An Aberration Corrected (S)TEM Microscope for Nanoresearch

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Introduction

The continued focus on improving materials, combined with the fact that it is now commonly understood that material properties are affected by characteristics at the atomic level, give rise to the need to characterize and image at the best resolutions possible. The (Scanning) Transmission Electron Microscope ((S)TEM) has the capability to image structures with atomic resolution and provides, at the same time, information on the chemical composition, bonding and electronic structure of the material. The nanoresearcher's continued need for the ultimate resolution has accelerated the development of next generation electron optics and technology. The utilization of aberration correctors and monochromators in the electron microscopes enables new insight into the relationship of nanostructures and their physical properties, which is pivotal for a deeper understanding of the fundamental principles of nanotechnology devices. Aberration corrected electron microscopy will mean tighter, brighter beams, yielding a stronger signal, higher imaging contrast, greater analytical sensitivity and unprecedented spatial and spectral resolution. The new platform, the Titan™ 80-300 corrected (S)TEM (see figure 1), will open doors for researchers to study morphology, crystallography, elemental and chemical composition, as well as electronic structure at resolution levels not demonstrated before in one advanced analytical instrument:

Tackling objective lens aberration with correctors

Until recently, ultra-high resolution imaging in (S)TEM applications were limited, in a large measure, by the spherical aberration of the objective lens. Correctors have the capability to remove this aberration thereby removing the barrier to better resolution. Typically, in any round magnetic lens, those parts of the object wave that carry the most interesting information pass through the outer zone of the objective lens and suffer from large phase shifts. The additional wave phase shifts are mainly introduced by the spherical aberration (Cs) of the objective lens. Scherzer proved in 1936, that spherical aberration cannot be avoided in rotationally symmetric electromagnetic fields [1]. This aberration can only be corrected by means of multipole fields, which produce a third order negative spherical aberration. In 1990, Rose described a hexapole corrector that provides for the compensation of the Cs-aberration of an objective lens [2, 3] (see figure 2). By changing the hexapole excitation, the value of the total Cs-aberration of the (S)TEM imaging system can be set to any value, as well as zero. This gives an additional parameter for tuning the microscope for optimal imaging conditions.

Figure 1: FEI Titan™ 80-300. (Unless noted otherwise, all images below were acquired with the Titan™ 80-300)

In classical (S)TEM uncorrected microscopes, the image intensity originating from an object point is spread over an area whose radius is dependent on the spherical aberration and the defocus. This is called delocalization and hampers direct interpretation of (S)TEM images. It is quantified by the radius of the point-spread function and minimizes in the so-called Lichte focus to $Z_L = -1/4\, Cs\, \lambda^3 g^3_{max}$ with λ the wavelength of the imaging electrons and g_{max} the maximal transferred spatial frequencies [7]. In a Cs corrected TEM, the contrast delocalization can be minimized by correcting the spherical aberration or by tuning it to an optimal value. As a result, delocalization is suppressed and images are directly interpretable.

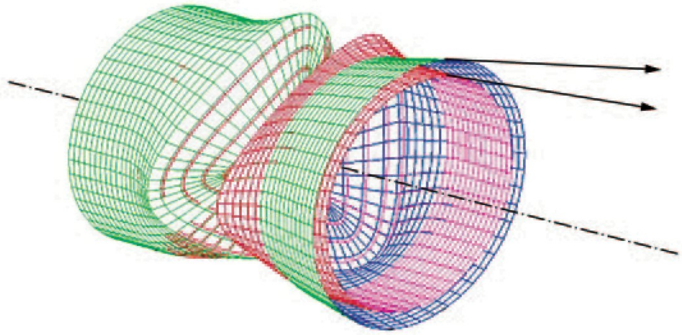

Figure 2 : Ray path through the hexapole Cs-corrector consisting of two hexapoles (rectangular boxes), which are separated by a telecentric round lens couple. It can be seen that the outer ray-path (green) is tilted away from the optical axis in order to compensate for the spherical aberration of the objective lens. Figure courtesy of Dr. S. Uhlemann, CEOS GmbH.

In addition to coherent spherical aberration, incoherent disturbances limit the information transfer of the TEM. The chromatic aberration of the objective lens, which is a function of the energy spread of the illuminating electron beam, dampens the contrast transfer of the TEM at high spatial frequencies. This damping can be overcome by reducing the energy-spread of the illuminating electrons with a monochromator [4]. A (S)TEM equipped with a monochromator in the illumination system is capable of doing this, as it allows for the selection of the energy spread of the illuminating electron beam [5], (see figure 3). A smaller energy spread will also improve the analytical capabilities of the (S)TEM. The clear benefits and scientific opportunities of combined corrector and monochromator technology have encouraged FEI to develop an entirely new (S)TEM platform that provides aberration correction, flexibility in the high-tension range from 80 to 300kV, and ultra-high resolution in imaging and spectroscopy.

A newly designed microscope

The new electron microscope can transfer information with deep sub-Ångstrom lateral resolution and ultra-high spectral resolution,

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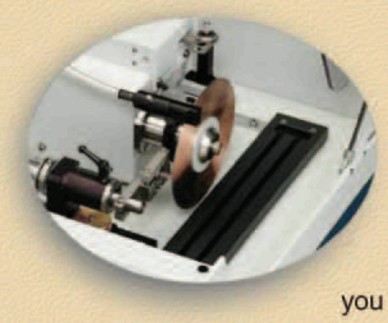

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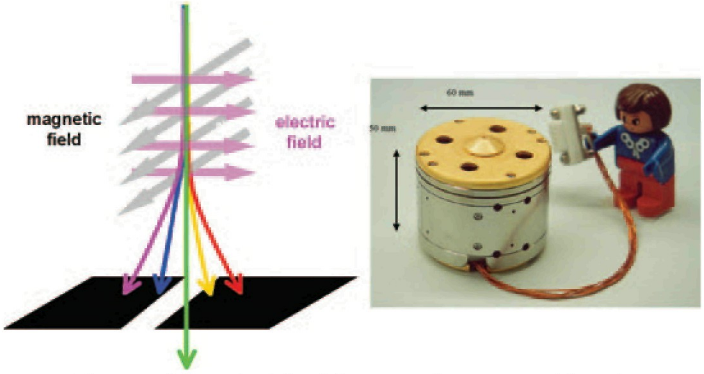

Figure 3: The principle of the monochromator with a picture of a Wien-filter type monochromator

(examples are given in figures 8 and 9). Stability is clearly key to ultimate performance. The system's design complies with the stringent needs for ultimate mechanical, electronic, and thermal stability, as well as precise alignment of the advanced components that build this microscope. Disturbances due to mechanical vibrations have been avoided by introducing a design concept that separates the magnetic flux circuit from the mechanical forces in the column. Additionally, the column's stiffness has been improved by increasing its diameter to 300 mm, making use of the 4th power dependency of stiffness on column diameter. To avoid damping of the information transfer of the (S)TEM due to alignment aberrations, the microscope encompasses a new (patent pending) invention that aligns optical components of the column automatically to each other while assembling with extreme precision. This invention also makes a field upgrade of Cs-correctors possible, thus allowing a step-by-step approach to aberration corrected microscopy in the field.

Like the chromatic aberration, noise in the drive current of the magnetic lens and deflectors reduces the temporal coherence of the imaging electrons, which leads to a limitation of the lateral resolution as well. Newly developed electronics in the microscope reach a noise level below 0.1 ppm RMS. This proves to be mandatory to achieve information transfer down to the deep sub-Ångstrom level. For optimal use of the monochromator, the high tension (HT) has to provide a noise/ripple below 0.1ppm RMS. Only by using a stable HT can energy resolution of 0.1eV be reached in energy loss spectra.

For a robust and reliable working (S)TEM not only short term stability but also long term stability is important; mechanical drift, and especially optical alignment drift, play a much more significant role. In addition to the precise mechanical alignment of the optical components to each other during assembly, the thermal drift behavior of the (S)TEM needs to be reduced to negligible values to avoid long stabilization times after changes in optical settings. In a lens, magnetic flux depends linearly on lens current while thermal power generation depends on the square of the current. By introducing at least two coils per lens it is possible to offset one against the other to maintain a constant temperature in the column for all optical working modes. This design is intended to comply with future requirements for chromatic aberration correction currently under development in the TEAM project of the Department of Energy (DOE) in the USA [6].

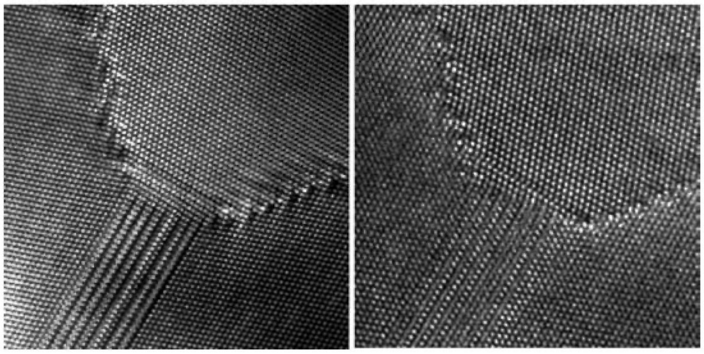

Figure 4. Comparison of a not Cs-corrected (left) and a Cs-corrected (right) HR-TEM image on the same area of a polycrystalline gold sample in <110> direction. The delocalisation due to spherical aberration is visible at the grain boundary in the uncorrected image (left). The exact positions of the atoms on the grain boundary can be clearly seen in the Cs-corrected image (right), while Móire patterns are degrading the resolution in the uncorrected image (left). Sample courtesy: C. Kisielowski, from the National Center of Microscopy in Berkeley, USA

Benefits to Nanoresearch

In NanoResearch, critical dimensions have decreased, so that the structure of the materials being studied must be understood down to the atomic level so as to modify their physical properties. For the macroscopic properties of a nanocrystalline metal, grain sizes and the grain boundaries of the metals play a dominant role. Hence, the size of the grains needs to be measured accurately and the atomic structure at the grain boundary must be understood to improve the properties of these materials.

The benefit of an image Cs-corrector can be demonstrated

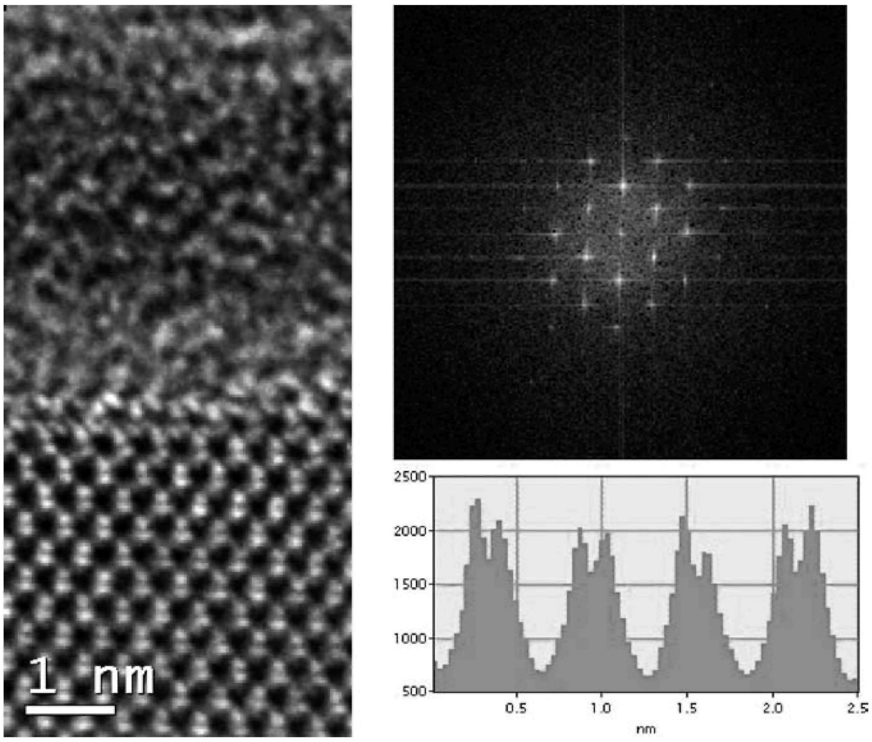

Figure 5. Silicon <110> sample with SiO_2 and HfO_2 layer on top of the silicon. Image was recorded with spherical aberration corrector on a Tecnai F20 S-Twin. The image and the intensity profile show clearly the silicon dumbbell structure. The interface between the silicon and the amorphous layers can be clearly resolved. The Fourier transform indicates the transfer of the column with the Cs-value corrected down to less than 3 microns.

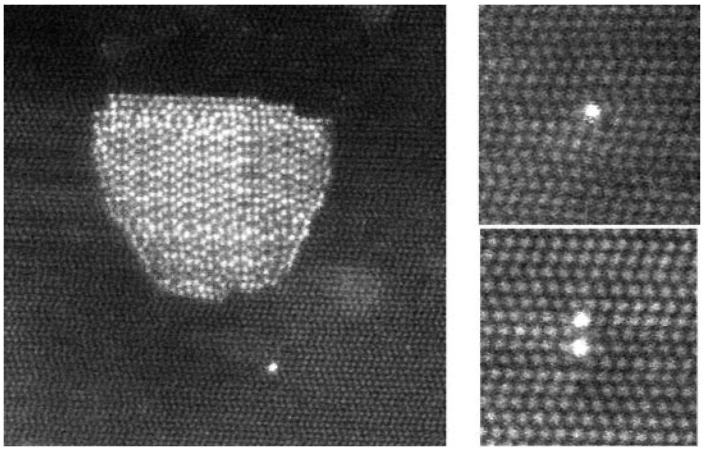

Figure 6 : HR-STEM image of erbium atoms in SiC. The images show the unprocessed data. A precipitate of erbium and single atomic columns of erbium can be visualized in the STEM images. Single erbium columns create stacking faults in the SiC lattice (upper right image). Sample courtesy: Prof. U. Kaiser, University of Ulm, Germany

by comparing an atomic resolution TEM image without spherical aberration using the corrector technology and an atomic resolution TEM image with spherical aberration of the imaging system. An example on nano-crystalline gold acquired with image Cs-corrected TEM is shown in figure 4. The improved image quality of the grain boundary is clearly visible since the exact positions of the atoms at the boundary can be obtained, while in the uncorrected image the blurring of the image information makes the interpretation at the grain boundary difficult or even impossible.

In electronic devices the atomic structure of interfaces and the thicknesses of layers determine the electronic properties of the device. The dimensions of these layers have reached the sub-nanometer level. Therefore, images with the highest atomic resolution are needed to understand the material's properties and critical dimensions so as to improve the quality of these devices. Figure 5 shows an atomic resolution image of a high-*k* dielectric nanoscale device acquired with spherical aberration correction. The dumbbell structure of the silicon, with a spacing of 0.136 nm, can be clearly seen. Moreover, the structure of the silicon-silicon oxide interface can be visualized without noticeable delocalization effects. A darker amorphous HfO_2 layer is visible on top of the amorphous silicon oxide layer. In a transistor the dimensions of these layers are intimately linked with the performance of the device and understanding their dimensions is crucial.

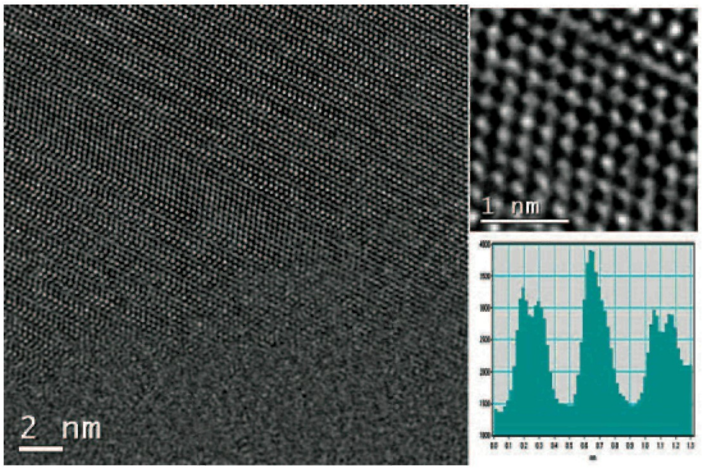

Figure 7: HR-TEM image of cubic and hexagonal SiC. In the magnified area the splitting between the silicon and carbon is visible. The intensity profile is taken from this area and shows a splitting around 0.085nm. Hence sub-Angstrom resolution is proven with this image. Sample courtesy: Prof. U. Kaiser, University of Ulm, Germany.

Z-contrast imaging in STEM mode is a powerful method to visualize changes in atomic composition down to the atomic level. Figure 6 shows an example of HR-STEM acquired on the new platform. The sample is SiC that has been doped with erbium. The erbium has segregated and precipitated in the SiC and has formed single atomic columns in the direction of the electron beam. These single columns, which have a brighter contrast in the image due to their high atomic number (z-contrast), also create stacking faults in the surrounding SiC matrix.

The atomic distance between the Si and C atoms in silicon carbide is smaller than 0.1 nm and cannot be resolved in HR-STEM without Cs probe correction. In contrast, in HR-TEM, the information limit is capable of resolving this distance. Figure 7 shows an uncorrected HR-TEM image of SiC. The image shows SiC grown in the cubic and hexagonal modifications. A distance of better than 0.09 nm (Si-C distance) can be resolved in the image, proving the sub-Ångstrom resolution of the instrument in HR-TEM mode. Preliminary results on the probe Cs-corrector prove a resolution better than 0.1nm in HR-STEM mode. An example of this resolution is given in figure 8 on a silicon sample oriented in the <110> direction. In the Fourier transform of the image, a distance of 0.083 nm can be inferred. Additionally, the probe Cs-corrector system increases the currents in focused probes compared to an uncorrected (S)TEM, allowing the acquisition of EDS or EELS spectra with higher speed or better statistics.

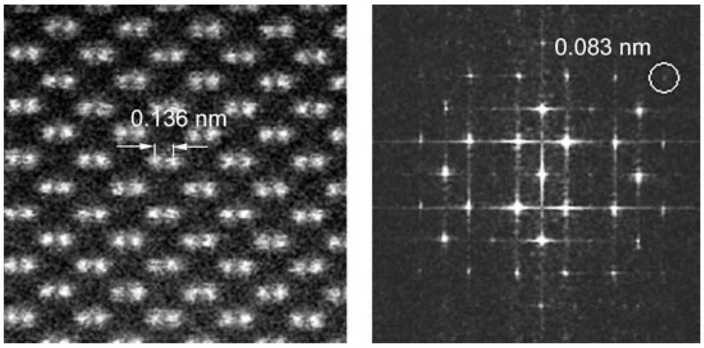

Figure 8 : HR-STEM image of silicon in a <110> direction with a probe Cs-corrector. In the left image, the splitting between the silicon atoms (0.136 nm) is clearly resolved. In the Fourier transform (right) a distance of 0.083 nm can be observed.

In an advanced (scanning) transmission electron microscope, not only can the specimen's structural properties be measured with imaging techniques like STEM and TEM, but electronic properties via electron energy loss spectroscopy (EELS) can be determined as well — *e.g.*, band-gap energies or bonding state information as revealed in the fine structure of inner shell excitations. Band gap energies are low and are typically in the energy range of 1-3 eV. Therefore, the band-gap edges in EELS are close to the peak of the electrons that have not lost energy when passing through the sample (zero-loss peak). Hence energy resolutions on the order of 0.2 eV (FWHM) are needed to separate the peak of the elastically scattered electrons from the absorption edge of the electrons scattered on the band-gap to obtain this kind of information reliably.

The monochromator improves the energy resolution of the Schottky-emitter to the level required for high resolution EELS. The

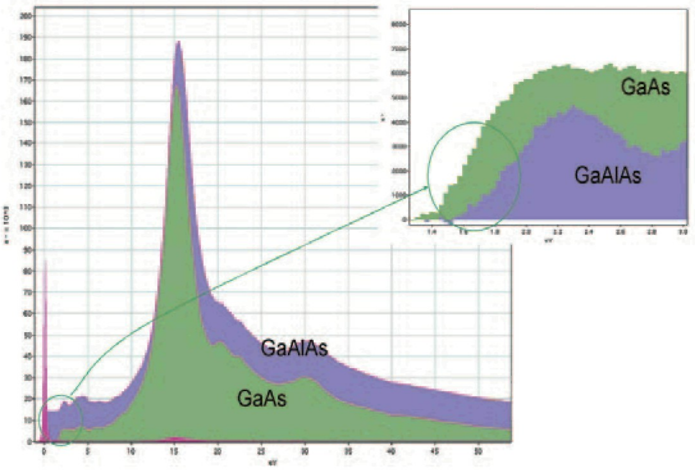

Figure 9 : Low loss EELS spectra of GaAs and GaAlAs acquired with a monochromator at 300 kv acceleration voltage. A shift in the band-gap energy between pure GaAs and GaAlAs can be observed. The measured band-gap energies are 1.42eV (GaAs) and 1.73eV(GaAlAs). Notice the shift in the plasmon peak, which indicates a change in the electronic properties.

benefit of having this capability on the TEM is not only to measure band-gap energies in spectroscopy, but to do this with high lateral resolution as well, which is needed in small semi-conducting devices. An example of a band gap measurement on a GaAs, GaAlAs multilayer is shown in figure 9. In the EELS spectra an energy shift of the band gap can be measured that is caused by the change in the chemical composition of the layers. In this nanoresearch application, a change in the physical property that is essential for the proper operation of the device, can be directly measured with high lateral resolution.

Conclusion

On the cutting edge of nanoresearch as well as materials science, the need to gain knowledge at the single atomic level, including understanding inter-atomic bonding electron densities for the characterization of chemical composition, electronic structures and mechanical properties on the nanoscale, has accelerated the development of new electron-optics technology for ultra-high resolution electron microscopy. Until now, aberration correction technologies in electron microscopes have been treated as accessory components for (S)TEM systems that were not truly optimized for this type of advanced technology. Thus, the integration of these types of correctors for breaking the next resolution barrier and for high usability has been met with limited success. The FEI Titan^{TM} 80-300 is a new microscope designed as a dedicated aberration-corrected system that will enable corrector and monochromator technology to enter into nanotechnology research and industrial markets. ■

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