Recent advances in electron optics and electron microscopy

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ABSTRACT. Electron optics was born in 1927, when Hans Busch showed that the elementary lens equation is applicable to electron image formation. Interest was soon awakened in France and a book on the subject by Louis de Broglie appeared in 1950. We draw attention to the highlights of electron optics and microscopy over the decades, with particular reference to aberration correction, and to some little known French contributions to the story of the scanning electron microscope and to Fourier optics.

The natural starting-point for this occasion is the book *Optique électronique et corpusculaire* by Louis de Broglie, published in 1950 and based on lectures delivered in the Institut Henri Poincaré during the academic years 1946–47 and 1947–48. This is a remarkably complete work, all the more impressive in that de Broglie had not contributed to geometrical electron optics and, according to his preface, had not had access to any recent books on electron optics and electron microscopy; the only works on electron optics cited are the early text of Brüche and Scherzer (1934) and the dissertation of Paul Chanson (1947). He appears not to have been aware of the important collection of articles published in book form by Busch and Brüche in 1937 (which had first appeared in the *Zeitschrift für Technische Physik*) nor of the book by von Ardenne (1940), which was reasonably widely available and is among the references in Chanson's thesis. It is less surprising that de Broglie had not seen the excellent book published in England in 1939 by Myers (though this too is listed by Chanson) but surely copies of Zworykin *et al.* (1945) would have reached post-war France. We know from the bibliography of Grivet's lecture at the Réunions d'Etudes (1946) that he had seen several of the major German publications, though he too cites neither Myers nor Zworykin *et al.*

Electron optics has its beginnings more than 20 years earlier. Surprisingly, wave electron optics preceded geometrical electron optics: the association of a frequency and hence a wavelength with charged particles by de Broglie (1925) antedates the recognition that a magnetic coil has the same effect on a beam of electrons as a glass lens on light rays. It was not until 1927 that Hans Busch (1884–1973) showed that the elementary lens relation is applicable in both cases. With hindsight, it seems odd that the discovery was not made much sooner. The seed of such an idea is to be found in the work of Hamilton, nearly a century earlier, and at any time since then, and especially since the demonstration by Thomson in 1897 and 1899 that cathode rays are light charged particles, someone might have asked himself the question: 'If the same basic principle governs mechanics and optics, what is the mechanical analogue of a lens?' But apparently no-one did.

The thirty years that separated Thomson's first paper on the electron (a word that he himself eschewed) and Busch's lens equation saw repeated attempts to improve the concentration of electrons in the spot of the cathode-ray tube, which Braun had invented in 1897, before the nature of the rays had been elucidated! We note in passing that Dennis Gabor (who was later to invent electron holography) worked on this CRT problem and in fact constructed a real electron lens, a coil enclosed in an iron yoke, to concentrate the beam but, as he ruefully admitted later, he did not realise at the time how his 'lens' worked.

In the research unit of Professor Matthias in the Berlin Technical University, Ernst Ruska (1906–1979) was given the task of verifying Busch's lens formula experimentally by his research supervisor, Max Knoll (1897–1969). In 1931, he formed a magnified image by combining two primitive lenses and the electron microscope came into being. That story has been told in detail by Ruska himself (1979, 1980) and we therefore move on to a result obtained in 1936 by Otto Scherzer (1909–1982), which has had an immense influence on electron optical research ever since. The resolution of the electron microscope is limited by the geometric and chromatic aberrations of the objective lens of the instrument (once all parasitic aberrations caused by mechanical imperfections have been eliminated). Only the aberrations that do not vanish close to the optic axis are of importance in a magnifying system because only the region of the specimen close to the axis is observed. The relevant aberrations are the *spherical* and *chromatic* aberrations. The spherical aberration blurs the image since rays that travel far from the optic axis are focused more strongly than those close to the axis; the chromatic aberration again blurs the image since electrons with slightly different wavelengths are focused more or less strongly. This latter aberration is so large that highly monochromatic beams must be used (today, the energy spread of the electron beam in a microscope does not exceed an eV or a few eV for a beam energy of 200–300 keV). These aberrations are characterized by two coefficients, the spherical aberration coefficient C_s and the chromatic aberration coefficient, C_c. The unwelcome result established by Scherzer (during the long night hours when he was on Home Guard duty, next to a telephone that never rang) is expressed by the following formulae:

$$C_s = \frac{1}{16}\int_{z_o}^{z_i}\left\{b^4h^4 + 2\left(hb' + h'b\right)^2 h^2 + 2b^2h^2h'^2\right\}dz$$

$$C_c = \frac{1}{4}\int_{z_o}^{z^i} b^2h^2 dz$$

in which *b(z)* is a multiple of *B(z),* the magnetic induction on the optic axis, and *h(z)* is a solution of the paraxial equation of motion. It is immediately obvious that the integrands are positive definite (or strictly, non-negative definite) and hence that these aberrations can never be eliminated by clever lens design. Moreover, their values for real lenses are so huge that electron lenses must be operated at very small numerical apertures.

Loopholes in Scherzer's derivation were immediately sought, notably by the great electron optician Walter Glaser (1906–1960), who observed that the C_s integral can be transformed by partial integration into the form

$$C_s = \frac{1}{48}\int_{z_o}^{z_i}\left(5b^2 - bb'' + 4b^4\right)h^4 dz = \int_{z_o}^{z_i} f(B)h^4 dz$$

The function *f(B)* contains only the field distribution *B(z)* and its derivatives. Glaser argued that the field obtained by setting the integrand equal to zero and solving the resulting differential equation for *B(z)* would yield a field distribution free of spherical aberration. Moreover, he did indeed succeed in finding a solution (Glaser, 1940), subsequently exploited for beta-ray spectrometers but useless for microscopy since it does not admit the existence of a pair of real conjugates. In passing, we might wonder whether a referee today would recommend publication of Glaser's paper, as Scherzer's formula (above) shows that such an attempt could never succeed for all the terms in the integrand must vanish if the integral is to be equal to zero, but Glaser was undeterred by such negative thinking. Many years later, Werner Tretner, a pupil of Scherzer's, established the lower limits on C_s and C_c in practical lenses (Tretner, 1959).

Just after the war, a series of Réunions d'Etudes et de Mises au Point was presided over by de Broglie. The theme was 'L'Optique Electronique' and the lectures give a vivid snapshot of the state of electron optics in France in 1945. They open with an account of 'Mécanique ondulatoire en optique électronique' by de Broglie himself (22 May 1945), in which we are reminded that "L'optique électronique aurait pu avoir en France son développement initial. Dès 1927–28, je signalais à l'un de mes premiers élèves l'intérêt qu'il y aurait à développer l'optique géométrique des électrons. Malheureusement, il n'a pas poursuivi son travail dans cette direction et moi-même, absorbé par des recherches plus générales sur la mécanique ondulatoire, je n'ai pas approfondi ces questions." Contributions by C. Magnan on the electrostatic microscope of the Collège de France, destined to be converted into an ion or proton microscope and by André Lallemand on his 'electron telescope' bring us to a noteworthy review by Emmanuel Fauré–Frémiet (29 May 1945) of the 'Applications du microscope électronique à l'étude des problèmes de la biologie cellulaire et bactérienne'. The bibliography is astonishingly complete, so far as work in the USA and Germany is concerned and it is worth noting that Fauré–Frémiet had apparently seen the American facsimile reproduction of von Ardenne's book of 1940 and the collection *Das Übermikroskop als Forschungsmittel* (de Gruyter, Berlin 1941). Paul Chanson and André Ertaud then describe the properties of electrostatic lenses and their measurement with the aid of an electrolytic tank.

The following two contributions, by Pierre Grivet (1911–1992) and Gaston Dupouy (1900–1986), are landmarks in the history of the electron microscope in France (Grivet, 1946; Dupouy, 1946). The authors describe the construction of an electrostatic and a magnetic electron microscope during the wartime years and the results obtained with them. Grivet's chapter contains a good bibliography, revealing that he too had had access to some of the German and American publications of the previous years and in particular, to the special electron microscope issue of the *Jahrbuch der AEG Forschung* (1940) and to the book describing the achievements of AEI in this area published three years later (Ramsauer, 1943). Dupouy's lecture, a splendid example of his style, is followed by a 'Discussion', opened by de Broglie thus: "Je remercie Mr Dupouy de sa très belle conférence. Il nous a montré les beautés, les finesses de l'optique électronique magnétique, ainsi que les résultats intéressants qu'il a obtenus. Si la France arrive à ecrire quelques pages nouvelles dans les livres de l'optique électronique, ce sera en partie à Mr Dupouy qu'elle le devra".

The collection concludes with an essay by L. [*sic*] Léauté on 'Les applications du microscope électronique à la métallographie' (Léauté, 1946). For the examination of metals, Léauté (who was in fact André Léauté, 1882–1966, Professeur at the Ecole Polytechnique) mentions first the emission microscope and then reproduces an image of a replica obtained by Mahl, inventor of this technique. The third method mentioned is by far the most unexpected: Léauté, following the example of von Ardenne, developed a scanning electron microscope, now completely forgotten. The Notice sur la Vie et l'Œuvre[1] de André Léauté (1882–1966) by Jacques Pomey (1969) tells us a little more about this instrument and I therefore quote it at length: "Pour un esprit comme le sien, l'enseignement ne peut pas se concevoir sans un laboratoire, qu'il crée à l'Ecole polytechnique, et où il initie ses élèves à la méthode expérimentale. Ceux-ci étudient et réalisent de nombreux appareils originaux d'électronique, conçus par leur maître. Parmi ces chercheurs il convient de citer MM. Boisot, R. Brachet, Cl. Brachet, F. Davoine, Fouretier, G. Mayer, Taillade et surtout L. Cartan, qui fut victime de la cruauté nazie, ce dont A. Léauté n'a pas pu se consoler! A ceux-là il convient aussi d'ajouter sa fille aînée, Madame J. Guigan, qui avait reçu une formation scientifique complète et qui a travaillé pendant dix ans auprès de son père, dans ce laboratoire de l'Ecole Polytechnique.

Le principe du microscope électronique à balayage et à émission électronique secondaire est dû à Von Ardenne, mais celui-ci ne parvient pas à le réaliser et abandonne [in fact, von Ardenne did build such an instrument, which was destroyed in the wartime bombing of Berlin]. A. Léauté reprend cette idée, car il en mesure tout l'intérêt pour la métallurgie, à une époque où les métallographes ne disposaient ni du procédé d'amincissement des échantillons par polissage électrolytique de Jacquet, ni de faisceaux électroniques de très haute énergie; à cette époque la technique des répliques débutait à peine. Ce microscope à émission secondaire devait donc jouer par rapport au microscope électronique classique par transparence le même rôle que le microscope de H. Le Chatelier par réflexion vis-à-vis du microscope classique histologique utilisant les coupes minces transparentes. Il présente aussi pour autre intérêt de pouvoir en principe avoir un pouvoir résoluteur et une

[1] I am most grateful to Mlle Claudine Billoux, archivist at the Ecole Polytechnique, who kindly sent me this document and unearthed much interesting information.

profondeur de champ comparables à ceux du microscope électronique classique, ce qui permettrait de l'utiliser directement en microfractographie. Dans son laboratoire de l'Ecole Polytechnique, A. Léauté réalise d'abord un appareil préliminaire à une seule lentille, puis en 1946 il réalise le premier microscope de ce type avec capteur à multiplicateur d'électrons et reproducteur d'image à tube cathodique qu'il construit dans son laboratoire avec les conseils de R. Barthélémy. Il obtient ainsi les premères micrographies. Il est arrêté faute de crédits, mais son élève F. Davoine, qui dans son laboratoire a poursuivi des études sur l'émission électronique secondaire des métaux et sur l'influence des contraintes et de l'écrouissage, sur celle-ci, poursuit ses recherches à la Faculté des Sciences de Lyon et grâce à l'appui du CNRS et à l'expérience acquise auprès de A. Léauté, construit un nouveau microscope basé sur le même principe. Dans ce domaine, A. Léauté a joué le rôle de précurseur, car maintenant les microsondes électroniques Castaing–ONERA sont munies d'un microscope à balayage et à émission électronique secondaire et les deux grands principes de A. Léauté sont repris dans d'autres appareils: d'une part le balayage dans un microscope anglais à optique classique et à comptage électronique, et d'autre part l'émission secondaire dans un microscope électronique à photographie directe aux U.S.A." No other publication in which this microscope is mentioned has been traced, apart from a passing reference to it in the discussion at the end of a paper by Charles Brachet (1946). The work of François Davoine at Lyon is, on the other hand, better known and referred to in Grivet's *Electron Optics*, for example, in the bibliography assembled by Oliver Wells (1972) and in the list of references included in Oatley *et al.* (1985). In his doctoral thesis[2], Davoine (1957) tells us "J'ai fait mes premières armes en microscopie par balayage, au laboratoire de l'Ecole Polytechnique où MM les Professeurs Léauté et Vignal m'ont accueilli avec beaucoup de bienveillance". The chapter devoted to the scanning electron microscope built at Lyon opens with a brief account of earlier work on the SEM: Knoll's proposal of 1935, von Ardenne's and Zworykin's instruments and then a brief description of Léauté's microscope: "En France, au Laboratoire de Physique de l'Ecole Polytechnique, un microscope à balayage a été réalisé en 1944–46 sous la direction du Professeur Léauté. Dans cet instrument, les électrons secondaires étaient collectés par une simple plaque; les fluctuations dans le réseau d'entrée limitaient considérablement le pouvoir séparateur, bien que les vitesses de balayage aient été

[2] Extracts from this thesis were kindly supplied by Mme Delorme, librarian at the Université de Lyon.

extrêmement réduites et l'enregistrement effectué sur l'écran rémanent d'un tube cathodique." No reference to any publication concerning Léauté's microscope is given.

A year later, the microscopes built at the CSF Laboratory and near to Toulouse were presented to a wider audience at an electron microscope conference held in Oxford in September 1946. Henri Bruck, a colleague of Grivet's, described the electrostatic instrument and Dupouy himself talked about the magnetic instrument. At the same meeting, the Dutch and UK efforts were presented by J.B. Le Poole (1917–1993), A.C. van Dorsten and W.J. Oosterkamp and by M.E. Haine respectively, thereby encouraging cross-fertilization.

Before leaving the immediate post-war years, we draw attention to three publications that had repercussions for many decades. The first is a book entitled *L'Intégrale de Fourier et ses Applications à l'Optique* (1946), published at his own expense by Pierre-Michel Duffieux (1891–1976). This extraordinary book, by a no less extraordinary man (see Duffieux, 1972/73 and Hawkes, 1983), laid the foundations of what we now call Fourier Optics and of optical transfer theory. Duffieux (1970) has told us how the idea came to him: "Un matin où j'étais très libre, je reçus un ingénieur de l'Institut d'Optique qui représentait la maison Mader–Ott. Il me montra des appareils de mathématiques et les catalogues de la maison Mader–Ott. J'ai pris tout de suite un très beau planimètre d'Amsler. Il me proposa un dispositif nouveau qui en faisait un analyseur harmonique, plus lent que le Corradi, mais beaucoup moins cher et en réalité plus rapide quand on ne demandait que les premiers harmoniques. Tandis qu'il montait les deux appareils, je feuilletais la notice qu'il m'avait ouverte et j'eus tout de suite une révélation hallucinante…J'ai écrit en 1963 dans *L'Education Nationale* : "Il y a eu deux parts de ma vie ; j'ai d'abord cherché ma voie, puis un jour, comme cela, brusquement, je l'ai trouvée. J'ai eu quelques secondes pour choisir, j'ai choisi, et depuis cet instant-là, je travaille toujours dans la même direction. C'est à la lecture de ce catalogue que je faisais allusion". For many years, his work was known to only a tiny number of opticians but in 1959, when Born and Wolf's *Principles of Optics* appeared, justice was at last done to Duffieux: "We shall now describe some general methods based on the techniques of Fourier transforms, relating to imaging of extended objects. These methods were developed chiefly by Duffieux…", wrote Emil Wolf, and Duffieux's little-known book is cited. Many years later, the importance of the book was recognised in France and a second edition was published (1970); an English translation appeared in 1983. The Société Française des Microscopies marked the fiftieth anniversary of the first publication with a Duffieux Symposium at its annual meeting, which was held in Rennes in 1996 (see *Microsc. Microanal. Microstruct.* **8**, 1996, No. 1).

The second publication is by Otto Scherzer, who had established the formulae discussed above in 1936. Here, he re-examined the proof and showed how, by relaxing one or other of the necessary conditions, aberration correction could be achieved (Scherzer, 1947). In particular, he suggested correctors based on departure from rotational symmetry, space charge, the use of high frequency excitation and discontinuity in the potential or field on the optic axis. Each of these suggestions has been investigated and we shall see that one at least of Scherzer's proposals has been successful, though it took just half a century to make it work! (For a full account of all these attempts, see Chapter 41 of Hawkes & Kasper, 1989.)

Lastly, we recall that it was in 1948 that Dennis Gabor (1900–1979) published his first paper on holography, which he intended as a method of circumventing the adverse effect of spherical aberration in electron microscopy. He had already been thinking about this problem for some time but this completely new idea, of recording an image blurred by spherical aberration and restoring a sharp image in a second step, came to him as a bolt out of the blue: he was sitting with his wife on a garden seat beside tennis courts in Rugby, where he worked, when holography sprang into being in his mind (Mulvey, 1995).

Theory and instrumentation progressed rapidly in the next two decades. In the 1950s and early 1960s, experimental evidence accumulated showing that the principle of Scherzer's corrector based on cylindrical lenses (subsequently, quadrupole lenses) and octopoles was sound (Seeliger, 1951; Deltrap, 1964). However, this was not sufficient for the production of a working corrector that could be incorporated into an electron microscope. The first family of correctors employed four quadrupoles and three octopoles and the task of aligning and adjusting such a system proved too difficult in pre-computer days, for these correctors are inherently unstable: the objective lens of the electron microscope to be corrected is already a highly perfected optical component and the object of the exercise is to reduce the aberration coefficient of a lens on which considerable design skill has been expended. But the principle of the corrector is to create asymmetry by means of quadrupoles, which have much larger and many more aberrations than the round lens and then correct the new (large) aberrations and the (much smaller) inherent aberration by means of octopoles. It is therefore disappointing but not surprising that none of the very ambitious attempts to build correctors during the 1970s and 1980s was successful.

We shall return to aberration correction below but let us first return to the 1960s, a decade in which several major microscope projects came to fruition. In 1960, the first very high voltage electron microscope, built under the direction of Gaston Dupouy in the Laboratoire d'Optique Electronique in Toulouse, furnished its first images and it continued to operate with no serious difficulties for many decades. In about 1989, a new director of the laboratory chose to sell it for scrap. In a historical article, Dupouy (1985) wrote: "I planned to build a microscope working at one million volts, and more if it was possible. This was a bold enterprise and forecasts on all sides were absolutely pessimistic. Many difficulties were advanced; lens design, stabilization, lack of contrast.

Naturally I had, for my part, thought a lot about all these problems before I tried this bold experiment. Many people believed my plans would lead to failure; they turned out very well indeed."

The main justification for striving for such high voltages came from the life sciences. Dupouy's ambition was to image living specimens, enclosed in an environmental chamber in which the conditions necessary for survival were maintained. Only very high-energy electrons could penetrate the windows of the chamber, the atmosphere inside it and the specimen itself. Although the specimens examined did not in practice survive – the x-ray dose received was undoubtedly lethal – the ability of high-energy electrons to penetrate relatively thick layers was quickly appreciated in other areas of research and the success of the Toulouse instrument led several companies in the UK (AEI), the USA (RCA), France (GESPA) and Japan (JEOL, Hitachi) to put such instruments on the market (see Allen and Dorignac, 1998 for an assessment of the situation).

In Cambridge, a series of research students under the direction of Charles Oatley had been perfecting prototype scanning electron microscopes and, after considerable hesitation, the Cambridge Instrument Company launched the first commercial scanning electron microscope, the Stereoscan, in 1965 (Stewart and Snelling, 1964; Oatley, 1982; Oatley *et al.*, 1965, 1985). Here, a fine electron probe explores a thick specimen, the incident electrons generating one or more signals characteristic of the topography of the surface or the chemical nature of the sample: secondary electrons, back-scattered electrons, x-rays, fluorescence. The Stereoscan was the culmination of nearly twenty years of research and development in the Cambridge University Engineering Department and was at first regarded as a very risky venture, for it was far from sure that this new type of microscope would catch on. Very soon, however, demand outstripped the initial production capacity. (See

Breton *et al.*, 2004 for a rich account of the genesis of the SEM and much related material.)

That same year, 1965, an English accelerator physicist working in the USA, whose name was unknown to the electron microscope community, caused a furore at a small Institute of Physics meeting on 'Non-conventional Electron Microscopy' held in Cambridge at which many of the SEM pundits were present. Here, Albert Crewe described his new scanning transmission electron microscope, an instrument in which, as in the SEM, a fine probe explores the specimen but now, the specimen is as thin as in a TEM and it is the electrons of the incident beam that emerge from the far side of the sample that are used to form the image. At the time, this was thought to be quite impracticable, as the time needed to record an image with the thermionic guns in regular use would be prohibitive. Crewe's major innovations were the utilisation of a field-emission source, much brighter than the thermionic gun, and above all, attainment of the very high vacuum necessary for stable field emission. Three years later (Crewe *et al.*, 1968), the microscope was working well and three companies launched commercial models: Siemens, who put a fully engineered STEM (the ST104) on the market shortly before the company withdrew from the electron microscope market; AEI, who did not persist in their project; and VG Microscopes, who equipped many laboratories with STEMs before discontinuing production.

The 1960s also witnessed other major developments in electron optics. The first holographic reconstructions were made by Akira Tonomura in Japan (Tonomura *et al.*, 1968; Tonomura, 1998, 1999) and H. Wahl (Möllenstedt and Wahl, 1968; Wahl, 1974) in the laboratory of G. Möllenstedt, where the electron biprism had been introduced (Möllenstedt and Düker, 1955, 1956). Later developments in Tübingen and more recently in Dresden are described by Lichte (1982, 1995, 2002); see also Pozzi (2002). Karl-Joseph Hanszen translated Fourier optics into the language of electron optics and introduced the notions of phase and amplitude transfer functions, which give a clear understanding of the meaning of resolution and are at the heart of much of today's digital image processing (Hanszen and Morgenstern, 1965; Hanszen, 1966, 1971; 1982). In Cambridge, the first three-dimensional reconstructions were being made by de Rosier and Klug (1968) while in the early 1970s, Ralph Gerchberg and Owen Saxton obtained the first practical solution of the 'phase problem', the problem that arises from the fact that electron microscope samples are *phase* objects (no electrons are halted within the specimen) but the fluorescent screen or photographic medium display only *amplitude* information (Gerchberg & Saxton, 1972).

We now take a large step forward to the final stage in the history of aberration correction but first, we need to mention a new type of non-rotationally symmetric corrector that was discovered in the late 1970s by Beck and Crewe in Chicago (Beck, 1979; Crewe, 1982) and by Harald Rose in Darmstadt (1981). In order to use non-rotationally symmetric elements to correct the spherical aberration of a round lens, it is clearly essential that the corrector should suffer from the same aberration as the round lens but with opposite sign. I had noticed many years earlier that sextupoles possess this property (Hawkes 1965) but since these have second-order effects as well as the third-order spherical aberration, I had dismissed them as potential correctors. However, by suitably combining two sextupoles with different orientations, a device that has the desired properties can be designed and the resulting corrector is much easier to align and excite than the quadrupole–octopole version. This type of corrector was studied and perfected (Rose, 1990) and in 1995, Joachim Zach and Max Haider showed that the size of the probe in a low-voltage SEM (Zach, 1989) could be reduced with the aid of such a device. This was essentially a proof-of-principle experiment, since the lens being corrected was comparatively poor one, but it showed that it would be worth trying to use such a device to correct the spherical aberration of a high-resolution electron microscope objective. This was attempted (Haider *et al.,* 1995) and in 1998, the first results form a corrected TEM were published (Haider *et al*., 1998a, 1998b). For some years now, the corrected microscope at the Forschungszentrum in Jülich has been producing stunning micrographs. Delivery of commercial microscopes equipped with such correctors begins in 2004 (FEI–Philips, JEOL, …).

Meanwhile, Ondrej Krivanek, working in the Cavendish Laboratory in Cambridge, had returned to the quadrupole–octopole configuration for correction not of a TEM objective, which is required to form an extended image, but of the probe-forming lens of a STEM. For this, a very flexible system was built, with all the excitations and alignment under computer control and, for the first time, correction of a high-quality lens was achieved (Krivanek *et al*., 1997). Krivanek announced his achievement at a meeting of the Electron Microscopy and Analysis Group (EMAG) of the Institute of Physics at their meeting in Cambridge to coincide with the 'discovery' of the electron in 1897. Many STEMs have now been fitted with an improved version of this corrector and the British SuperSTEM project will make two such instruments available to the scientific community.

We have seen that the widespread availability of fast computers with ample memory was an essential element in the successful development of these correctors. Computing power has also made it possible to calculate the properties of most of the electron optical elements with great accuracy in a very short time on desktop computers. Evidence for this can be found in the proceedings of the Charged-particle Optics Congresses (see Orloff and Dragt, 2004 for the most recent meeting) and especially in the work of Eric Munro and Bohumila Lencová (Munro, 1997; Lencová and Lenc, 1986; Lencová, 1997).

The introduction of the sextupole corrector generated new work for the theoreticians of electron optics and new results of the highest interest have indeed emerged as a result. The importance of system symmetries to cancel intrinsic aberrations has become clear, and this is exploited in the omega-shaped energy analyser, in which the device not only has symmetry about the mid-plane but also exhibits anti-symmetry about intermediate planes. For a systematic account of this, see Rose and Krahl (1995) or Rose (2003). Before leaving correctors, we mention that the defeat of spherical aberration has acted as a catalyst for work on overcoming the deleterious effect of chromatic aberration, notably by means of monochromators, and even more ambitiously, on correcting all the geometrical aberrations of rotationally symmetric systems (Munro *et al*., 2001; Rose, 2004).

We have mentioned image processing in passing in this survey, which is confined to a small part of the history of the subject, and to conclude, I should like to draw attention to a mathematical tool that has been added to the image processing paraphernalia and to an unsolved but fascinating problem. A glance at any image processing textbook reveals immediately that such works are more like cookery books than scientific treatises and that the vocabulary of the subject is quite different in the widely separated areas of application (microscopy, astronomy, medicine, geology, forensic science,…). In an attempt to harmonize all this work and to put it on a sound mathematical footing, an image algebra has been created, in terms of which the various image processing algorithms can be written compactly (Ritter, 1991; Ritter and Wilson, 2001; Ritter *et al*., 1990). The elements of the algebra are images (not individual pixel values) and the word 'image' is interpreted very broadly. An image is a *set* of values (pixel values in the simplest case) and addresses. It may be one-dimensional (an energy-loss spectrum, for example) or two-dimensional (an everyday image) or of higher dimensions. Several values may be associated with a given address (SEM signals, colour images). In one extremely important case, each 'pixel value' may itself be an image; this is useful in the mathematics to handle such operations as convolutions and other linear transforms and is so ubiquitous that such image-valued images are given a special name: *templates*. They are, however, much more than mere mathematical constructs, for the scanning transmission electron microscope generates such images. Since the object and image planes are not conjugates in such instruments, a far-field diffraction pattern is formed in the detector plane of the STEM from each point of the specimen (Hawkes, 1994). Moreover, the use of templates provides a remarkable and unexpected unification of the families of linear convolutional filters and the nonlinear operations of mathematical morphology. These have a very similar appearance in image algebra, the sum and product operators that appear in convolutions being replaced by max (sup) and sum, respectively. The structuring elements of mathematical morphology are represented by templates, just like the filter functions of the convolutional procedures.

This algebra was introduced, as we have seen, to simplify and harmonize the many image processing algorithms. But why should we stop there? It would be very satisfying if we could express the whole chain – image formation plus image processing – in terms of this algebra. The idea is all the more attractive in that a microscope contains stops that truncate the wavefunctions characterizing the light or electron beam and these are therefore functions with finite support; they can hence be represented by a (finite) set of sample values. It ought to be possible to describe the propagation of the wavefunction through the instrument by means of a *difference* equation (instead of a *differential* equation) and this difference equation should show us that the succession of planes of the microscope (source, specimen, diffraction pattern, image) are related by a sequence of finite Fourier transforms. Work on this is in progress (Hawkes, 2002).

References

Note: In addition to the cited material, this list contains a few historical surveys, which offer a much fuller and perhaps more balanced picture of the history of the subject: Agar (1996); Hawkes (1985); Mulvey (1996); Rasmussen (1997).

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