From a fluorescent patch to picoscopy, one strand in the history of the electron

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The particle nature of the electron was established in 1897 and the complementary wave nature in 1923. Four years later, it was shown that the behaviour of the electron in a rotationally symmetric electrostatic or magnetic field bears a close analogy to that of light in a lens, a finding that led to the notion of an electron lens and, soon after, to the first electron microscopes. The development of the three main families of microscopes and the struggle to overcome the optical aberrations are described; with the arrival of aberration correctors in the late 1990s, electron microscopes capable of furnishing information in the 50–100 pm range are now in current use.

1. The first decades. The electron and its optics

The electron has a hazy prehistory[1] but the first recorded sighting of the particles that today we call electrons was made in Bonn in 1858 by Julius Plücker (1801–1868),[2] a mathematician and natural philosopher who had settled there after halting at many other German universities and become interested in electrical discharge phenomena, as had some of his contemporaries, notably Faraday. On applying a voltage across the electrodes inside a partially evacuated glass envelope, he observed a fluorescent patch on the wall, apparently generated by some kind of emanation from the cathode; a magnetic field altered the position and shape of the patch. His observation aroused considerable speculation and many more experiments were performed in order to establish the nature of these cathode rays but it was nearly 40 years before the correct answer was obtained. Why was Plücker the first to see this fluorescent patch? By a happy coincidence, Johann Heinrich Geissler, a remarkably skilful glassblower, also lived in Bonn and was able to furnish Plücker with exceptionally highly evacuated tubes, not available to rival

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[1] The story is told in great detail, very readably, in *Flash of the Cathode Rays* by P.F. Dahl (Institute of Physics Publishing, London & Philadelphia 1997).

[2] J. Plücker, Ueber die Einwirkung des Magneten auf die elektrischen Entladungen in verdünnten Gasen. *Ann. Physik Chemie* **103** (1858) 88–106 and (Nachtrag) 151–157; Fortgesetzte Beobachtungen über die elektrische Entladung durch gasverdünnte Räume. *Ibid.* **104**, 113–128, **105**, 67–84 and **107** (1859) 77–113; Ueber einen neuen Gesichtspunkt, die Einwirkung des Magneten auf den elektrischen Strom betreffend. *Ibid.* **104** (1858) 622–630.

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students of discharge effects. Geissler was awarded an honorary doctorate in 1868, on the 50th anniversary of the University of Bonn.

Many investigations into the nature of these rays were made in Germany and in England, and to a lesser extent elsewhere. The German school inclined to the idea that they were some new kind of wave phenomenon in the æther while the British thought they were more likely to consist of particles. It was known that they were deflected by magnetic fields but an unfortunate experiment of Hertz failed to reveal that they were also deflected by a transverse electric field; this misleading result was no doubt caused by the relatively poor vacuum in Hertz's tube and strongly influenced thinking in Germany. It is widely (but not universally[3]) accepted that the dilemma was resolved by J.J. Thomson (1856–1940), Cavendish professor in Cambridge, who showed in 1897 that the rays consisted of charged particles, either very highly charged (not very likely) or much lighter than any known particle.[4] Two years later, he was able to demonstrate that the second alternative was correct: "...we have clear proof that the ions have a much smaller mass than ordinary atoms; so that in the convection of negative electricity at low pressures, we have something smaller even than the atom, something which involves the splitting up of the atom, inasmuch as we have taken from it a part, though only a small one, of its mass.... From what we have seen, this negative ion must be a quantity of fundamental importance in any theory of electrical action; indeed, it seems not improbable that it is the fundamental quantity in terms of which all electrical processes can be expressed."[5] Thus, cathode rays consisted of some new, very light, singly-charged particle, which Thomson persistently referred to as a "corpuscle", long after the rest of the world had adopted the word "electron". We owe this name to George Johnstone Stoney, a versatile Irish scientist who enjoyed coining new words. (How long does it take light to travel 0.1 mm? Answer, one jot.) In 1881, Stoney had suggested that the relatively new metric units, the metre, the gramme and the second should be replaced by units based on the velocity of light, *c*, the gravitational constant, *G*, and the elementary charge, *e*, thus anticipating the current deliberations at the Bureau International des Poids et Mesures (BIPM) by some 130 years. These would form "a series of systematic units that in an eminent sense are the units of Nature, and stand in an intimate relation with the work that goes on in Her mighty laboratory"—can we hope that the conclusions of the BIPM will be expressed in such splendid language? Ten years later he wrote "...in electrolysis, a definite quantity of electricity, the same in all cases, passes for each chemical bond that is ruptured...the amount of this very remarkable quantity of electricity is about the twentiethet (that is, $1/10^{20}$) of the usual electromagnetic unit of electricity...This is the same as three-eleventhets ($3/10^{11}$) of the much smaller C.G.S electrostatic unit of quantity. A charge of this amount is associated in the chemical atom with each bond...These charges, which it will be convenient to call *electrons*, cannot be removed from the atom; but they become disguised when atoms chemically unite." The word caught on quickly. Larmor used it in the *Philosophical Transactions* in 1894 and, in his introduction to Thomson's first account of his findings in *The Electrician*, G.F. Fitzgerald repeatedly referred to

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3 French historians insist on the contributions of Perrin, and the claims of Emil Wiechert (1861–1928) are also vigorously defended.

4 J.J. Thomson, Cathode rays. *The Electrician* **39** (1897) 104–109; Cathode rays. *Phil. Mag*. **44** (1897) 293–316.

5 J.J. Thomson, On the masses of the ions in gases at low pressures. *Phil. Mag*. **48** (1899) 547–567.

electrons—but then, Stoney was Fitzgerald's uncle. The usual reason given for J.J. Thomson's preference for "corpuscle" and avoidance of "electron" (even in his Nobel lecture) is that Stoney used the word electron as a quantity of charge, like coulomb, and not for a particle carrying such a charge but I am tempted to believe that Thomson could not bring himself to follow the lead of a scientist who had published articles "On the experiment of Mahomet's coffin", "On the energy expended in propelling a bicycle" (an Xtraordinary) and on "A dimerous form of pansy", even though he was President of the Irish Academy of Sciences. By 1913, the word was so familiar that the *Empire Review* could speak of "The imponderable electrons of sentiment and feeling which allow our far-away peoples and clans to cohere."

By a happy coincidence, 1897 saw not only the clarification of the character of the electron but the invention of the cathode-ray tube (or Braunsche Röhre) by Ferdinand Braun,[6] who exploited the deflexion of the rays by a magnetic field even before knowing what the rays were made of. It rapidly came into use (e.g., Wehnelt and Donath[7]).

The next few years generated a voluminous literature on the behaviour of electrons in magnetic and electrostatic fields, in particular in an attempt to understand the properties of cathode-ray oscillographs and to improve the quality of the spot in such devices. At first, however, progress was made in a very different domain, the study of the Northern Lights or aurora borealis by Störmer, Birkeland and Villard; Störmer came close to discovering geometrical electron optics, as he pointed out ruefully in 1933: "...so habe ich in meiner Arbeit zur Theorie des Polarlichtes auch den Fall eines axialsymmetrischen elektromagnetischen Feldes behandelt";[8] and in fact, the geometrical optics of charged particles was not discovered until 1927, when Hans Busch observed that "Eine kurze Spule hat also die Eigenschaft, die Kathodenstrahlen nach der Achse zu um einen Winkel γ abzulenken, der proportional der Achsenentfernung...des Strahles ist".[9] Electron ballistics had given way to electron optics. This was four years after Louis de Broglie had argued that a frequency, and hence a wavelength, should be associated with electrons in motion, a suggestion that was confirmed by the observation of electron diffraction patterns by several scientists, among whom was G.P. Thomson, JJ's son. It is a nice irony of history that J.J. Thomson should have received the Nobel Prize for showing that electrons are particles while his son received it for showing that they are waves! Earlier attempts to explain spot formation in oscillographs had been based on electron ballistic calculations but in 1927, Busch showed that, in a first order approximation, the effect of a rotationally symmetric electrostatic or magnetic field on a beam of electrons can be described in the same language as the focusing of light by a glass lens and characterized in terms of a few simple parameters: focal lengths, the positions of focal and principal planes. The notion of an electron lens followed soon after and indeed, one magnetic lens was built even before the term was introduced: in 1927/8, Dennis Gabor enclosed a coil in a magnetic yoke but did not then

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6 F. Braun, Ueber ein Verfahren zur Demonstration und zum Studium des zeitlichen Verlaufes variabler Ströme. *Ann. Physik Chemie* (Wied.) **60** (1897) 552–559.

7 A. Wehnelt and B. Donath, Photographische Darstellung von Strom- und Spannungscurven mittels der Braun'schen Röhre. *Ann. Physik* **305** (1899) 861–870.

8 C. Störmer, Ueber die Bahnen von Elektronen im axialsymmetrischen elektrischen und magnetischen Felde. *Ann. Physik* **408** (1933) 685–696.

9 H. Busch, Ueber die Wirkungsweise der Konzentrierungsspule bei der Braunschen Röhre. *Arch. Elektrotech.* **18** (1927) 583–594.

realise that it possessed the properties of a lens and was not a mere concentrating coil. At the same date, Louis de Broglie suggested to one of his research students that electron optics would be worth exploring but there were (ostensibly) more exciting subjects in the air and the idea was not followed up.

2. Microscopes and aberrations

In 1928, the young Ernst Ruska (1906–1988) was working in the Electrotechnical Institute of the Technische Universität in Berlin, where Max Knoll led a team working on the technical development of the cathode-ray oscillograph. This led him to study the electron optical properties of magnetic and electrostatic lenses and in 1931, two lenses were connected in tandem to form the first primitive electron microscope. By 1933, in an improved instrument, a resolution comparable to that of a light microscope had been reached and was surpassed soon after. Funding was so elusive in the 1930s that Ruska was obliged to leave the university and joined Fernseh AG but, together with Bodo von Borries, a former fellow student and by now his brother-in-law, he continued to seek support for further development of the electron microscope, warmly encouraged by his brother Helmut, a doctor who saw the promise of such an instrument for medicine. In 1936, Carl Zeiss and Siemens both expressed interest and, in view of Siemens' experience and expertise in the area of electrical engineering, Ruska and von Borries joined forces with that company. "In a disused building in Berlin–Spandau that had previously served as a wholesale bakery and before that as a small-arms factory, we set up in the Spring of 1937 the Laboratory for Ultramicroscopy of Siemens and Halske AG. In this laboratory we were able, together with H.O. Müller and a few, mostly very young, co-workers to carry the development of a commercial electron microscope to the point where at the beginning of 1938 the first of two similar instruments was put into operation". By the end of the decade, serial production had been launched; in all, 38 instruments were delivered, their fate is chronicled in a fascinating historical paper by Carl-Heinrich Wolpers.[10]

This was not the only work on the electron microscope in that first decade. Electrostatic lenses were studied in the AEG research laboratory, also in Berlin, and it is curious to note that this appears to have impressed some external observers more than the work of Ruska at the Technical University and Siemens. In 1939, an excellent and well-documented book on electron optics by L.M. Myers appeared,[11] showing great familiarity with the literature of the subject. "Without doubt," writes Myers in his Preface, "the greatest proportion of experimental and theoretical work during the last few years has been carried out at the AEG Forschungsinstitut in Berlin, under the supervision of Dr Brüche, consequently it is understandable that this introduction to the subject should follow closely the writings of this investigator and his numerous collaborators"; Knoll and Ruska's work is nevertheless well covered in the bibliography. A third attempt to develop the electron microscope was made by Baron Manfred von Ardenne,[12] who built the first scanning electron microscope (SEM) in his private laboratory. This was, strictly speaking, a scanning transmission electron microscope

[10] C. Wolpers, Electron microscopy in Berlin 1928–1945. *Adv. Electron. Electron Phys*. **81** (1991) 211–229.
[11] L.M. Myers, *Electron Optics, Theoretical and Practical* (Chapman & Hall, London 1939).
[12] M. von Ardenne, *Elektronen-Uebermikroskopie* (Springer, Berlin 1940).

(STEM), since the signal used to produce the image was collected beyond the specimen (we return to the difference between the SEM and the STEM later). Von Ardenne's laboratory was destroyed during World War II and he turned to other subjects. The early publications on the electron microscope stimulated interest outside Germany as well. In England, the Metropolitan–Vickers Company constructed a microscope for L.C. Martin, whose lecture on electron optics and microscopes, delivered before the Television Society in 1934, was printed in the first volume of the society's journal. This instrument was installed in Imperial College, London, and among those who helped to get it working was G.P. Thomson. Other prototype instruments were built by Ladislaus Marton[13] in Brussels, by P.A. Anderson and K. Fitzsimmons in Washington State University and by E.F. Burton, J. Hillier and A. Prebus in Canada.

The 1930s also witnessed vast developments in the theory of electron optics. Walter Glaser, then a professor in Prague, made major contributions to lens theory as did Otto Scherzer and his colleagues at the AEG Laboratory. Glaser worked on all aspects of geometrical and wave optics and his work is embodied in his magisterial *Grundlagen der Elektronenoptik* (Springer, Vienna 1952) and its condensed but more readable version in the *Handbuch der Physik* (33 (1956) 123–395).[14] Before the advent of computers, he was well-known for the Glasersche Glockenfeld, a bell-shaped magnetic field model that represents the axial magnetic field in a magnetic lens quite well and had the huge attraction that all lens properties, including aberration coefficients, could be written explicitly in terms of circular functions. Scherzer[15] is best known for his 1936 paper[16] in which he demonstrated that the formulae from which two of the most damaging aberrations, the spherical and chromatic ones, are calculated cannot change sign. Each aberration coefficient has the form of an integral and in both cases Scherzer showed that the integrand can be cast into a form that contains only squared terms. This was a cruel blow to the defenders of the new electron microscope, who had tacitly assumed that adroit lens design would be capable of rendering aberrations negligible as is the case for glass lenses. These aberrations limit the attainable resolution because they affect all points in the object plane equally, including those close to and on the optic axis (the axis of symmetry). Moreover, the smallest value of the spherical aberration coefficient is so high that whereas the resolution of a light microscope is comparable to the wavelength of the light used, that of an electron microscope is of the order of 100 wavelengths.

Glaser found Scherzer's result so unpalatable that he sought loopholes in the proof for the remainder of his life. The most memorable (and most preposterous if we accept that Scherzer's formula was correct) was published in 1940. The coefficient of spherical aberration, C_s, takes the forms

$$C_s = \int_{z_o}^{z_i} A h^4 \mathrm{d}z \tag{1}$$

[13] L. Marton, Electron microscopy of biological objects. *Nature* (Lond.) **133** (1934) 911.
[14] H. Grümm and P. Schiske, Reminiscences of Walter Glaser, *Adv. Imaging Electron Phys*. **96** (1996) 59–66.
[15] M. Marko and H. Rose, The contributions of Otto Scherzer (1909–1982) to the development of the electron microscope. *Microsc. Microanal*. **16** (2010) 366–374.
[16] O. Scherzer, Ueber einige Fehler von Elektronenlinsen. *Z. Physik* **101** (1936) 593–603.

in which $A = \frac{1}{48}\left(5\frac{\eta^2 B'^2}{\hat{\phi}} - \frac{\eta^2 BB''}{\hat{\phi}} + 4\frac{\eta^4 B^4}{\hat{\phi}^2}\right)$

or

$$C_s = \frac{1}{16}\int_{z_o}^{z_i}\left(\frac{\eta^4 B^4}{\hat{\phi}^2}h^4 + 2(hB' + h'B)^2\frac{\eta^2 h^2}{\hat{\phi}} + 2\frac{\eta^2 B^2}{\hat{\phi}}h^2 h'^2\right)\mathrm{d}z \qquad (2)$$

in which $B(z)$ denotes the magnetic field distribution on the optic axis of the lens; η is a constant and $h(z)$ is a particular solution of the paraxial ray equation, a linear, homogeneous second-order ordinary differential equation, which describes the paths of electrons in a first approximation. $\hat{\phi}$ is the relativistically correct accelerating voltage. Equation (2) shows that the coefficient cannot become negative but it was the first form that interested Glaser, who argued that, if a field distribution could be found for which A vanished, the spherical aberration coefficient would also vanish. Astonishingly, Glaser did find a solution, even though equation (2) shows that C_s can become zero only if $B(z) \equiv 0$. It soon emerged that Glaser's lens would be incapable of forming a real image of a real object, a necessary condition for the correctness of expression (2), thus resolving the apparent contradiction. A similar result was obtained for electrostatic lenses. As late as 1956, only a few years before his death, Glaser thought that he had found the much-desired loophole but alas, he detected an error in his calculation, which destroyed his hopes. Scherzer's formula continues to be challenged occasionally (e.g., Nomura[17]), always without success. Scherzer was also the author of another key paper[18] in electron optics, in which he described strategies for circumventing his 1936 result. The derivation of expression (2) makes a number of assumptions about the corresponding lens, namely, that it has rotational symmetry about the optic axis, is static, has no elements on the optic axis and acts as a lens, not a mirror. Scherzer examined the effect of relaxing one or other of these requirements and in each case, described a system capable of aberration-free imaging. All of these were investigated both theoretically and experimentally over the years but only two survive today: the use of optical devices that do not possess rotational symmetry about the axis and electron mirrors. Before going more deeply into these, however, we record some important stages in electron microscope development.

3. Proliferation

We have seen that commercial development of a transmission electron microscope began in Germany in the late 1930s. During the wartime years, companies in the USA and Japan began producing such instruments and several laboratory models formed the basis of later commercial developments. In France, for example, Gaston Dupouy in Toulouse and Pierre Grivet in Paris built microscopes with magnetic and electrostatic lenses respectively, which were later

[17] S. Nomura, Design of apochromatic TEM composed of usual round lenses. In: M. Luysberg, K. Tillmann and T. Weirich (eds), *Proc. 14th European Microscopy Congress, Aachen*, vol. 1, pp. 41–42 (Springer, Berlin 2008); P.W. Hawkes, Can the Nomura lens be free of spherical aberration? *J. Microscopy* **234** (2009) 325.

[18] O. Scherzer, Sphärische und chromatische Korrektur von Elektronenlinsen. *Optik* **2** (1947) 114–132.

marketed by Optique de Précision de Levallois (OPL) and the Compagnie Générale de Télégraphie sans Fils (CSF). In Holland, Jan Le Poole built a prototype instrument which was later taken up by Philips. An extremely thorough account of microscope development in Europe has been prepared by Agar[19] and for many highly readable and often moving personal accounts, see *The Beginnings of Electron Microscopy*.[20] These were all "conventional" transmission electron microscopes, in which a relatively broad beam of electrons falls on the specimen and generates an image on a fluorescent screen or recording medium. Another approach, in which a small probe is scanned across the specimen and a signal collected from each point sequentially, attracted interest in Germany in the 1930s and in the USA in the wartime years but it was only in the late 1940s and 1950s that the scanning electron microscope as we know it today was developed, by C.W. (later Sir Charles) Oatley and a series of research students in the Cambridge University Engineering Department. Unlike the microscope built by von Ardenne, this was a reflexion instrument: the small electron probe falls on a massive specimen and various signals are generated by the interaction of the incident electrons and the specimen. Detectors capture these signals and display the resulting images on monitors, scanned in synchronism. Among the possible signals are backscattered electrons, secondary electrons, X-rays and cathodoluminescence but this by no means exhausts the list. The first publication by Oatley's first PhD student, Dennis McMullan, appeared in 1953 and a succession of gradually improved models were constructed over the years, thanks to the efforts of K.C.A. Smith, Oliver Wells, P.R. Thornton, W.C. Nixon and many others.[21] These culminated in the first commercial scanning electron microscope, the Stereoscan, launched in 1965 by the Cambridge Instrument Company, originally founded by Darwin's son Horace and in the 1950s still on the original site in Cambridge. The success of this first model owed much to the skill and enthusiasm of A.D.G. Stewart, who left Oatley's laboratory to oversee the work at the Cambridge Instrument Co.

At the same period, Raymond Castaing[22] in Paris was busy converting a CSF microscope into the first X-ray microanalyser, an instrument in which information about the chemical nature of the specimen was obtained by analysing the electrons generated by a small electron probe. In Castaing's instrument, the specimen was moved under a stationary beam. Soon, using the experience gained in Cambridge on scanning techniques and the associated optics, a scanning X-ray microanalyser was assembled by Peter Duncumb in the Cavendish Laboratory and subsequently marketed by the Cambridge Instrument Co.

The resolution of the scanning electron microscope is of course limited by the size of the smallest probe that can be produced and here it is not just the spreading caused by aberrations that determines the lower limit. In order to generate a useful image in a reasonable time, the current in the probe must reach a certain value and this in turn means that the brightness of the source of the electrons is important. Brightness, an intrinsic property of a source, is a measure of

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[19] A. Agar, The story of European commercial electron microscopes. *Adv. Imaging Electron Phys*. **96** (1996) 415–584.

[20] P.W. Hawkes (ed.), The beginnings of electron microscopy. *Adv. Electron. Electron Phys*. Supplement 16 (1985).

[21] C.W. Oatley, The early history of the scanning electron microscope. *J. Appl. Phys*. **53** (1982) R1–R13.

[22] R. Castaing, Applications des sondes électroniques à une méthode d'analyse ponctuelle chimique et cristallographique. Thèse, Paris (1951).

the current emitted per unit area and per unit solid angle. Thermionic sources (heated tungsten or, later, lanthanum hexaboride filaments) have a limited brightness, sufficient for scanning microscopy as described above. But in the 1960s Albert Crewe, whose background was in accelerator optics and who was at the time director of the Argonne National Laboratory near Chicago, set out to construct a high-resolution scanning transmission electron microscope. The specimen is now thin, as in the TEM, and the scattered electrons that generate the signals are collected beyond the specimen. Crewe soon realised that higher brightness than in the traditional SEM would be needed and that this could be achieved by using a field-emisson source instead of a thermionic emitter. The drawback was that such sources require a vacuum several orders of magnitude better than that for a thermionic gun. He presented his first results at a small conference on Non-Conventional Electron Microscopy in 1965, in the Cambridge University Engineeering Department where so much of the early SEM development had taken place; many of the SEM pioneers were in the audience and the first reactions were cool. But by 1968, the new scanning transmission electron microscope with its field-emission gun was working well and was soon taken up by several of the major electron microscope manufacturers of the period (notably Siemens and AEI). However, it was a company new to electron microscopy but with long experience in high-vacuum technology, Vacuum Generators (VG), that captured the market and many of their instruments are in use today, often equipped with the aberration corrector that we shall meet below.

The 1960s were a particularly active decade for our subject. Not only were the SEM and the STEM developed but the first successful holographic reconstructions were made, in Tübingen (where Gottfried Möllenstedt and H. Düker had introduced the electron biprism in 1954) by Herbert Wahl and Möllenstedt[23] and in the Hitachi research laboratory in Japan by Akira Tonomura,[24] H. Watanabe, Hiroshi Tomita and colleagues. Holography had originally been invented by Dennis Gabor in 1948 as a means of circumventing Scherzer's finding that spherical aberration was inevitable; his idea was to record the image in such a way that the deleterious effect of spherical aberration could be cancelled in a second light-optical step. At that time, the method could not be made to work since electron sources did not possess the necessary coherence (the spatial coherence, closely related to the source size, was inadequate and the temporal coherence, related to the energy spread of the illuminating beam, was even worse); moreover, the laser (needed for the optical reconstruction) had not yet been invented! The first experiments in Tübingen and Tokyo were extremely encouraging and since then the techniques have been steadily improved, largely thanks to Tonomura and Hannes Lichte, formerly in Tübingen and now in a custom-built laboratory in Dresden. Digital processing has now replaced the optical reconstruction step and holography is in regular, if not routine use for imaging magnetic fields, strain fields and other phase distributions. Ironically, holography is now performed in aberration-corrected instruments and is no longer regarded as a method of correction.

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[23] G. Möllenstedt and H. Wahl, Elektronenholographie und Rekonstruktion mit Laserlicht. *Naturwissenschaften* **55** (1968) 340–341; H. Wahl, Experimentelle Ermittlung der komplexen Amplitudentransmission nach Betrag und Phase beliebiger elektronenmikroskopischer Objekte mittels der Off-Axis-Bildebenenholographie. *Optik* **39** (1974) 585–588.

[24] A. Tonomura, A. Fukuhara, H. Watanabe and T. Komoda, Optical reconstruction of image from Fraunhofer electron-hologram. *Japan. J. Appl. Phys*. **7** (1968) 295.

The earliest three-dimensional reconstructions were made by David de Rosier and Aaron Klug[25] in the MRC Laboratory of Molecular Biology in Cambridge, while in Munich Walter Hoppe too was working on reconstruction. Although he felt that his contribution to this subject was unfairly neglected, he will go down in history for a slightly later but much more original contribution, which he called ptychography.[26] This was not likely to succeed in the transmission electron microscope, for it called for structured illumination, but in recent years it has been adapted to the STEM, principally by John Rodenburg (now at the University of Sheffield) and huge progress is being made in what is called "lensless microscopy", as we shall see in Section 5.1. Back in the 1960s, the notion of optical transfer function was introduced into electron imaging theory by Karl-Joseph Hanszen and colleagues in Brunswick, nearly a quarter of a century after the appearance of the seminal *L'Intégrale de Fourier et ses Applications à l'Optique* by Pierre-Michel Duffieux (1891–1976), the founder of what we now call Fourier optics. This colourful figure merits a small digression. Professor of physics in the University of Rennes in the 1930s, Duffieux was perplexed by a problem posed by Charles Fabry; eventually, he found a solution but none of his contemporaries could understand it! In 1941 he presented his idea to the French Physical Society but no-one showed any interest in his presentation—again, no-one had been able to follow it! Pierre Fleury encouraged him to write a book about it and the result was privately printed at his own expense in Rennes and hence very little known—in order to obtain a copy, one wrote to the author, who moved to Besançon in 1945 (a CNRS laboratory there bears his name), and soon after, an untidily wrapped package arrived with the address written in Duffieux's spiky hand. Fortunately a copy reached Emil Wolf and credit for the theory, set out formally and lucidly in *Principles of Optics*, was given to Duffieux in Born and Wolf's best-selling treatise.[27]

Another instrumental development that had a wide impact was the high-voltage transmission electron microscope, which we owe to Gaston Dupouy (1900–1986). His interest in the instrument had been awakened in the 1930s and during the wartime years, when he was professor in Toulouse, he managed to construct a simple electron microscope, completed in 1942, which was taken up commercially after the war by OPL, as we saw earlier. Biological specimens could be observed only after many chemical transformations and cutting into thin sections and Dupouy (Director-General of the CNRS from 1950 to 1957), decided to try to observe living material in the electron microsope by inserting a small chamber containing water vapour into the microscope column. The electron energy would have to be increased considerably to enable the beam electrons to pass through the walls of the chamber, the water vapour and the specimen and Dupouy therefore determined to construct a million-volt electron microscope (at the time, 100 kV was the usual upper limit).[28] A new institute was built in

[25] D.J. de Rosier and A. Klug, Reconstruction of three dimensional structures from electron micrographs. *Nature* **217** (1968) 130–134.

[26] W. Hoppe, Beugung im inhomogenen Primärstrahlwellenfeld, I. *Acta Cryst*. A**25** (1969) 495–501; W. Hoppe and G. Strube, ibid., II. *Acta Cryst*. A**25** (1969) 502–507; W. Hoppe, ibid. III. *Acta Cryst*. A**25** (1969) 508–514; W. Hoppe, Trace structure analysis, ptychography, phase tomography. *Ultramicroscopy* **10** (1982) 187–198.

[27] M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1959).

[28] G. Dupouy, Electron microscopy at very high voltages. *Adv. Opt. Electron Microsc*. **2** (1968) 167–250; Megavolt electron microscopy. *Adv. Electronics Electron Phys*., Suppl. 16 (1985) 103–165.

Toulouse to house the instrument and the associated scientists and technicians, the Laboratoire d'Optique Electronique du CNRS. The microscope column and the Cockcroft–Walton accelerator were housed in a huge silvery sphere, the accelerator in the northern hemisphere and the column in the southern hemisphere; the microscope worked perfectly for several decades until it was sold for scrap by a later director. Dupouy's original dream of seeing living material never came to fruition for the environment inside the instrument was too hostile for survival, but the HVEM was used to study thick specimens of many kinds and once again, several commercial models were produced, notably in England (by AEI), the USA and Japan. Ten years later, in 1970, Dupouy built an even higher voltage microscope, which reached 3 MV. Recent years have seen a revival of interest in high-voltage electron microscopy, notably in Japan for high-resolution studies.

Before leaving the 1960s, we should mention the arrival of digital image processing of electron images. We have already mentioned the 3D reconstructions made by Erickson and Klug; just afterwards, W. Owen Saxton and Martyn Horner, working in the Cavendish Laboratory and equipped with a PDP-8 computer with 16 kbytes of memory, developed image processing languages, one of which evolved into the widely used SEMPER program suite, and Ralph Gerchberg and Owen Saxton devised the algorithm for solving the "phase problem" that bears their names.[29] This problem, by no means peculiar to electrons, arises when information is contained in the amplitude and phase of a complex-valued signal but only the intensity (and hence the amplitude) can be recorded. For electrons, the complex-valued signal is the wave function and the recorded intensity is the current distribution in the image: we know where the electrons arrive but not their direction of motion on arrival. In the Gerchberg–Saxton algorithm, the image and diffraction pattern of the same area of the specimen are the input data and an iterative sequence of Fourier transforms yields the missing phase. Many improvements have been made to the original algorithm but the basic idea is always the same: obtain two or more sets of information about the same specimen area and iterate between them, using some appropriate constraint (in the Gerchberg–Saxton case, the fact that the complex signals are related by a Fourier transform).

4. Aberrations and their correction

Electron lenses suffer from three families of aberrations: geometrical, chromatic and parasitic. Geometrical aberrations reflect the fact that lens behaviour is studied in terms of a power series in the position and angle coordinates of the electron at the object plane (some other choices may be preferable but the argument is the same). In a first (linear) approximation, lens behaviour is described by the usual cardinal elements (focal length, etc.) and to the next higher approximation (terms of third degree in the coordinates for a system with a straight axis), this is perturbed by aberrations. For a rotationally symmetric electrostatic lens, these are five in number (spherical aberration, coma, astigmatism and field curvature and distortion) while for magnetic lenses, three additional "anisotropic" aberrations must be added (anisotropic coma, astigmatism and distortion). Chromatic aberrations arise because the focusing strength of

[29] R.W. Gerchberg and W.O. Saxton, A practical algorithm for the determination of phase from image and diffraction plane pictures. *Optik* **35** (1972) 237–246.

electron lenses varies rapidly with the energy (and, hence, the wavelength) of the electrons in the beam. This effect is so strong that fluctuations in the accelerating voltage must be limited to a few parts per million. Finally, parasitic aberrations, always present, arise because no system is perfect. The assumed rotational symmetry will never be exact, some parts may be slightly misaligned or tilted and the metal from which the yoke of a magnetic lens is made may not be perfectly homogeneous.

We have seen that Scherzer described possible aberration correctors based on relaxation of each of the necessary conditions in the derivation of equation (2). In particular, he showed that, by using electron lenses without rotational symmetry, it would in principle be possible to eliminate the most harmful of the geometric aberrations, the spherical aberration. (As we saw earlier, this is the principal resolution-limiting aberration because it remains large for points on or close to the optic axis. At high resolution, this is the only region of interest and the other aberrations are negligible or can be removed.) Among his other proposals, Scherzer showed that a configuration including an electron mirror can correct C_s, an idea that had already been mentioned by Zworykin et al. in their influential texbook.[30]

All Scherzer's proposals of 1947 were explored thoroughly during the following decades, with particular emphasis on the use of elements without rotational symmentry. Scherzer had suggested the use of cylindrical lenses (that is, the electron analogues of glass lenses with cylindrical rather than spherical surfaces) together with octopoles but Geoffrey Archard soon suggested that these cylindrical lenses could be advantageously replaced by quadrupole lenses. Moreover, the latter could be combined with octopoles into a single unit. Early experiments, which showed that Scherzer was on the right lines, were made by Seeliger in Darmstadt and, a decade later, Hans Deltrap in the Cavendish Laboratory showed that real correction could be achieved with a system of four quadrupoles and three octopoles. Enormous efforts were expended to make such a system operational in a high-resolution electron microscope, notably by Scherzer himself and his team in Darmstadt and by Albert Crewe's team in the University of Chicago. All failed. Even though some simplifications are possible, a quadrupole–octopole corrector contains so many poles (or electrodes) that alignment and adjustment are just too complicated. Efforts persisted nonetheless, and in 1994 Joachim Zach and Max Haider in the European Molecular Biology Laboratory in Heidelberg showed that the spherical aberration of the probe-forming lens in a low-voltage SEM (LVSEM) could be reduced by means of a quadrupole–octopole system.[31] Here, the challenge is not so steep as in the TEM, where a highly perfected lens must be coupled to a set of quadrupoles, the spherical-type aberrations of which are much greater than that of the round lens to be corrected; the octopoles are then required to correct both the large aberrations of the quadrupoles and the small aberration of the round lens, a highly unstable arrangement. In the LVSEM, however, the lens to be corrected is (relatively) poor and some degree of correction is therefore welcome. In the same period Ondrej Krivanek made a new attempt, together with L.M. Brown and others in the Cavendish Laboratory, to use quadrupoles and octopoles to correct the spherical aberration of the probe-

[30] V.K. Zworykin, G.A. Morton, E.G. Ramberg, J. Hillier and A.W. Vance, *Electron Optics and the Electron Microscope* (Wiley, New York and Chapman & Hall, London, 1945).

[31] J. Zach and M. Haider, Aberration correction in a low voltage SEM by a multipole corrector. *Nucl. Instrum. Methods Phys. Res. A* **362** (1995) 316–325.

forming lens of a STEM. Here, the problem is less acute than for the TEM because correction is only needed at the point where the probe is formed and not, as in the TEM, over an extended area. His efforts were at last successful, as he reported at the 1997 meeting of EMAG[32] (the Electron Microscopy and Analysis Group of the Institute of Physics), which itself celebrated the centenary of J.J. Thomson's first paper on the electron (or corpuscle!). Why was he successful where so many others had failed? They had been defeated largely by the complexity of such systems but since then, ever faster and more powerful computers had entered the laboratory and Krivanek could now harness the power of the computer to provide rapid feedback and automatic control of the many settings involved. Improved STEMs appeared and the NION Company, set up by Krivanek, now sells a wholly new corrected instrument as well as correctors for existing STEMs.

But quadrupoles and octopoles are not the only non-rotationally symmetric lenses capable of providing correction. As long ago as 1965, I had noticed that sextupoles (devices with six poles or electrodes regularly spaced around the axis, also known as hexapoles) should be capable of providing correction but at the time I could not see how their intrinsic asymmetry could be removed. The geometrical aberrations of round lenses and quadrupoles and octopoles are of third order; that is, they depend on the third power of the coordinates of position and angle of the electron trajectory in the specimen plane. Sextupoles, however, have a second-order deflexion effect and third-order aberrations; conversely, they have no linear paraxial effect, which is an attraction. In Crewe's laboratory in Chicago, Vernon Beck realized in the late 1970s that a combination of two sextupoles could be made free of the unwanted second-order effect, leaving the third-order aberrations available for use as a corrector of the spherical aberration of a round lens. Soon after, Harald Rose, who had been deeply involved in aberration studies since the 1960s, described an improved configuration and this forms the basis of the sextupole corrector developed by Max Haider and colleagues, who created the firm CEOS to market it. They too obtained their first true correction in 1997 and submitted their results to *Nature* but the paper appeared only in 1998[33]—details of this saga are to be found in my survey article.[34] Their sextupole corrector now equips many commercial transmission electron microscopes.

With TEMs and STEMs free of spherical aberrations (and of many other geometrial and parasitic aberrations) the task is, however, still incomplete. The chromatic aberration remains inacceptable for the highest resolutions. In a first step, reasonably easy to put into practice, the energy spread of the illuminating beam can be narrowed by means of a monochromator, a device that allows only a small range of energies to pass. Electrons with energies outside the window are intercepted. This has the disadvantage that the beam current is reduced but is otherwise effective. A better solution is to correct the chromatic aberration of the electron lens at the same time as the spherical aberration but, unfortunately, this obliges us to use quadrupole–octopole correctors instead of sextupoles since the only practical form of chromatic aberration

32 O. Krivanek, N. Dellby, A.J. Spence, R.A. Camps and L.M. Brown, Aberration correction in the STEM. In J.M. Rodenburg (ed.), *Proc. EMAG 1997, Cambridge*, pp. 35–39 (Institute of Physics, Bristol 1997).

33 M. Haider, S. Uhlemann, E. Schwan, H. Rose, B. Kabius and K. Urban, Electron microscopy image enhanced. *Nature* **392** (1998) 768–769.

34 P.W. Hawkes, Aberration correction past and present. *Phil. Trans. R. Soc*. A**367** (2009) 3637–3664.

corrector requires us to use a hybrid electrostatic–magnetic quadrupole. Such correctors are currently preoccupying the electron opticians.[35]

As a final degree of sophistication, we mention the even more complex designs that are capable of cancelling most or all of the aberrations of electron lenses, not just the spherical aberration. Harald Rose, in particular, has proposed several configurations for the American TEAM project and for the SALVE (sub-ångström low-voltage electron microscope) project directed by Ute Kaiser in Ulm.[36] An idea of their complexity can be gained from the fact that in one design, two multipole quintuplets separated by a round lens doublet are involved.

The crucial importance of the computer in the successful accomplishment of aberration correction reminds me of another important thread in the electron optical skein. Theoretical electron optics has always been important and has attracted many talented mathematicians and mathematical physicists. We have already met Walter Glaser and Otto Scherzer, the giants of German electron optics, but they were by no means alone. In Russia, for example, P.A. Grinberg, P.P. Kas'yankov and O.I. Seman made many valuable contributions; O.I. Seman in particular devised a very ingenious procedure that replaced the hit-or-miss method of transforming aberration integrals based on partial integration (to show that equations (2) and (3) are equivalent, for example) by a systematic approach based on differentiation. A later Russian group in the A.F. Ioffe Institute in Leningrad, as it then was, worked extensively on the form of the aberration coefficients of quadrupoles and octopoles and produced the standard work in Russian on electron optics. In Alma Ata, in Kazakhstan, a new and powerful way of analysing mirror systems was developed. In England, P.A. Sturrock at the Cavendish Laboratory reformulated the Hamiltonian approach to aberration studies, pioneered by Glaser, and gave the corresponding perturbation theory a formal elegance. In Germany, Friedrich Lenz in Tübingen added much to our understanding of electron lens properties and created a brilliant school of pure and applied electron opticians; the same could be said of Otto Scherzer in Darmstadt and his pupil Harald Rose. But with the increasing power of computers, pure theory joined or was even replaced by numerical studies. In two centres in particular, London and Brünn, highly sophisticated program suites for electron optics have been generated while in Michigan State University, Martin Berz has developed very advanced programs based on differential algebra for accelerator optics.[37] In London, Eric Munro, who was one of the pioneers of computer electron optics, created MEBS (Munro's Electron Beam Software) and a speciality of his software is the inclusion of routines employing differential algebra.[38] In Brünn, Bohumila

[35] M. Haider, P. Hartel, H. Müller, S. Uhlemann and J. Zach, Information transfer in a TEM corrected for spherical and chromatic aberration. *Microsc. Microanal.* **16** (2010) 393–408.

[36] U. Kaiser, J. Biskupek, J.C. Meyer, J. Leschner, H. Rose, M. Stöger–Pollack, A.N. Khlobystov, P. Hartel, H. Müller, M. Haider, S. Eyhusen and G. Benner, Transmission electron microscopy at 20 kV for imaging and spectroscopy. *Ultramicroscopy* (2011) (in press).

[37] M. Berz, Modern map methods for charged particle optics. *Nucl. Instrum. Meth. Phys. Res.* A**363** (1995) 100–104; K. Makino and M. Berz, Optimal correction and design parameter search by modern methods of rigorous global optimization. *Nucl. Instrum. Meth. Phys. Res.* **645** (2011) 332–337.

[38] E. Munro, Numerical simulation methods for electron and ion optics. *Nucl. Instrum. Meth. Phys. Res.* **645** (2011) 266–272; L. Wang, J. Rouse, H. Liu, E. Munro and X. Zhu, Simulation of electron optical systems by differential algebra method combined with Hermite fitting for practical lens fields. *Microelectron. Engng* **73–74** (2004) 90–96.

Lencová's company SPOC (Software for Particle Optics Calculations) offers programs such as EOD (Electron Optical Design), a feature of which is the ability to extract aberration coefficients from exact ray tracing as well as from aberration integrals; another important tool enables the user to check the accuracy of the results of the calculations.[39] In the future, it seems certain that these approaches will complement and even supplant the calculation of more aberration integrals. Those for all the primary (third-order) aberrations of round lenses, quadrupoles and octopoles and many other optical elements such as deflectors and Wien filters are well established;[40] formulae are known for the fifth-order aberration coefficients of round lenses but for any other high-order aberrations and for parasitic aberrations, exact ray tracing seems the better approach. The days of sheets of double foolscap covered with handwritten expressions are long since past, replaced first by computer algebra and now by fast computer routines.

5. Phase

5.1 Are lenses necessary?

Although aberration correction occupied the lion's share of electron optical endeavours during the first decade of the century, a very different approach was challenging the very need for such correctors. The object of electron microscopy is of course to obtain structural information down to scales of the order of 50 pm and another way of reaching this is a new variant of ptychography. Here, overlapping areas of the specimen are illuminated in turn and the far-field diffraction pattern from each area is recorded. In principle, no lens is needed between the specimen and the recording plane and this technique has hence become known as lensless microscopy. A sequence of computer operations on the set of signals recorded yields the desired image. This procedure was inspired by an idea of R.H.T. Bates but has been developed essentially by John Rodenburg, at the Cavendish Laboratory and more recently at the University of Sheffield. It has now reached the point where it is being successfully used in light microscopy and great progress is being made with electrons.[41] This is by no means the only form of lensless microscopy, as a long survey by John Spence eloquently shows.[42]

5.2 Phase plates

The specimens studied in transmission electron microscopes are essentially phase objects: all the electrons incident on the specimen emerge from the far side and information about the structure is coded as changes of direction—scattering —of the electrons. In routine use, such

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39 B. Lencová and J. Zlámal, A new program for the design of electron microscopes. *Phys. Procedia* **1** (2008) 15–324; J. Zlámal and B. Lencová, Development of the program EOD for design in electron and ion optics. *Nucl. Instrum. Meth. Phys. Res.* **645** (2011) 278–282.

40 P.W. Hawkes, Aberrations. In: J. Orloff (ed.) *Handbook of Charged Particle Optics*, pp. 209–340 (CRC Press, Boca Raton, 2009).

41 J. Rodenburg, Ptychography and related diffractive imaging methods. *Adv. Imaging & Electron Phys.* **150** (2008) 87–184; F. Hüe, J.M. Rodenburg, A.M. Maiden and P.A. Midgley, Extended ptychography in the transmission electron microscope, possibilities and limitations. *Ultramicroscopy* (2011, in press).

42 J.C.H. Spence, Diffractive (lensless) imaging. In P.W. Hawkes and J.C.H. Spence (eds), *Science of Microscopy*, vol. 2, pp. 1196–1227 (Springer, New York, 2008).

phase changes are converted into visible contrast at the image by two effects: first, a stop in the back-focal plane of the objective lens intercepts certain electrons, and their absence is a source of contrast; secondly, the combination of some defocusing of the objective lens and its natural spherical aberration impress a phase change on the elecron wave function, which acts rather like a phase plate in a light microscope. For many years, sporadic attempts have been made to introduce real phase plates into the microscope, in order to control the phase changes introduced. Until recently, these met with only moderate success, owing to charging of the plate and damage caused by the irradiating beam. New attempts to use such phase plates are proving much more promising.[43] The phase plates may be thin films or "virtual" plates, which are in fact local electrostatic fields. It is too soon to guess how widely these will be used. A good idea of the present situation can be gained from the papers of Typke,[44] and of Schultheiss, Schröder and their group,[45] in which earlier work is cited.

6. Concluding remarks

It will, I hope, be apparent that electron optics and electron microscopy are in a state of effervescence and that this survey can be at best a progress report and not a history of the subject. Before coming to a halt, I should like to draw attention to some very high points in the story. At least three Nobel Prizes have been awarded for one aspect or another of electron optics, not to mention those that have been awarded to electron microscopists for their findings obtained with this instrument. First, of course, we must mention the Nobel Prize awarded to Ernst Ruska in 1986, not long before his death. Nobel Prizes were also awarded to Dennis Gabor (1971) and to Aaron Klug (1982). A glance at the list of Nobel Prizewinners shows that many other prizes are related to the history of the electron and its optics—J.J. Thomson (1906), Braun (1909), Wien (1911), W.H. and W.L. Bragg (1915), Louis de Broglie (1929), C.J. Davisson and G.P. Thomson (1937) and Kai Siegbahn (1981) come to mind. I cannot list here all the related national prizes or other distinctions but a glance at the list of Fellows of the Royal Society, honorary members of national microscopy societies and similar bodies shows the enormous influence of this instrument worldwide.

What can we expect in the next few years? Further improvements in correction undoubtedly and, in particular, aberration correctors that combine geometrical and chromatic correction.[46] The

[43] R. Danev and K. Nagayama, Transmission electron microscopy with Zernike phase plate. *Ultramicroscopy* **88** (2001) 243–252.
[44] D. Typke, Zernike phase contrast electron microscopy with a spherically corrected foil lens. *Microsc. Microanal.* **16** (2010) 441–444.
[45] B. Gamm, M. Dries, K. Schultheiss, H. Blank, A. Rosenauer, R.R. Schröder and D. Gerthsen, Object wave reconstruction by phase-plate transmission electron microscopy. *Ultramicroscopy* **110** (2010) 807–814; K. Schultheiss, J. Zach, B. Gamm, M. Dries, N. Frindt, R.R. Schröder and D. Gerthsen, New electrostatic phase plate for phase-contrast transmission electron microscopy and its application for wave-function reconstruction. *Microsc. Microanal.* **16** (2010) 785–794; M. Dries, K. Schultheiss, B. Gamm, A. Rosenauer, R.R. Schröder and D. Gerthsen, Object-wave reconstruction by carbon film-based Zernike- and Hilbert-phase plate microscopy: a theoretical study not restricted to weak-phase objects. *Ultramicroscopy* **111** (2011) 159–168.
[46] R. Leary and R. Brydson, Chromatic aberration correction: the next step in electron microscopy. *Adv. Imaging Electron Phys.* **165** (2011) 73–130.

ptychographic approach will surely be developed and dominate some fields of application. The phase plates evoked in the closing section may well play a gradually larger role. Another aspect of electron microscopy has not been mentioned here: the use of very short exposure times. This is likely to become more widespread, as the book by Zewail and Thomas suggests.[47]

This is not the place to examine the "vexed question" of the patenting of the electron microscope but nor can it be entirely neglected. For a text by the legal inventor, Reinhold Rüdenberg and commentary by his son and grandson, see Rüdenberg[48] and Rudenberg and Rudenberg.[49] The background to the story is summarized in the preface to the same volume of *Advances in Imaging & Electron Physics* and related documents are reproduced in Hawkes.[50] Ruska's study of the patenting of the electron microscope is cited in "Further reading" below.

7. Further reading

This survey is inevitably incomplete and unwittingly invidious, for I have had to omit the names of many of those who have made significant contributions, not to mention the contributions themselves. The following books and collections provide far more information.

On the history of the electron, several volumes appeared for the centenary in 1997, see Davis and Falconer (1997),[51] Dahl (1997),[52] Arabatzis (2006),[53] Buchwald and Warwick (2001)[54] and Kirkland and Brown (1998).[55]

For the early history of the electron microscope and the relations between some of the pioneers, see the publications of Ruska (1980, 1984, 1986),[56, 57] the historical essays of Mulvey (1962, 1973),[58, 59] the biographical article by Bodo von Borries' widow

[47] A.H. Zewail and J.M. Thomas, *4D Electron Microscopy* (Imperial College Press, London, 2010).

[48] R.R. Rüdenberg, Origin and background of the invention of the electron microscope. *Adv. Imaging Electron Phys*. **160** (2010) 171–205.

[49] H.G. Rudenberg and P.G. Rudenberg, Origin and background of the invention of the electron microscope: commentary and expanded notes on memoir of Reinhold Rüdenberg. *Adv. Imaging Electron Phys*. **160** (2010) 207–286.

[50] P.W. Hawkes, Complementary accounts of the history of electron microscopy. *Adv. Electron. Electron Phys*. Supplement 16 (1985) 589–618.

[51] E.A. Davis and I.J. Falconer, *J.J. Thomson and the Discovery of the Electron* (Taylor & Francis, London and Bristol PA, 1997).

[52] P.F. Dahl, *Flash of the Cathode Rays* (Institute of Physics Publishing, London and Philadelphia 1997).

[53] T. Arabatzis, *Representing Electrons: a Biographical Approach to Theoretical Entities* (University of Chicago Press, Chicago, 2006).

[54] J.Z. Buchwald and A. Warwick (eds), *Histories of the Electron. The Birth of Microphysics* (MIT Press, Cambridge MA and London, 2001).

[55] A. Kirkland and P.D. Brown (eds), *The Electron* (IoM Communications, London, 1998).

[56] E. Ruska, *The Early Development of Electron Lenses and Electron Microscopy* (Hirzel, Stuttgart 1980; also published as Supplement 5 to *Microscopica Acta*, 1980).

[57] E. Ruska, Die Entstehung des Elektronenmikroskops (Zusammenhang zwischen Realisierung und erster Patentmeldung, Dokumente einer Erfindung). *Arch. Geschichte Naturwiss*. (1984) 525–551; English translation: The emergence of the electron microscope. Connection between realization and first patent application. Documents of an invention. *J. Ultrastruct. Molec. Struct. Res*. **95** (1986) 3–28.

[58] T. Mulvey, Origins and historical development of the electron microscope. *Brit. J. Appl. Phys.* **13** (1962) 197–207.

[59] T. Mulvey, Forty years of electron microscopy. *Phys. Bull*. **24** (1973) 147–154.

(von Borries, 1991)[60] and the most enjoyable little book by Marton (1994);[61] Mulvey has also written biographies of Ruska (Lambert and Mulvey, 1996)[62] and Jan Le Poole (Mulvey and van de Laak–Tijssen, 2001).[63] Many autobiographical accounts are to be found in the volumes edited by Hawkes (1985)[20] and Mulvey (1996).[64] The first decade of the subject has been re-examined by Müller (2009)[65] and a full account is planned by the same author. The history of the instrument in the USA has been recapitulated in great detail in a fascinating book by Rasmussen (1997);[66] the history of Japanese electron microscopes is presented in Hawkes (1985)[20] and Mulvey (1996).[64]

The history of the scanning electron microsope has been traced in many publications—the volume edited by Breton, Smith and McMullan (2004)[67] is probably the best and certainly the fullest account. However, the first extensive account of the SEM, by Oatley, Nixon and Pease (1965),[68] remains an excellent introduction to the subject. A volume on the scanning transmission electron microscope (Hawkes, 2009)[69] charts the history of this instrument and includes two accounts by A.V. Crewe of the first years; the stories of the commercial STEMs from AEI, Siemens and VG are recounted at length. A chapter by Pennycook in a book edited by Pennycook and Nellist (2011)[70] brings the story up to date.

Aberration correction is studied at length in two thematic volumes (Hawkes, 2008; Cockayne et al., 2009)[71, 72] and a textbook, more suitable for newcomers, has recently been devoted to the subject (Erni, 2010).[73] For the latest developments, see Brydson (2011)[74] and the

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[60] H. von Borries, Bodo von Borries: pioneer of electron microscopy. *Adv. Electron. Electron Phys*. **81** (1991) 127–176.

[61] L. Marton, *Early History of the Electron Microscope* (San Franciso Press, San Francisco, 1994).

[62] L. Lambert and T. Mulvey, Ernst Ruska (1906–1988), designer extraordinaire of the electron microscope: a memoir. *Adv. Imaging Electron Phys*. **95** (1996) 2–62.

[63] T. Mulvey and D.J.J. van de Laak Tijssen, Jan Bart Le Poole (1917–1993), pioneer of the electron microscope and particle optics. *Adv. Imaging Electron Phys*. **115** (2001) 287–354.

[64] T. Mulvey (ed.), The Growth of Electron Microscopy. *Adv. Imaging Electron Phys*. **96** (1996).

[65] F. Müller, The birth of a modern instrument and its development during World War II. Electron microscopy in Germany from the 1930s to 1945. In: A. Maas and H. Hooijmaijers (eds), *Scientific Research in World War II. What Scientists did in the War*, pp. 121–146 (Routledge, London and New York, 2009).

[66] N. Rasmussen, *Picture Control, The Electron Microscope and the Transformation of Biology in America, 1940–1960* (Stanford University Press, Stanford, 1997).

[67] B.C. Breton, D. McMullan and K.C.A. Smith (eds.), Sir Charles Oatley and the Scanning Electron Microscope. *Adv. Imaging Electron Phys*. **133** (2004).

[68] C.W. Oatley, W.C. Nixon and R.F.W. Pease, Scanning electron microscopy. *Adv. Electron. Electron Phys*. **21** (1965) 181–247.

[69] P.W. Hawkes (ed.), Cold Field Emission and the Scanning Transmission Electron Microscope. *Adv. Imaging Electron Phys*. **159** (2009).

[70] S. Pennycook and P.D. Nellist (eds), *Scanning Transmission Electron Microscopy* (Springer, New York, 2011).

[71] P.W. Hawkes (ed.), Aberration-corrected electron microscopy. *Adv. Imaging Electron Phys*. **153** (2008).

[72] D. Cockayne, A.I. Kirkland, P.D Nellist and A. Bleloch (eds), New possibilities with aberration-corrected electron microscopy. *Phil. Trans. Roy. Soc*. A**367** (2009) 3631–3870.

[73] R. Erni, *Aberration-corrected Imaging in Transmission Electron Microscopy* (Imperial College Press, London, 2010).

[74] R. Brydson (ed.), *Aberration-corrected Analytical Electron Microscopy* (Wiley, Chichester, 2011).

Handbook of Nanoscopy (van Tendeloo et al., 2012).[75] *The Handbook of Charged Particle Optics* edited by J. Orloff (2009)[76] contains abundant information, notably about electrostatic lenses (Lencová, 2009),[77] magnetic lenses (Tsuno, 2009)[78] and aberrations (Hawkes, 2009);[40] *Science of Microscopy* (Hawkes and Spence, 2008)[79] is likewise a rich resource. The books by Tonomura (1998, 1999)[80, 81] on electron holography are very readable; for a recent account of the work in Dresden, see Lichte et al. (2010).[82] Ptychography is in a state of rapid progress; see the articles cited above and watch out for new developments in the electron optical literature, notably in *Ultramicroscopy*.

[75] G. van Tendeloo, D. van Dyck and S.J. Pennycook (eds), *Handbook of Nanoscopy* (Wiley, Chichester, 2012).
[76] J. Orloff (ed.), *Handbook of Charged Particle Optics* (CRC Press, Boca Raton, 2009).
[77] B. Lencová, Electrostatic lenses. In: J. Orloff (ed.), *Handbook of Charged Particle Optics*, pp. 161–208 (CRC Press, Boca Raton, 2009).
[78] K. Tsuno, Magnetic lenses for electron microscopy. In: J. Orloff (ed.), *Handbook of Charged Particle Optics*, pp. 129–159 (CRC Press, Boca Raton, 2009).
[79] P.W. Hawkes and J.C.H. Spence (eds), *Science of Microscopy* (Springer, New York, 2008).
[80] A. Tonomura, *The Quantum World Unveiled by Electron Waves* (World Scientific, Singapore, River Edge NJ & London, 1998).
[81] A. Tonomura, *Electron Holography* (Springer, Berlin & New York, 1999).
[82] H. Lichte, M. Linck, D. Geiger and M. Lehmann, Aberration correction and electron holography. *Microsc. Microanal*. **16** (2010) 434–440.