Special Contributions

Development of Aberration Correctors for the HD-2700, the HF3300S, the 1.2 MV FIRST Program, and Future Prospects

Prof. Dr. Max. Haider
Dr. Heiko Müller

EDITOR'S SUMMARY: For electron microscopes, the spherical aberration of the electron lenses has long been an obstacle in the way of improving resolution. Finally, in the mid-1990s, a system for correcting spherical aberration was developed. However, there were many technical challenges for implementing it in a high voltage STEM/TEM. In coope-ration with CEOS GmbH which had been developing a spherical aberration corrector (Cs-corrector) for practical use, Hitachi, Ltd. and Hitachi High-Technologies Corpo-ration developed a 200 kV STEM and a 300 kV TEM that implement spherical aberration correctors, and achieved a significant improvement in resolution. In addition, the companies succeeded in incorporating a Cs-corrector into a 1.2 MV atomic-resolution holography electron microscope. This made a great contribution to achieving world-leading resolution.

In the development of these instruments, in addition to the development of spherical aberration correctors that support high-voltage and ultra-high-voltage, the electron microscope itself required a significant improvement in stability. Through close cooperation, the companies have solved these technical issues one by one. In this paper, Prof. Dr. Max. Haider, who founded CEOS and has led the development of spherical aberration correctors, summarizes the development CEOS carried out in cooperation with Hitachi, Ltd. and Hitachi High-Technologies Corporation.

INTRODUCTION

THE successful implementation of a Cs-corrector first for a high resolution 200-kV transmission electron microscope (TEM)(1) and later on also for the scanning transmission electron microscope (STEM) has stimulated the development of a new generation of high-resolution TEM and STEM instruments. With the research prototypes of the Cs-correctors it became quite clear that the previously existing microscopes were simply not prepared to provide sufficient stability in electronics and mechanics to allow for routine atomic-resolution imaging. In this paper we summarize the efforts and development we carried out in cooperation with Hitachi, Ltd. and Hitachi High-Technologies Corporation in order to provide novel aberration-corrected instruments. After first discussions at the Microscopy & Microanalysis meeting 2003 in Quebec/Canada we initially concentrated on the development of a hexapole-type probe corrector for Hitachi's dedicated STEM HD-2700. Almost thirty systems of this type have been installed since then. Later, with the availability of Hitachi's new 300-kV high-resolution (S)TEM HF3300S, equipped with a cold field-emission gun (C-FEG), we continued with the development of a three-hexapole imaging-corrector with large field of view and dedicated capabilities for field-free (Lorentz) imaging. Just recently we supplied an imaging corrector for the 1.2 MV atomic-resolution holography electron microscope within the framework of Tonomura's FIRST program. Together with Hitachi and Hitachi High-Technologies our next step now is to equip the HF3300S instrument with an advanced hexapole-type probe corrector in order to fully exploit the high-resolution and analytic capabilities of this 300-kV C-FEG instrument.

PROBE CORRECTOR FOR A 200-kV DEDICATED STEM

The hexapole-type Cs-corrector as proposed by Rose as a theoretical concept(2) at first has been successfully

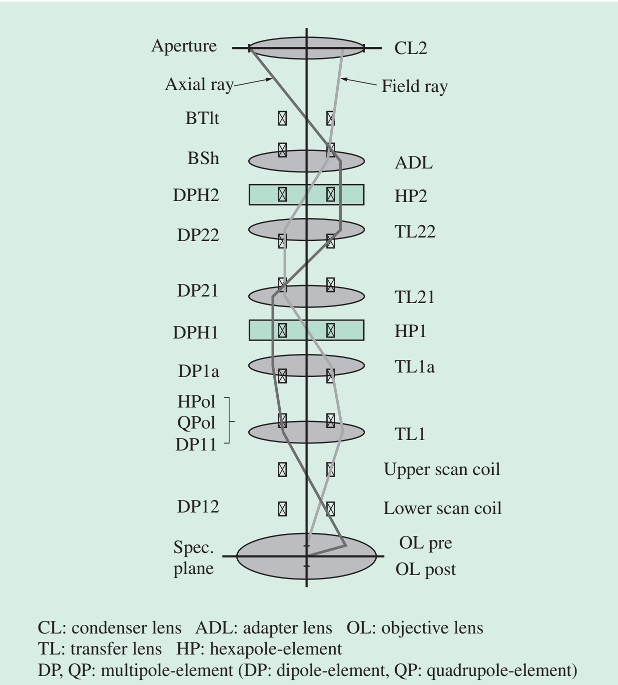

Fig. 1—Schematic Drawing of a Hexapole-corrector for STEM.
This corrector has two transfer lenses (TL1, TL1a) between the objective lens (OL) and the first hexapole-element (HP1) and a 2nd transfer-lens doublet (TL21, TL22) between the two hexapole-elements (HP1, HP2). With additional weak multipole-elements (DP, QP) the required precise ray-path and focus planes can be aligned. The dark gray ray shows the axial ray and the light gray ray the field ray.

employed as an imaging corrector in a 200-kV TEM[1]. This corrector (see Fig. 1) is based on two strong hexapole elements which generate magnetic fields with three-fold symmetry. The first hexapole field produces a strong threefold astigmatism which has to be compensated by the second hexapole field. This can be achieved by placing two round lenses in between the two hexapole elements. Additionally, the correction unit must be matched optically with the objective lens in a proper way. This again is done by additional round lenses between the objective lens and the correction unit. The production and compensation of the strong three-fold astigmatism by long hexapole elements placed at two optically conjugated planes generates - as a secondary effect - a negative third-order spherical aberration. This effect is used to compensate for the spherical aberration of the objective lens. A very similar design can be used in a probe-forming system too. Fig. 1 shows a sketch of a hexapole-type Cs-corrector for STEM. The simplicity of this design is the basis of its success over the last few years. The primary benefit of Cs-correction in TEM is to enable proper phase-contrast without artifacts from spatial delocalisation or phase reversal up to the information limit of the instrument. For STEM the most important parameter is the usable aperture size and the available total current in the probe-forming system. A Cs-corrector allows the probe semi-angle to be greatly increased while the probe size is maintained or even reduced. In this case resolution is not limited by diffraction anymore but by the brightness of the electron source. Achievable probe profiles and probe diameters are illustrated in Fig. 2. Besides the brightness, the energy width of the gun is also an important parameter for

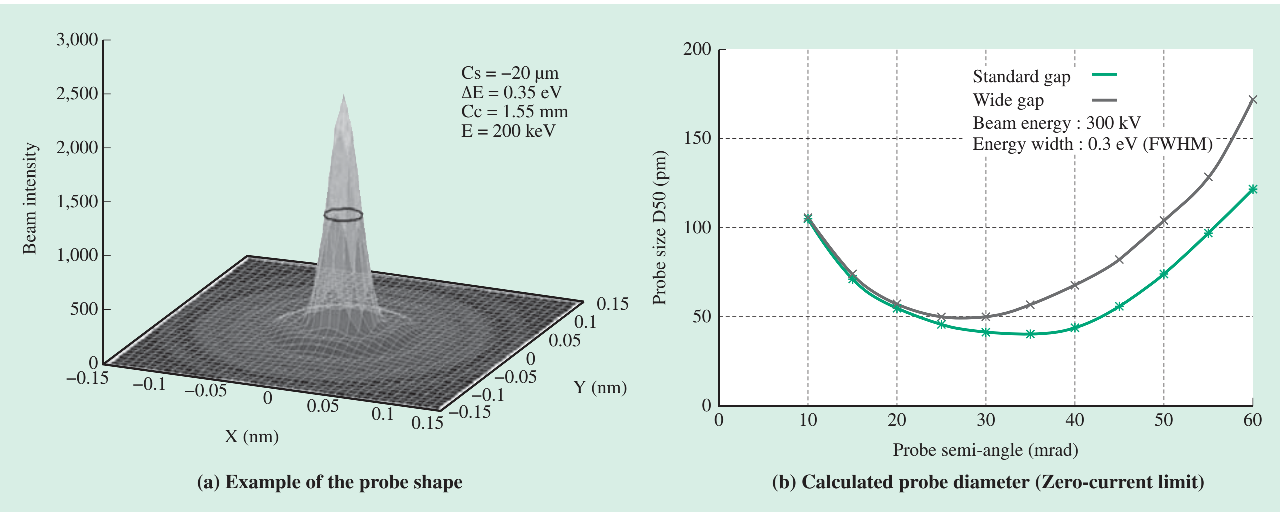

Fig. 2—Example of the Probe Shape and Calculated Probe Diameter.
In (a), as an example, the probe shape is given for a 200 kV electron probe. The small ring indicates the FWHM. On (b), the calculated probe diameter is given for various illumination angles. One easily can observe the influence of the chromatic aberration and the energy width of the electron probe on the probe diameter.

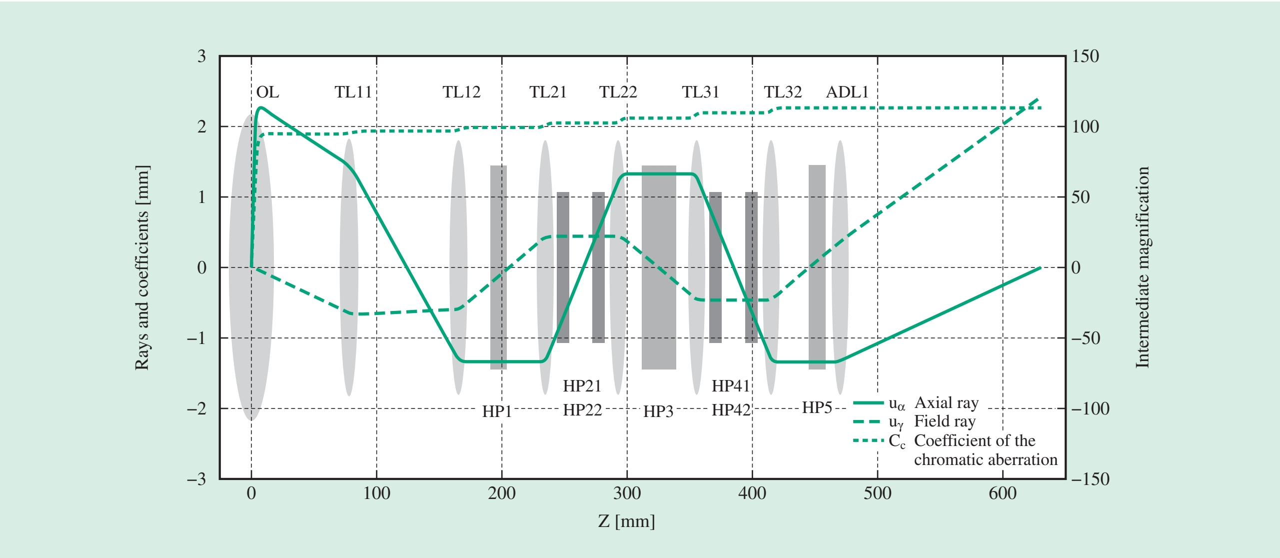

Fig. 3—Beam Path for the B-COR Design from the Objective Lens (OL) to the First Intermediate Image Plane below the Corrector.
The axial ray uα, the field ray uγ, and the coefficient of the chromatic aberration Cc are plotted. The light-gray boxes indicate the strong hexapole triplet HP1/HP3/HP5 and the dark-gray boxes show the position of the anti-symmetric weak hexapole doublets HP21/HP22 and HP41/HP42. Additionally, all seven transfer lenses are shown. The given intermediate magnification value shows the increase of the field of view with respect to the object plane at various intermediate planes within the correction system.

the sharpness of the electron probe especially when working at energies below 80 kV. Therefore, Hitachi's HD-2700 equipped with a C-FEG that has a small intrinsic energy width provides a good platform for sub-Angstrom resolution with analytic capabilities[3]. Even by detecting the signal from secondary electrons (SE), atomic resolution could be demonstrated[4].

APLANATIC IMAGING CORRECTOR FOR A 300-kV TEM

A TEM equipped with a two-hexapole imaging Cs-corrector by design is semi-aplanatic. For a fully aplanatic system, the remaining off-axial aberrations of the objective lens also have to be compensated and the parasitic off-axial aberrations must be controlled. This can increase the number of equally-well resolved image points from a few hundred to some thousand image points. The most critical off-axial aberrations are the off-axial coma and the variation of the two-fold astigmatism across the field of view. A three-hexapole design can be used to compensate for all axial aberrations up to fifth order including the six-fold astigmatism and for all off-axial aberrations up to third order[5], [6]. A schematic drawing of such an aplanatic corrector is depicted in Fig. 3. This novel design has been implemented successfully as an imaging corrector for the HF3300S TEM. It allows for aberration-free imaging for an effective imaging aperture of 30 mrad or larger (see Fig. 4). At 300 kV an information limit better than 70 pm could be demonstrated[7]. As an important feature, the corrector allows easy tuning of the relevant axial aberrations and the lower-order off-axial aberrations including the azimuthal off-axial coma. Fig. 5 illustrates the effect of parasitic off-axial astigmatism before and after correction. This improvement helps to guarantee

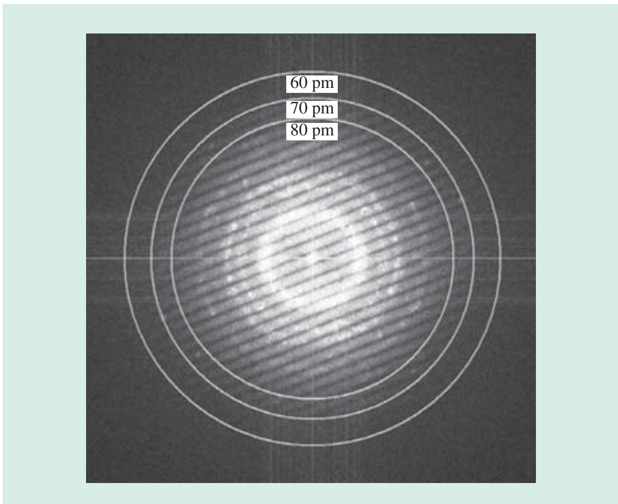

Fig. 4—Central Region of a Young's Fringe Pattern from a Thin Tungsten Specimen.
A Young's fringe pattern is recorded at 300 kV with 4 s illumination time. The Nyquist frequency in the original image is 38.3 nm^{-1}. The fringe pattern spreads outside of 70 pm.

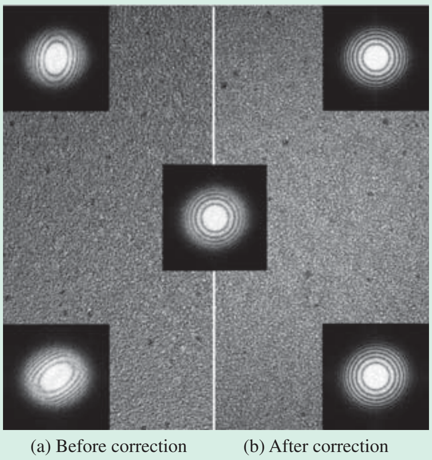

Fig. 5—Comparison Before/After Off-axial 2-fold Astigmatism A_{1G} Correction.
An amorphous Tungsten foil has been imaged at 60 kV with a field of view of 70 nm. The left side (a) shows the residual phase shift due to an off-axial 2-fold astigmatism in the diffractograms at the left corners and at the right hand side (b) the compensation of this off-axial astigmatism is demonstrated by the diffractograms at these positions. The phase shift by this aberration is much smaller and almost not visible by eye.

a large field of view for aberration-free imaging even with a 4k x 4k or 8k x 8k camera. This feature makes the system particularly attractive for holography and large field-of-view applications. Optionally, for the HF3300S, the corrector can be used for Cs-corrected field-free (Lorentz) imaging with the specimen placed at the upper stage or at the objective lens stage position. An example is given in Fig. 6 which was provided by the group of E. Snoeck at CEMES/Toulouse. All holography options of the HF3300S are supported by the corrector too.

ADVANCED PROBE CORRECTOR DCOR

The Hitachi Cold-FEG provides a high spectral brightness. In STEM this allows for a large probe semi-angle since the effects from the chromatic aberration of the probe-forming system are reduced. With the classical design of the hexapole corrector in this case, limitations have to be expected due to its intrinsic six-fold astigmatism and uncompensated parasitic fourth-order aberrations. This provided the motivation for an advanced design of the hexapole-type probe corrector. By incremental changes, we could eliminate the effects of the six-fold astigmatism and add means to correct for all parasitic fourth-order axial aberrations. This has been achieved by using an optimum length and excitation of the two hexapole elements and by exploiting a combination aberration of the hexapole fields with the transfer lenses in between[8]. Just recently, this design has been implemented for Hitachi's HF3300S[9]. The aim is to allow for considerably larger probe semi-angles and accordingly larger probe currents for analytic work at atomic resolution. The system is designed to enable deep sub-Ångstrøm resolution with wide-gap type pole pieces and Cs-corrected STEM imaging with the specimen placed at the upper stage in field-free (Lorentz) mode. Both are very attractive for modern in-situ and lab-in-the-gap applications. This probe corrector also helps to improve the quality of the illumination system in TEM and fully supports holography with a double bi-prism in the condenser system for split-beam illumination.

IMAGING Cs-CORRECTOR FOR A 1.2 MV HOLOGRAPHY ELECTRON MICROSCOPE

The late Akira Tonomura, who passed away much too early in 2012, initiated the FIRST program for which he received funding from the Japanese government. His idea was to combine the world's most advanced techniques to achieve an unprecedented resolution in transmission electron microscopy. However, the highest resolution was not only meant to resolve the

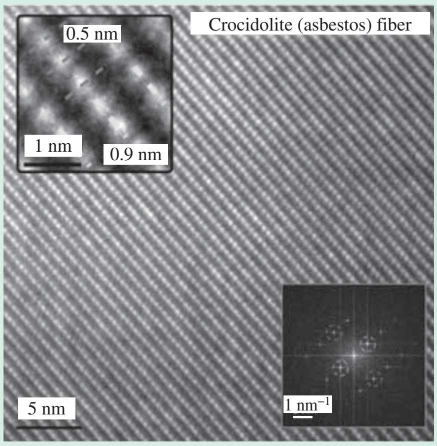

Fig. 6—Imaging of Crocidolite with this Object in a Field Free Area (Lorentz Mode).
The small insets show an enlarged view of the smaller object area (top left corner) and the diffractogram (lower right corner) indicating the achieved resolution. In the diffraction pattern one can see reflections out to about 5 Å (courtesy of C. Gatel & E. Snoeck, CEMES-CNRS, Toulouse France).

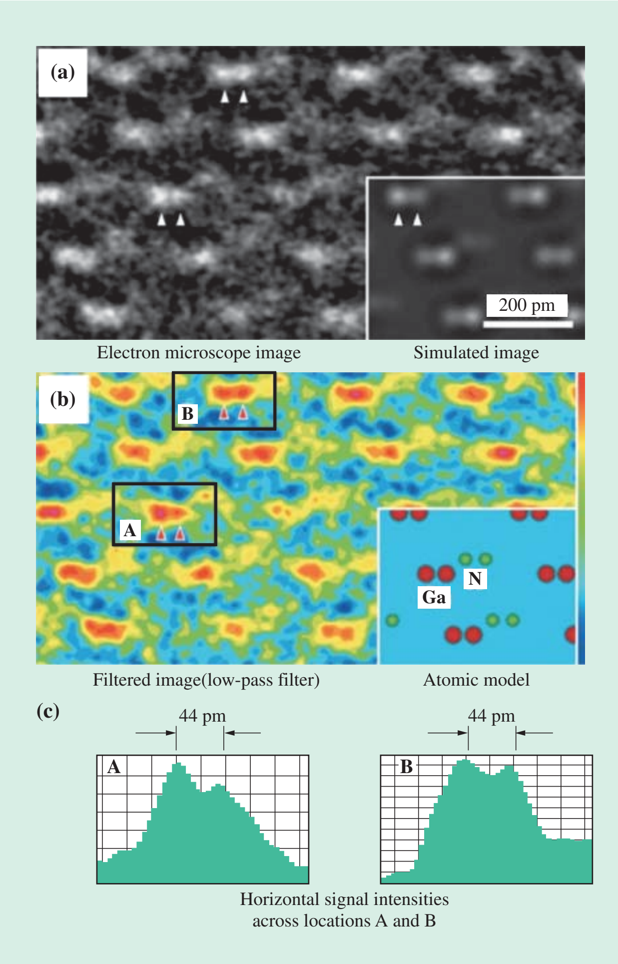

Fig. 7—Imaging of Ga Atom Spacing in GaN.
(a) shows a high-resolution TEM image of a GaN [411] thin sample. Projected Ga atom positions (white arrows) with 44 pm separation were clearly observed. (b) shows the corresponding Gaussian low-pass filtered image. The color key on the right shows the image intensity. (c) shows the line profiles of the Ga atom pairs indicated by black rectangles A and B in (b). Ga atom pairs were also clearly resolved in these profiles. [from T. Akashi et al., Appl. Phys Let. 106 (7), 074101, 2015].

smallest details, he also wanted to measure phase shifts of electrons by electromagnetic fields with atomic resolution, a so far unavailable precision. For this goal he started the development of a new high voltage TEM equipped with a cold field emitter – for highest coherence – and decided to use a Cs-corrector to avoid delocalisation of information due to the spherical aberration of the objective lens. Due to the extremely high beam energy and the size of the instrument, the development of the Cs-corrector has been a true challenge.

It employed the standard two-hexapole design, but the necessary strength, power dissipation and possible magnetic saturation effects of all optical elements had to be considered very carefully. After a theoretical feasibility study and detailed design, the manufacturing could finally be started. A critical point was that the proper function of the corrector and also the correctness of the underlying theory could only be tested very late in the final stage of the implementation. Due to the excellent cooperation between Hitachi and CEOS, this project was finished successfully without any delay caused by the corrector technology.

The initially anticipated resolving power of the system finally could be demonstrated for high-resolution imaging and in Cs-corrected Lorentz mode for field-free imaging. Here, a new technique has been used with an extra dedicated Lorentz lens in between the upper stage and the objective lens. As shown in Fig. 7 the achievable resolution is at least 44 pm and the adjacent Gallium columns in GaN <114> are clearly resolved[10]. This currently is considered a new world record in TEM resolution. The FIRST program microscope is now in operation and used for holography and other precision measurements.

Today, the long-lasting, fruitful and successful cooperation between Hitachi and Hitachi High-Technologies in Japan and CEOS in Germany is looked back on with gratitude. Not only in terms of technological challenges, but also at the personal level, the relationship between the companies became very close. During more than one decade together the companies were able to successfully finalize four challenging development projects, one further project currently is a work in progress and some other ideas are expected to become reality in the future.

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ABOUT THE AUTHORS

Prof. Dr. Max. Haider
Co-founder of CEOS GmbH, former Managing Director of CEOS GmbH, and today Senior Advisor and authorised officer, Honorary Professor at KIT (Karlsruhe Institute of Technology), Germany.

Dr. Heiko Müller
Managing Director, CEOS GmbH. He is currently involved in theoretical studies of advanced charged particle systems and the project management of several projects.