce a signal at least ten to twenty times above the noise level of the amplifier. The first condition for this, according to equation (10) of chapter 2, is an electron source with high current density. After some experiments with thermionic cathodes the R.C.A. tried to use cold fine-etched tungsten points which by field emission can yield currents of the enormous density of 10,000 amp/cm^2, or even more. But these proved too unstable, and they had to return to incandescent tungsten cathodes.

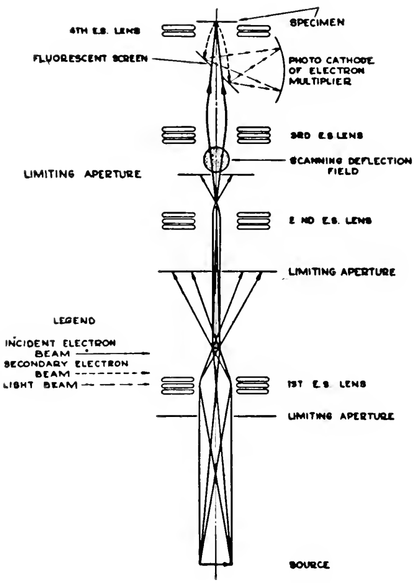


FIG. 35. Ray diagram of R.C.A. scanning electron microscope

A first electrostatic lens produces a cross-over which has been discussed in connection with figure 13. The cross-over serves as electron source for a second electrostatic lens which produces a strongly reduced image of it. An aperture cuts out rays beyond a certain angle, to reduce the spherical aberration. This is followed by a magnetic scanning field, similar to that used in television tubes, and by a third lens system which reproduces the first image on the specimen, with approximately unit magnification. On the way to the specimen the oscillating electron pencil passes an apertured fluorescent screen, and finally a fourth lens, which is placed immediately in front of the specimen. This lens has little influence on the scanning beam, but spreads out the secondary electrons released in the specimen over a wide area of the fluorescent screen which has a suitable positive potential against the specimen.

The beam current and the secondary electron current are extremely small, of the order of 10^{-13}–10^{-14} amp. The amplification of such minute currents with ordinary amplifiers is hopeless, the signal would be drowned by the noise which is mainly caused by the thermal fluctuations of voltage in the input

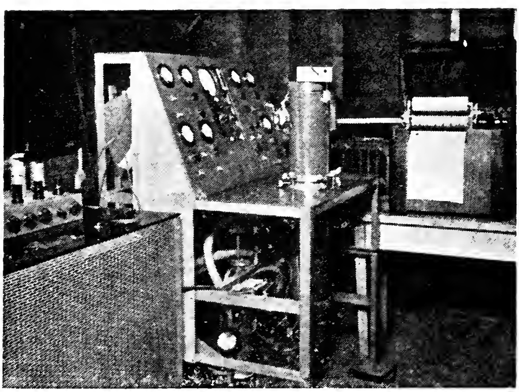


Fig. 36. R.C.A. scanning microscope, experimental form

resistor, parallel with the first grid. Let this resistance be R, the fluctuation voltage v, Boltzmann's constant k, the temperature of the resistor T, and the frequency band transmitted by the amplifier ΔF. Nyquist's well-known formula gives for the mean square voltage fluctuation

$$\bar{v^2} = 4kTR\Delta F \qquad (28)$$

The R.C.A. reduces this noise by two ingenious artifices. The first is employing an electron multiplier as the first stage of the amplifier. The multiplier receives on its photo-cathode the varying light intensity emitted by the fluorescent screen. The second artifice is to reduce the frequency band ΔF of the amplifier to 300 cycles, and take the micrograms extremely slowly, in about 8–10 minutes, so as not to lose any details of the picture. The amplifier operates on a facsimile recorder, which can be seen at the right of figure 36, and which prints a seven hundred fifty lines image every 10 minutes. Figure 37 is a sample of a scanning micrograph. The resolving power is of the order of 500 Å, which is sufficiently below the resolution limit of the optical metallographic microscope to promise the new instrument a wide and useful field of application.

FIG. 37. Scanning electron microgram of the surface of slightly annealed brass obtained with R.C.A. scanning electron microscope. Magnification $\times$ 5,000

CHAPTER 12

POSSIBILITIES OF FUTURE DEVELOPMENT

THE future development of the electron microscope is likely to proceed in two general directions. One line of development will aim at a simple, safe, and yet powerful instrument which will be as useful, and probably as indispensible in the routine tests of virus diseases, as the optical microscope is in bacteriological tests. The simplified R.C.A. electron microscope can be considered at present as the nearest approach to this ideal, but its resolution limit of about 100 Å still falls rather short of the limit of about 20 Å which appears desirable.

The second line of development will aim at a reduction of the resolution limit considerably below 20 Å, and to create a new tool for fundamental investigations in physics, chemistry, and biology. A discussion of these possibilities will be attempted here, which will be divided into two parts. First, it will be convenient to discuss possible improvements in the *uncorrected* type of microscope, that is, improvements more or less along the line which has led to the present stage. The second part is a survey of the possibilities for creating electron optically corrected microscope objectives.

Possible Improvements in the Uncorrected Electron Microscope

In chapter 7 we have derived equation (24) for the resolution limit of the uncorrected microscope, which results from the optimum balance of spherical aberration and diffraction. We write this now, somewhat more conveniently,

$$d_{sA} = 750C^{\frac{1}{4}}\left(\frac{f}{V}\right)^{\frac{1}{4}}V^{\frac{-1}{8}}\text{Å} \qquad (24b)$$

C is the constant of spherical aberration, which depends only on the shape of the field, not on its linear dimensions or intensity. The focal length, f, and the accelerating potential, V, are preferably used in the combination $\left(\frac{f}{V}\right)$ as in a magnetic lens field of given strength f is proportional to V, *i.e.*, $\left(\frac{f}{V}\right)$ is a characteristic constant of the lens. In electrostatic lenses $\left(\frac{f}{V^2}\right)$ should be chosen as a characteristic quantity, leaving $V^{\frac{1}{8}}$ in the final factor instead of $V^{\frac{-1}{8}}$. But this varies so slowly with V that we can leave it out of account in the following discussion, noting that we are free to chose V in such a way as to suit the thickness and scattering or absorbing properties of the object best. For very thin objects it may be best to make V only 10,000 or even 5,000 volts. Electrons with energies of this order have photographic efficiencies considerably below the optimum, which for most emulsions is somewhere between 50 and 100 kev.[65] But, at least in principle, it is easy to accelerate the electrons up to the optimum velocity before they reach the photographic plate. In practice, this introduces some complications as compared with present instruments in which all parts behind the object can be earthed. But the complication may be well worth while, as considerable improvement could be expected in the contrast and detection limit of small particles of low atomic weight, especially organic ones.

There are, therefore, only two effective possibilities for reducing the resolution limit: reducing the aberration constant C, or increasing the *lens strength* $\frac{V}{f}$, in electrostatic lenses $\frac{V^2}{f}$. Let us first consider the second possibility. The simplest method to increase the lens strength appears to be to scale down the lens dimensions, and scale down f in the same ratio by keeping constant the magnetic potentials between the poles, or the electric potential differences between the electrodes of the lens. In the case of magnetic lenses, this is of no avail, as the pole pieces of present-day magnetic objectives are already practically saturated, and the magnetic intensities cannot be further increased without the field spreading out over a larger region, thus frustrating the reduction of the pole piece dimensions. In electrostatic lenses, reduction of the dimensions by one order of magnitude is still possible, if the technique of precision watchmakers is used, but the problem is how to prevent breakdown of the voltage by auto-electric discharge. It has been suggested in German and American patent applications to avoid this by operating the microscope with surges of very short duration. (A.E.G., 17th Dec., 1938; S. Ramo and G.E. Co., 1st April, 1941, and 11th Sept., 1942.) But as f figures in equation (24b) only with the $\frac{1}{4}$ power even a reduction by a factor of $\frac{1}{10}$ would reduce the resolution limit only in the ratio $\frac{1}{1.78}$, and this would not be sufficient to bring the best electrostatic microscopes in line with the best magnetic ones. Surge operation is likely to produce potential differences by oscillations between the electrodes, therefore, the gain, if any, is likely to be even less.

E. G. Ramberg[52] was the first to point out that more could be gained by increasing the strength of existing lenses than by scaling them down, as this would reduce C as well as f, though he left it open how to achieve this in face of the limitations discussed above. But from the material computed by Ramberg it is easy to conclude that appreciable progress could be achieved not by scaling down present-day lenses, but, rather surprisingly, by scaling them up. Ramberg calculates a magnetic lens formed by two cylinders of infinite permeability, of diameter D, separated by a narrow gap, and gives f and C as functions of a parameter

$$R = \frac{(DH)^2}{V} =$$

$$\frac{(\text{lens diameter} \times \text{maximum magnetic field on axis})^2}{\text{accelerating potential}} \quad \frac{\text{cm}^2\text{gauss}^2}{\text{volt}}$$

Some values calculated from Ramberg's data are:

R	65	130	260	520	1040 $cm^2 gauss^2 volt^{-1}$
$\frac{f}{D}$	1.54	0.81	0.50	0.34	0.26
$\frac{c}{D^2}$	7.6	2.4	1.12	0.68	0.51 cm^{-2}
$\frac{D}{D_o}$	1	1.414	2	2.828	4
$\frac{f}{f_o}$	1	0.74	0.65	0.62	0.67
$\frac{C}{C_o}$	1	0.63	0.59	0.71	1.06
$\frac{Cf}{(Cf)_o}$	1	0.47	0.38	0.43	0.71

Consider a typical objective lens with $D = 0.25$ cm, $H_{max} = 10{,}000$ gauss with $V = 100{,}000$ volt. This gives $R = 62.5$, *i.e.*, it corresponds very nearly to the first column of the table. We take now this lens as unit, and refer the other columns of the table to it. Assuming that H_{max} is kept constant, these correspond to proportional increase of the linear dimensions in the ratios $\sqrt{2}$, 2, $\sqrt{2} \times 2$ and 4, as shown in the table below the line. It may be seen that both f and C decrease first, go through a minimum, and increase again. The most important parameter, the product Cf, which figures in equation (24b), has a minimum at twice increased dimensions, but this is so flat that even a three to four fold increase gives appreciably the same gain.*

As the dimensions of the lens increase, starting from small values, the object must be approached to the center of the field, and finally, at large values of R, it has to be positioned beyond the maximum, if it is to be focused at infinity. The optimum is reached when the object coincides with the maximum of the field, as was first pointed out by W. Glaser. Such an arrangement has been also considered in careful detail by L. Mar-

* The connection between lens strength and resolution has been discussed in detail by V. E. Cosslett, "The variation of resolution with voltage in the magnetic electron microscope," Proc. Phys. Soc., **58**, 443 (1946).

ton and R. G. E. Hutter.[66] But it appears that it has been put into practice for the first time in the new Philips commercial electron microscope which was developed on the basis of research work extending for several years by Dr. le Poole at Delft University. In the new Philips microscope the bore of the pole pieces is 11 mm. The advantage in the product $(Cf)^{\frac{1}{4}}$ as compared with typical lenses of narrow bore is about 20 per cent, a very valuable gain. But even greater advantages have been derived from the comfortable manipulating space provided by the 11 mm bore, which makes it possible to accommodate a movable objective aperture. It is also to be noted that the wide bore makes it much easier to achieve axial symmetry, and thus to approach the theoretical resolving power, which for this instrument is estimated at 3.5 Å.

Suggestions for Corrected Microscope Objectives

As the scope for improvements in electron microscopes with uncorrected lenses is very limited, it appears of interest to explore the possibilities of corrected objectives. Though the way to them bristles with difficulties, the prize may be worth the effort.

1. High frequency operated objectives

Two lens errors cannot be corrected by methods as used in present-day electron optics: the spherical aberration, and the chromatic aberration. R. Kompfner[67] made the very attractive suggestion to use these two enemies of resolution in such a way as to eliminate one another. We have seen in chapter 2 that spherical aberration makes the outer zones of a lens stronger than the inner ones, whereas the lens strength in every zone decreases with increasing velocity by *chromatic aberration.* Let us, therefore, make the outer electrons by E electron-volts faster than the axial ones, which have an energy V ev. From equations (20) and (21), we obtain for the apparent diameter of a point object, produced by the combination of spherical and chromatic aberration in a magnetic lens

$$d_{sc} = fa\left(Ca^2 - \frac{2E}{V}\right) \tag{29}$$

and this is zero if we make

$$E = \tfrac{1}{2}Ca^2V \tag{30}$$

i.e., if we give the outer electrons so much excess energy. As C is usually near unity this means about E = 3 ev if V = 60 kev at $a = 10^{-2}$ radian. From the point of view of geometrical electron optics the idea is entirely correct, and it could be realized, *e.g.*, by imaging on the object a cathode with a suitable radial potential drop. But in the electron microscope the wave nature of the electrons manifests itself very noticeably, and if we irradiate the object with different electrons at different angles, we must expect very strong diffraction, as each electron uses only, as it were, a small fraction of the aperture. Kompfner suggested to overcome this difficulty by producing *coherent* electrons of different velocities. His project is explained in figure 38a. The electron swarm emitted by the cathode is *chopped* into thin slices, and these slices traverse an accelerating space between two concentric grids, which is operated with the chopping frequency in such a phase that the outer electrons, which reach the grids later, are more accelerated than the inner ones. But, unfortunately, even if we assumed that the great technical difficulties could be overcome, the diffraction error in this arrangement would be much larger than Kompfner's estimate of 1.5 Å.

Heisenberg showed, in 1928, that the basic facts of quantum theory could be derived from what he called the Principle of Indeterminacy. It is meaningless to talk of an electron as a particle with a certain position and a certain velocity, as there is no possible or thinkable experiment by which these two quantities could be ascertained simultaneously. The mathematical formulation of this principle in a general form, and the proof that it gives results identical with the wave picture are rather difficult. But in its original and simplest form, it can be used very conveniently to account at least for the order of magnitude of diffraction effects, as can be seen by its application to Kompfner's problem.*

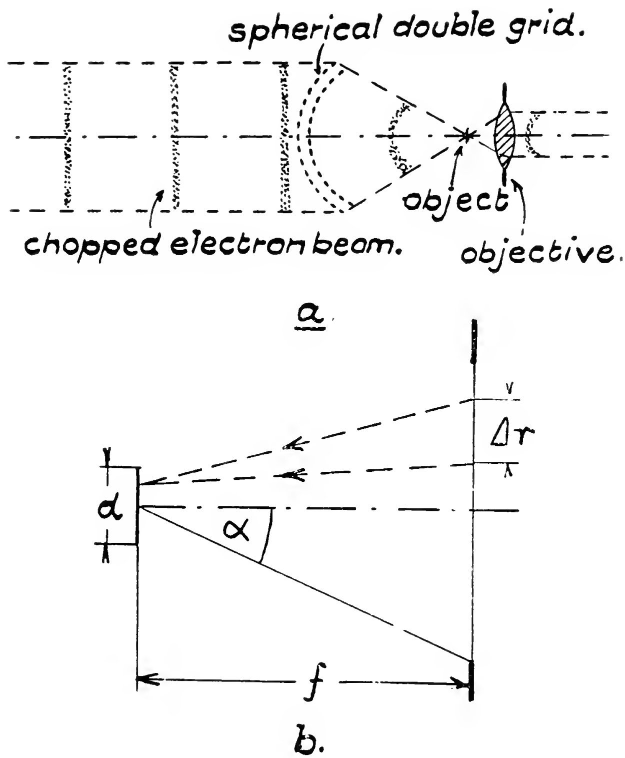


Fig. 38

As we always reduce errors to the object plane, we can conveniently imagine the lens replaced by a mirror, at a distance f from the object, as shown in figure 38b. The photographic

* *Cf.* also the very simple proof of the Abbé limit by Henriot, quoted by Marton, *et al.*[27, 49]

plate is therefore in the same plane as the object. The error disk may have a diameter d, which is the measure of indeterminacy of the position of an electron when it arrives at the plate. By Heisenberg's principle there is a corresponding indeterminacy Δmv_r in its radial momentum mv_r, connected with d by

$$d \, . \, \Delta mv_r = h \tag{31}$$

where h is Planck's constant. This, however, means that the zone r of the lens or mirror, from which this electron came is known only with an inaccuracy

$$\Delta r = f\frac{\Delta v_r}{v} \tag{32}$$

which, however, cannot be larger than the physical aperture $2af$ of the lens, as we know that the electron has passed the aperture.

By the indeterminacy contained in (31) and (32) we have replaced the *wave picture* and we can go on considering the electron as a particle of which we know certain data within certain limits only. We can therefore equate d to the error in d_{sc}, which would be produced by an electron straying by $\pm\frac{1}{2}\Delta r$ from the zone which it should pass in order to arrive at the geometrical image point. Writing $a = \frac{r}{f}$ we obtain by differentiating (29) and by eliminating $\frac{E}{V}$ by means of (30)

$$d = \left(\frac{3Cr^2}{f^2} - \frac{2E}{V}\right)\Delta r - 2r\frac{\Delta E}{V} = \left(\frac{2Cr^2}{f^2}\right)\Delta r - 2r\frac{\Delta E}{V} \tag{33}$$

For completeness we have added the error arising from an electron passing a zone with the wrong energy, but neglect it in the following. From (31), (32) and (33) we obtain Δr

$$\Delta r = \sqrt{\frac{\lambda f^3}{2Cr^2}} \tag{34}$$

where $\lambda = \frac{h}{mv}$ is the de Broglie wavelength. The diameter of the confusion disk becomes, if we express r by the aperture angle α

$$d = \alpha\sqrt{2C\lambda f} \tag{35}$$

This shows that diffraction has not entirely destroyed Kompfner's correction of the spherical aberration. The error diameter d increases only with the first power of the aperture instead of with the third. But α cannot be decreased below any limit, because, as remarked above, Δr cannot be larger than the physical aperture $2\alpha f$. Substituting $r = 2\alpha f$ into equation (34) we obtain for the smallest, *i.e.*, optimum size of the aperture

$$\alpha_{\text{opt}} = \left(\frac{\lambda}{8Cf}\right)^{\frac{1}{4}} \tag{36}$$

For 60 kev electrons, $f = 0.5$ cm and $C = 2$, this is 4.7×10^{-3} radian. The optimum error diameter becomes

$$d_{\text{opt}} = 0.85(Cf)^{\frac{1}{4}}\lambda^{\frac{3}{4}} \tag{37}$$

This, apart from the numerical factor, is the same formula as equation (24) which we obtained for the uncorrected microscope. If the relation between λ and V is taken into account, the numerical coefficient of equation (37) is found to be 0.73 of the factor of equation (24). But in the derivation of equation (24) we have allowed a factor of 0.67 for the ratio of resolution limit to confusion diameter. Making the same allowance in equation (37) we see that the maximum gain by Kompfner's suggestion is about $0.73 \times 0.67 = 0.5$. Though this appears an appreciable improvement, it would be almost certainly more than outweighed by the technical difficulties arising from the complication of the scheme.

2. Space charge corrected objectives

Equation (37), like equation (24), reveals the principal difficulty in improving the electron microscope. Not only are C and

f quantities which by means, known at present, cannot be reduced below certain limits, but they figure in the $\frac{1}{4}$ power, so that only very radical improvements can appreciably reduce the resolution limit. The suggestion of the author, to be now described, seems to offer at least a possibility of reducing Cf very considerably. Though the factors C and f are intimately connected, it will be convenient to start with a discussion of the focal length f by itself.

It may be noted that a reduction of f appears not only desirable but also necessary in any scheme for an efficient reduction of the resolution limit, as the error diameter caused by fluctuations of the driving voltage in the magnetic lens

$$d_c = 2af\frac{\Delta V}{V} \tag{20}$$

is proportional to f. If we want to reduce d_c, and at the same time increase the aperture a in order to reduce the diffraction error, we must very strongly reduce the fluctuation $\frac{\Delta V}{V}$ or f or both. But it appears from Vance's investigations that a substantial reduction of the fluctuation will be very difficult, and progress in this direction is bound to stop anyway once the fluctuation becomes of the order of the initial velocities of the electrons emitted by thermionic cathodes. We must therefore look for ways to reduce f.

The problem of the short focus lens is illustrated in figure 39. The focal length is given by the abscissa of the intersection between the initial tangent, starting from the focal point, and the final tangent, which is parallel to the axis. Between these two limits the trajectory can have different shapes, and these will strongly influence the spherical aberration. R. Rebsch has shown,[48] that if there were no limit to the field intensities which we can produce, the spherical aberration could be reduced below any limit. It would be sufficient to produce an infinitely strong field in an infinitesimally small interval near the focal point, which could be followed by a weak lens field to bring the focal length f to any prescribed value. It has been shown in the foregoing section that recent developments of the uncorrected microscope are making use of strong fields near the object in the small margin which is still left for this type of instrument, but for radical improvements it is of no value. We have rather to go the opposite way, as illustrated in the third part of figure 39. Instead of producing a very strong, short condensing lens near the object, it is suggested to produce a short focus *by following up an ordinary condensing lens by a long diverging lens.*

It has been proved in chapter 11 that trajectories with an inflection can never be realized with magnetic lenses, as the trajectory always bends toward the axis. Are they also impossible with electrostatic lenses? The answer is: Not impossible, but impracticable. This is illustrated in figure 40, which shows several trajectories of the desired type, with the corresponding lens field. The lens potentials had to be drawn on a logarithmic scale in order to accommodate the enormous values which are neces-

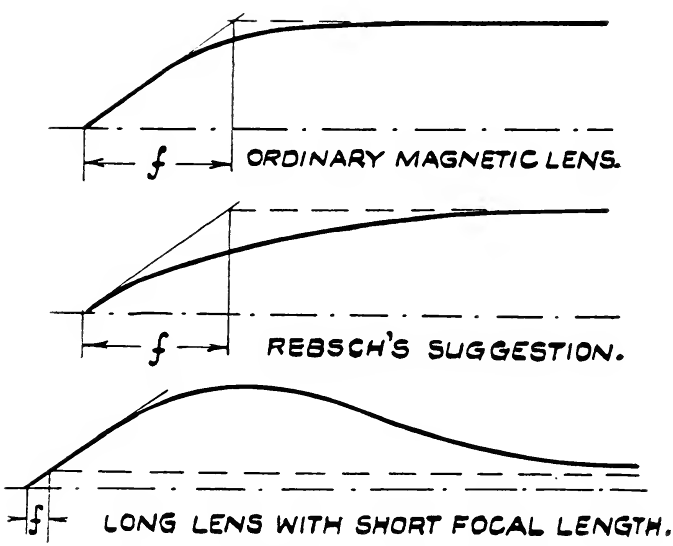


FIG. 39. The problem of the short-focus lens

sary even for a moderate reduction of the focal length. The asymptomatic values of the potentials, measured in units of the object potential, are marked on the trajectories. Only very weak diverging lenses can be therefore realized, at a prohibitive cost in voltage.

We must now remember the clause which accompanied the theorems on the impossibility of diverging lenses and corrected lenses: without space charge. With negative space charges it would be obviously possible to produce diverging lenses, as the electrons nearer the axis would repel those outside. The problem is only how to realize negative space charges of considerable magnitude. An elementary estimate shows that we require charge densities of the order of a hundred electrostatic units per cm^3, which could be realized with electron concentrations of the order 10^{11}–$10^{12}/cm^3$. Concentrations of this order produce by their space charge voltages of the order of 100–1,000 volts in devices of a few cm, and if they are in motion with velocities of this order, they produce currents of the order of hundreds of amp, which, if they were allowed to flow to the electrodes would soon convert the device into a vacuum furnace. Therefore, it is imperative to keep these huge space currents away from the electrodes, and to make them circulate inside the vacuum space. A device which realizes this to some extent is known for a long time. It is the *static magnetron* of A. W. Hull.[68] Figures 41–44 illustrate a suggestion by which the principle of the static magnetron could be adapted for the solution of the problem of corrected objectives with very short focus.

Figure 41 is a longitudinal section of the projected objective, with no claim as regards correct dimensions. The movement of the electrons which image the object, and which will be called for short *beam electrons,* is from left to right. The objective consists of two lenses, a condensing lens at the left, and a diverging, *space charge lens* at the right. The condensing lens is a combination of a magnetic lens, and of a *unipotential* electrostatic lens. The reason why such a combination has to be used will be explained later. The magnetic and the electrostatic field are shown separately. The middle electrode "E" of the unipotential lens is connected with the cathode as usual, whereas the outer electrode, preferably earthed, is formed by the pole pieces of the magnetic lens. The object is placed outside this lens, shielded from the strong electrostatic fields by an apertured diaphragm. The focal length of this first lens is supposed to be of the same order as in present-day microscopes, *i.e.*, about 0.5 cm for 60 kev electrons.

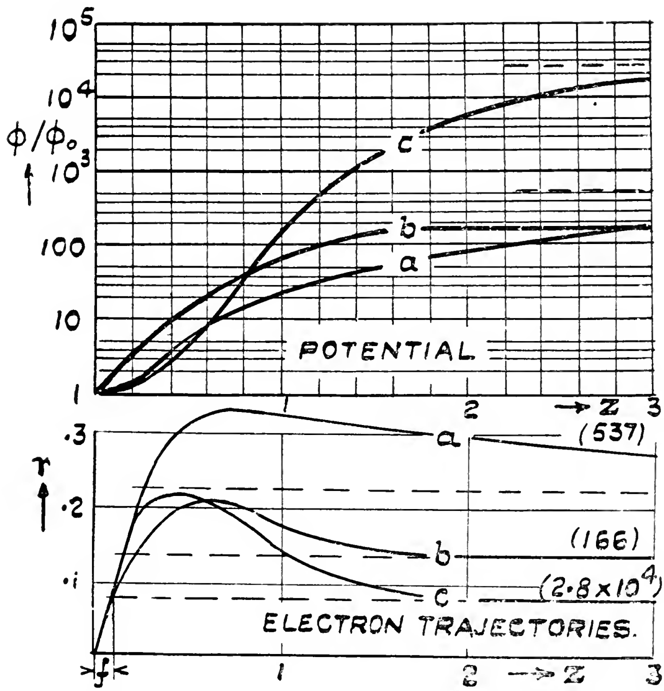


FIG. 40. Potential and trajectories in electrostatic lenses

The space charge lens is essentially a cylindrical magnetron, but with the important difference that it contains no axial cathode. Its main parts are a long coil, which produces, at least in the middle part, a nearly homogeneous magnetic field, a tubular anode or *sheath electrode* "A," and a cathode "C," which is a filament, bent to a coaxial circle. The currents are flowing in the two coils in opposite directions, so that they produce on the axis magnetic fields H_0 of opposite signs. If the field at the right is somewhat weaker than the field at the left, the axial point in which H_0 changes its sign will be at the right of the partition. A field line of roughly parabolical shape starts from this point. The cathode is placed at or near to this field line, so that there is no magnetic flux between the cathode and the axis.

The reason for this particular arrangement of the cathode can be understood from equation (2) in chapter 2, which states that an electron moving in an axially symmetrical magnetic field acquires an angular momentum around the axis proportional to the flux between the circle from which it started and the circle at which it arrives. The angular momentum of an electron which reaches the axis is zero, and for the present we will neglect the initial velocities at the cathode. This means that an electron starting from the cathode can reach the axis only if it has acquired no momentum, that is, if the flux between the cathode and the axis is zero. Collisions between electrons and between electrons and gas molecules would make the condition less stringent, but we will neglect these for the present.

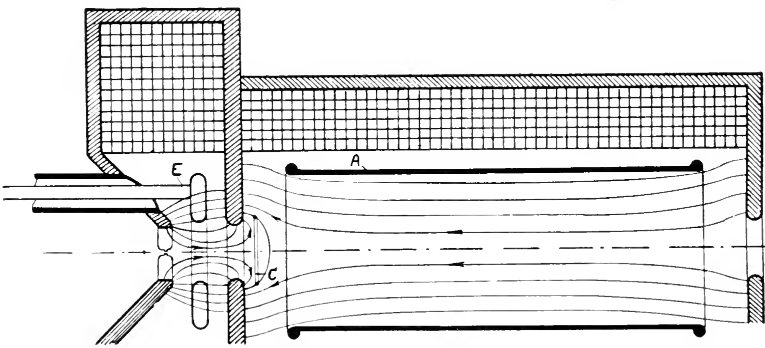
The boundaries of the space which is accessible to electrons can be easily obtained from the *equivalent electrostatic potential*, U, which was introduced in chapter 2. Equation (8) was derived for exactly the case which applies here, *i.e.*, for electrons which need not cross a magnetic flux to reach the axis. In the accessible space U must be positive, *i.e.*,

$$U = \phi - \frac{e}{2mc^2}A^2 \geqq 0 \tag{38}$$

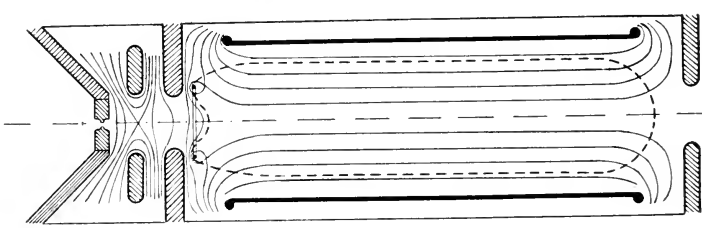
We can restrict this space therefore either by making ϕ negative, or by making A (which is defined by equation (3) in chapter 2) large. It is suggested to use limitation by strong magnetic fields on the cylindrical part of the boundary, and limitation by negative potentials at the ends. In the middle part where the magnetic field H is nearly uniform the condition is, as here $A = \frac{1}{2}Hr$

$$\phi = \frac{e}{8mc^2}(Hr)^2 \tag{39}$$

and the electrons will not be able to reach the sheath electrode "A" if its potential V against the cathode is less than a certain value V_c which is obtained by substituting the radius of "A" into equation (39). V_c is Hull's critical potential. It is known that in practice V has to be made somewhat smaller than V_c if anode currents are to be avoided. This effect is probably due to electron collisions, and has never been fully explained.



41a
Magnetic Field



41b
Electrostatic Field
Corrected Objective

FIG. 41

At the ends, where the magnetic field goes to zero, limitation is obtained by negative potentials, which means that the potential of the cathode "C" must be positive against the magnetic casing by a suitable amount. By these two measures the electrons are trapped in a space, the approximate outlines of which are shown in the second figure 41 in dotted lines. They cannot escape anywhere, except back to the cathode, and as the filament is very thin, this will happen very rarely, in general only after the electron has traversed the accessible space many times. Therefore, according to this rather over-simplified picture, the negative space charge could be maintained in a stationary condition with no input at all.

The actual distribution of space charge within the accessible space presents a very difficult problem. The static magnetron is usually explained by Hull's simple theory, which has been only slightly modified by Pidduck [69] and Brillouin.[7] According to these theories, the distribution is the same as if the electrons were all rotating on coaxial circles, held in equilibrium by the radial electric field and by the inward directed force exerted by the magnetic field; the law of the electric field is given by the condition that this equilibrium should be possible at any radius. This gives a quadratic increase of the potential with the radius, and by Poisson's equation a constant space charge density

$$-\rho = \frac{eH^2}{8\pi mc^2} \tag{40}$$

Numerically this is $2.33 \times 10^{-5}H^2$ electrostatic units, if H is substituted in gauss. At $H = 2{,}000$ gauss, this gives 93.2 elst.units/cm^3, or 1.94×10^{11} electrons/cm^3, which is of the desired order of magnitude. But in fact, the effect would be zero, as the concentrating effect of the magnetic field would exactly counterbalance the dispersing effect of the space charge. This can be immediately understood from the way in which Hull's equation was derived. It is also shown explicitly by the path equation of paraxial electrons which is conveniently written in a form due to Scherzer:

$$R'' = -\left[\frac{3}{16}\left(\frac{\Phi'}{\Phi}\right)^2 + \frac{1}{\Phi}\left(\frac{eH^2}{8mc^2} + \pi\rho\right)\right] R \tag{41}$$

Here Φ is the potential on the axis produced by the electrodes alone, H and ρ are the values of the magnetic field and space charge density along the axis, the primes denote differentiation with respect to the axis z, and R is

$$R = r\Phi^{\frac{1}{4}} \tag{42}$$

It can be seen that the magnetic field is compensated by the space charge if Hull's value ρ_H is substituted. Therefore, if Hull's were the true or only possible distribution of charges in a static magnetron, the diverging lens as proposed would be an impossibility. But there are good reasons to doubt this.

The author has carried out a theoretical investigation of the problem of stationary distribution of electrons in combined electric and magnetic fields, to be published elsewhere,* which starts from more general assumptions. Instead of considering equilibrium orbits, the theory assumes that the electron trajectories inside the accessible space are so long, so complicated, and so irregular, that statistical considerations can be applied to them. This leads to a distribution, illustrated in figure 42, which is essentially different from Hull's solution. In figure 42 it is assumed that the cathode is not placed exactly into a position of zero magnetic flux, so that the electrons cannot reach the axis, and the accessible space becomes annular, bounded inside and outside by two radii, r_1 and r_2. It can be easily shown from the momentum theorem (2) that these will be the two roots of the equation

$$\phi = \mathcal{A}^2 \equiv \frac{e}{2mc^2}\left(A - \frac{A_0 r_0}{r}\right)^2 \tag{43}$$

where A_0 and r_0 are the values of A and r at the cathode. Statistical mechanics gives the result, that between these two limits the electron density will distribute itself in inverse ratio to the distance from the axis, *i.e.*, according to $\rho = \text{const.}/r$. The con-

* D. Gabor, Stationary Electron Swarms in Electromagnetic Fields, *Proc. Roy. Soc. A.*, **183**, 436 (1945).

stant is determined by the ratio beween electron injection and removal. If the removal is negligible, electrons will accumulate in the accessible space until they have depressed the potential at the inner radius to the level of the cathode potential. The inner radius itself moves outward during this accumulation, until it reaches the magnetic field line passing through the cathode. From this moment on, no further electrons can leave the cathode; the cloud is saturated. The initial velocities of the electrons will produce near the boundary departures from the $\frac{1}{r}$ law, which results in blurring of the edges.

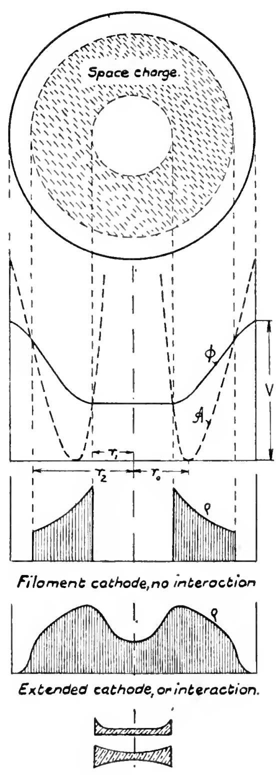


Fig. 42

The calculation shows that if the outer radius r_2 is large in comparison with r_1, the saturation value of the density at r_2 will be very nearly one-quarter of Hull's density ρ_H. Inside, it can be very much larger, according to the approximate law

$$\rho = \rho_H \frac{2r^2}{r} \qquad (44)$$

An electron cloud of this type *can* therefore act as a divergent lens. This can also be immediately understood. The electrons in the cloud are not in radial equilibrium, as in Hull's picture of the magnetron, but they are reflected to and fro between the inner and the outer boundary. The forces near the inner boundary are repellent, and it is easy to show that if r_1 is sufficiently small in comparison with r_2, a certain inner zone will act as a divergent lens. But the law of repulsion is not of the type required for an electron lens. Instead of increasing linearly with r, it falls off with increasing radius, and changes sign at a certain point. This is particularly marked if we arrange the cathode so that the electrons can reach the axis. In this case the density at the axis becomes infinite. This, of course, is a consequence of certain simplifications, notably the fact that we have assumed the cathode to have zero radial extension, and neglected the initial velocities.

If extended cathodes are used, the distribution of density can be modified in a way as is shown in figure 42 (*d*). The interaction of electrons in the cloud is very difficult to treat quantitatively, but it can be expected that moderate interaction would produce a similar distribution, even if the cathode is made with as small radial extension as can be practically realized. Its first effect will be a smoothing of the corners, and filling up of the *prohibited space* with electrons, approximately as shown in the figure. In order to keep the interaction sufficiently small, it may be necessary to remove the electrons fairly rapidly, before they have time to establish perfect equilibrium by collisions. In order to avoid heat development, this will be done preferably by means of collector electrodes connected with the cathode, and arranged near the ends of the accessible space. (The collectors are not shown in figure 41.) Determining their size and shape will be best left to experiment, for electron interaction is the most complicated and the least explored problem in the whole field of electronics.

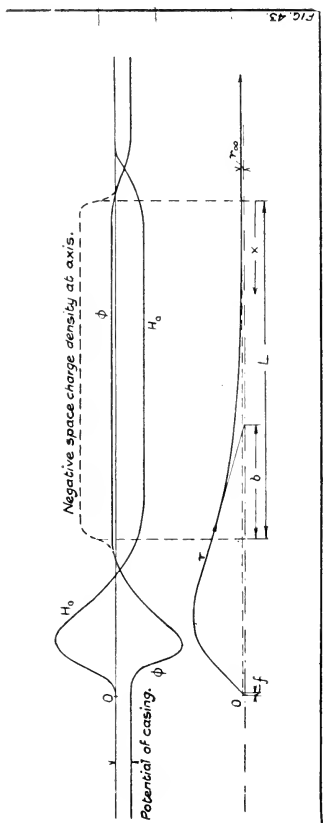


FIG. 43. Electromagnetic field and electron trajectories in corrected lens

The roughly parabolic shape of electron density distribution which has been assumed in figure 42 is suitable not only for producing a divergent lens, but also for correcting the spherical aberration introduced by the first, condensing lens of the system. An approximate explanation is contained in the bottom figure of figure 42. In a homogeneous electron cloud the radial electric field intensity is proportional to the radius. Therefore, it acts very nearly like an ordinary spherical lens. The parabolic distribution superimposed on it, however, has an effect approximately equivalent to a lens with a surface figured according to a parabola of the fourth order. Such correcting plates are used in various optical systems, of which the Schmidt camera is the first, and the best known.

The compensation of spherical aberration by space charges can be also understood by considering Scherzer's formula for the aberration of a purely magnetic lens. In our notation

$$Cf = \frac{1}{4\Phi}\frac{e}{mc^2_0}\int\left[\left(H' + H\frac{r'}{r}\right)^2 + \left(H\frac{r'}{r}\right)^2 + \frac{e}{4mc^2}H^4\right] r^4 dz \quad (45)$$

r denotes a trajectory, calculated from the paraxial equation (41), which starts from the focal point with $r' = 1$, *i.e.*, at 45°. The integration has to be carried out over the axis z from the object to the image. The expression can never be zero, as the integrand is a sum of positive squares. But it can be seen from equation (41) that a negative space charge has the same effect as an *imaginary magnetic field,* therefore, the integrand of (45) can well become negative in space charge lenses

Equation (45) shows that the short focal length of the suggested lens system does not by itself ensure a reduction of the spherical aberration. The factors outside the bracket are of the same order of magnitude as in the usual uncorrected lenses, therefore improvements can be effected only by reducing the factor in brackets. This is bound to be hard work, as the resolution limit depends on $(Cf)^{\frac{1}{4}}$, therefore, a compensation of the spherical aberration to 1 per cent would reduce the resolution limit to little less than one third of the uncorrected value. But it may be remembered that the optical microscope has improved since Abbé's time by less than a factor 2, and yet the progress in bacteriology has amply repaid the efforts spent in developing the ultraviolet miscroscope. It appears that the present limit of about 10 Å conceals as important details of viruses as 2,000 Å did in bacteria. Moreover, the short depth of focus resulting from increased aperture and improved resolution promises to reveal far more detail than could be expected from a comparison of the resolution limits. It will be, therefore, of interest to obtain an estimate of these quantities.

Figure 43 is a diagram of the fields in the lens combination according to figure 41, together with an electron trajectory. For simplicity let us consider the space charge density ρ_0 at the axis as constant along a length L. As the potential along the axis, Φ_0, can be also considered as constant, the paraxial equation (41) simplifies to

$$r'' = -\frac{1}{\Phi_0}\left(\frac{eH^2}{8mc^2} + \pi\rho_0\right) r = \frac{r}{L_e^2} \qquad (46)$$

L_e is a real length if, as we have assumed, the space charge term outweighs the magnetic term and the second lens acts as a diverging lens. The solution for the trajectory which leaves the system parallel to the axis becomes

$$r = r_\infty \cosh\left(\frac{x}{L_e}\right) \qquad (47)$$

If L_e is appreciably shorter than L, the space charge lens will reduce the distance of the trajectory from the axis approximately in the ratio

$$2 \exp\left(-\frac{L}{L_e}\right)$$

We can therefore reduce r_∞ in any desired ratio by making the space charge lens longer. Lengthening it by L_e means a reduction in the ratio $\frac{1}{e} = 0.368$. The focal length is reduced in the same ratio. In practical units we can write equation (46)

$$\frac{1}{L_e^2} = 0.022\left(\frac{\rho_0}{\rho_H} - 1\right)\frac{H^2}{V} \qquad (48)$$

where the energy of the beam electrons, V, has to be substituted in volts. As an example, if we realize at the axis a charge density twice Hull's value, and assume $V = 60$ kV, $H_0 = 1{,}650$ gauss will be required to make $L_e = 1$ cm. Therefore, if $L = 5$ cm, the off-axis distance of the trajectory will be reduced to $2e^{-5} = \frac{1}{75}$ with $H = 3{,}300$ gauss even to $\frac{1}{11{,}000}$. The reduction of the focal length of the composite lens as compared with the focal length of the first lens is of the same order. Therefore, it does not appear impossible to realize focal lengths of the order of one μ, and magnifications of the order of 100,000 with a throw of only 10 cm.

The focal depth of an objective can be defined as the axial interval or object thickness Δz which can be simultaneously focused within the resolution limit d_r. A simple consideration gives.

$$\Delta z = \frac{d_r}{a} \qquad (49)$$

In present-day microscopes d_r is not less than 25 Å, and a not more than 5×10^{-3}, therefore, the focal depth is not less than about 5,000 Å or half a micron.* But if Cf can be reduced to $\frac{1}{100}$, d_r will be reduced to $\sqrt{0.1} = 0.316$ and a increased in the same ratio, which means that the focal depth is reduced by a factor 10, *i.e.*, to about 500 Å. This is a considerable advantage in exploring details of objects, but it raises certain difficulties. It might appear that it is an easy matter to keep an objective focused within about $\frac{1}{20}$ of its focal length, as it is possible to

* Practically, often of the order of 10 μ.

keep present-day objectives constant within $\frac{1}{10{,}000}$. But it must be remembered that the short focal length of the suggested objective has been obtained not by a reduction of geometrical dimensions, but by a sort of compensation between two large systems. Fluctuations in the driving voltage will produce in it a new phenomenon which deserves special attention.

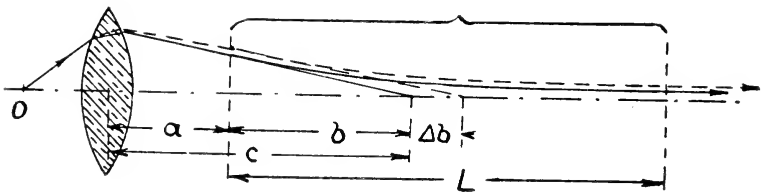


FIG. 44. Achromatic condition

It can be easily shown that fluctuations of the driving voltage will produce relative variations of the focal length of the composite lens of the same order as the relative fluctuation of the voltage. Because of the very short focal length, its absolute variation will be entirely negligible. But the new phenomenon arises that unless special measures are taken, the focal point will *shift* along the axis by about the same amount as the variation of the focal length of either of the two component lenses, taken by itself. Assuming that the constancy of the driving voltage is not better than in present-day microscopes, the *shift* in the above example would be about ten times the focal depth, and would frustrate the superior resolution. Instead of calculating the shift, we can at once indicate the measures to compensate it. The intercept of the tangent to the trajectory where it enters the space charge lens is

$$b = \left(\frac{r}{r'}\right)_{(x=L)} = L_e \coth \frac{L}{L_e} \tag{50}$$

and if $\frac{L}{L_e}$ is large, this will be very nearly

$$b \approx L_e = \Phi^{\frac{1}{2}} \cdot \left(\frac{eH^2}{8mc^2} + \pi\rho_0\right)^{-\frac{1}{2}} \tag{51}$$

The second factor depends on the magnetic field and the space charge which in turn depends on H and on the potential of the sheath electrode "A." As there is no current flowing to "A," and as the time constant of the second coil can be made very long, it appears possible to keep the second factor constant within much finer limits than the first. Φ_0 is the driving voltage V, only expressed in electrostatic units instead of in volts. We obtain, therefore, for the relative fluctuations of b which are produced by inconstancy of the driving voltage

$$\frac{\Delta b}{b} = \frac{\Delta V}{2V} \tag{52}$$

The meaning of this is explained in figure 44. If the driving voltage increases by ΔV, the point "O" will still remain in focus, provided that ΔV produces such a change in the first lens that it forms now an image of "O" shifted to the right by Δb, as given by equation (52). If the first lens were all electrostatic, the shift would be zero, if it were all magnetic, equation (52) would lead to inconvenient dimensions. This is the reason why it is suggested to make the first lens a combined lens. Application of the elementary lens equation, combined with the fact that the focal length of a magnetic lens is proportional to V gives

$$\frac{\Delta c}{c^2} = \frac{\Delta f_m}{{f_m}^2} = \frac{\Delta V}{V f_m} \tag{53}$$

where f_m is the focal length of the magnetic part of the first lens. As $\Delta c = \Delta b$, we obtain by combining (53) with (52)

$$f_m = \frac{2c^2}{b} \tag{54}$$

By itself this would be a somewhat weak lens, it therefore appears preferable to add a unipotential electrostatic lens to it. This complication could be avoided if it should prove possible to reduce $b \approx L_e$ to about 2 mm or less. This appears by no means impossible especially if use is made of the suggestion in the previous chapter, to operate the lens system with a beam voltage of only 5–10,000 volts, and bring up the electrons by after-acceleration to optimum photographic efficiency.

It may be noted that though by condition (54) the suggested lens system is made insentitive to fluctuations of the driving voltage, it is *not* achromatized, in the sense that it will not correctly focus electrons which have suffered inelastic collisions in the object. This is neither necessary nor desirable in view of the beneficial effect of the chromatic aberration on the contrast.

There is one more question which is naturally provoked by the suggestion of using an electron cloud as a lens: Will the cloud not act on the beam like frosted glass? This is a very difficult question to decide, as the only experimental data known to the author are the classical experiments of I. Langmuir [70] on the scattering of electron beams in ionized gases. But these were carried out with 100-volt electrons, and in a plasma, in which the ions might also play a part, difficult to estimate. It can be said only, that both, from a somewhat bold extrapolation of Langmuir's results and from an unpublished theoretical investigation of the author, it appears that the effect will not cause serious interference. It appears highly desirable to settle this point by the simple experiment of shooting a fast electron beam parallel to the axis through a static magnetron. It may be mentioned that such an experiment would also at once settle the question how nearly Hull's simple theory agrees with actual conditions in a magnetron, as on Hull's theory the beam would suffer no radial deflection.

The Proton Microscope

A most interesting attempt toward higher resolutions is being made in France, at the Collège de France, Paris, under direction of M. Magnan and the general and theoretical guidance of Louis de Broglie. A microscope with protons, that is to say atomic hydrogen ions instead of electrons, is in the course of construction, after careful preliminary studies, both theoretical and experimental, extending over several years.

It is easy to see that the proton microscope should have, in principle, a resolution limit appreciably better than any uncorrected electron microscope. The de Broglie wavelength λ of a particle of mass m and charge e which has dropped through a potential V and has acquired a velocity v is, by equation (12) on p. 15

$$\lambda = \frac{h}{mv} = \frac{h}{\sqrt{2meV}}$$

and is, therefore, for particles of equal charge and equal accelerating voltage, inversely proportional to the square root of the masses. The mass ratio of protons and electrons is 1840, and its square root 42.9.

Introducing λ instead of V in equation (24) on p. 39, the resolution limit as determined by spherical aberration and diffraction, without chromatic error becomes

$$d_{sA} = 1.14C^{\frac{1}{4}}\lambda^{\frac{3}{4}}f^{\frac{1}{4}}$$

This formula is valid for protons as well as for electrons. Thus if it were possible to use in the proton microscope objective lenses of the same quality and strength as in the best electron microscopes, that is to say with the same values of C and f, and at the same voltage V, the middle factor would give an advantage of $42.9^{\frac{3}{4}} = 16.8$ in favor of the proton microscope. But not all of this huge gain can be realized. It is not possible to use magnetic lenses for protons, as for the same strength, at the same accelerating potential, the magnetic fields would have to be 42.9 times stronger. Therefore, proton microscopes can be operated only electrostatically. But the spherical aberration coefficient of the best electrostatic lenses is about ten times larger than for the best magnetic lenses, so that the gain is reduced by a factor of about $10^{\frac{1}{4}} = 1.78$ to about 9.5. Moreover, it will be necessary according to calculations of Louis de Broglie to raise the operating voltage to at least 300 kv, if sufficient penetrating power is to be achieved. It appears rather doubtful whether at such high voltages lenses of sufficient power can be realized without autoelectronic breakdown.

Another consideration, which reduces the gain further, is that it may be very difficult to keep the chromatic error at the required low level, as protons have to be generated by gas discharges, which may not yield currents of as homogeneous velocities as thermoelectric cathodes, in the case of electrons. But it appears that this capital difficulty has been satisfactorily solved, and the inventors of the proton microscope have powerful sources of thermal protons at their disposal. They expect to realize a gain of about 8, which would mean a resolution limit below 1 Å.

One grave objection naturally presents itself. Will the protons with their relatively huge masses not smash up the structures which they are intended to explore? This is rather generally expected outside the circle of the Collège de France, although the inventors themselves are optimistic. Perhaps the situation will repeat itself which existed, round about 1927–28, with regard to the electron microscope. Many physicists turned away from the project, as they expected the electrons to burn fine organic structures to a cinder. Perhaps the optimism of the Collège de France circle will be similarly rewarded as that of Knoll and Ruska.

CHAPTER 13

THE ULTIMATE LIMIT OF ELECTRON MICROSCOPY

IT will have become sufficiently clear from the last chapter, that future development of the electron microscope is bound to be slow and arduous. This is mainly due to the factor $(Cf)^{\frac{1}{4}}$ which promises only an improvement by a factor of 3.16, even if 99 per cent of the spherical aberration could be successfully eliminated. There are also other difficulties to be considered, such as the question of mechanical stability, and especially the question of intensity. There is no difficulty in providing sufficiently intense illumination. If the resolution is improved say k times, this means that in order to get all the available detail the object has to be imaged on a k^2 times larger area of the photographic plate. But as by the Airy-Abbé relation the aperture has also increased in the ratio k, the solid angle of the illuminating beam increases in the ratio k^2, which means that the exposure remains the same. But the electronic bombardment of every part of the object is now k^2 times stronger, and the danger arises that beyond a certain point progress must stop, as we destroy the details that we want to study more closely. This is particularly true in the case of highly complex organic molecules. As regards living matter, as von Ardenne [71] has shown on the basis of experiments by C. P. Haskins, the limit is already reached in present-day electron microscopy. Even the most tenacious spores would be killed if we tried to take high resolution pictures of them. This reminds one of the somewhat pessimistic saying by N. Bohr, that we shall never know life entirely, as we have to destroy it in order to study it. But though we may never know everything, this does not mean that we cannot learn a great deal more. Even if electron bombardment should reduce the highly complex organic substances to a sort of carbon skeleton, the chemist may be able to reconstruct the original, as the palaeontologists have reconstructed extinct animals from their skeletons.

But there are many simpler substances, in particular the metals and all the elements which, unless they are melted or volatilized, will never be destroyed by electron bombardment, however prolonged. How far can we hope to proceed in their exploration by the electron microscope? In particular, can we expect to see single atoms, and to what detail? Let us for the moment abstract from the spherical aberration. However great the difficulties in the way of reducing it, they are of a technical order. But there are other limitations of a more absolute nature, inherent in the exploring agent, the electron, and in the object, the atom.

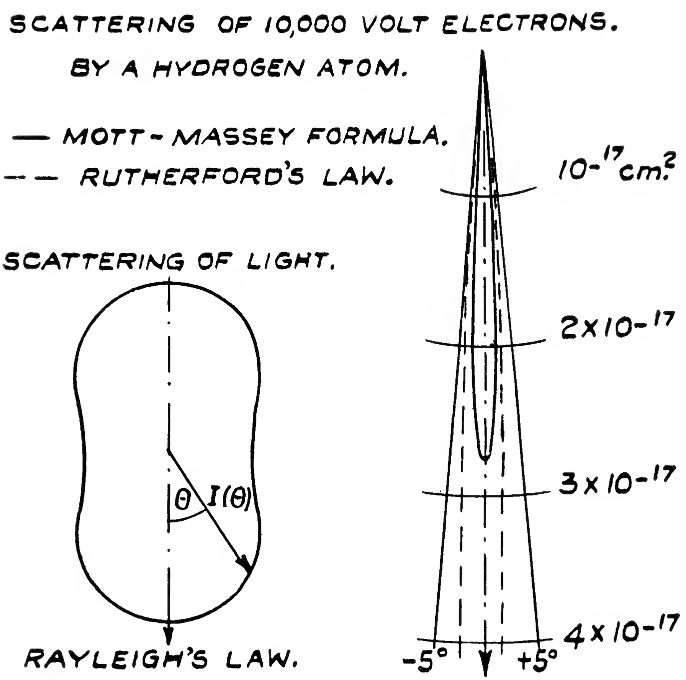


Fig. 45. Scattering of light and of electrons

Scattering of electrons by single atoms is considerably different from the dispersion of light by small particles. Figure 45 shows, side by side, the scattering of a parallel light wave by a body, very small in comparison with a wave-length, and the scattering of 10,000 ev electrons by a hydrogen atom. This example has been chosen, because rigorously calculated data are available. (Mott and Massey,[39] p. 120 and 174.) The light intensity scattered at an angle θ follows Rayleigh's law

$$I(\theta) = I_0(1 + \cos^2\theta)$$

which is symmetrical to the plane of the wave, *i.e.*, it is the same forward as backward. Electrons, however, are scattered almost exclusively forward, and practically all within an angle of 5°. The angle of the cone of scattering is inversely proportional to $\sqrt{V}$. The reason for this different behavior is the very much larger momentum of electrons as compared with photons of comparable energy.

Figure 46 shows the same phenomenon in more detail. The scattering is here divided into elastic and inelastic components. It can be seen that the inelastically scattered electrons are in general deflected by smaller angles. This can be roughly understood from the classical picture, as these electrons have collided with the hydrogen electron, while the elastically scattered ones have collided with the nucleus. They are far less numerous. At 100,000 ev, for instance, they represent only 5.1 per cent of the total scattering, while the other 94.9 per cent suffer an average loss of 21.7 ev. At 10,000 ev the figure is 6.5 per cent, at 1,000 ev 8.7 per cent.

Figure 46 shows also for comparison scattering by the hydrogen ion, *i.e.*, by a proton. This is the well-known Rutherford formula

$$I(\theta) = \left(\frac{Ze^2}{2mv^2}\right)^2 \sin^{-4}\tfrac{1}{2}\theta$$

where Z is the atomic number; in the case of hydrogen, $Z = 1$. This is a law of very different character, as it goes sharply to infinity for $\theta = 0$, while $I(0)$ for hydrogen, or any other neutral atom is large but finite. Consequently the *total scattering cross section* of an ion is infinite, because the long-range Coulomb force affects electrons appreciably at any distance. This is the explanation of Hillier's paradoxial result,[38] that an atom, for which he assumed Rutherford's Law, ought to produce the best contrast at zero aperture.

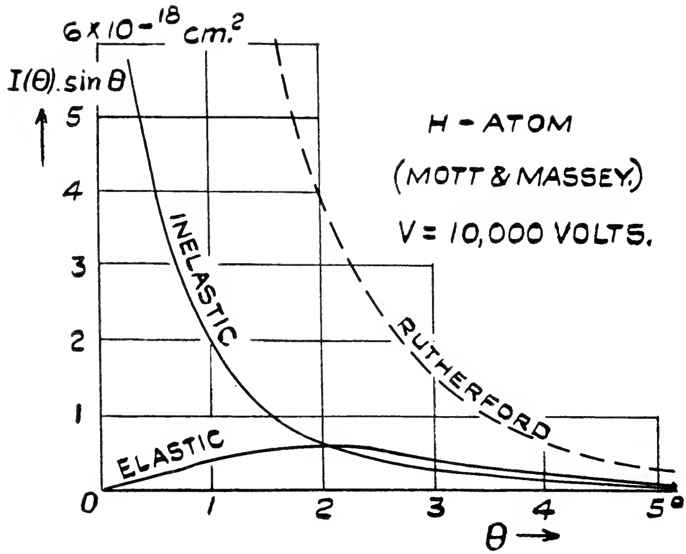


FIG. 46. Scattering by hydrogen atom

The total scattering cross section of a neutral atom on the other hand is finite. In the instance of a hydrogen atom and 10 kev electrons it is 4.26×10^{-18} cm^2, corresponding to a disk of 0.23 Å diameter. Applying the considerations in chapter 8, we can say at once that a single hydrogen atom can become visible, *i.e.*, can produce a contrast of at least 10 per cent in an electron microscope of the transmission type only if the resolution limit is $\frac{0.23}{0.3} = 0.77$ Å or better. This would mean improving the microscope about twenty times beyond the limit of the uncorrected type. At 60 kev, for instance, a corrected objective would have to be constructed with an aperture of about 0.1 radian or 5.7°. At 10 kev it would have to be about 14°. But even if this could be achieved, a hydrogen atom would still remain invisible. This was first pointed out by L. I. Schiff,[72] though by an argument which requires some modifications. Schiff assumed a perfect lens, corrected for spherical aberration, though not necessarily for chromatic aberration, as he takes only elastically scattered electrons into account. In such an objective, contrast could be produced only by those electrons which fail to pass the physical aperture. But in order to produce the required definition, the aperture at 10 kev would have to be opened up to about 14°, and it can be seen from figure 46 that under such conditions practically no contrast would be produced, as only a negligible fraction of the electrons is deflected by more than 5°. This could be remedied by oblique illumination, but the difficulty arises that the effective aperture of the lens is only the part actually filled by scattered electrons, and this is not sufficient for the required resolution.

At this point the argument requires modification. It is hard to see how the fact that the aperture is not filled by electrons could produce a diffraction error, as diffraction in the aperture arises from cutting out a part of the wave. Yet the result is correct, as can be seen from the following reasoning.

It has been explained in chapter 6 that in the transmission type of electron microscope, small objects are not detected by scattered electrons, but by the electrons which are missing from the original beam. Inelastically scattered electrons are distributed by the chromatic aberration over an area considerably larger than the resolution disk, and can be considered as absorbed. Elastically scattered electrons on the other hand will contribute to the contrast in a perfect microscope only if they are absorbed by a physical diaphragm. Without going into the details of the very complicated problem which involves diffraction, both at the object and at the aperture, we can obtain an approximate estimate by a direct application of the Principle of Indeterminacy.

If an electron is scattered by a hydrogen atom, we know with a probability amounting almost to certainty that it will be deflected by an angle θ_m less than about $\frac{7}{\sqrt{V}}$ radian. For 10,000 ev electrons this is about 4° and the uncertainty $2\theta_m$ is 8°. The corresponding uncertainty in transversal momentum is $2mv\theta_m$, and with this is associated, by Heisenberg's principle an uncertainty d_H in the position from which the electron appears to originate. This is given by

$$d_H . 2mv\theta_m = h \tag{55}$$

With the de Broglie wavelength

$$\lambda = \frac{h}{mv}$$

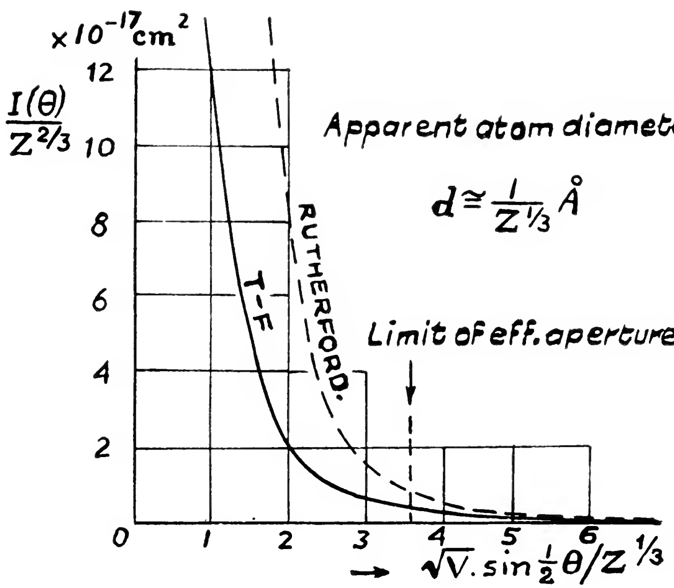
this can be written

$$d_H = \frac{\lambda}{2\theta_m}$$

which is the same as Abbé's relation, with the difference that θ_m is not the aperture of the objective, but the half-angle of diffraction *at the object.* This explains the difficulty which we have met in the case of beams not filling the aperture. The *diffraction error* is no error at all, but a measure of the size of the object. This becomes even clearer if we substitute the values of λ and θ_m

$$d_H = \frac{\lambda}{2\theta_m} = \frac{\frac{12.2}{\sqrt{V}}}{\frac{14}{\sqrt{V}}} = 0.87 \text{ Å} \tag{56}$$

This is the apparent diameter of the hydrogen atom. It is a little less than the *Bohr diameter* of the older quantum theory, the diameter of the nearest orbit on which the electron was supposed to circle, which is 1.064 Å. In an electron micrograph the *missing electrons* would produce a deficit inside a disk of approximately this diameter. As the missing electrons are mostly inelastically scattered electrons which constitute also more than 90 per cent of the total scattering, the contrast will be about $\left(\frac{0.25}{0.87}\right)^2 = 0.08$ which falls just a little short of 10 per cent. We can therefore say that in an electron microscope with lenses perfect as regards spherical aberration but uncorrected for chromatic aberration, a hydrogen atom would be just below the limit of visibility. We see also that the reason for this is not so much the smallness of the hydrogen atom, but rather that it is too large for the electric charges which it contains, in other words, its *density* is too low.



ELASTIC SCATTERING
BY THOMAS-FERMI ATOM.

FIG. 47

Atoms with an atomic number larger than $Z = 2$ ought to be above the visibility level. This estimate is more optimistic than Schiff's, who puts the limit to $Z = 7$, because he takes only the elastic scattering into account.

What is the apparent size of heavier atoms? The discussion is much simplified if we consider only the elastic part of the scattering, and replace the atoms by a convenient model, such as the Thomas-Fermi atom.* (Mott-Massey,[39] p. 126.) This is

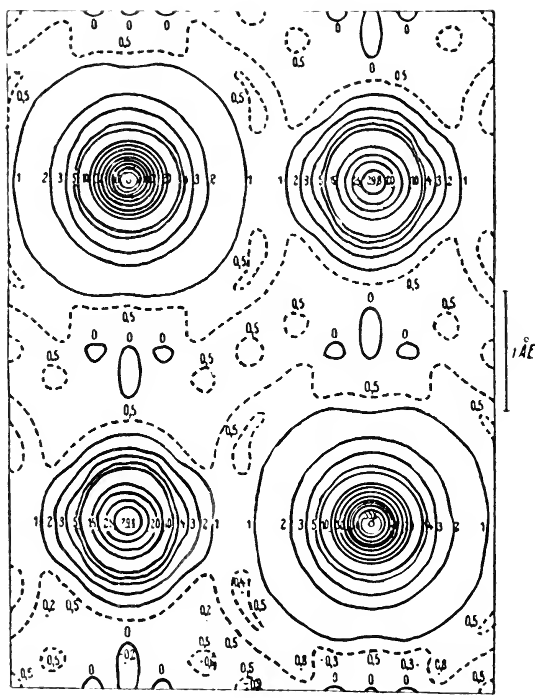


FIG. 48. Electron density diagram of NaCl crystal, obtained by W. L. Bragg's method of Fourier analysis from a great number of diffraction photographs. (H. G. Grimm, R. Brill, C. Hermann and Cl. Peters, *Naturwiss*, **26**, 479, 1938)

* This part was written and figure 47 drawn before I was aware that Schiff[72] had discussed the problem in almost exactly the same way.

a fair approximation to the elastic scattering of all atoms of the periodic system, particularly the heavier ones. The results are summed up in figure 47. It can be seen that practically all scattered electrons are confined inside a cone for which the parameter

$$\sqrt{V} \, . \, \sin \tfrac{1}{2} \frac{\theta}{Z^{\frac{1}{3}}}$$

assumes a value roughly between 3 and 4. If we fix the limit, somewhat arbitrarily, at 3.7, and repeat the same calculation as in the case of the hydrogen atom, we obtain an apparent atom diameter $d_z = \frac{0.82}{Z^{\frac{1}{3}}}$. As the estimate is rather rough it is preferable to say that even in the most perfect electron microscope an atom will appear as a rather blurred disk with a diameter

$$d_z \approx Z^{-\frac{1}{3}} \text{ Å} \qquad (57)$$

Thus heavier atoms appear smaller than lighter ones. The heaviest, uranium, will be about 0.22 Å. As the scattering cross section increases with Z and the apparent area decreases with $Z^{\frac{2}{3}}$, the contrast will increase with $Z^{\frac{5}{3}}$, *i.e.*, it will be about nineteen hundred times better for uranium than for hydrogen. Thus it ought to be rather easy to identify atoms of different kinds.

It is evident from this discussion that, although it may be possible to resolve atomic lattices, the detail visible in them would be much inferior to the wonderful electron density diagrams obtained by W. L. Bragg's indirect methods of structure analysis [73] of which figure 48 gives an example. Without going into details, it can be said that these diagrams have been obtained by the interference of diffracted beams from millions of unit cells.* The reason for this inferiority is not the electron micro-

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* G. P. Thomson and W. Cochrane suggest the useful word *interfraction* for phenomena of this sort, as opposed to *diffraction* which ought to be reserved for what goes under this name in optical treatises.

scope, but the fundamental plan on which nature seems to be built. It is perhaps more correct to say that *individual* atoms *have* blurred features than to assume that they are blurred by taking photographs of them. Sharp features come out only when they are investigated in millions. This is just the opposite of what we would expect if mass-photographs were taken of humans, or even of sheep, and it conveys an idea how ill-suited our everyday concepts are for the understanding of the fundamentals of atomic physics.

Very recently Louis de Broglie * has advanced an interesting argument which, it appears, destroys all hope of ever seeing an atomic lattice with an electron or proton microscope. He starts from the consideration that in order to see a particle it is not sufficient to probe it with one beam corpuscle, but with several. Will the next corpuscle find the object in the same place? De Broglie's result is that in order to locate an atom with sufficient accuracy it is necessary to knock it sideways, thus imparting to it a motion which will necessarily blur the picture. But what is worse, in the case of atoms the energy imparted by the first collision will be sufficient to knock it out of the lattice. Thus it appears that while the lattice as a whole is a solid piece of physical reality, the single atom in the lattice belongs into metaphysics rather than into physics!

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* Louis de Broglie, *Comptes rendus,* **222**, 1017 and **223**, 490–493, 1946.

CHAPTER 14

CHEMICAL AND STRUCTURAL ANALYSIS BY THE ELECTRON MICROSCOPE

ELECTRON microscopy possesses no equivalent of color discrimination. This puts it at a certain disadvantage against light microscopy in which substances can be often identified by their characteristic color or dyed with selective dyes. Different substances under the electron microscope are distinguished only by their density, or more exactly by their electronic density which is almost the same, except if they have characteristic shapes, as in the case of crystals. This natural disadvantage has been more than compensated by several recent improvements which promise to make the electron microscope the most powerful tool for micro-analysis.

In 1942, Hillier, Baker and Zworykin [73] equipped the Type B electron microscope of the R.C.A. with a diffraction adapter which has become a standard component of the new universal E.M.U. model. The adapter is a unit of the same size and interchangeable with the projection lens. It contains a specimen chamber in to which the object can be introduced through an air-lock. Diffraction diagrams can be taken within 2–3 minutes of taking a micrograph of the same object. The ray diagram is shown in figure 49. The objective lens forms a reduced size image of the cross-over which acts as source for the diffraction camera. The previous projection lens has been divided to contain two coils, the upper of which acts as a projection lens when electron micrographs are taken, but is de-energized for diffraction photographs. Instead, the lower coil is used as a lens for focusing the diffraction images on the photographic plate. This arrangement allows considerably higher intensities and shorter exposures than the conventional type of diffraction camera, with parallel beams, collimated by pinholes.

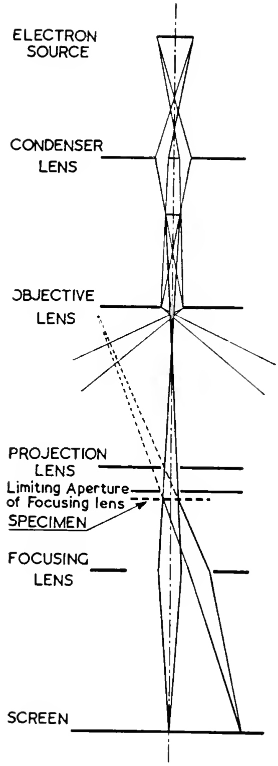

FIG. 49. Electron optical arrangement of R.C.A. diffraction adapter

Figure 50 shows an example of a diffraction pattern and micrograph obtained within a few minutes of one another. In many cases, especially if one has to deal with mixtures of substances, it would be desirable not only to shorten this time, but also to have means for identifying the particle which produced the diffraction pattern. Von Ardenne and collaborators [74] have constructed a universal electron microscope in which a very fine electron probe can be directed at different parts of the specimen and diffraction patterns can be taken without removing it from the position in which it was micrographed. Under favorable circumstances such diffraction patterns can be obtained with any conventional electron microscope with sufficiently wide physical aperture. Hillier and Baker [75] noticed sharp black lines in electron micrographs of minute crystals which on closer investigation were shown to be images of crystallographic planes, very nearly parallel to the optic axis. They appeared blank because they reflected the beam outside the paraxial zone of the objective. The trace of the diffracted beams could also be found at a certain distance from the crystal because of spherical aberration, as explained in figure 51. It is an obvious conclusion that such evidence of the structure of the particles investigated could be found in most electron micrographs, if the contrast were not insufficient. It may be suggested that the contrast could be greatly improved by cutting out the central beam by means of a small shield or a thin wire, as indicated in figure 51. Electron micrographs and diffraction patterns could be taken in immediate succession, the first without a shield, the second with one, and with longer exposures.

A second method for the identification of substances is based on the characteristic line spectrum of energy losses, discovered by Ruthemann.[41] J. Hillier [76] has described a special electron microscope in which this effect is utilized. By a two-stage magnetic

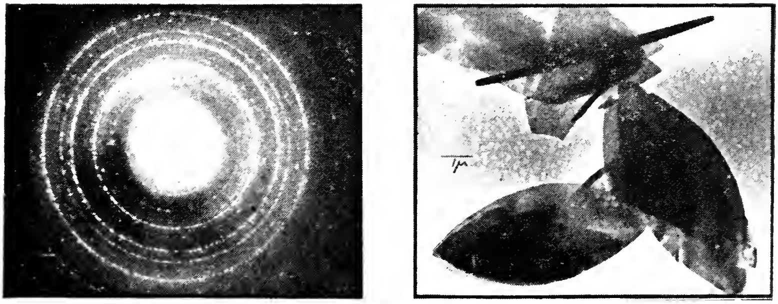

Fig. 50. Diffraction pattern and electron micrograph of the same sample of monohydrated aluminum oxide, taken with R.C.A. diffraction adapter.

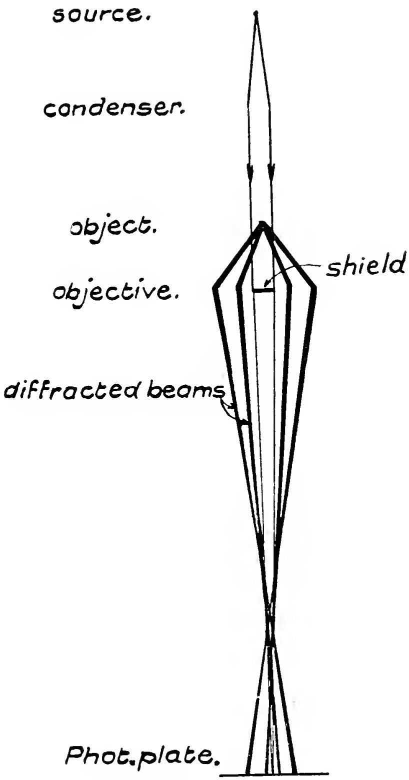

Fig. 51

lens arrangement a very strongly reduced image of the electron source or the cross-over is focused on a small part of the specimen. The transmitted electrons, some of which have suffered energy losses, are refocused on the slit of an electron velocity analyzer. This is of the *half-circle type* introduced by Ramsauer. A homogeneous magnetic field at right angles to the beam bends the electron trajectories to circles, and it is well known that the slit will be approximately focused on a photographic plate which is arranged at 180° from the slit, in radial direction. If operated with the extremely constant voltages and currents produced by feedback stabilizers, such electron spectographs can have resolutions $\frac{\Delta V}{V}$ of the order 10^{-4} or better. In Hillier's instrument a micrograph can be taken of the same object by switching off the magnetic field which deflects the beam and focuses it on the analyzer. The very small image of the electron source is withdrawn for a short distance from the specimen, and projects, without lenses, a magnified shadow of the object on a photographic plate. Such *shadow microscopes,* first described by H. Boersch,[77] can have resolutions of the order of 200 Å or somewhat better.*

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* The micro-analyzer has been fully described by J. Hillier and R. F. Baker, *J. Applied Phys.* **15**, 663 (1944).

APPENDIX

DIFFRACTION THEORY OF THE ELECTRON MICROSCOPE

THE diffraction theory of microscopic vision deals with limitations inherent in the wave nature of light or electrons, and assumes an optical system perfect in the geometrical sense. While in light optics this assumption is fully justified, it is not even approximately true in electron optical systems; therefore, we must never forget that we are dealing with a high degree of idealization.

Objects can be self-luminous, or illuminated by an external source. In self-luminous objects every point is supposed to emit radiation independently of all others, and, as the phases of the light emitted by them are distributed at random, the resulting intensities in the image will be the same as if only one point emitted light at a time, and the elementary intensity components were summed up. Therefore, the theory of the microscope for self-luminous objects is essentially contained in the Airy figure, which is the intensity distribution corresponding to a single luminous point, shown for reference in figure 52. It is produced by diffraction of a spherical wave at the edge of the objective aperture.

Non-self-luminous objects, or as it is more correct to say in the case of transmission type microscopes, the interstices between the objects, receive light from the same elementary sources, therefore, the waves issuing from these interstices are *coherent, i.e.,* capable of interference. The importance of this fact was first noticed by Ernst Abbé, in 1870. Abbé published the resolution limit which bears his name, the elementary considerations which led to it, and some supporting experiments, in 1873, but he never published the mathematical details of his theory. This was done only 10 years after his death, in 1910, by his pupil, O. Lummer, and by F. Reiche [78] who tried to establish it on the most solid foundation known at that time.*

Abbé considered the simplest object, a grid or grating with equal distribution of black and white, and the simplest method of illumination: parallel light, *i.e.*, a plane wave. This will give rise to the well-known Fraunhofer diffraction phenomenon: At a distance large in comparison with the grid constant, the transmitted wave will consist approximately of a number of plane waves of which one is in the original axial direction and the one nearest it will be deflected by an angle θ, given by

$$\sin\theta = \frac{\lambda}{d} \tag{58}$$

if d is the grid constant, as illustrated in figure 53. This is the wave diffracted *in the first order.* Abbé showed by theory and by experiment that if the aperture of the objective is so small as to admit only the axial, *zero order* wave, the image will show no trace of the grid structure of the object. Likeness to the original begins to emerge only if the first order diffracted beam is added to the zero order beam. If the first order is admitted, but the zero order cut out, a spurious structure appears, the image has twice as many bars as the original. This is illustrated in figures 54 *(a–c)* which relate to the case of the grid with ten bars. Interesting photographs, showing the apparent distortion of diatomae by microscope objective apertures of various shapes have been taken by Beck.[79]

Whereas a microscope with axial illumination cannot resolve a grid with a period finer than $\frac{\lambda}{\sin\alpha}$, the resolution can be improved if the illuminating beam forms an angle α with the axis, so that, in addition to the undeflected beam, *one* of the first

* Rather incomprehensibly, Lummer and Reiche omitted any reference to the violent controversy on Abbé's theory, though they list in the bibliography without comments several books and papers which contradict their contentions.

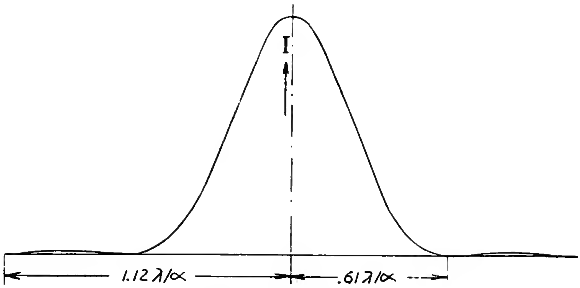

Fig. 52. Airy figure

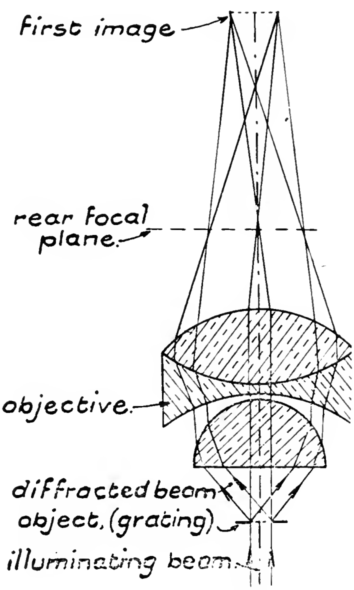

Fig. 53. Abbé's theory

order diffracted beams can enter the objective. This gives for the finest detail resolvable with a given objective

$$d_A = \frac{\lambda}{2 \sin \alpha} \tag{59}$$

which is usually called the Abbé limit.

By proving the advantage of oblique illumination, Abbé's theory proved to be of great heuristic value, as it led to the wide angle condenser which was one of the most important improvements in the history of the microscope. But in pointing toward wide angle illumination, the theory pointed beyond itself. It was soon found experimentally that it is better to fill the cone of the objective aperture entirely or almost entirely * with light, than to use hollow cones with nearly marginal rays, and here the simple assumptions of the original Abbé theory were obviously not fulfilled. It was suspected that Abbé had over-emphasized the importance of coherence, and this view received strong support, in 1896, when Lord Rayleigh showed that if the light source is sharply focused on the object, it will become almost exactly equivalent to a self-luminous object, as different

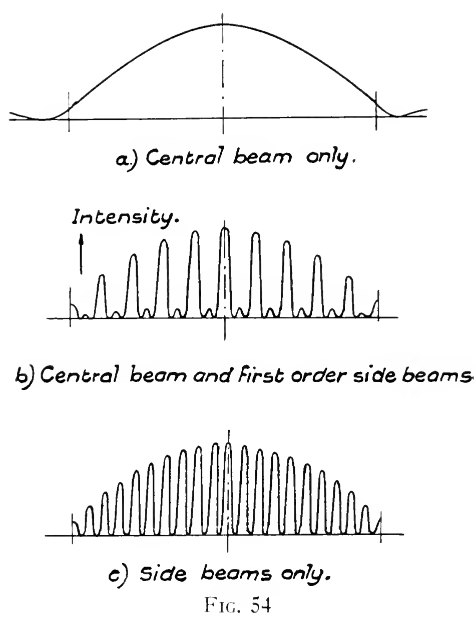

Fig. 54

* In practice it is best to fill the cone about two thirds with light, as this gives better contrast than the full cone.

points of the object are now illuminated with waves incoherent between themselves; and yet this makes very little difference to the microscopic image. This is the so-called *critical illumination.* The violent controversy which followed between supporters of the *equivalence theory* and Abbé's theory * was decided, in 1926, by M. Berek who showed that not only in the case of critical illumination, but also in most other methods of illumination used in practice, it is justified to consider the object, or rather its interstices, as self-luminous, and that Abbé's original theory gives accurate results only if the illuminating beam is narrow in comparison with the objective aperture. L. C. Martin [81] showed, in 1931, that the two theories are not irreconcilable, and that Abbé's method can be applied also to wide angles of illumination, by analyzing the illuminating light into plane, coherent waves. But as this *generalized Abbé theory* has not yet been fully worked out, *Abbé's theory* will be used here in the restricted, original meaning of the word.

In electron microscopy the dividing line between the *equivalence theory* and the *Abbé theory* is somewhere near a half-illuminating angle of 5.10^{-4}. If the cathode is imaged on the object the angle will be about ten times wider, as has been shown in chapter 5, and the instrument operates entirely in the region of the *equivalence theory*. But the modern tendency in electron microscopy is to restrict the half-illuminating angle to 5.10^{-4} or even less. Though this should not affect the resolving power, the best resolutions have been found in fact with very restricted illuminating angles, presumably because the contrast was much better. The reason for this is not quite clear, but it appears very likely that incipient Fresnel fringes are a great help in emphasizing, and even overemphasizing small contrasts. Another possible explanation is that phase contrast plays a certain part, and this phenomenon is strongest in almost parallel illumination.

Whereas these simple outlines of the theory are sufficient for the understanding of most fundamental phenomena in present-

* For a concise history of this controversy, *cf.* Sir Herbert Jackson and H. Moore.[80]

day electron microscopes, and can be transferred without much alteration from light optics, it would be desirable to possess a more complete diffraction theory which would take full account of the peculiarities of electrons and may be a help in the interpretation of doubtful structures, near the limit of resolution. No attempt will be made here to develop such a theory, but it may be useful to call attention to some features of the problem, and to explain some statements made in previous chapters.

In some ways electron wave optics is simpler and more complete than light wave theory. As polarization effects, if they can be detected at all, are practically negligible, free electrons can be described by *scalar* waves, similar to sound waves. Optical diffraction problems are also treated most often by substituting scalar fields for the electromagnetic waves, but whereas in light optics this substitution remains an often doubtful approximation, it is entirely justified in electron optics. Another feature of optical diffraction theory which is open to objections is the assumption of black, or at least partially absorbing *surfaces,* to which reference has been made at the end of chapter 7. This difficulty is very fully discussed in a monograph by B. B. Baker and E. T. Copson.[82] The orthodox optical method is to treat diffraction as a field problem with boundary conditions. But it is easy to see that a boundary in the mathematical sense, *i.e.*, a surface, can be transmitting or reflecting or both, but never absorbing. For absorption a certain *depth* is needed, for full absorption of the order of half a wavelength. The theory of electron diffraction, however, is built up from the beginning as diffraction by spatially extended fields. In principle we could therefore expect more accurate solutions of the problems of the electron microscope as in the case of the optical microscope, if the problem were not complicated by the very large geometrical errors.

It has been shown in chapter 6 and other places, how the very magnitude of these errors can be used to construct a simple theory, by emphasizing the fact that in the case of uncorrected objectives, contrast is mostly produced by the *"missing electrons."* It will be now attempted to give a wave-theoretical interpretation of this geometrical concept.

Let us consider a small object under the microscope illuminated by a parallel beam of electrons with energy E, which may be represented, as usual, by a plane wave in the z-direction

$$\psi_i e^{\frac{2\pi jht}{E}} = I_0 e^{2\pi j\left(\frac{ht}{E} - \frac{z}{\lambda}\right)}$$

where ψ_i is Schrödinger's complex amplitude, $j = \sqrt{-1}$ the imaginary unit, and λ the de Broglie wavelength. The suffix i means *incident* or *illuminating*. When the electron beam is scattered at the object, two sorts of waves will originate from it, both falling off at large distances like $\frac{1}{r}$. One shall be represented by the amplitude ψ_c, and called the *coherent wave*. It corresponds to the same value of energy, E, as the incident beam. The other shall have the amplitude ψ_s, and the energy constant E′. This corresponds to electrons scattered with losses appreciable enough to make them incapable of interfering with the incident wave. The criterion of *incoherence* will be discussed later. The resulting wave at some point, *e.g.*, at the photographic plate, will be represented by an expression

$$(\psi_i + \psi_c)e^{\frac{2\pi jht}{E}} + \psi_s e^{\frac{2\pi jht}{E'}}$$

The resulting electron density is obtained by taking the square of the absolute value and averaging it over a long time interval. The electron current or intensity is given by a formula of similar build. As we want to indicate the essentials only, we replace this by squaring, as if the ψ_s were real, and obtain the following scheme for the resulting intensity or photographic density:

$$(\psi_i + \psi_c)^2 + \psi_s^2 = \underset{\text{(Background)}}{\psi_i^2} + \underset{\text{(Missing electrons)}}{(2\psi_i\psi_c + \psi_c^2)} + \underset{\text{(Incoherent scattered electrons)}}{\psi_s^2} \quad (60)$$

Filling in this scheme promises to be rather laborious. The large material on atomic collisions worked out and collected especially by Mott and Massey [39] is not quite sufficient for this purpose. In collision experiments, and even in the greater part of electron diffraction or rather *interfraction* experiments, only the scattered electrons matter. *Kikuchi lines* are an exception.

Therefore, the second term is either not calculated at all, or not given, and it is just this term which is of the greatest importance in microscopy.

It has been stated before and will be proved now that for the purpose of microscopic image formation not only the inelastically but also the elastically scattered electrons can be considered as *incoherent*. It is possible to decide this without going into the somewhat obscure question of *coherence length* or *length of wave trains* of electrons, though obviously the answer depends on this to some extent. If the wave trains were of infinite length, *i.e.*, if the energy constant, E, of the electrons were defined without uncertainty, *any* difference between E and E′ would mean incoherence. But if the wave trains are extremely short, *e.g.*, consisting only of a few waves, incoherence will arise only if we can expect phase shifts between electrons in the beam of the order 2π. This is therefore the *minimum* condition for incoherence, and will be applied in the following.

If an electron of mass m collides with an atom of mass M and suffers a deflection by an angle θ, its energy loss will be approximately

$$\frac{\Delta E}{E} = \theta^2 \frac{m}{M} \qquad (61)$$

and its relative change of wavelength will be half as much,

$$\frac{\Delta\lambda}{\lambda} = \tfrac{1}{2}\theta^2 \frac{m}{M} \qquad (62)$$

A phase shift of π will be produced on the length L between object and photographic plate between the original beam and a deflected electron if

$$\frac{\Delta\lambda L}{\lambda^2} = \tfrac{1}{2} \qquad (63)$$

Combining this with (62) we obtain an upper limit for the divergence θ inside which the phase shift, and therefore, by the minimum criterion, the incoherence is negligible. Let the atom for example be a carbon atom, $\frac{m}{M} = \frac{1}{1,840} \times 12 = 4.5 \times 10^{-5}$, $\lambda = 0.05$ Å $= 5.10^{-10}$ cm, and $L = 100$ cm. This gives 3.3×10^{-4} radian for the coherent bundle. As this is about ten times smaller than the aperture or effective aperture of electron microscopes, we can consider the coherence of the elastically scattered electrons as negligible.

It appears that metallic objects are an exception to this rule. According to M. von Laue, electrons which are elastically scattered by metals can be considered as if they had collided with the whole metal crystal, therefore, in this case the scattering can be treated as coherent.

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