One-Ångstrom Microscope Update

Report to the External Steering Committee of the NCEM

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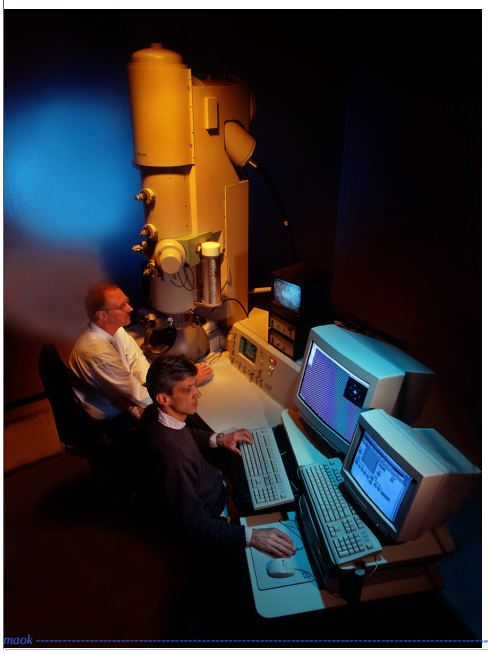
One-Ångstrom Microscope Update



- **Three-fold astigmatism on the Philips CM300 FEG/UT electron microscope has been corrected from 2.25µm to 50nm.**
- **We have achieved information transfer to below 1Å.**
- **We have imaged the C-C dumb-bells in diamond. The 0.89Å (400) spacing is present in diffracto-grams.**
- **The one-Ångstrom microscope (OÅM) project has used focal-series reconstruction (Thust) to extend the CM300 resolution from its standard 1.7Å to near 1Å.**
- **The CM300 is now available to users on-line as part of the MMC.**

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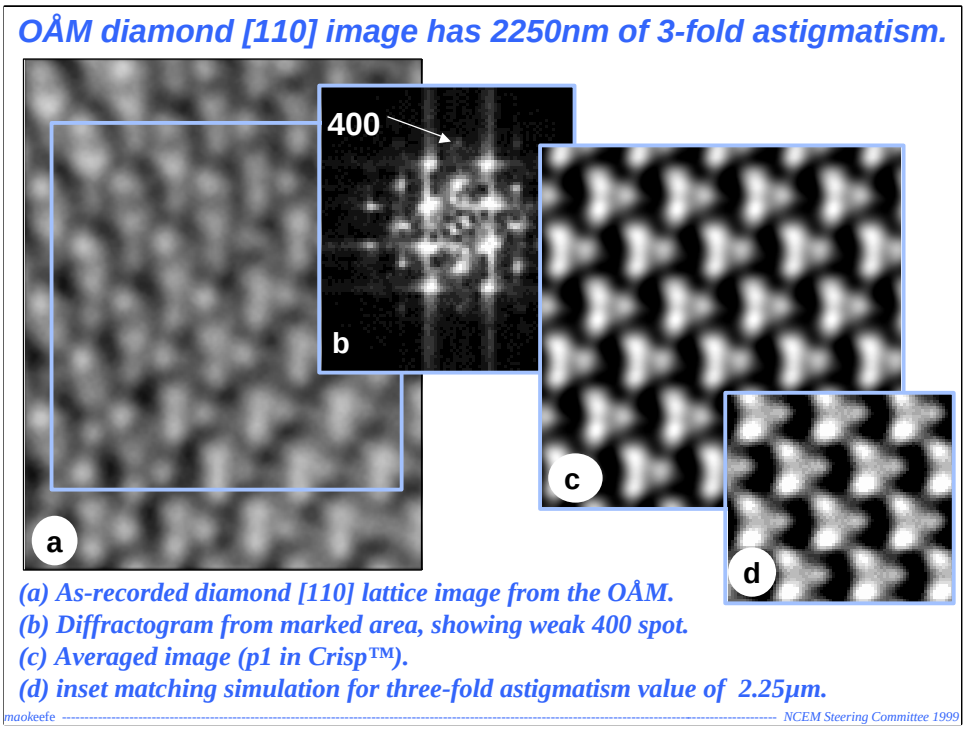
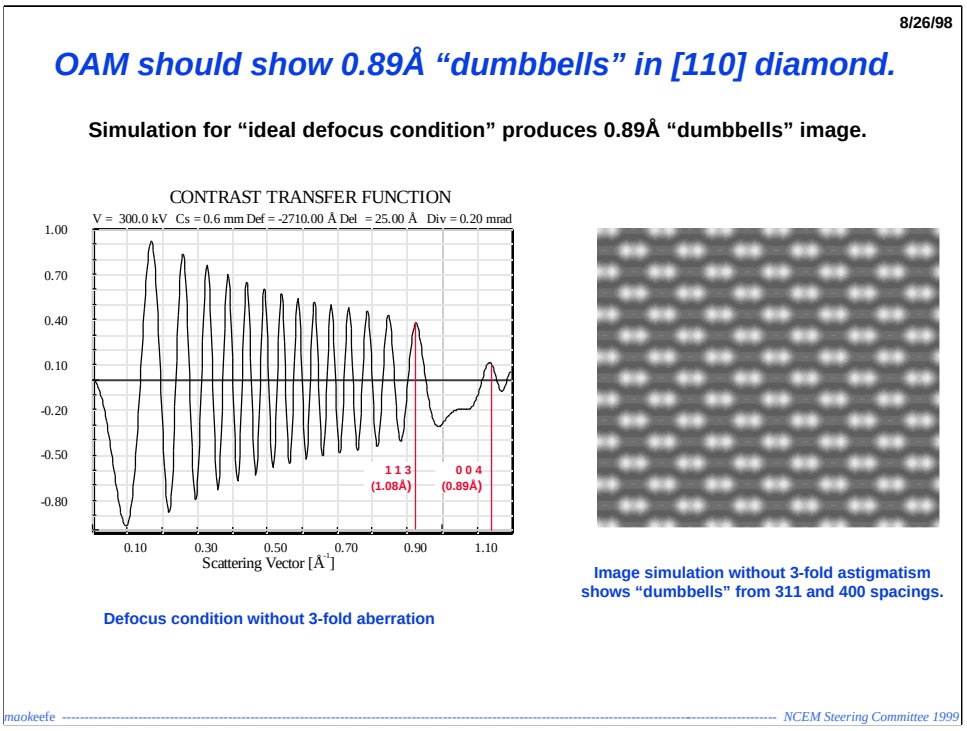
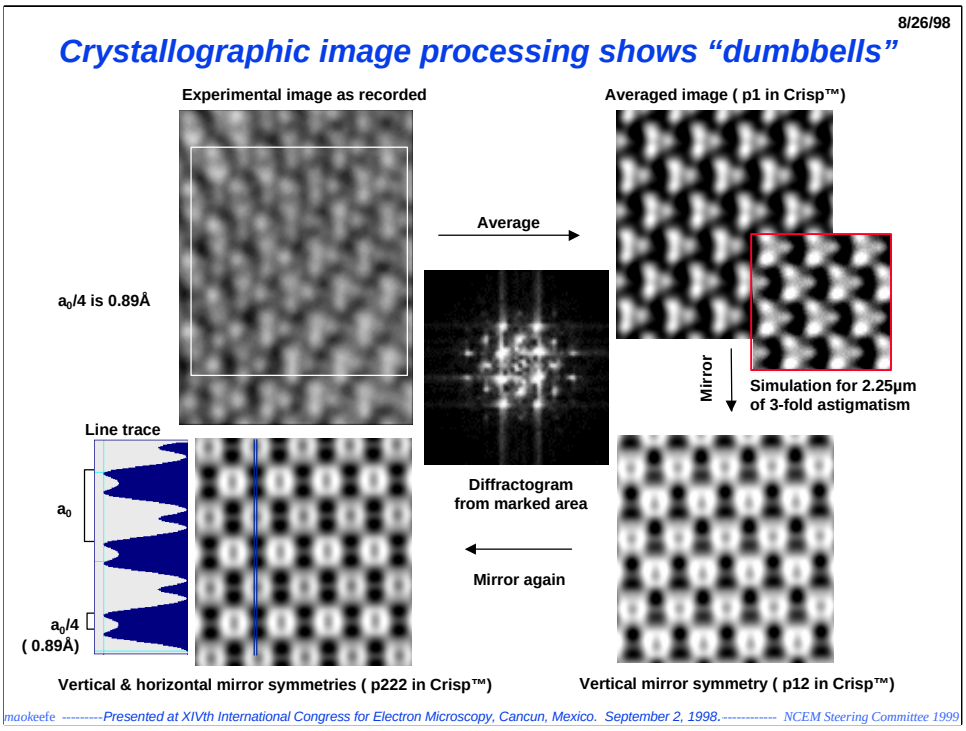
- The One-Angstrom Microscope project has attained its goal, and is now producing images down to 1Å resolution.
- Transmission electron microscopy of defect structures to a resolution of 1.1Å has been achieved, with evidence that 0.89Å will be possible. This level of resolution will soon be made available to all those NCEM users who have a requirement for 1Å resolution.
- Improvements since the previous meeting of the NCEM Steering Committee include:
  - correction of the CM300FEG objective lens 3-fold astigmatism to a value meeting the original OÅM specifications of 500Å.
  - acquisition of the MAL software for focal-series reconstruction of the specimen exit-surface wave from Philips -- as specified in the original OÅM specifications.
  - we have successfully implemented remote-access capability for the CM300 and it is now available to users via the Materials Microcharacterization Collaboratory.
  - in addition to the remote-access capability for the CM300, remote access will soon be available for the MAL software -- thus providing full remote capability for the OÅM.



- The OÅM project is designed to achieve sub-Ångstrom resolution by focal-series reconstruction from images obtained at a better information limit than any microscope previously available
- For sub-Ångstrom resolution, I foresaw a need for correction of an aberration (3-fold astigmatism, or A_2) that no one has corrected before. I showed Philips how to correct it by modifying the existing 2-fold stigmator coils. The CM300 was delivered with the stigmator coils modified to my specification, but with the control circuits still under construction.
- While he was working with a specimen of CV diamond in [110] orientation, YC Wang obtained a “weird” image -- it shows triangular shapes instead of showing the carbon atoms arranged in “dumbbell” configuration.
- When I Fourier transformed the image, the diffractogram showed that the orientation is [110], with the 400 spot just visible. Averaging of the image brought out the triangular shapes of 3 white dots per atom pair.
- I ran some simulations of diamond in [110] orientation, using the microscope parameters Crispin and I had measured for the OÅM, but with the addition of 3-fold astigmatism. Since I had to test direction as well as magnitude, it took several hundred simulations in steps of 5 degrees and 50nm.
- My best match with the averaged experimental image occurred when I used two and a quarter microns of 3-fold astigmatism.
- It is encouraging that the 400 spot is visible, with a spacing of 0.89Å.

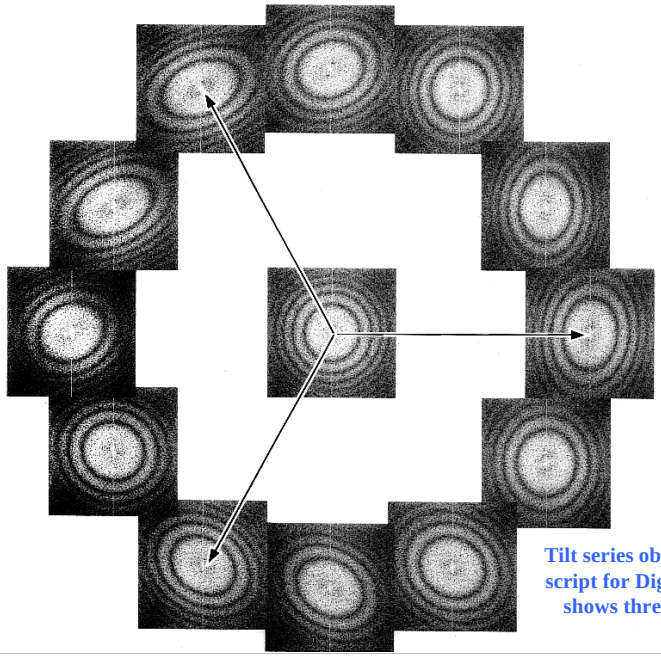


- The simulation shows the diamond "dumbbell" image we expected the OÅM to be able to produce -- once the corrector controls had been supplied by Philips and A_2 had been reduced to meet the OÅM specification of 500Å.
- The OÅM contrast-transfer function on the left shows how transfer should be possible out to 0.89Å with a spread of focus as high as 25Å (the spread of focus should be closer to the specified 20Å once Philips meets the voltage ripple spec by replacing our HT tank with the improved model).
- The simulated image corresponding to the OÅM parameters shows clear separation of the carbon atoms spaced at 0.89Å.
- The many oscillations in the contrast transfer function demonstrate why focal-series reconstruction of the image (actually of the electron wave at the specimen exit surface) is required for single-image resolution of complex structures to the OÅM's sub-Ångstrom information limit.
- Focal-series reconstruction (FSR) of the exit-surface wave (ESW) is done with the Brite-Euram software purchased from Philips as a component of the OÅM order. The software is a combination of the PAM (parabolic method) and MAL (maximum likelihood) developed at Antwerp, and now maintained and extended by Andreas Thust working as a consultant for Philips.
- The IBM workstation originally specified for this software was superceded in the OÅM order by the Fischione plasma cleaner, but Andreas Thust agreed to my purchase of a DEC Alpha (Durango) as a suitable replacement, and will install his software on that.



- The OÅM should be capable of imaging C-C "dumbbells" at 0.89Å spacing in diamond. This is supported by the presence of the 400 spot in experimental diffractograms as well as image simulations for OÅM parameters with $A_2 = 0$.
- To confirm the correct spacings in the astigmatic image, I used Crisp™ to "correct" the phasings by imposing known the symmetries of the specimen.
- Imposition of a left-right mirror on the averaged image produces slightly asymmetric black pairs of spots, which can then be symmetrized into perfect 0.89Å dumbbells by the addition of a top-bottom mirror. Of course this result is too "over processed" to be generally applicable to other structures.
- I processed the image to show that imposition of the correct diamond symmetry would change it into an image with 0.89Å spacings present in the correct positions for the diamond structure, and to confirm that the "weird" symmetry was due to the 3-fold astigmatism I had predicted would limit OÅM resolution.
- I showed this slide in my presentation at the XIVth International Congress for Electron Microscopy in September 1998. At this meeting I also met with Philips and used this result to urge them to supply the 3-fold controls I had been asking for since the delivery of the OÅM (the microscope was delivered with the standard 2-fold stigmator coils modified as I had instructed, but the 3-fold control circuits had not yet been built and tested).
- In October (1998), the controls were delivered and installed and we were able to correct the 3-fold astigmatism!

Pre-corrected OÅM diffractograms with 2250nm of 3-fold astigmatism.



Tilt series obtained with NCEM script for Digital Micrograph™ shows three-fold symmetry.

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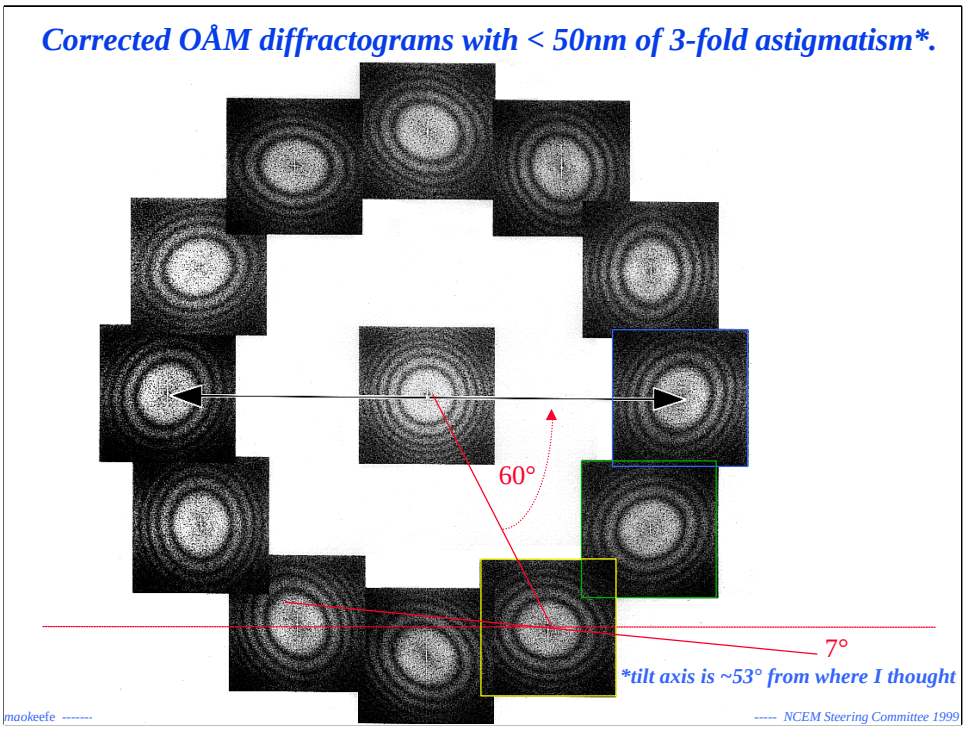
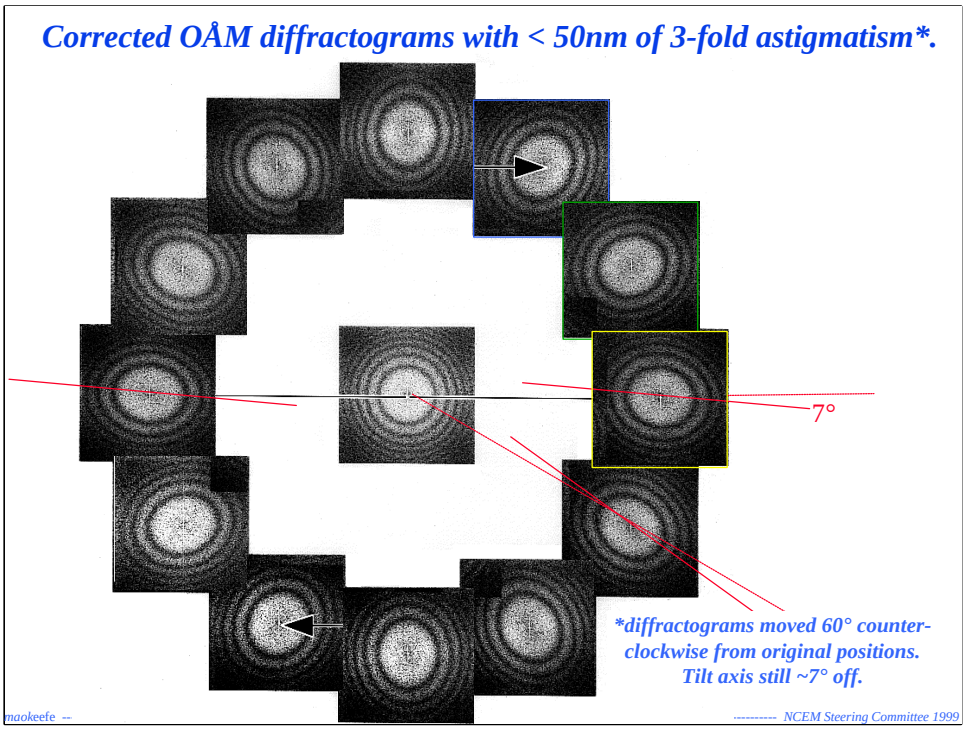
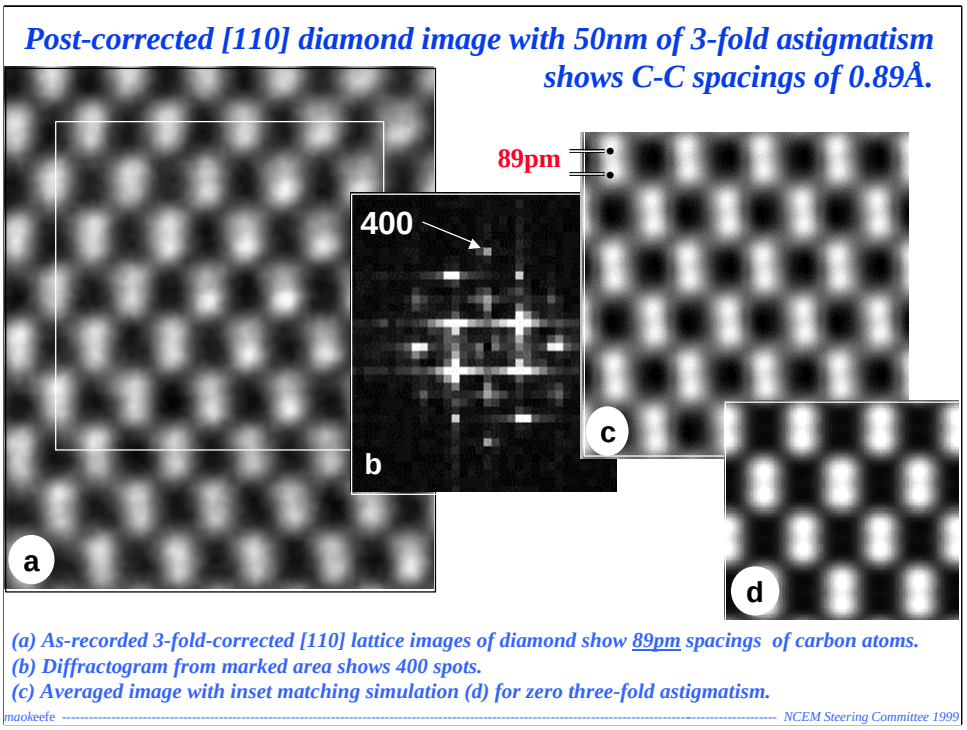
- As well as using the 3-fold astigmatic diamond image, we also measured the value of 3-fold astigmatism by using amorphous specimens of carbon. I wrote a Digital Microscope™ script to control the microscope and CCD camera and produce a series of tilted diffractograms. Then I worked with Crispin Hetherington to debug it – Crispin operated the microscope to find the beam as my initial attempts at tilting it sent it off the screen!
- The script tilts the beam, captures the image on the CCD, Fourier transforms it, then places the center area of each diffractogram in the correct position within the composite.
- The slide shows a tilt series before correction of the 3-fold astigmatism. The arrows show the 120 degree rotational symmetry.
- Using Gatan's Digital Microscope™ plug-in, YC and I measured the 3-fold astigmatism six times for a mean value of 2.46 micron. This value is some 9% higher than the value of 2.25 micron that I found to best match the diamond image.



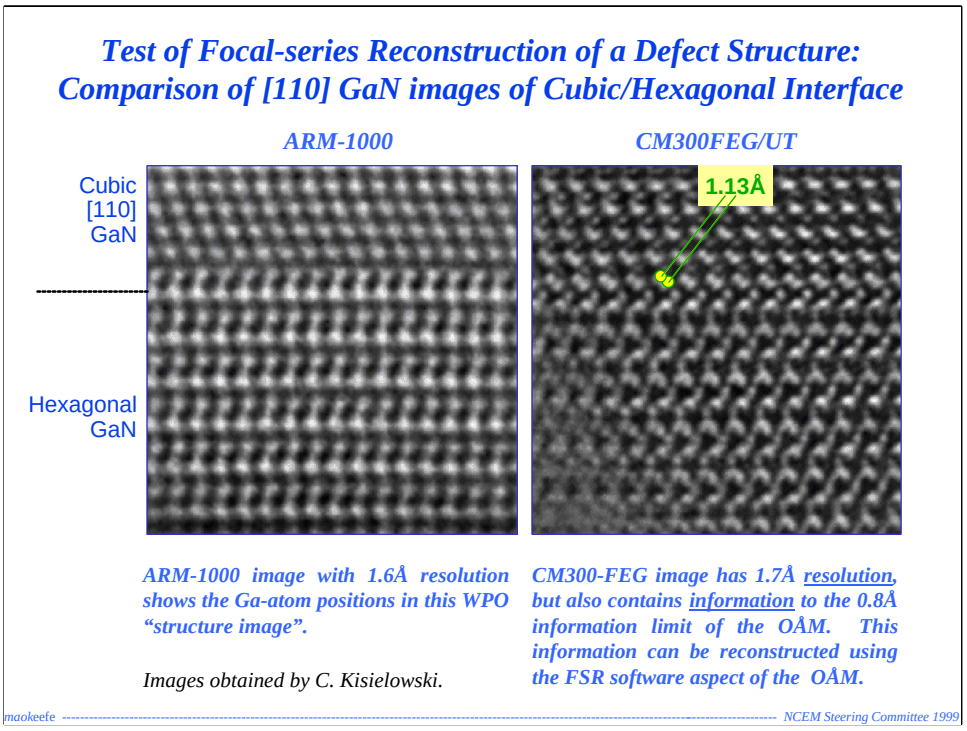
- The 3-fold corrector box arrived one month after my Cancun meeting with Philips, at which I showed them my processed version of YC's diamond results and stressed the possibilities for sub-Å resolution with the OÅM.
- I scheduled Bob Mueller to install the corrector, but there was a delay while Bob had to modify the box to add eight potentiometers to control the coils directly. Then Bob and Jan Ringnalda made the adjustments while I ran the Gatan software and measured the changes in the value of A_2.
- Final results showed 50nm of residual 3-fold astigmatism -- exactly meeting the specification I had placed into the original microscope specification list, and allowing spacings at 0.8Å to contribute to the image with a phase distortion of $\pi/4$.
- After correction, I worked with YC and Ming Pan (Gatan) to obtain and display a series of amorphous-carbon diffractograms using my DM™ script.
- Although the series showed no 3-fold character (120 degree symmetry), it was not clear that the axes of the ellipses had the correct symmetry. In fact, it turned out that my DM™ script positioned the diffractograms on the "page" at about 53 degrees from the correct positions.



- The incorrect positioning of the diffractograms can be fixed (approximately) by moving each diffractogram two positions counter-clockwise (by 60 degrees).
- Now the diffractograms all have their long axes pointing toward the center of the tilt "clockface" (actually 7 degrees off).
- Three-fold astigmatism limits resolution by applying a rogue phase change that distorts the image (as for the triangular "dumbbells"). The phase change is given by $(2\pi/3)\ A_2\ \lambda^2\ |\mathbf{u}|^3 \cos 3(\theta-\theta_2)$ where A_2 is the coefficient of three-fold astigmatism and $\cos 3(\theta-\theta_2)$ represents the 120º azimuthal periodicity. For a value of A_2 of 500Å, the phase change at 1Å is 0.13π.
- Generally, the resolution limit from three-fold astigmatism is recognized to be where the phase change equals $\pi/4$. This limit is given by $(8A_2\lambda^2/3)^{1/3}$. For a value of A_2 of 500Å, the limit is 0.80Å.



- With the 3-fold astigmatism now corrected to a level of 0.8Å, the OÅM should be able to image structures in the sub-Ångstrom range, so I looked for a test specimen with atoms separated by 0.8 to 0.9Å.
- The diamond "dumbbell" spacing is 0.89Å, so diamond is obviously the test specimen of choice. To test the OÅM, I had YC obtain a series of images from the [110] diamond specimen. We used defocus values out near -3000Å so as to optimize transfer at spatial frequencies close to 0.89Å (as I showed in the CTF describing the conditions I used for the simulation).
- One of YC's images is shown. It clearly demonstrates the 0.89Å splitting of the white areas into two white dots separated by 0.89Å.
- I was also very happy to see a much stronger 400 spot when I transformed the image to get the diffractogram. The spot is stronger because the interference of the 400 beam with the central 000 beam now has the same phase as the interference of the -400 beam with the 000 -- previously the 3-fold astigmatism contributed an opposite phase to the 400 and -400, since they are 180 degrees apart and 3-fold means a 120 degree azimuth.
- The averaged image is clearer than the original, suggesting that the specimen is damaged or has a substantial amorphous contribution. In addition, the averaged image shows a little clearer resolution than the simulation -- suggesting that the spread of focus may be even less than 25Å, making the information limit better than 0.89Å.



- Although a single image from a simple structure like [110] diamond can demonstrate the resolution of the OÅM, it takes a reconstruction from a focal series to realize the full resolution in the case of a defect structure.
- The left image shows an image of the interface between cubic and hexagonal GaN taken by Christian Kisielowski using the JEOL ARM-1000. The 1.6Å resolution of the ARM is insufficient to separate the Ga and N atoms (spaced at 1.13Å), so we see them as single dots with positions close to the "center of gravity" of the Ga-N pair (i.e. close to the Ga atom).
- The image on the right was obtained by Christian on the CM300FEG/UT. It obviously contains higher-frequency information (details are closer together). However, this information cannot be related directly to the structure, except in some areas (marked with yellow dots) in which important reflections are transferred with the correct phases by the highly-oscillatory CTF. In particular, the exact position of the interface is unclear.

Reconstruction from [110] GaN focal series shows Ga and N atoms at a resolution of better than 1.13Å

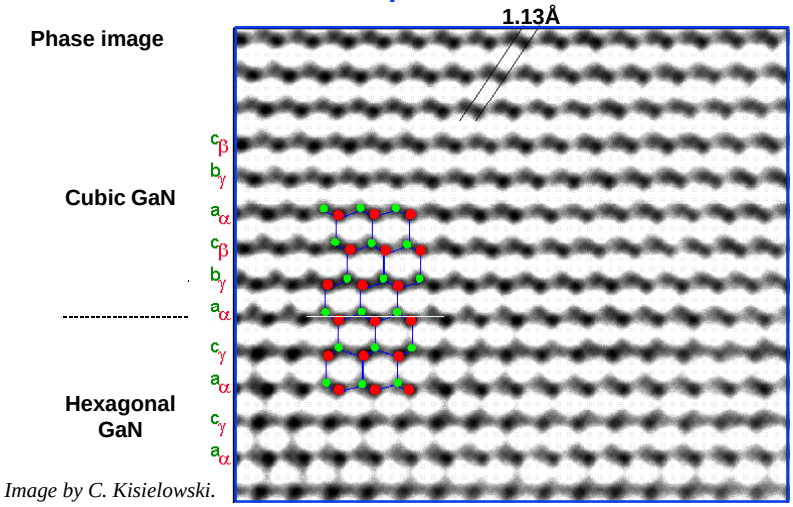


Image by C. Kisielowski.

Reconstruction of the phase of the electron exit wave of the 3C/2H GaN interface from a focal series of 20 images obtained on the One-Ångstrom Microscope. Ga and N sub-lattices are clearly resolved at a separation of 1.13Å, and are distinguished by different contrast at the (darker) Ga and (lighter) N sites. This contrast is maintained across the 3C/2H interface.

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- The mixed-phase CM300 result can be converted to a simple projection of the defect structure at sub-Å resolution by using the OÅM's software component.
- For a thin specimen (phase object), the scattered electron wave can be written as $\psi(x,y) = \exp\{i \sigma \phi_p(x,y)\}$, where σ is the cross-section for scattering and $\phi_p(x,y)$ is the specimen potential projected in the incident beam direction. The phase of the scattered wave is directly proportional to the projected potential.
- The "image" shows the phase of the exit-surface wave reconstructed by the MAL software from 20 images obtained over a range of focus on the OÅM. The positions of all the atoms (both the heavy Ga and the lighter N) can now be seen, and the interface structure precisely determined.
- This is the first result from the OÅM using its combined resources -- the CM300 (hardware component) and the MAL reconstruction (software component). Although the reconstruction step was slow (15mins), it will become much faster once the software is transferred from Kisielowki's Silicon Graphics workstation to the much faster (3x) OÅM computer. This computer (Durango) also provides much more storage for image data than the SGI.
- Since the CM300 has been made available for remote use by NCEM users, it makes sense to provide the full OÅM capability by making the MAL similarly accessible. Philips has agreed to my providing remote access by NCEM users to the MAL once the software has been moved to Durango.
- With the MAL working, the next test of the OÅM will be to obtain and reconstruct a focal series of diamond images to demonstrate that 0.89Å resolution is possible from non-periodic and defect structures.

Notes on the Acquisition and Disposition of the Brite-Euram Software for Focal-series Reconstruction of the Exit-surface Wave

- The Brite-Euram software for focal-series reconstruction combines the PAM (parabolic method) and MAL (maximum likelihood). Developed at Antwerp as part of the Brite-Euram project, it is maintained, and is being extended, by Andreas Thust in Jülich working as a consultant for Philips.
- Whilst working to get the OÅM's 3-fold astigmatism corrected, I was also working to have the software installed on the NCEM's OÅM computer.
- Originally, the OÅM computer was to be an IBM workstation provided by Philips to run the reconstruction software, and was so specified in the original OÅM order. However, funds for the IBM were subsequently traded for a Fischione plasma cleaner to combat contamination problems. In 1996, I found a way to fund a replacement OÅM computer by using $15K that I had available from a DOE prize. I consulted with Andreas, and he agreed to my purchase of a DEC Alpha (Durango) as a suitable OÅM computer to run his software.
- However, in November last year, Andreas told me he was unable to install the software on Durango. I then arranged with Andreas and Philips (Sheri Kurland, Ben Bormans and Frank de Jong) to meet in January of 1999 to arrange a way to get the NCEM copy of the FSR software working on the OÅM computer.
- At the meeting, I found out why there was a problem and agreed to install Unix as the operating system on Durango instead of the current WNT so that the FSR software could run using some routines that were only available under Unix. At this point Kisielowski made the suggestion that we temporarily install the software on his personal SGI since it ran Unix. Philips and Thust agreed to a short-term installation on the SGI with a transfer to the OÅM computer once I had it converted to run under Unix, just as long as we had only one copy of the software at any one time.
- Unfortunately, Uli Dahmen has not yet authorized the funds for conversion of the operating system of the OÅM computer from WNT to Unix. At the moment, the OÅM computer remains idle, and the only access to the NCEM's copy of the FSR software is via Christian Kisielowski's much slower SGI. Dahmen has told me that access to the software will remain this way for one year. Once this year has passed, we will be able to move the software to the OÅM computer and enjoy much faster reconstructions with fewer storage constraints for image data, as well as remote access to the OÅM's Brite-Euram reconstruction software.

Acknowledgement

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