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Future directions in high-resolution electron microscopy: Novel optical components and techniques

L'avenir de la microscopie électronique à haute résolution : Éléments et techniques innovantes

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ABSTRACT

Aberration-corrected electron microscopes currently dominate the high-resolution scene but they are not the only instruments that can provide such information. Other techniques are attracting attention, notably ptychography and the use of phase plates. Moreover, operation of these aberration-corrected microscopes at their ultimate performance raises questions that are still under discussion. We note too that correctors can be useful for tasks other than correction, such as vortex beam creation. To conclude, the specialized role of electron mirrors is recalled.

RÉSUMÉ

La microscopie à haute résolution est actuellement dominée par les microscopes électroniques équipés de correcteurs d'aberration, mais ces instruments ne sont pas les seuls capables d'atteindre le domaine de la haute résolution. D'autres techniques les concurrencent, notamment la ptychographie et l'utilisation des dispositifs à phase. De plus, l'opération des microscopes corrigés soulève des incertitudes. Notons que les correcteurs peuvent jouer d'autres rôles que la correction, la création de faisceaux vorticiels, par exemple. En conclusion, la correction par miroir électronique est évoquée.

1. Background

The history of electron optics has been punctuated by a small number of extremely important publications, notably those of Busch, Scherzer, Beck, Crewe, Rose, Krivanek et al. and Haider et al. To Hans Busch, we owe the realisation that electrons in rotationally symmetric electrostatic or magnetic fields behave like the light rays in a glass lens [1]. This led to the development of the electron microscope by Max Knoll and Ernst Ruska [2–4] and to a paper by Otto Scherzer showing that the resolution-limiting aberrations, spherical aberration (C_s) and chromatic aberration (C_c), cannot be eliminated by skillful lens design as they can for glass lenses [5]. A later paper by Scherzer [6] described several ways of correcting these aberrations, notably by abandoning rotational symmetry in favour of cylindrical lenses or by exploiting the properties of electron mirrors. It is not possible to identify a single paper on the origin of sextupole correctors. They were not mentioned

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by Scherzer but in 1965, I pointed out that sextupoles possess an aberration of the same nature as that of spherical aberration of round lenses, though I did not propose any practical corrector design [7]. Then in 1979 Beck [8] did recognise the potential of sextupoles for spherical aberration correction and this was taken up by Crewe soon after [9–12]; a student of Crewe's tested the device a few years later [13]. At the same time, Rose proposed a sextupole corrector configuration [14,15], which is at the heart of the present commercial model. We shall not discuss electron holography here but we recall that Gabor invented holography in 1948, only a year after Scherzer's second paper, as another way of circumventing the undesirable effect of spherical aberration [16,17].

It was not until the 1990s that any of these proposals for corrector design was successfully put into practice, though numerous attempts to follow them up were made in the intervening decades [18]. In 1997, Krivanek et al. proved that quadrupoles and octopoles could correct the spherical aberration of the probe-forming lens of a scanning transmission electron microscope (STEM) and soon after they were able to surpass the resolution of an uncorrected instrument [19–23]. Meanwhile, Haider et al. [24–28] had employed a sextupole device to correct the spherical aberration of the objective of a transmission electron microscope (TEM). New ways of creating the corrector fields continue to appear [29].

Spherical and chromatic aberration are not the only aberrations that afflict these instruments but while they remained uncorrected, the others were relatively unimportant. With the advent of correctors, however, it became necessary to reconsider the situation. Leaving aside chromatic aberration for the moment, the aberrations are of two kinds: the other third-order geometrical aberrations and the higher order geometrical aberrations, notably fifth-order spherical aberration; and the parasitic aberrations, which arise as a result of imperfect alignment, symmetry and adjustment in general. However, once non-rotationally symmetric elements have been incorporated into these instruments, they can be made sufficiently flexible to cancel parasitic effects. See Haigh and Kirkland [30] or Section 6.5 of [31] for systematic studies of and many references to these aberrations.

2. Chromatic correction

Two major questions remain: what is the best way of attacking the effects of chromatic aberration and how can the reliability of corrected systems be ensured? The chromatic aberration could be overcome in two ways: either the energy spread of the electron beam could somehow be made so narrow that the chromatic aberration of the objective lens has little effect. Alternatively a corrector capable of cancelling both spherical and chromatic calibration could be designed. Until recently, the only known way of reducing the energy spread is by including a monochromator of some kind in the column; an energy resolution of 30 meV has been achieved with a STEM equipped with a monochromator. Such a device, whatever principle is adopted, selects only those electrons with energies close to the mean beam energy and rejects the remainder. The beam current is inevitably reduced, which is the only real disadvantage of monochromators. However, they are very much easier to design and implement than chromatic aberration correctors. But very recently, Janzen et al. [32] have found a way of narrowing the energy spread of the electron beam by accelerating the slower particles and retarding the faster ones. Preliminary tests are promising.

It has been known since 1961 that quadrupole systems with negative C_c can be designed and could hence be used to combat the positive chromatic aberration of round electron lenses. But these quadrupoles are more complex than those used for spherical correction since both electrostatic and magnetic quadrupole fields must be present for chromatic correction. This is easily understood when we examine the formulae for the various quadrupole chromatic aberration coefficients (Eq. 29.48 in [33]). Each contains a term representing the difference between the electrostatic and magnetic field contributions. Clearly, the signs of the coefficients can be changed by altering the relative strengths of the two contributions. The inevitability of quadrupoles has upset the earlier trend to use quadrupole and octopole correctors for STEMs and sextupole correctors for TEMs. The latter have the big attraction that they exhibit no linear focusing and their intrinsic aberrations are not unduly large. Quadrupoles, on the other hand do exert linear effects and a quadrupole–octopole corrector works by adding three large quadrupole aberrations to the small aberration of the round lens and using the octopoles to correct the total aberration, clearly an unstable arrangement. It is acceptable for a STEM, where only the axial zone is of importance but much less suitable for a TEM. But when C_c correction is required, designers have no option but to use quadrupoles. Recent thinking about chromatic correction is to be found in Zach [34], Leary and Brydson [35] and Section 9.1 of [36] and, in connection with the SALVE and TEAM projects, in [37,38].

3. Do correctors really work?

This question may seem an impertinence given the rapidly growing number of aberration-corrected microscopes in use but it reflects a real preoccupation of practising microscopists. Certainly, the control software that selects the corrector settings is capable of providing genuine correction, with which many very high-resolution images have been obtained. But a corrected microscope is not a static system: for many reasons the corrector settings need frequent readjustment and some sceptics doubt whether the diagnostic information available to the control software will always be exact enough to maintain the correction. This is a contentious subject, and this is not the place to enter the lists, but microscopists will certainly be following closely the arguments of Schramm et al. [39] and Barthel and Thust [40] and the responses to their apprehensions. Schramm et al. have examined the sensitivity of any correction to small changes in the corrector settings and have found that the required accuracy of the corrector settings increases dramatically with the degree of correction demanded of the

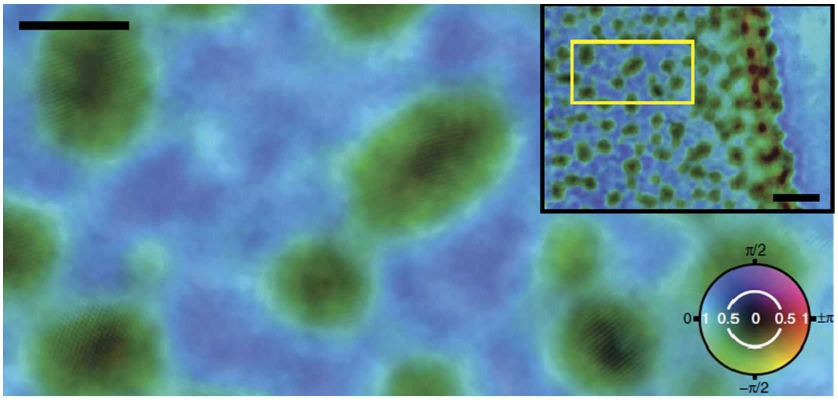

Fig. 1. (Colour online.) Ptychographic reconstruction of gold particles showing the atomic fringes. Scale bar on inset = 5 nm. Modulus and phase are combined in the image: phase is indicated by colour, modulus by brightness. After Humphry et al. [50], courtesy of the authors and Macmillan.

system. Certainly this is what intuition would suggest but Schramm et al. find that the situation is much more critical and hence the correction more fragile than had been anticipated; see too [41]. Barthel and Thust have considered the stability in time of three microscopes: an FEI Titan with chromatic and spherical correction; an FEI Titan with spherical aberration correction; and an uncorrected FEI Tecnai instrument. Extremely careful analysis of the performances of these microscopes suggests that, even in the rigorously controlled conditions of their experiments, the user cannot hope to work in aberration-corrected conditions for more than a few minutes, and that even the uncorrected microscope is not immune to such problems. However, these pessimistic opinions are not shared by all practising microscopists [42] and automation of the correction sequence is being actively studied [43]. The fact that the far-field current distribution of a STEM probe close to focus, the ronchigram, contains so much diagnostic information is a vital weapon in the struggle for correction [44].

A new obstacle has recently been identified: thermal magnetic-field noise. The importance of this emerged when the performance of a new TEAM instrument to which a chromatic-aberration corrector had been added was found to be worse than that of its predecessor! Extremely skillful detective work by Stephan Uhlemann and colleagues at CEOS, the firm that had supplied the corrector, reveals that the cause of the deterioration is magnetic-field noise arising from weak thermal currents in the conductive part of the column [45]. This very recent finding will inevitably influence future thinking on ultimate aberration correction and especially on the role of chromatic correctors.

4. Are correctors inevitable?

The object of high-resolution microscopy is to recover the phase distribution of the wave function at the exit surface of the specimen. In a TEM, the role of the microscope is to map this distribution into an amplitude (current density) distribution in some image plane. Traditional microscopy is not, however, the only way of recovering the phase distribution. The first solution of the phase problem was obtained in 1972 by Gerchberg and Saxton and has given rise to a huge literature [33,46,47]. In the 1970s Walter Hoppe described a very ingenious technique, which he named ptychography, and this has been rethought and put into practice by John Rodenburg and his team, now in the University of Sheffield [48,49]. Here, structural information about the specimen is obtained not from an image created by the objective lens and succeeding lenses but from several versions of the diffraction pattern of the specimen. Many small areas of the specimen are illuminated in turn, and the diffraction pattern of each is recorded. Care is taken to ensure that the illuminated areas overlap and the ensuing redundancy is sufficient for calculation of the phase of the wave function. This in turn yields the wave function at the exit plane of the specimen. Despite its promise, and the fact that all the hardware problems of aberration-corrected microscopes vanish, this technique is only slowly penetrating into electron microscopy. So far, its principal successes have been in x-ray studies. Nevertheless, a recent paper by Humphrey et al. [50] indicates that electron ptychography may well become a real (and less expensive) rival to the other approaches. In their experiments, an FEI Quanta 600 scanning electron microscope operating at 30 kV was used, though the method works for any accelerating voltage – the latter can be chosen to minimize radiation damage, according to the nature of the specimen. A patch of the object (20–40 nm in diameter), was illuminated by slightly defocusing the probe and the resulting diffraction pattern recorded on a CCD detector. The area outside the central bright disk contains information about the high-frequency (and hence, high-resolution) components of the specimen. Provided that the relative phases of these high-angle diffraction intensities can be recovered, a high-resolution image can be reconstituted. It is the ptychographic principle that provides this phase information: by ensuring that the illuminating patches overlap, there will be a substantial degree of redundancy in the recordings, and it is this that can be exploited to yield the missing phase information (Fig. 1). For details and extensive discussion, we refer to the publications of J.R. Rodenburg and his team [48–54]; for a very lucid account of the subject see [48]. The technique is not of course without drawbacks. The illuminating patch must be very accurately positioned and the other geometrical parameters must be calibrated very exactly. Preliminary tests suggest that the use of a defocused probe offsets the requirements on spatial

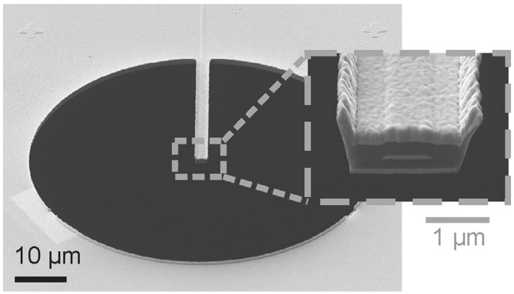

Fig. 2. Scanning electron micrograph of the "Zach" plate, a form of Boersch plate. A rod extends into the back focal plane as far as the optic axis.

coherence to some extent. Novel features of ptychography continue to emerge. Thus Maiden et al. [55] have extended the technique to three-dimensional reconstruction. In this very ingenious proposal, the multi-slice method routinely used in electron image simulation is employed in an iterative sequence to yield information about the three-dimensional structure of the specimen. Even if multiple scattering occurs, the reconstruction procedure can produce a structure.

5. Another use for correctors

The role of geometrical-aberration correctors is to modify the phase distribution over the electron wave in a beneficial fashion. However, once it is realised that a corrector is itself a kind of phase "plate", other uses of the corrector than simple correction come to mind. A recent and very original suggestion concerns the creation of electron vortex beams. Such beams have helical wavefronts, for which the wave function is of the form $A(r)\exp(il\phi)\exp(ikz)$, where $l(\neq 0)$ is an integer. They were first considered theoretically by Bliokh et al. [56] in 2007 and experiments followed in 2010 [57,58] and 2011 [59]. A book on *Linear and Chiral Dichroism in the Electron Microscope* has been edited by Peter Schattschneider [60], co-author of several papers on the subject [61,62]. Very recently, a radically new way of creating electron vortex beams has been introduced and tested by Clark et al. [63]. Here, an aberration corrector is adjusted in such a way that the lower orders of astigmatism are non-zero and take values determined by the dimensions of the annular aperture that also forms part of the system. The other aberration coefficients are rendered as small as possible. The authors have tested their proposal (in a double aberration-corrected FEI Titan transmission electron microscope) and managed to obtain a Bessel-function beam at the object plane of the microscope for which $l = 1$ was dominant. (The Bessel function arises from the presence of the annular aperture.) For some related work, see [64–68] and a forthcoming review by Herring and McMorran [69]. For many more details, see the article by Verbeeck et al. in this issue.

6. Phase plates

Since all the information about the specimen is coded in the phase of the wave function, the electron microscope is required to convert this phase distribution into an amplitude distribution in the image plane. At the crudest level, this is accomplished by the objective aperture, which removes electrons scattered through relatively large angles – their absence, one might say, creates the image. Closer to the limit of resolution of the (uncorrected) instrument, a suitable balance of spherical aberration and defocus behaves as a phase plate, thereby creating the contrast in the image described by the contrast transfer function. But even before this mechanism was fully understood, microscopists had begun to speculate about real phase plates, analogous to those used in light microscopy. Of the many ideas that have been tested (see Section 66.5.3 of [33] for numerous references), three are still attracting attention: in one case, an electrostatic (or even magnetic) field is created in the back focal plane of the microscope, which alters the phase distribution of the electron wave (Boersch phase plates). In another, a true phase plate is inserted in this plane, for the same purpose (Zernike phase plates). And finally, a half-plane aperture is introduced there (Hilbert phase plates). None of these is new but interest in all of them has revived in recent years.

The Boersch plate was proposed by Hans Boersch in 1947 [70] and has been reconsidered by Schultheiss et al. [71], Edgcombe [72–74] and Hettler et al. [75]. Zernike plates have always seemed the most attractive, despite the difficulties of producing very thin films with a prescribed thickness variation and a very careful study was made by Dieter Willasch [76,77] as long ago as the 1970s; there, earlier work, which tends to fall into oblivion, is reviewed. For recent progress, see [78–83]. Hilbert plates were first mentioned in the aftermath of the publication of Gerchberg and Saxton's solution of the phase problem. It was quickly realised that the key feature of their algorithm was the use of two (or more) recordings of the same specimen area in different conditions; images at different defocus values were suggested and another idea consisted in forming images with a half-plane aperture in two complementary orientations; see [84,85] for the roles of K.-J. Hanszen, W. Hoppe, D.L. Misell and A. Lannes. The papers of Gamm et al. [86], Dries et al. [82,87], Glaeser et al. [88] and Danev and Nagayama [89] give a good picture of recent attempts to use such plates in practice. A related idea is advanced by Buijsse et al. [90]. Fig. 2 shows a variant of the Boersch plate, a thin rod in the back-focal plane that protrudes to the centre of the microscope column and Figs. 3 and 4 illustrate results obtained with it by Schultheiss et al. [71] and very recently, by Frindt

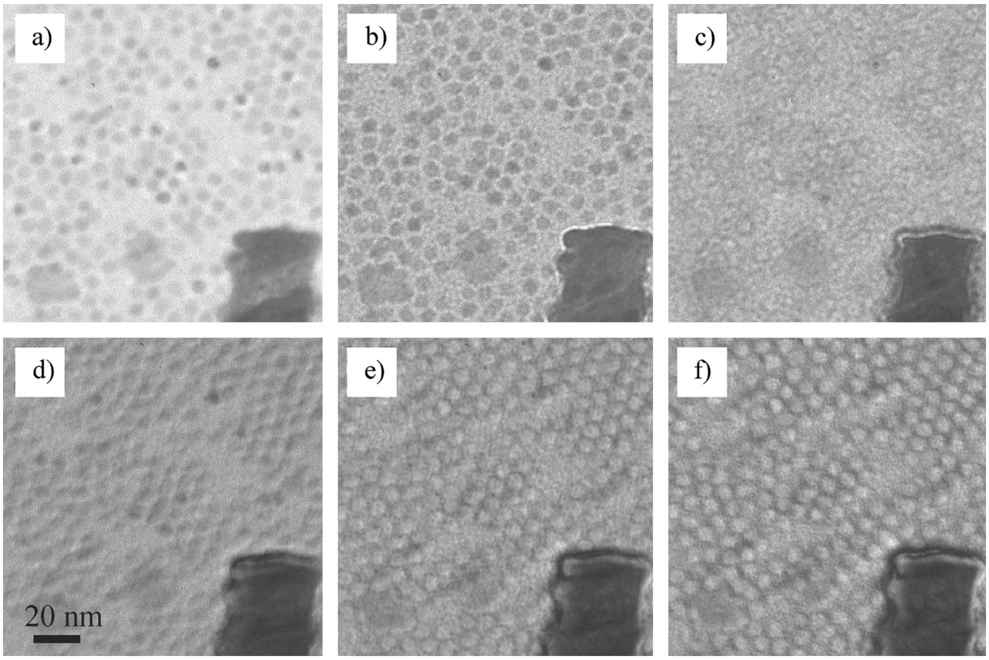

Fig. 3. Conventional images and images obtained in the presence of the phase plate of Fig. 2 showing PbSe nanoparticles about 6 nm in diameter on an amorphous carbon film. (a)–(c), CTEM images at Gaussian focus and at defocus values of −970 nm and +1980 nm. (d)–(f), images obtained with the phase plate, for three different phase shifts. Figs. 2 and 3, after [71], courtesy of the authors and Cambridge University Press.

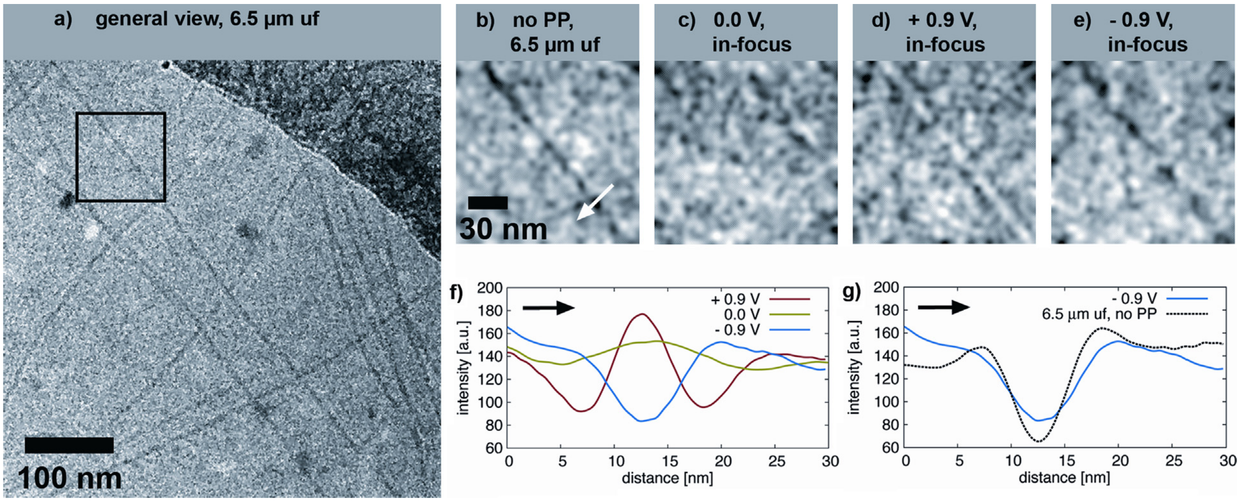

Fig. 4. (Colour online.) Imaging of a cryo-sample of filamentous actin embedded in vitreous ice with the aid of a Boersch (Zach-type) phase plate. (a). General view taken at 6.5 µm underfocus without any phase plate. The black box shows the area examined in subsequent images. (b). As (a), underfocus 6.5 µm. (c)–(e). Image contrast for three values of the voltage applied to the phase plate, 0 V, +0.9 V and −0.9 V. (f)–(g), line scans across the filament zone indicated by a white arrow in (b). In (f), we see the contrast reversal that occurs when the phase-plate voltage is switched from +0.9 V to −0.9 V. (g) shows that the phase-contrast intensity obtained with the phase plate at −0.9 V is comparable with that achievable without a phase plate at a high underfocus value (6.5 µm). Courtesy of Nicole Frindt; for full details, see [91].

et al. [91], who have succeeded in obtaining images of actin filaments. An entirely different way of introducing an electric field at the centre of the back focal plane has been proposed by Müller et al. [92]. Here, a laser beam crosses the path of the electron beam and the ensuing interaction alters the phase of the central zone.

7. Electron mirrors as correctors

The proof that the spherical aberration coefficient of round electron lenses, magnetic or electrostatic, cannot change sign rests on several conditions, one of which excludes electron mirrors. The idea of using an electron mirror to cancel the spherical aberration of an electron lens has a very long history. It appears in Scherzer's paper of 1947 [6] and a possible

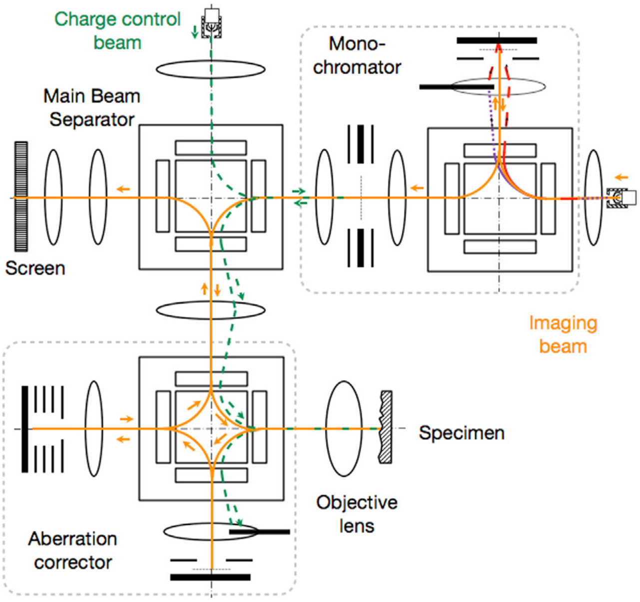

Fig. 5. (Colour online.) The MAD-LEEM column, incorporating a monochromator and an electron mirror to cancel the aberrations of the objective lens. Since the instrument is intended for use with biological and insulating specimens, a second beam is provided to eliminate charging. The beam emitted by the electron source (top right) first passes through a monochromator, after which the upper beam separator consisting of magnetic prisms directs it down to the lower beam separator. Here it is directed to the specimen via the objective (bottom right) and returns to the lower separator, where it is deflected down to a mirror, which returns it to the separator. It then enters the mirror aberration corrector (bottom left), after which it is directed up to the upper separator and finally arrives at the detector (top left). At the same time, a second beam (top centre) with a lower energy than the primary beam travels down to the specimen and prevents any accumulation of charge. This secondary beam is subsequently intercepted in the lower part of the device. After [107], courtesy of the authors and Elsevier.

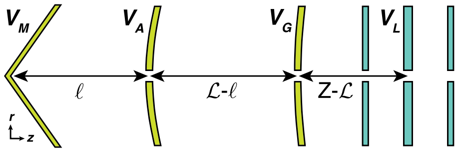

Fig. 6. (Colour online.) A triode mirror combined with an einzel lens, for use in a PEEM. Diode mirrors have the disadvantage that the spherical and chromatic aberration coefficients cannot be varied independently. This limitation vanishes if a third electrode is included, as shown here. The optics of this combination is fully described in [109]. After [109], courtesy of the authors and Elsevier.

configuration is to be found even earlier in *Electron Optics and the Electron Microscope*, published in 1945 [93]. For many years, the fundamental problem of separating the electron beam travelling towards the mirror from that reflected by the mirror found no satisfactory solution, though several configurations were suggested. The somewhat clumsy approaches to the theory of mirrors likewise hindered progress. However, a very elegant formulation of the theory of image formation in the electron mirror and the aberrations of mirrors has been developed by Kel'man, Yakushev, Bimurzaev and Sekunova (reviewed in detail in [94,95] and summarized in [31]). At last, Rempfer and colleagues in Portland (Oregon) [96–99], Crewe et al. in Chicago [100–102] and Hartel et al. [103] devised practical systems and many of the difficulties have been overcome. Recent efforts have been concentrated on low-energy electron microscopes (LEEM) and photo-emission electron microscopes (PEEM) [104–109]. Fig. 5 shows the optics of the MAD-LEEM designed by Mankos et al. [106,107]. This is an aberration-corrected, dual-beam, low-energy electron microscope, which incorporates a monochromator, a mirror corrector to compensate the aberrations of the objective lens and a second beam, the role of which is to eliminate charging when the specimen is an insulator or a poor conductor. The image-forming beam emerges from the gun (right) and its energy spread is narrowed by the monochromator. It is then deflected (down) to a second deflection unit, where it encounters the specimen and the mirror that provides aberration cancellation. It then returns (up) to the first deflection unit, where it is directed to the screen (left). At the same time, a second beam (top) wends its way down to the specimen, where it neutralises any charge deposited by the imaging beam. Fig. 6 illustrates a recent development in the work launched by Gertrude Rempfer at Portland State University. It was known that a two-electrode mirror can correct spherical and chromatic aberration simultaneously but not independently. A way of avoiding this inconvenience has been found by Fitzgerald et al. [108,109]; here a triode mirror is combined with an einzel lens, as shown in Fig. 6. This provides the desired flexibility.

8. Concluding remarks

How are the unconventional approaches to high-resolution electron microscopy likely to affect the future of microscopy? Ptychography and the use of phase plates of one kind or another will surely rival or complement aberration-corrected microscopy as they become more widespread, especially in the light of the difficulties that are being encountered at the very highest resolutions. John Rodenburg, the doyen of the ptychographers, is perplexed by the reluctance of electron microscopists to try out this technique, in routine use for synchrotron imaging. At a workshop on ptychography held in Castle Hohenkammer in May 2013, he was the only electron microscopist present. But once it has been given a fair trial, we shall know whether it deserves to take its place beside (or even ahead of) the other high-resolution techniques. The future of phase-plate-assisted microscopy is not quite so rosy. Fabrication of Zernike plates is not easy and requires a very special expertise. Boersch plates have the disadvantage that part of the back focal plane is obscured, though clever ways of reducing the effect of this have been proposed. Schröder et al. [110] even used a quadrupole to compress the current distribution into a line focus, which avoided the support structure of the phase plate. Perhaps these plates will continue to be used to image exceptionally difficult specimens without becoming part of the average microscopist's toolbox.

The news that thermal magnetic field noise imposes a limit on the performance of aberration correctors reached me as I was about to submit these lines and should certainly stimulate work on alternative ways of extracting high-resolution information from electron microscopes, such as those described here. It is ironic that it was the incorporation of a chromatic-aberration corrector into the TEAM design that revealed that correctors may themselves be sources of chromatic aberration. But after all, the resolution of about half an angstrom of the TEAM microscope with no C_c corrector is already impressive.

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