The Otto Scherzer Memorial Symposium on Aberration-Corrected Electron Microscopy

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Introduction

The year 2009 marks the centenary of the birth of Otto Scherzer, one of the early pioneers of electron microscopy. Scherzer, shown in Figure 1, was the originator of the famous microscopy theorem that the spherical and chromatic aberrations of rotationally symmetric electron lenses were unavoidable [1]. In honor of this centennial occasion, we organized a special memorial symposium during the Microscopy & Microanalysis 2009 meeting, which was held in Richmond, Virginia, in late July. The introductory talks of the symposium presented a fascinating mix of firsthand accounts about working with Scherzer in Darmstadt and descriptions of the correction concepts and the early corrector prototypes that emerged from his group. Placed in this historical context, the latest advances in aberration correction for scanning and fixed-beam instruments that were presented in this symposium were all the more impressive and conveyed a vivid sense of history in the making. Representative applications of aberration correction to a broad range of materials were also highlighted in platform and poster presentations. Here we give a short account of the emergence of aberration-corrected electron microscopy (ACEM) and very briefly summarize some of the prospects and challenges for this burgeoning field. Further information about these developments, including details of applications, will be found in selected papers from the symposium, which will be published in a forthcoming issue of the journal *Microscopy and Microanalysis* due to appear in mid-2010.

Aberration Correction

Spherical aberration is a focusing defect that is inherent to circularly symmetric electron lenses, and it prevents off-axis electrons from all being focused to the same point. Aberration correction makes it possible to "tune" spherical aberration (Cs) by making its value vanishingly small or even negative [2]. Because the Scherzer resolution limit—as given by $d{\sim}0.7\lambda^{3/4}C_S^{1/4}$, where λ is the electron wavelength—is proportional to the spherical aberration, one might reasonably ask why the microscope resolution can't be made arbitrarily good by making the Cs value arbitrarily small. The problem is that there are many lens aberrations: spherical aberration is only the tip of the iceberg. This situation is similar to the case for the human eye where spherical aberration leads to blurred vision, astigmatism leads to distorted vision, and chromatic aberration leads to color blurring. Once the spherical aberration of the eye is corrected with a pair of glasses or contacts, then other aberrations such as astigmatism become important. The distortions present in the objective lens of the electron microscope can be visualized in terms of a magnetic field that transforms the incoming plane wave into a highly irregular shape. This shape can be described as a power series of waves, each with its own symmetry (2-fold, 3-fold, 4-fold) and order (1st order, 2nd order, 3rd order, etc.). This leads to a veritable "zoo" of aberrations of different symmetry and order, which in turn could limit the microscope performance. Latest aberration correctors take care of aberrations up to 5th order, and these have succeeded in improving the resolution limits of the transmission electron microscope (TEM) to about 50pm for both fixed-beam and scanning-beam TEMs.

Emergence of ACEM

The early days of electron microscope construction and development were mostly spent struggling to overcome the many serious limitations to instrument performance that were caused by mechanical and/or electrical defects. Although resolution was already much better than that achievable with a light microscope, it was still far removed from the theoretical limit, and even further from the electron wavelength. Little

Figure 1: Image of Otto Scherzer (courtesy of Dieter Typke).

doi: 10.1017/S1551929509000303

thought was probably given to the possibility of actually correcting lens aberrations. The situation changed dramatically following the publication in 1947 of another seminal article by Scherzer [3], which pointed out several possible avenues for overcoming spherical *and* chromatic aberration. Most workers in the field of electron optics pursued Scherzer's suggestion of using asymmetric imaging correctors based on multipole elements, but scant success with aberration correction was achieved over the following years despite many valiant attempts. Although mechanical imperfections and electrical instabilities were undoubtedly major contributors to the lack of progress [4], the absence of any systematic procedure for carrying out electrical and mechanical adjustments based solely on image appearance meant that correction of spherical aberration came to be regarded as a task so complex that it was well beyond the skills of an unaided human operator [5]. Something much faster and more routine was needed before online aberration correction could become a reality.

Online computer control of microscope parameters was first implemented for the scanning electron microscope [6], and iterative procedures based on image contrast analysis that were suitable for online focusing and stigmating in high-resolution electron microscopy soon followed [7]. The development of automated diffractogram analysis, sometimes termed "autotuning," was made possible by the quantitative recording capability of the slow-scan CCD camera [8], and this technique led to rapid and reproducible adjustment of focus, beam tilt, and image astigmatism. Similar to the more recent procedures that are applicable to aberration correction, this autotuning approach for the fixed-beam TEM mode relied on the presence of a small region of amorphous material in or near the field of view. This specimen requirement obviously represents a serious restriction when investigating certain types of materials. By analyzing a systematic set, or "tableau," of diffractograms taken from images recorded with axial and tilted-beam illumination, aberration coefficients can be determined to the high degree of accuracy that is necessary for subsequent correction of aberrations [9]. An alternative approach to aberration assessment, based on the acquisition and analysis of far-field images, also termed Ronchigrams, is used during the aberration correction procedure normally applied in the scanning TEM mode [10].

Initially, spherical aberration was the primary target for correction efforts because it was widely perceived as the dominant factor that predetermined the microscope resolution. However, as the impact of incoherent effects such as vibrations, noise, and stray fields was progressively reduced, thereby extending resolution limits, it slowly came to be realized that other difficult-to-measure lens aberrations such as misalignment coma and three-fold astigmatism were in turn limiting image interpretability [11]. These additional aberrations can be measured accurately using either the diffractogram or Ronchigram approaches, and they can then be easily accounted for during the process of aberration correction.

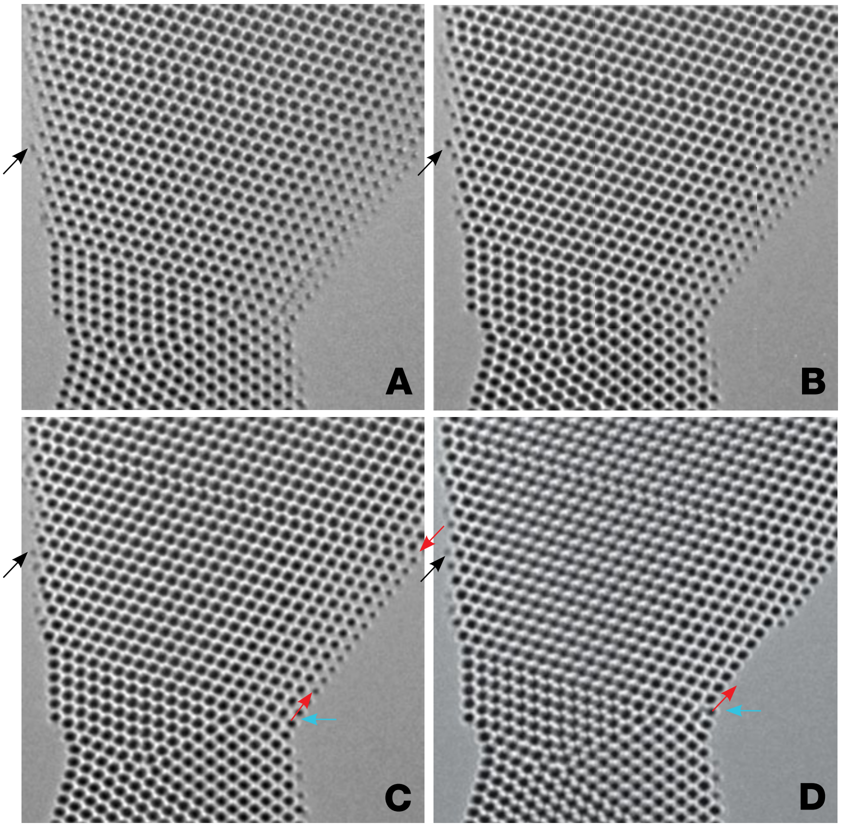

Figure 2: Four sequentially recorded TEM lattice images of gold [110] nanobridge connecting two grains that are rotated relative to each other by 90 degrees around [110] axis. The four images shown are part of a 15-member focal series, recorded in time intervals of 1.5 s. Black arrows: (a,b) 2-atom column and (c,d) single atom. Red arrows: thirteen 2-atom columns, some of which disappear in (d). Turquoise arrows: Rearrangement of atom columns at the intersection of a dissociated grain boundary with the surface. The focus difference on both sides of the bridge is negligible because the film was grown onto a flat single crystal substrate. Images from Kisielowski et al., Microsc. Microanal. 14, 469–477, 2008 (courtesy of the Microscopy Society of America).

Approaches to ACEM

Aberration correction can be achieved nowadays in several different ways. Online approaches in either fixed-beam TEM [9] or scanning-beam STEM [10] follow the principles outlined by Scherzer and use hardware corrector systems installed on the instrument, whereas off-line correction techniques use special software programs to reconstruct off-axis electron holograms [12] or exit-surface wavefunctions [13]. All of these approaches involve computer-aided analysis to produce a phase plate that effectively combines the effects of all prevailing lens aberrations as a function of scattering angle. An inverse phase plate for either off-line or online correction purposes can then be easily generated.

The electron holography technique was initially proposed by Gabor in 1949 as a path towards improving microscope resolution beyond the traditional spherical-aberration limit [14], but little practical progress towards this goal was made until the high-brightness, high-coherence, field-emission electron gun (FEG) was developed [15]. The clear visibility of individual Si atomic columns in reconstructed phase and amplitude images was an early validation of the efficacy of this approach [12]. High-resolution electron holography has since been shown to be facilitated, and phase sensitivity greatly improved, when online correction hardware has been used for compensation of most aberrations *before* commencing detailed holography observations [16]. Reconstruction of the exit-surface wavefunction in the fixed-beam TEM combines images recorded at several different defocus values to avoid any loss of information due to zero crossovers in the objective-lens transfer function—and image interpretability is pushed out as far as the microscope information limit, which is defined by incoherent effects or by the envelope functions that are due either to beam divergence (spatial coherence) or focal spread (temporal coherence).

Online corrector systems involve various combinations of multipole elements that serve to overcome the symmetry constraints recognized in Scherzer's original 1936 theorem. In his later 1947 paper, Scherzer had suggested using quadrupoles to distort the beam from its rotational symmetry and octopoles to cancel the aberrations [2]. Based on this concept, the original corrector for the scanning TEM consisted of a combination of six quadrupole-octopole elements that preceded the normal objective lens [10]. Ronchigram analysis and beam-induced image shift were used for alignment of the corrector elements and for aberration measurement, which was then followed by the correction of third-order spherical aberration, axial coma, and astigmatism (two-fold and three-fold). For fixed-beam TEM, the first corrector combined two hexapoles and two additional round-lens doublets that were inserted into the microscope lens column as a single unit immediately below the objective lens [8]. The correction procedure then consisted of diffractogram analysis, followed by computer-controlled feedback to the various corrector, stigmator, and lens focusing coils. More complicated corrector systems have since been developed to correct the prevailing higher-order aberrations, but the basic symmetry-breaking principles remain in place.

Benefits of ACEM

The successful online correction of spherical aberration in the electron microscope is an exciting and praiseworthy feat that has deservedly attracted much attention. Representative examples of aberration-corrected TEM and aberration-corrected STEM images are shown in Figures 2 and 3, respectively. Direct TEM image interpretation has been pushed into the sub-Ångstrom regime, and aberration-corrected probes promise a veritable revolution for atomic-scale microanalysis. However, these improvements in resolution *per se* for imaging and analysis are not the only important contributions of aberration correction; the improved signal quality for dynamic studies in aberration-corrected images, the greater current available for analysis with the corrected probe, and the higher sensitivities to variations in atomic position or chemical composition are all significant for materials investigations. These advances have already resulted in a number of striking applications of ACEM, many of which were also represented in the Scherzer symposium.

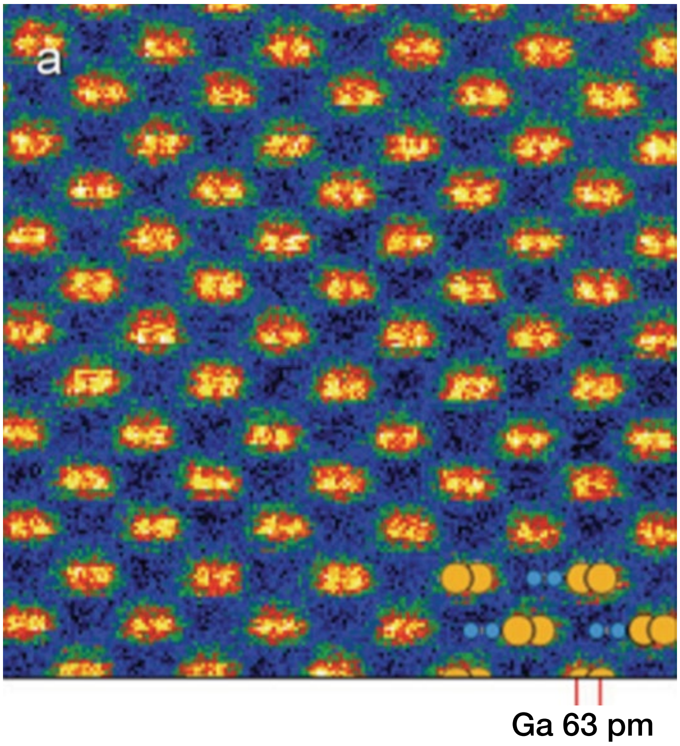

Figure 3: Aberration-corrected STEM image of [211] GaN recorded with the TEAM 0.5 ACEM, showing clear resolution of individual Ga atomic columns with separations of 0.63Å (from Ref. 17).

There are additional benefits of aberration correction that have yet to be fully appreciated by the broader materials community. For example, one simple advantage of using a corrected TEM is the complete removal of the displacement error present in selected-area diffraction patterns for higher-order diffracted beams that is caused by spherical aberration. Another advantage is that axial coma is markedly reduced so that slight beam tilts by electronic controls rather than crystal tilting by mechanical means can be used to achieve more accurate alignment of the incident-beam direction with the required crystal zone axis. A further benefit is that TEM imaging with a small but slightly negative Cs value, combined with a slight overfocus lens setting, can greatly enhance the visibility of oxygen atomic columns that are closely adjacent to much heavier metal-atom columns [2]. An additional advantage is the potential for depth sectioning due to the reduced depth of field that results from the larger convergence angles achievable with aberration corrected probes [18]. Furthermore, as already pointed out, the off-line holography and exit-wave reconstruction methods are greatly simplified because determination of the aberration phase plate, which is an essential step for these techniques, is an integral part of the correction procedure. In the case of scanning TEM, much larger collection angles can be used for bright-field (BF) imaging because aberration correction reduces the impact of the spatial coherence envelope: signal intensities can be increased by factors of 100 or more, and aberration-corrected

BF images of exceptional quality can now be recorded [19].

Meanwhile, it is not intuitively obvious that TEM imaging with the spherical aberration coefficient set at exactly zero is necessarily beneficial because amplitude contrast rather than phase contrast will dominate the image characteristics. Moreover, slight changes in sample thickness can have a remarkable impact on TEM image appearance when using very small Cs values, so that image simulations must still be considered as highly desirable in order to avoid possibly erroneous conclusions about very fine image features. Further investigation of microscope and specimen parameter space is urgently required to determine the imaging conditions most appropriate for studying specific types of materials.

Prospects and Challenges

The field of aberration correction is expanding rapidly. All of the major TEM manufacturers are aggressively developing and marketing ACEMs, and novel applications of these instruments are being reported with increasing frequency, as will be readily evident by reference to the proceedings of recent electron microscopy meetings. Even more sophisticated ACEMs that incorporate correction of lens aberrations of up to fifth order have been designed and are being tested [17, 20]. The first promising results from a prototype system for chromatic aberration correction developed under the DoE-supported TEAM project have just been reported [21, 22]. The prospects seem excellent for exciting times ahead for both the microscopy and materials communities. Otto Scherzer would surely be delighted to see the fruits of aberration correction!

The capabilities of the new aberration-corrected instruments greatly exceed those of previous generations of microscopes, opening up new opportunities to explore materials at the atomic scale. Some of these opportunities can be realized immediately without the need for further developments, but others pose challenges for the development of novel instrumentation or new techniques. The notion of a tunable electron-optical observatory for materials research, which was the underlying vision of the TEAM project, includes the ability to tune the electron energy to the material or problem at hand. The range of 80-300 keV covers an important operating regime that is determined by the need to find a balance between usable sample thickness, tolerable radiation damage, and achievable resolution. However, this balance is shifting because aberration correction, especially when Cc is included, makes lower-energy microscopy more accessible for atomic-resolution imaging. In addition, the interface between hard and soft matter is an anticipated growth area where the new developments in electron microscopy instrumentation and technique can potentially have a great impact, even though sensitivity to electron-beam irradiation will always remain a problematic issue. Likewise, real-time observations of such atomic processes as quasi-melting, crystal growth, atomic diffusion, and phase transformations are among the more important scientific needs for aberration-corrected microscopy. For advances in all of these areas, faster, more sensitive detectors will make a substantial contribution.

Challenges remain. The increased sensitivity of TEM image appearance to slight variations in orientation, thickness, or lens defocus underscores the need for image simulations in support of image interpretation. Sample preparation also becomes more demanding as resolution limits improve. On the one hand, clean sample surfaces are needed to ensure that oxide or contamination layers do not degrade image quality. On the other hand, the absence of any amorphous material makes accurate focusing and astigmatism correction much more difficult to achieve. Also, specimen regions suitable for assessment and correction of aberrations using Ronchigrams or diffractogram tableaus are still required. Thinner crystals are needed for higher-resolution imaging, but local crystal bending is then more likely to occur. These somewhat conflicting and demanding specimen requirements will need concerted attention if the potential of the ACEM for solving real materials problems is to be fully realized.

Finally, despite the considerable recent technical advances, many other problems remain to be addressed. These include improved instrumental stabilities, better detectors, image and signal quantification, better sample preparation, and capabilities for *in situ* chemical studies. Perhaps the ultimate goal is three-dimensional tomography at atomic resolution to locate and identify all the atoms and to determine how they are bonded. MT

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