Chromatic correction: a revolution in electron microscopy?

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When the development of correctors started in the 1970s, chromatic correction was already the main goal. The first corrector that could improve the resolution of an electron microscope was a chromatic corrector for a scanning electron microscope.

Within the last three decades, the development of transmission electron microscopes (TEMs) was to a large extent driven by the attempt to improve the resolution in the presence of chromatic aberration. The major technical developments were high acceleration voltages, highly excited objective lenses with short focal length and field emission guns.

Meanwhile, chromatic correction has reached the TEM world. Now, the question arises as to whether chromatic correction will make some of the aforementioned developments obsolete for high-resolution TEM, thereby opening up new imaging possibilities, which are nowadays prevented by instrument constraints. We show some examples for a 0.1 nm resolution TEM with unconventional microscope designs: very low voltages, far-field objective lenses and inexpensive electron guns.

Keywords: chromatic correction; atomic resolution; electron microscope design

1. Electron microscopy without correctors

From the beginning, in the 1930s, the development of the electron microscope was driven by the idea that the diffraction limit of light optics could be circumvented by using electrons for imaging. Owing to their short wavelength of only a few picometres, the resolution of single atoms in a solid-state body should be possible.

Unfortunately, it became clear that electron lenses have very poor optical quality. Therefore, the imaging aperture has to be reduced to such an extent that the diffraction limit again prevents atomic resolution.

Mainly two axial aberrations limit the resolution of the electron microscope: spherical and chromatic aberrations. Their influence on the achievable resolution can be estimated as

\[ d_s = B \sqrt[4]{\lambda^3} C_s \]  \hspace{1cm} (1.1)

for the spherically and

\[ d_c = A \sqrt{\lambda \frac{\Delta E}{E}} C_c \]  \hspace{1cm} (1.2)

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Figure 1. Achievable resolution in spherical aberration limited microscopes according to equation (1.1).

for the chromatically limited resolution. $A$ and $B$ are the numerical values of the order of 1, $\lambda$ the electron wavelength, $E$ and $\Delta E$ the mean energy and energy width of the beam, respectively, and $C_c$ and $C_s$ the chromatic and spherical aberration coefficients.

These equations were steering the development of electron microscopes for seven decades. The attempt to approach atomic resolution as far as possible led to instrumental improvements that were targeted at manipulating the free parameters in the equations. We will illustrate this strategy and its consequences with the help of figures 1 and 2.

In the case of a spherical aberration-limited system, the only parameter to optimize is the aberration coefficient. As a rule of thumb, the coefficient has at least the same order of magnitude as the focal length. If the specimen is outside the magnetic field of the objective lens, $C_s$ is significantly larger than the focal length, but if the specimen is immersed in the field, less than half the focal length can be reached. However, as the focal length cannot be made much smaller than 1 mm owing to mechanical constraints, $C_s$ can hardly be smaller than half a millimetre. In this case, figure 1 shows that true atomic resolution, e.g. point resolution of approximately 0.1 nm, can be achieved only at very high beam energies. So, the strategy is clear: build very strong objective lenses and increase the beam voltage.

The microscope manufacturers followed this strategy for a long time. The results were the giant megavolt microscopes of the last century and the big objective lenses with minimum free space for the specimen, which are still used in modern instruments.

In a chromatically limited instrument, the characteristic quantity to optimize is the product $Q = dU \ast C_c$, where $dU = \Delta E/e$ is the energy width of the beam divided by the electron charge.
Chromatic correction

The chromatic aberration coefficient has about the same magnitude as the focal length. So, the same lower limit as for the spherical aberration coefficient is valid, resulting in a minimum value of approximately 1 mm. Additionally, the chromatic resolution limit can be reduced by reducing the energy width of the electron source. This led to the development of LaB$_6$, Schottky and cold field emission sources. With these guns, the quality factor $Q$ could be reduced to approximately 0.5 V mm. Therefore, atomic resolution would be possible already at 200 kV according to figure 2, if no spherical aberration were present. The drawback to this improvement is the rather complicated vacuum system for generating the ultra-high vacuum that is required by these sources.

2. Electron microscopy without chromatic and spherical aberration

Correctors for spherical aberration have been available now for about 2 years and have, meanwhile, become a standard feature in high-end transmission electron microscopes (TEMs) and scanning transmission electron microscopes.

Although Scherzer (1947) had already shown that chromatic and spherical aberration can be corrected in non-rotationally symmetric systems, all efforts were first concentrated on the correction of spherical aberration (Seeliger 1951; Archard 1954). In the 1960s electric–magnetic quadrupoles were investigated theoretically (Kelman & Yavor 1961) and experimentally (Hardy 1967). It became clear that they were the key optical elements for chromatic correction. Then, running for more than 10 years, the Darmstadt corrector project proved the principal chromatic and spherical correction capabilities of a quadrupole–octupole...
corrector, as published in numerous papers. See Rose (2008) for a historical overview of this project. The first chromatic corrector which could improve the resolution of a microscope was built by Zach & Haider (1995).

In October 2008, the first chromatic and spherical aberration corrector for a TEM within the Transmission Electron Aberration-Corrected Microscope (TEAM) project of the US Department of Energy Office of Science was shipped to Argonne National Laboratory. One target of this project is to provide aberration-free imaging (AFI) at a resolution of 0.05 nm. Here AFI means that the aberrations of the microscope should not produce a disturbing phase shift of the electron wave. The limit is defined as follows: the sum of all aberrations of a certain annular multiplicity should not generate a phase shift of more than $\pi/4$ over the entire spatial frequency range that is required for the target resolution. We will adopt this concept of AFI for further discussion within this article.

Since spherical and chromatic correctors are available now, the question arises as to whether the microscope concepts that were developed while aberrations were unavoidable are still appropriate. We will discuss this question on the basis of a few examples in the following sections for a model TEM set-up.

(a) The model corrector in high-resolution mode

For discussion, we will use a model spherical and chromatic corrector set-up. The model corrector differs in various aspects from the corrector, which was built for the TEAM project. This corrector model has been chosen because it seems to be slightly more flexible for the range of microscope set-ups discussed subsequently. However, it should be understood that the basic conclusions of this article do not depend very much on the specific corrector model.

The basic set-up of the corrector is depicted in figure 3. The set-up is mirror symmetric with respect to a central plane to reduce off-axial aberrations. If only the first half were used as a corrector, the number of image points would typically be less than 250, whereas it will be approximately 1800 or more for the symmetric set-up. The corrector has four correction pieces for chromatic and spherical corrections. They consist of a multipole element with superimposed electric–magnetic quadrupole fields, octupole and twelve-pole fields. Four pure quadrupoles control the astigmatic beam path, and a central multipole element with an octupole and twelve-pole element corrects for the internal four- and sixfold astigmatisms. Note that the grey areas just indicate the axial position of the multipole fields. Their amplitudes have arbitrary units.

Generally, the excitations for this corrector type can be kept quite low. Even at 200 kV the magnetic quadrupoles have an excitation below 25 Ampturns and the twelve-poles below 4 Ampturns, although the ratio of bore-to-beam diameter is between 7 and 14. This high ratio ensures that the corrector is not too sensitive to field instabilities.

The beam path shows four astigmatic images in the correction pieces. Thus, the corrector uses the correction principles of a first-order Wien filter together with octupole correction as was used in the first successful spherio-chromatic corrector for an SEM (Zach & Haider 1995).

If we were to incorporate this corrector in the TEAM instrument as a replacement for the current TEAM corrector, we would get the phase shifts due to the residual aberrations listed in table 1. Of course, for chromatic aberrations,
Figure 3. Basic set-up of the model corrector showing its multipole elements and the stigmatic beam path.

Table 1. Phase shifts due to residual aberrations at 200 kV high-resolution mode. The phase shift is given for a spatial frequency limit of 20 nm$^{-1}$.

<table>
<thead>
<tr>
<th>type of aberration</th>
<th>symbol</th>
<th>phase shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>spherical aberration of seventh order</td>
<td>$C_7$</td>
<td>0.0654</td>
</tr>
<tr>
<td>chromatic aberration of third order</td>
<td>$C_{3k}$</td>
<td>0.0163</td>
</tr>
<tr>
<td>chromatic star aberration of third order</td>
<td>$S_{3k}$</td>
<td>0.0261</td>
</tr>
</tbody>
</table>

this is the amplitude of a phase spread rather than a static phase shift. The phase shift or spread is in units of $\pi$. Only those aberrations that contribute more than a phase shift of $\pi/100$ are listed. Obviously, this set-up fulfils the AFI criteria for a target resolution of 0.05 nm.

A detailed list of the aberration notation used in this paper can be found in Müller et al. (2006). A different notation for aberrations used in several other papers is described in Krivanek et al. (1999).

(b) Low energies

In a corrected microscope, there is no longer any need to use high beam energies to achieve atomic resolution. But how far can we reduce the energy and still see single atoms? As in the introductory discussion, we will assume that a resolution of 0.1 nm is sufficient for atomic resolution. We will now scale
Table 2. Phase shifts due to residual aberrations at 30 kV. The phase shift is given for a spatial frequency limit of 10 nm\(^{-1}\).

<table>
<thead>
<tr>
<th>type of aberration</th>
<th>symbol</th>
<th>phase shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>spherical aberration of seventh order</td>
<td>(C_7)</td>
<td>0.224</td>
</tr>
<tr>
<td>star aberration of seventh order</td>
<td>(S_7)</td>
<td>0.443</td>
</tr>
<tr>
<td>Rosette aberration of seventh order</td>
<td>(R_7)</td>
<td>0.060</td>
</tr>
<tr>
<td>eightfold astigmatism</td>
<td>(A_7)</td>
<td>0.114</td>
</tr>
<tr>
<td>chromatic aberration of third order</td>
<td>(C_{3k})</td>
<td>0.113</td>
</tr>
<tr>
<td>chromatic star aberration of third order</td>
<td>(S_{3k})</td>
<td>0.230</td>
</tr>
<tr>
<td>chromatic fourfold astigmatism</td>
<td>(A_{3k})</td>
<td>0.022</td>
</tr>
</tbody>
</table>

Table 3. Phase shifts due to residual aberrations at 100 kV assuming a LaB\(_6\) source. The phase shift is given for a spatial frequency limit of 10 nm\(^{-1}\).

<table>
<thead>
<tr>
<th>type of aberration</th>
<th>symbol</th>
<th>phase shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>chromatic aberration of third order</td>
<td>(C_{3k})</td>
<td>0.0182</td>
</tr>
</tbody>
</table>

down the high-resolution system described in §2a to 30 kV and investigate its properties for a target resolution of 0.1 nm. Table 2 shows the residual phase shifts. All individual aberrations already fulfil the AFI criterion, except for the star aberration of the seventh order. However, its phase shift can easily be reduced by a factor of 2 or more by counterbalancing it with another aberration of multiplicity 2, like the well-known twofold astigmatism. This is in full agreement with the AFI specifications. So, even at 30 kV, the chromatically corrected system provides AFI at 0.1 nm.

c) A simple standard transmission electron microscope

For a long time, the standard TEM has been an instrument that runs at an acceleration voltage around 100 kV and has a comparatively simple gun such as the LaB\(_6\) source. One could not expect higher performance from these instruments, but they served as the standard work horse.

We have assumed an energy width of 1 eV for this type of source. Table 3 lists the phase shifts for this case. There is just one entry above a limit of \(\pi/100\). So we can expect a resolution clearly below 0.1 nm. Table 3 also indicates that atomic resolution in the AFI sense would still be possible with an energy width of the beam that is about 14 times higher than the assumed 1 eV of the LaB\(_6\) gun.

Together with a cost-effective chromatic corrector, the standard type instruments could be pushed to the atomic resolution level. This might in the future provide something like ‘atomic resolution for everyone’, opening up a wide range of research capabilities to scientists, who have difficulties accessing such a quality of information nowadays.

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For a long time, one could not expect reasonable resolution in a TEM without a heavily excited objective lens. The specimen had to be squeezed into a tiny gap between the two objective lens polepieces and had thereby to be immersed into a very high magnetic field.

We have investigated the possibilities of using small magnetic lenses with low excitation as objective lenses. The specimen was placed outside the magnetic field. Such lenses are standard parts, which are used in several places within correctors or condenser and projector arrangements. They have a comparatively simple mechanical design and can be produced very cost-effectively.

Figure 4 shows the arrangement of the model corrector in a system with such a lens as an objective lens. Owing to the small size of this lens, there is no need for a transfer lens system between corrector and objective lens. Such a transfer lens system is typically used in corrected systems to image the coma-free point of the objective lens, which is located near the maximum of the imaging field, properly into the corrector. This is required because the bulky design of traditional objective lenses prevents them from bringing the corrector close enough to this coma-free point. For the small far-field objective lens, the corrector can easily be placed in its proper position without interfering with the magnetic circuit of the lens. This results in a very compact arrangement, as can be seen from the figure: the entire assembly of the corrector and objective lens has a size of only approximately 35 cm and is therefore not much bigger than a standard objective lens alone.

In this case, a resolution of 0.1 nm is difficult to achieve in practice, because it would require detailed counterbalancing of various aberrations of the seventh order by aberrations of a lower order. However, already for a target resolution of...
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Table 4. Phase shifts due to residual aberrations for a far-field OL at 100 kV. The phase shift is given for a spatial frequency limit of 9 nm$^{-1}$.

<table>
<thead>
<tr>
<th>type of aberration</th>
<th>symbol</th>
<th>phase shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>spherical aberration of seventh order</td>
<td>$C_7$</td>
<td>0.362</td>
</tr>
<tr>
<td>pentacle aberration of seventh order</td>
<td>$G_7$</td>
<td>0.042</td>
</tr>
<tr>
<td>eightfold astigmatism</td>
<td>$A_7$</td>
<td>0.255</td>
</tr>
<tr>
<td>chromatic aberration of third order</td>
<td>$C_{3k}$</td>
<td>0.053</td>
</tr>
<tr>
<td>chromatic star aberration of third order</td>
<td>$S_{3k}$</td>
<td>0.241</td>
</tr>
<tr>
<td>chromatic fourfold astigmatism</td>
<td>$A_{3k}$</td>
<td>0.024</td>
</tr>
</tbody>
</table>

0.11 nm, the residual phase shifts reduce to the values listed in Table 4. Again, the AFI criteria are fulfilled. Only the spherical aberration of the seventh order needs some small counterbalancing with ordinary defocus.

This arrangement represents the most extreme deviation from the present high-resolution development:

(i) only 100 kV,
(ii) a LaB$_6$ gun, and
(iii) a cheap and simple objective lens.

Nevertheless, this strange microscope concept can provide atomic resolution.

Of course, there are also some drawbacks to this concept: owing to the large focal length of the objective lens, the stability requirements become quite severe. The relative stability of the objective lens current has to be $10^{-7}$, and one of the quadrupoles within the correction pieces has to be as low as $2.5 \times 10^{-8}$. However, the requirements for the quadrupoles are of the same order as those that were fulfilled for the TEAM corrector, and the requirements for the lens current seem to be in the range of typical high-quality objective lens power supplies. The large focal length also has the consequence that the magnification until the first intermediate image behind the corrector is rather small. Therefore, the projection system has to provide a large overall magnification, which may require an additional lens there.

3. Conclusion

The development of electron microscopes was driven for a long time by the attempt to suppress the effects of aberrations, especially the chromatic aberration. As chromatic correctors are now available, a new degree of freedom in the design of electron microscopes has been reached. Many of the solutions that had been found in the past may now be relinquished without affecting the resolution of the microscope.

We have discussed some possible microscope designs here, which all provide atomic resolution but do not use some or all of the components and which were mandatory for high resolution until now. Other combinations are possible, thus allowing the design of application-specific instruments as well as general purpose microscopes for a wide community of electron microscopists.

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It should be noted that already today the microscope manufacturers offer very cost-effective general purpose microscopes, which give a lattice resolution of the order of 0.14 nm. Since some fraction of this limitation has to be attributed to chromatic aberration and lateral incoherence, the basic stability of the instruments with respect to stage, deflectors and stray field sensitivity seems to be very close to the target of 0.1 nm, which was used as the limit of ‘atomic resolution’ within this paper. The stability requirements for the corrector supplies are quite relaxed for this target compared with 0.05 nm at the 200–300 kV target of the TEAM project. Relative stabilities slightly better than $10^{-7}$ are sufficient in many cases. Therefore, adding a corrector to such microscopes can provide atomic resolution without needing too many improvements to the microscope.

If such proposals are adopted by the microscope and corrector manufacturers, we may have atomic resolution for everyone in the near future.

References


