# **ABERRATION CORRECTION IN ELECTRON MICROSCOPY\***

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## Abstract

Spherical and chromatic aberrations limit the resolution of conventional electron microscopes. Both defects are unavoidable in the case of static rotationally symmetric electromagnetic fields (Scherzer's theorem). To compensate for these aberrations, multipole correctors or electron mirrors are required. The correction of the resolution-limiting aberrations is demonstrated for the hexapole corrector, the quadrupole-octopole corrector and the tetrode mirror. Electron mirrors require a magnetic beam separator, which must be free of second-order aberrations. The multipole correctors are highly symmetric telescopic systems compensating for the defects of the round lenses. The hexapole corrector has the simplest structure yet eliminates only third-order spherical aberration and coma, whereas the mirror and the quadrupole-octopole (QO) corrector are able to correct the chromatic aberration as well. The QO corrector eliminates chromatic aberration by means of crossed electric and magnetic quadrupoles and the third-order spherical aberration by octopoles. At present, aberration-corrected electron microscopes obtain a resolution limit of about 1 Angstroem.

## INTRODUCTION

The ultimate goal of high-performance electron microscopy is the acquisition of detailed information on the atomic structure, the chemical composition and the local electronic states of real objects. This knowledge will give information on correlations between the macroscopic and the microscopic properties of solid materials and biological objects. Non-periodic atomic structures in materials - such as interfaces, dislocations and other defects - can only be visualized by means of electron microscopes with atomic resolution. In order to prevent radiation damage, the energy of the electrons must stay below the knock-on threshold for atom displacement. This threshold is in the range between 100 and 300 keV for most materials. Biological objects can tolerate only very low doses regardless of the energy of the electrons, because the electronic excitations destroy the atomic bonds. Therefore, we cannot directly visualize individual atoms in biological objects regardless of the voltage and the resolution of the electron microscope.

Diffraction and spherical and chromatic aberrations limit the resolution of conventional electron microscopes to about  $d = 100\lambda$ , where  $\lambda$  denotes the wavelength of the image-forming electrons at the object plane. Hence, conventional magnetic electron lenses are about hundred times poorer than the objective lenses of light microscopes. The wavelength of the electrons at an acceleration voltage of 200 kV is roughly 25 pm giving a resolution limit of about 0.25 nm, which does not suffice to resolve individual atoms of non-periodic objects. Since we cannot significantly increase the acceleration voltage, we must eliminate or sufficiently reduce spherical and chromatic aberrations. One can largely suppress the chromatic aberration by means of (a) a monochromator, which reduces the energy spread of the incident electron beam to 0.1 eV and (b) an imaging energy filter placed between object and image plane. This filter removes the inelastically scattered electrons without affecting the path of rays of the zero-loss electrons forming the image.

Unfortunately, we cannot suppress the third-order spherical aberration by reducing the aperture because this procedure increases the diffraction effect. Therefore, we must compensate for the spherical aberration in order to achieve atomic resolution without unduly increasing the acceleration voltage. However, we cannot achieve spherical correction for rotationally symmetric systems. This property, derived by Scherzer in 1936 [1], is so important in electron optics that it is known as *Scherzer theorem*. In particular the theorem states: "Spherical and chromatic aberrations are unavoidable for static rotationally symmetric charged-particle lenses free of space charges". Mirrors do not belong to this class of lenses because they reverse the direction of flight.

The reason for the Scherzer theorem originates from the Laplace equation for the electromagnetic potentials. The solutions of this equation adopt an extremum at the boundaries, which puts a constraint on the achievable spatial distribution of the electromagnetic potentials. Since the index of refraction for charged particles depends on these potentials, the outer zones of solenoids focus the electrons always more strongly than the paraxial zone. This behavior differs from that of a multipole because its potential adopts at the boundary either a maximum or a minimum depending on the polarity of the potential at the electrode or pole piece, respectively. This is the reason why we can compensate for the spherical aberration by abandoning rotational symmetry. Scherzer [2] himself has shown that one can eliminate chromatic and spherical aberrations by lifting one or other of the constraints of his theorem. Among the various possibilities, the use of multipoles and mirrors is most suitable in practice.

The correction of both chromatic and spherical aberration becomes mandatory at voltages below 10 kV because the chromatic aberration increases rapidly with decreasing energy whereas the spherical aberration does not. Hence, in order to improve the resolution of photoemission and/or low-voltage electron microscopes, we must eliminate both aberrations. Correction of chromatic aberration is also advantageous if we want to use the inelastically scattered electrons for spectroscopic

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imaging. This imaging mode yields information about the chemical composition of the object on the sub-nanometer scale [3].

Since electron microscopes have a straight optic axis, we cannot use sextupoles for correcting chromaticity, as it is possible for systems with curved axis because the dipoles introduce dispersion. In order to retain rotational symmetry as largely as possible, we deviate from this symmetry only within a short region behind the round objective lens. The multipoles placed in this region form a corrector, which acts like correcting glasses compensating for the aberrations of the objective lens without affecting the paraxial path of the rays outside the corrector. Owing to the close similarity between the propagation of light and particles, the properties of electron microscopes and their constituent components are described most appropriately in light-optical terms, which have been established at a time when charged particles were still unknown. Employing light-optical concepts is extremely helpful for designing and calculating electron optical instruments, corrected electron microscopes in particular.

#### CHROMATIC CORRECTION

We can compensate for the chromatic aberration of systems with straight optic axis by incorporating either an electron mirror or crossed electric and magnetic quadrupoles acting as first-order Wien filters. The chromatic aberration of entirely magnetic systems is unavoidable. Owing to the limitation of the maximum electric field strength, we can achieve chromatic correction of systems with straight optic axis only for acceleration voltages smaller than about 300 kV without unduly increasing the length of the corrector.



Figure 1: Electric and magnetic forces  $\vec{F}_e, \vec{F}_m$  acting on electrons located in the vertical and in the horizontal principal section of a first-order Wien filter.

Crossed electric and magnetic quadrupoles act as firstorder Wien filters if we choose the magnetic quadrupole strength  $\Psi_2$  and the electric quadrupole strength  $\Phi_2$  in such a way that the lateral components of the Lorentz force

$$F_x \approx 2e(v\Psi - \Phi_2)x, \quad F_y \approx -2e(v\Psi_2 - \Phi_2)y$$
 (1)

are zero for electrons with nominal velocity, as illustrated in figure 1. Accordingly, electrons whose velocity deviates by  $\Delta v$  from the nominal velocity  $v = v_n$  experience a lateral force whose components

$$F_x \approx e \kappa \Phi_2 x$$
,  $F_y \approx -e \kappa \Phi_2 y$  (2)

depend linearly on the position coordinates and on the relative energy deviation  $\kappa = 2\Delta v / v_n = dE / E_n$ .

The relations (2) demonstrate that the first-order Wien filter is focusing in one direction and defocusing in the other. In order to compensate for the axial chromatic aberration of round lenses, we need to deflect the faster electrons with  $\kappa > 0$  toward the optic axis and the slower electrons away from the axis. We achieve this for the horizontal x-z section by choosing a proper positive value for the electric quadrupole strength. Unfortunately, we simultaneously double the y-component of the axial chromatic aberration if we place the filter within the rotational symmetric paraxial domain. In order to obtain chromatic correction in the vertical and horizontal sections, we employ two filters and place each at one of two orthogonal line images within the astigmatic domain of a telescopic quadrupole corrector, as depicted in figure 2. One utilizes this system as the achromatic objective lens of a low-voltage scanning electron microscope [4].



Figure 2: Correction of the axial chromatic aberration of a round lens by a quadrupole corrector consisting of two magnetic outer quadrupoles and two inner crossed electric and magnetic quadrupoles, which are placed at astigmatic line images. The inner quadrupoles are excited such that each element acts as a focusing quadrupole and a first-order Wien filter compensating for the chromatic aberration of the entire system.; dE is the deviation of the energy from the nominal energy  $E_n$ .

#### CORRECTION OF THIRD-ORDER COMA AND SPHERICAL ABERRATION

Two different approaches exist for correcting the thirdorder spherical aberration. We can nullify the spherical aberration either by sextupoles or by octopoles in combination with quadrupoles, which are necessary to produce an astigmatic path of rays within the region of the octopoles. In order to image all points of the transferred object area with the same resolution, we must also compensate for or sufficiently reduce the off-axial aberrations, especially the coma, which depends linearly on the position coordinates of an object point. Systems, which are corrected for spherical aberration and coma, are called aplanats.



Figure 3: Arrangement of the elements and path of the paraxial fundamental rays, within the telescopic sextupole corrector, f is the focal length of the round lenses

The incorporation of multipole elements largely increases the number of aberrations. In order to minimize these additional aberrations, it is advantageous to impose symmetry conditions on the fields and the paraxial trajectories. The higher the degree of symmetry is, the more aberrations cancel out. Symmetric systems have the additional advantage that they can be aligned very precisely. Correctors, which are suited for electron optical aplanats, must exhibit double symmetry. This implies that the multipole elements must be symmetric with respect to the mid-plane  $z_M$  of the corrector and with respect to the mid-plane of each half of the arrangement. Although, the two subunits are identical, their elements can be exited symmetrically or anti-symmetrically with respect to these planes (Planes are perpendicular to the optic axis, while sections are surfaces, which embed the optic axis.).

The sextupole corrector has the smallest number of elements [5]. It consists of a telescopic round-lens doublet and two sextupoles one placed at the front focal plane of

the first lens and the other at the back focal plane of the second lens, as shown in figure 3. These focal planes represent the nodal planes  $N_1$  and  $N_2$  of the transfer doublet, which images the first sextupole with magnification M = -1 onto the second sextupole. As a result all second-order path deviations introduced by the first sextupole cancel out. The remaining secondary aberrations of the sextupoles are of third order and of exactly the same nature as those of round lenses. However, the sign of the spherical aberration of the sextupole corrector is opposite to that of round lenses. Since the sextupoles do not affect the paraxial trajectories, the corrector only introduces a divergent third-order path deviation, as illustrated in figure 4. Accordingly, we can use the corrector as "glasses", which counterbalance the too strong refraction power of the outer zones of the rotationally symmetric objective lens. The third-order spherical aberration introduced by the hexapole corrector is proportional to the square of the sextupole strength, which we can adjust to compensate for that of the round lenses.



Figure 4. Action of the sextuple corrector on the non-Gaussian trajectories of particles moving initially on the mantle of a tube centered about the axis. The second-order path deviations cancel behind the corrector, while the asymptotes of the third-order deviations are located on a diverging cone.

The nodal planes of the corrector coincide with its coma-free planes. In order to form an aplanat, we must match the front coma-free point  $N_1$  with the corresponding point of the objective lens. Unfortunately, this is not directly possible because the coma-free point of the round lens lies within the lens field and  $N_1$  is located at the center of the first sextupole. However, we match these points optically by means of another telescopic transfer doublet if we place its front nodal point  $N_0$  in the coma-free point of the objective lens and its back nodal point at the point  $N_1$  of the corrector, as depicted in figure 5. Electron microscopes equipped with this corrector yield routinely a resolution limit of about 1.2 Angstroem at 200 kV determined by the chromatic aberration [6]. This resolution limit decreases to about 0.7

Å if we reduce the energy width of the incident beam by means of a monochromator to 0.1 eV [7].



Figure 5: Coma-free arrangement of the sextupole corrector and the objective lens by means of a telescopic transfer doublet resulting in an electron optical aplanat.

We must employ quadrupoles and octopoles in order to construct an achromatic aplanat. Since the QO corrector compensates for the unavoidable aberrations of round lenses, its compensating aberrations must have rotational symmetry and be opposite to those of the round lenses. However, QO correctors also introduce aberrations with twofold and fourfold azimuthal symmetry. By employing a doubly symmetric corrector, we can excite the multipoles in such a way that all aberrations with twofold symmetry cancel out. Correctors, which meet these conditions, consist of at least 12 quadrupoles. We need three octopoles to compensate for the spherical aberration and for the fourfold axial astigmatism introduced by the quadrupoles. In order that the octopoles, which correct the third-order aberrations, do not introduce appreciable field aberrations limiting the field of view, we must place these elements at images of the coma-free-plane of the objective lens. These planes are defined by the mutual intersection of the field rays  $x_{\gamma}$  and  $y_{\delta}$  with the optic axis, as shown in figure 6 for the doubly symmetric QO corrector. This corrector consists of two telescopic quadrupole sextuplets, each formed by two identical symmetric quadrupole triplets. One separates the sextuplets such that the axial rays  $x_{\alpha}$  and  $y_{\beta}$  intersect the optic axis at the mid-plane  $z_M$  of the corrector. We place an octopole at the midplane of each subunit, where a strongly first-order distorted image of the point at infinity is formed. We focus this point by means of the adapter lens 1 located in front of the corrector into the coma-free point of the objective lens, as illustrated in figure 7. By exciting the two octopoles O1 with the same polarity and the quadrupoles of the second sextuplet with opposite polarity with respect to those of the first sextuplet, we cancel out all aberrations with twofold symmetry. Owing to the antisymmetric excitation of the quadrupoles, the course of the rays  $x_{\alpha}, x_{\gamma}$  in the horizontal x-z section of the first sextuplet coincides with that of the corresponding rays  $y_{\beta}, y_{\delta}$  in the vertical *y*-*z* section.



Figure 6: Arrangement of the elements and path of the fundamental paraxial rays within the doubly symmetric QO corrector

Due to the symmetries of the rays and the multipole fields with respect to the central plane of each sextuplet, none of the two subunits introduces coma and third-order distortions. To achieve these symmetries for the trajectories, we image the coma-free plane of the objective lens into the central plane of the sextuplet and the object plane into the front nodal plane  $N_1$  by means of the adaptor lens 1.



Figure 7: Arrangement of the elements and course of the fundamental rays of an achromatic aplanat employing a QO corrector compensating for the spherical and chromatic aberrations, the octopole  $O_2$  eliminates the fourfold axial astigmatism introduced by the quadrupoles

Since the objective lens and each sextuplet are free of coma, the fifth-order axial combination aberrations, which result from the concatenation of third-order coma with the axial aberration of different elements of the corrected system, are very small. The same holds true for the image curvature and the field astigmatism because we have placed the octopoles at positions where the field rays vanish. The adaptor lens 2 eliminates the coma of the first adapter lens 1. The two lenses form a telescopic transfer doublet because the front nodal point  $N_1$  of the telescopic corrector coincides with the back focal point of the first

lens and the back nodal point  $N_2$  with the front focal point of the second lens. Hence, the insertion of the corrector does not affect the primary aberrations of the transfer doublet

In order that the corrector also eliminates the chromatic aberration, we substitute crossed electric and magnetic quadrupoles for the quadrupoles adjacent to the octopoles **O1**. We excite these elements in such a way that they act as focusing quadrupoles **Q1** and as first-order Wien filters compensating for the axial chromatic aberration without introducing chromatic distortions. Then the entire system represents an achromatic aplanat. This system will serve as the objective compound lens of a sub-Angstroem analytical transmission electron microscope, which is presently under construction.

## MIRROR CORRECTOR

Electron mirrors are capable of introducing chromatic and spherical aberrations of arbitrary sign. Hence, we can utilize mirrors to compensate for the corresponding aberrations of round lenses. Although the mirror has rotational symmetry, we need a non-rotationally symmetric beam splitter to separate the incident electrons from the electrons reflected by the mirror. In order that the beam separator does not affect the resolution, it must be free of second-order aberrations [8].



(b) correction of spherical aberration



Figure 8: Path of rays illustrating chromatic and spherical correction by the mirror,  $v_n = 0$  at the potential  $\varphi_n$ 

Unlike a light-optical mirror, where the reflection occurs at the physical surface, the electron mirror represents a "soft" mirror, which allows the electrons to penetrate into the inhomogeneous refracting medium formed by the electrostatic potential. The depth of penetration depends on the energy and the direction of the electron in front of the mirror. We can conceive the total reflection as the sum of consecutive refractions on a continuous sent of equipotentials  $\varphi = const$  (Figs. 8, 9). In order that the mirror does not introduce field aberrations, we must image the back focal plane of the objective lens into the turning plane of the axial electron with nominal energy and place an image of the object plane at the focal plane of the mirror. In order to adjust the location of the focal plane, the chromatic aberration and the spherical aberration of the mirror, we need three free parameters, which are the voltages applied to the electrodes of a tetrode mirror [8, 9]. We cannot vary the potential of the fourth electrode because it must be at column potential.



Figure 9: Electrodes and equipotential lines of the mirror

Mirrors, which are suited for compensating the chromatic and spherical aberrations of round lenses, must be concave in the paraxial region and convex in the outer zones, as depicted in figure 8. We achieve this property by shaping the electrodes appropriately. Since the electrons stay a relatively long time in the domain of their turning points, the form of the equipotential lines in this region strongly affects the path of the reflected electrons.

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