WORKSHOP REPORT

ABERRATION CORRECTION IN ELECTRON MICROSCOPY

MATERIALS RESEARCH IN AN ABERRATION-FREE ENVIRONMENT

July 18–20, 2000

Materials Science Division
Argonne National Laboratory

Sponsored and Organized

by

Electron Microscopy Center (EMC)
Argonne National Laboratory

National Center for Electron Microscopy (NCEM)
Lawrence Berkeley National Laboratory

Center for Microscopy and Microanalysis (CMM)
Frederick Seitz Materials Research Laboratory

Shared Research Facilities Program (SHaRE)
Oak Ridge National Laboratory

Table of Contents

Acknowledgment

The organizers gratefully acknowledge significant contributions to the success of the Workshop from FEI Co. (Philips), LEO Electron Microscopy, Inc., Nissei-Sanjo America, Ltd. (Hitachi), and JEOL USA, Inc. We are also grateful for the participation of their representatives in the Workshop.

ABERRATION CORRECTION IN ELECTRON MICROSCOPY
MATERIALS RESEARCH IN AN ABERRATION-FREE ENVIRONMENT

Foreword

The following is taken from the Executive Summary of the National Transmission Electron Achromatic Microscope (NTEAM) Vision Document prepared for the Basic Energy Sciences Advisory Committee (BESAC) Subpanel Review in Fall 1999:

> "Thanks to advances in aberration correction and quantitative transmission electron microscopy, a new generation of microscope can be built, capable of sub-Ångstrom image-resolution and sub-electron-volt spectroscopic-resolution with adequate space to carry out a variety of important experiments on advanced materials. The project, to build a National Transmission Electron Achromatic Microscope (NTEAM), could involve a cooperative instrumental development at the four DOE National Centers for electron beam micro-characterization, with each contributing a complementary specialized facility, based on a common platform. The envisioned revolutionary combination of space and resolution will allow the electron microscope to be converted into a true experimental materials science laboratory. Scientific impacts to be expected include: the first 3-D atomic imaging of defect structures; the first atomic structure determination of a glass; microscopic understanding of magnetism and ferroelectricity in nanostructures; visualization of dislocation interactions in nanostructures under controlled stress; development of interface science to the level of surface science; understanding of grain boundary motion under stress in nanocrystals; understanding chemical reactions on highly-curved small catalyst particles; and imaging defects in the oxygen sub-lattice of complex oxides. Developments which we imagine here in electron beam microcharacterization would be crucial for proper implementation of the national thrust in nanotechnology. Furthermore, the project would help to revitalize the critically important electron optics industry in the United States."

In its final report of the review, the BESAC Sub-Panel subsequently endorsed the concept in principle.

Executive Summary

Over the past decades, emphasis in the improvement of electron beam microcharacterization instrumentation in general and of transmission and scanning transmission electron microscopes (TEM and STEM) in particular has been on improving electronic stability (lens currents and high voltage), electron source size and coherence (directly heated W, indirectly heated LaB_6 and thermally assisted and cold field emission sources), vibration isolation for mechanical stability, improving manufacturing tolerances and lens design (decreased focal length of objective pole pieces), and increased accelerating potentials. These evolutionary improvements have resulted in a wide variety of advances in the materials and life sciences, ranging from direct structure imaging and vastly improved microcharacterization of metals, semiconductors, ceramics and soft materials, to the discovery of carbon nanotubes. While we may expect further incremental improvements in electronic stabilities and in electron sources, especially cold field emission, the great frontier of electron beam instrumentation development is the correction of image and electron probe aberrations, which would ideally allow aberration-free imaging and microanalysis to the atomic scale. While such advances are often thought of in the context of high spatial resolution techniques, they are of no less importance in the context of in situ experiments requiring a reasonable volume within which a kind of dynamic microlaboratory can be installed and within which experiment and and analysis proceed simultaneously. Increased space within the TEM objective pole piece would be one important direct result of reduced instrumental aberrations for a given spatial resolution.

In the course of preparing in 1999 for a review of the four Electron Beam Microcharacterization Centers, supported by the U. S. Department of Energy, by a Sub-Panel of the Basic Energy Science Advisory Committee (BESAC), a Vision Document suggesting a national project for the development of a series of TEMs and/or STEMs which would be as fully corrected as possible for both spherical and chromatic aberrations was prepared, capitalizing on the increased available experimental space concept. Authored principally by J. Murray Gibson, the document was augmented and ratified by the management of the four Centers (Electron Microscopy Center, ANL; National Center for Electron Microscopy, LBNL; Shared Research Equipment Program—ShaRE, ORNL; Center for Microcharacterization of Materials, MRL-UIUC). In its final report of the review, the BESAC Sub-Panel subsequently endorsed the concept in principle. This Workshop is the first step toward implementation of such a national project. The full name of the Workshop (Aberration Correction in Electron Microscopy—Materials Research in an Aberration-Free Environment) emphasizes the two essential aspects involved in the development of such a project, the instrumental aspects and the impacts of such instrumentation on science.

Thus the purpose of this Workshop and of subsequent related gatherings is really threefold:

- To identify optical approaches for ideal in situ and high resolution electron microscopy and microanalysis
- To identify scientific imperatives for instrumentation development and
- To form partnerships of individuals and institutions and to establish procedural strategies.

The success of these three goals can result in the presentation of a very strong proposal for instrumentation development which will push the technological envelope and inspire scientific imagination for future materials research.

The technology exists now to completely correct spherical aberration in electron probes for STEM and in images formed in TEM. The next great instrumental challenge is the

correction of chromatic aberration which so far has been done only in a low voltage scanning electron microscope (SEM). This is precisely the challenge posed by the National Transmission Electron Achromatic Microscope (NTEAM) concept.

The Agenda for the Workshop is presented in Appendix A. Appendix B is an alphabetical listing of participants and their affiliation information. Appendix C reproduces the 1999 NTEAM Vision Document which was available to the participants during the Workshop.

The body of the Workshop Report includes a mixture of verbatim and paraphrased accounts of the participants' presentations and discussions. In addition, it contains a certain amount of interspersed editorial content which is intended to improve the document's readability and to promote its usefulness.

Overview of the Workshop: Murray Gibson

The Workshop opened with a wide-ranging overview talk by Murray Gibson in which he outlined the structure of the Workshop and addressed a large number of issues and possibilities related to aberration-corrected TEM and STEM. This section of the Report is based on Murray Gibson's talk, incorporating some information from various other participants as well, relating to the overview.

The mechanism for aberration correction was suggested about fifty years ago by Scherzer with pioneering attempts to reduce it to practice by Crewe, Rose, Haider, Krivanek and others over the past thirty years or so. Essential to successful C_S correction is precise alignment of the corrector elements, which finally is possible today largely because of advances in computer technology. Two distinct paths have been and are being pursued for C_S correction hardware: for TEM, systems of hexapoles (Haider, Rose...Crewe) and for STEM, systems of quadrupoles and octupoles (Krivanek, Delby...). While the hexapole design exhibits relative simplicity, it is not simply extendable for C_C correction and has larger intrinsic C_C. The quadrupole/octupole design can be extended to C_C correction with addition of electrostatic elements (Wien Filter), but the configuration is much more complex and exhibits large off-axis aberrations; the latter is more suitable for STEM for which the effects of C_C are mitigated by high angle annular dark field imaging. In addition, STEM has the attraction that TEM and STEM are complimentary; for example, TEM offers high speed, real time imaging, whereas STEM is ideal for spectroscopic imaging. Each of these current alternatives is discussed at length by Max Haider and Ondrej Krivanek during the Workshop. In order to decrease the effects of C_C, current applications focus on use of a monochromator to limit the energy spread of electrons after acceleration, and this topic was also addressed by several participants.

There are a number of compelling reasons for aberration correction in both TEM and STEM. In HRTEM applications, C_S correction results in improved interpretability due to the non-oscillatory contrast transfer function of the instrument, allowing also improved image localization (for example at interphase interfaces [Haider *et al.*, Nature, 392 (768) 1998]) as well as the ability to offset the higher order spherical aberration coefficient, C_5, by varying C_S slightly from zero. In STEM applications, C_S correction affects the electron probe in two ways, resulting in a finer probe size with a larger *total* probe current, both of which are advantageous for spatial resolution in high angle annular dark field imaging as well as in microanalysis. Furthermore, when $C_S = 0$, objective current centering becomes relatively unimportant within beam tilt angles of several milliradians, with coma also corrected, so that fine tuning of the incident beam orientation with respect to the specimen will not degrade image resolution; this allows very precise diffraction conditions to be established locally, independent of the problems associated with mechanical tilting of the specimen. This can be of considerable utility in both HRTEM and CTEM, including for in situ studies, strain field imaging, atomic scale tomography and the like. Finally, when C_S is fully corrected, the requirement of short focal length objective pole pieces to achieve high point-to-point image resolution is relaxed. This allows larger pole piece gaps which are especially appealing for a wide variety of in situ experiments. Kabius has shown, however, that, in the absence of C_S, C_C increases approximately linearly with objective pole piece gap dimension and thus remains an important consideration for correction, especially for in situ applications, for which a large gap is very important. For example, for a lens of focal length ~1 cm, point-to-point resolution ~ 0.5 nm results if the incident energy spread is 0.5 eV at 200 kV. (Because there was relatively little discussion of the details of C_C correction during

the Workshop, the question of C_c correction with or without monochromators remains one for extensive future consideration and certainly represents a major longer term challenge.) Further information is given in the thesis of K. Xiu (University of illinois—Urbana-Champaign, 2001).

As was mentioned above, a major thrust of the four DOE-sponsored Electron Beam Microcharacterization Centers is toward development of a project of national scope based on the National Transmission Electron Achromatic Microscope (NTEAM) concept. It is this provisional project which has lead to organization of this Workshop. The NTEAM Vision Document prepared for the BESAC Subpanel Review in Fall 1999 was made available to Workshop participants.

A series of modular instruments having 200 or 300 kV accelerating potentials is thus envisioned, initially taking advantage of current developments in spherical aberration correction in order to increase the objective pole piece gap for more complex in situ experiments and to accommodate more efficient detector systems for chemical and elemental microanalysis. This should allow a point-to-point resolution for imaging of 0.1 nm with a 1 cm gap. While the difficulties of designing a C_s corrected TEM/STEM have not been seriously examined, such versatility would appear to be very attractive to the user research communities involved. In order to record dynamic in situ information, TEM must usually be employed; on the other hand, for high spatial resolution elemental and chemical microanalysis, STEM must be employed, utilizing a very fine electron probe. For both TEM and STEM, subsequent incorporation of modular C_c correction could improve spatial resolution to the sub-Å level while allowing a several cm gap. To promote experimental innovation, parallel development of MEMS technology is also proposed, e.g., to null the lens field in a 1 mm^3 volume for high resolution magnetic imaging.

As a part of his overview presentation, Murray Gibson briefly compared several aspects of electron, neutron and photon scattering techniques and facilities, emphasizing their complementarity, the importance of which is not widely appreciated within the scientific community or its funding agencies. Several interesting characteristics of these three types of radiation are summarized in the Table 1.

Table 1. Comparison of characteristics: neutrons, X-rays and electrons

Radiation	Source Brightness (particles/cm^2/steradian/eV)	Elastic Mean-Free Path (Å)	Absorption Length (Å)	Minimum Probe Size (Å)
Neutrons	10^{14}	10^8	10^9	10^7
X-rays	10^{26}	10^4	10^6	10^3
Electrons	10^{29}	10^2	10^3	1

Again from the NTEAM Vision Document is the following:

> "The brightness of the electron sources is higher than that of undulators on third generation synchrotrons, and significantly higher than that of neutron sources. In addition, the electron signal from a tiny sample is increased even further because the electron elastic-scattering mean-free path is very short, so that one gets on the order of a million times greater signal from a single atom than with the brightest x-ray sources. The strong scattering of electrons is due to the Coulomb interaction, which is also the basis for powerful electron optics. This explains why electrons are uniquely useful for microcharacterization at the atomic scale using microscopy and spectroscopy.
>
> Of course, the weak atomic scattering for neutrons and x-rays has the advantage of straightforward interpretation because the simpler kinematical theory applies. But recent progress with computation has made inversion of dynamical theory for structure factors practical (see a beautiful recent example [J.M. Zuo, M. Kim, M. O'Keefe and J.C.H. Spence, "Direct observation of d-orbital holes and Cu-Cu bonding in Cu_2O", Nature 401 (1999) 49–52.] and dynamical scattering has the advantage that full symmetry information is preserved [Spence, 1992 #375]. And when one wants to study localized structure in three dimensions, microcharacterization by electron microscopy and electron microscope-based spectroscopy is the only choice. Improvements in quantitative measurement and fitting promise that electron scattering will take its full place as both a complementary and unique technique for materials characterization."

Gibson concluded that, with regard to aberration correction in general and the NTEAM project in particular, an exciting challenge exists which requires a medium scale effort analogous to the large scale effort involved in creation of a next generation synchrotron, but significantly less costly. To this end, we need to tap into the value of electron microscopy as a collection of complementary experimental tools in materials science which are not always off-the-shelf commodities. In such an effort of national proportions as NTEAM, the network of national laboratories and university and industrial partnerships must come together and move in a common direction.

Development of Hardware for Aberration Correction

Over the past five years the potential of aberration correctors incorporated into electron microscope columns has been clearly demonstrated to improve their spatial resolution beyond the theoretical, aberration-limited values of the uncorrected instruments: in 1994 with the correction of spherical aberration of a 200 kV Philips CM20 (LaB_6) [M. Haider, G. Braunshausen, E. Schwan, Optik 99 (1995) 167–179], in 1995 with the correction of spherical and chromatic aberration of a low-voltage scanning electron microscope [J. Zach, M. Haider, Nucl. Instr. and Meth. A 363 (1995) 316]; in 1997, of spherical aberration of a Philips CM 200 FEG ST at Jülich [M. Haider, H. Rose, S. Uhlemann, E. Schwan, B. Kabius, K. Urban, Ultramicroscopy 75 (1998) 53–60]; and in 1999, of spherical aberration of a dedicated STEM, a VG HB5 at Cambridge [O. L. Krivanek, N. Dellby, A. R. Lupini, Ultramicroscopy 78 (1999) 1–11]. In addition, shortly before this Workshop, testing of the VG HB501 at IBM Watson, equipped with a modified Krivanek corrector, began. Additional development projects at the time of the Workshop include spherical aberration correction of at least three VG HB5's or HB501's, of a VG HB603 and of a new TEM or

TEM/STEM in the USA and of the three evolutionary SESAMe TEMs and the SÅTEM TEM in Europe (The latter two projects are briefly described in the presentation by Bernd Kabius).

Two presentations were devoted to the application of aberration correction theory to hardware development, as represented first by the two principal players in the commercial arena, Max Haider (CEOS GmbH., Heidelberg) and Ondrej Krivanek (Nion, Inc., Kirkland, WA), both of whom presented technical overviews of their respective subjects. It is well beyond the purposes of this Report to attempt to present a review of the electron optical theory required to fully appreciate the complexity of factors limiting resolution in electron optical instrumentation, but we do attempt to stress those elements from these two presentations which appear to be particularly relevant to the purposes of this Workshop.

Correction Schemes for TEM with Comments on STEM: Max Haider

Of the myriad of aberrations which we usually distinguish in classical optics, microscopists have dealt with "defocus" (aberration coefficient $C_1 = \Delta f$) and (two-fold) "astigmatism" (aberration coefficient A_1) of images in the TEM for many years, optimizing focus by adjusting objective lens current and astigmatism by a small quadrupole lens following the objective. The corresponding operations in STEM (and SEM) are aimed at minimizing the size and asymmetry of the incident electron probe. Other important optical aberrations include "chromatic" aberration associated with the energy spread of the electron beam (C_c), three-fold astigmatism (A_2), axial coma (of second order, B_2 , and of fourth order, B_4) and spherical aberration ($C_3 = C_s$ and C_5). The influence of chromatic aberration has been considerably reduced by introduction of cold field emission and Schottky electron sources and by improved stability of high voltage and lens power supplies. Correction of three-fold astigmatism (A_2) and axial coma (B_2) are available commercially, generally as options. Just as in uncorrected high resolution TEM C_s is partially mitigated by appropriate defocus, C_5 can also be partially mitigated by small changes of C_3 and defocus from zero in a C_s-corrected TEM.

As the Scherzer theorem states (1936), spherical aberration cannot be avoided in rotationally symmetric electromagnetic fields (round lenses). In 1948, however, Scherzer proposed a hardware corrector for spherical and chromatic aberration, consisting of multipole lenses to which the theorem does not apply because the fields are not rotationally symmetric. Several attempts to improve the optical performance of a TEM employing Scherzer's suggestions failed, however, not the least of the reasons being the extraordinary complexity of aligning by hand the instrument including the corrector system [H. Hely, Optik 60 (1982) 307 and 353].

By way of introduction, Haider reminded the audience of the possible techniques for achieving 0.1 nm point-to-point image resolution, namely, focal series reconstruction, electron holography, reduction of electron wavelength (HVEM), and correction of C_s (=C_3) along with reduction of C_cDE. He then reviewed the two basic systems for aberration correction, which differ in principle, the original hexapole corrector system for STEM, essentially as proposed by Rose and Crewe [V. D. Beck, Optik 53 (1979) 241 first mentions third order axial aberration of hexapole fields; H. Rose, Nucl. Instr. and Meth. 187 (1981) 187; A. V. Crewe, Optik 55 (1982) 271] and the quadrupole(/octapole) system for TEM proposed by Rose [H. Rose, Optik 33 (1971) 1]. (It should be noted that the correctors described by Krivanek, Dellby and Lupini for STEM are not hexapole correctors but rather quadrupole/octapole correctors.) Dr. Haider then summarized the state of affairs

with respect to correction of C_3 (=C_S), the type of corrector and the type of electron microscopy, as shown in Table 2 which also includes three other corrector systems .

Table 2. Aberration correction for SEM, STEM, TEM and LEEM.

Correction of Aberrations

	SEM	**STEM**	**TEM**	**LEEM**
Spherical C_3	-	**+**	**+**	-
Spherical & Chromatic $C_3 + C_c$	**+**	+	**?**	**+**

Type of Corrector

		SEM	**STEM**	**TEM**	**LEEM**
Hexapole	C_3	**–**	+	**+**	**–**
Quadrupole	C_3	**–**	**+**	**+**	**–**
Electrostatic Mirror	C_3 C_c	+	**–**	**–**	**+**
Purely Electrostatic Quad.	C_3 C_c	+	**–**	**–**	**?**
Quadrupole Electr./Magn.	C_3 C_c	**+**	+	–?	**–**

Key: **+** = effective and already demonstrated;
+ = feasible, but not yet demonstrated;
– = not feasible or not useful;
? or **?** = questionable or very questionable

Focussing on TEM, Dr. Haider compared the expected point-to-point resolution of commercially available 200 kV TEMs, a prototype corrector which has been successfully developed, resulting in improvement of point resolution from 0.24 to 0.13 nm, and of 300 kV TEMs. At 200 kV achieving 0.1 nm would only be possible with addition of a monochromator. On the other hand for a Cs-corrected 300 kV TEM with FEG, 0.1 nm resolution should be achieved without a monochromator; however, the Cs corrector for 300 kV is more difficult to construct and does not yet exist.

The suitability of a particular TEM for addition of aberration correction for the attainment of resolution < 0.1 nm depends on a number of factors including the following:

1. Information limit of the uncorrected instrument should achieve its theoretical limit.
2. Lens and high voltage power supplies should be state-of-the-art with respect to regulation and stability.
3. Mechanical design of the instrument and its environment should be minimally sensitive to acoustical and other mechanical vibrations.
4. There should be an optimum number of alignment coils.

The operation of a corrector itself requires, in addition, a computer and slow scan CCD camera, electron optics simulation software and pattern recognition software for

diffractograms in order to perform the necessary routine, high precision alignments. Fig. 1 is a drawing of the objective and first intermediate lens section of the TEM (the Philips CM20 (FEG) at the European Molecular Biology Laboratory (EMBL), Heidelberg) with and without the hexapole-field

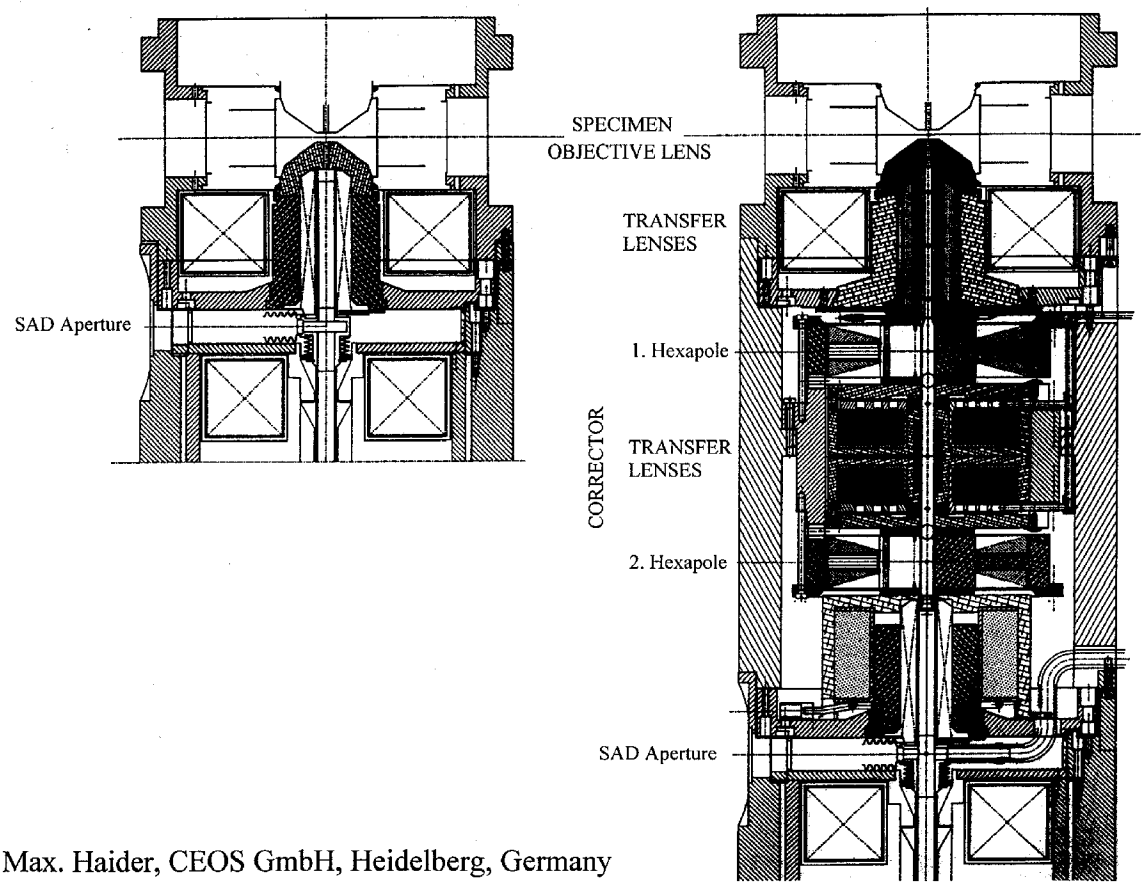


Fig. 1. Drawing of the column of the Philips CM 20 TEM showing the original set-up before and after insertion of the corrector. (Reprinted from Haider *et al.* , Optik 99 (1995) 167–179 with permission from Urban Fischer Verlag.)

corrector installed. The corrector consists of two "twelvepoles" and two sets of transfer lenses and increases the length of the column by 24 cm. A very interesting account of the design, benchtop testing and modification, and final testing as installed in the EMBL TEM has been presented [M. Haider, G. Braunshausen, E. Schwan, Optik 99 (1995) 167–179]. The very strict requirements on precision of alignment cannot be overemphasized. Employing a modification of a method suggested by Zemlin et al. [F. Zemlin, K. Weiss, P. Schiske, W. Kunath, K. -H. Herrmann, Ultramicroscopy 3 (1978) 49] to illustrate this point, Dr. Haider outlined a multiply iterative alignment procedure as follows:

1. Digital image acquisition (amorphous specimen) and calculation of diffractogram for initial alignments at zero beam tilts relative to the coma-free axis.
2. Deduction of defocus C1 and astigmatism A1 out of diffractogram
3. Repeat 1 and 2 for a set of small beam tilts. The set of diffractograms thus generated is arranged to form a "Zemlin tableau" in which the position of each diffractogram reflects the beam tilts for which the corresponding image was captured.
4. Calculation of aberration coefficients from the tableau data.

5. Calculation of new alignment settings based on electron optics simulations for minimizing the relevant aberrations.
6. Acquisition of a new tableau with larger beam tilts and so on depending on the required resolution.

The algorithm for analysis of tableau data covers a wide range of aberration magnitudes. Uhlemann and Haider have discussed this process in more thorough detail [S. Uhlemann, M. Haider, Ultramicroscopy 72 (1998) 109–119].

Dr. Haider next briefly addressed the issue of the Cc -corrector. To achieve high spatial image resolution or large energy windows for microanalysis a Cc -corrector will be required for TEM but is much less important for STEM. On the other hand if large pole piece gaps are to be employed to accommodate X-ray detectors and apparatus for in situ experiments, then a Cc -corrector becomes imperative for both TEM and STEM. This is highly relevant to the NTEAM-type instrument. One major problem is the very high stability required of current and voltage supplies for effective Cs-correction as well as for Cc-correction. For sake of comparison, he showed the following table of requirements of present and future instrumentation with respect to Cs -correction (Table 3).

The parameters in Table 3 refer to illumination half angle, precision in defocus and first order astigmatism correction, and the objective lens current and high voltage stabilities. The SÅTEM series of progressively improved C_S – corrected TEMs are currently under development by CEOS GmbH. The design

Table 3. Instrumentation requirements for C_s correction.

Microscope	q_A mrad	Ô DC_1Ô , Ô DA_1Ô nm	DI/I	Ô DF_2/F_2Ô
CTEM	10	10	$5x10^{-6}$	
SÅTEM I	30	1	$5x10^{-7}$	
SÅTEM II	36	0.8	$4x10^{-7}$	$8x10^{-8}$
SÅTEM III	50	0.4	$2x10^{-7}$	$2x10^{-8}$

of a C_C-corrector is non-trivial; however, such a corrector has been proposed for a medium voltage TEM by Rose [H. Rose, Optik 85 (1990) 19–24], and a functioning system for SEM already exists [J. Zach, M. Haider, Nucl. Instr. and Meth. A 363 (1995) 316] which cannot simply be scaled to STEMs of significantly higher energy, however. According to Haider, a minimum of four multipole elements is needed for a C_S and C_C -corrected STEM. There is a major improvement in the probe profile for a given probe convergence with C_C correction with the near elimination of the intensity in the long tail associated with the uncorrected probe. (Additional discussion of aberration correction in STEM was presented by Max Haider during Session 3 near the end of the Workshop.)

From an applications perspective, the bottom line is that C_S-correction accomplishes the following:

1. Point-to-point spatial resolution for imaging and diffraction (TEM and STEM) and for microanalysis (STEM) are improved.

2. An additional free parameter, C_3 (=C_S) is created, which, for example, can be employed to mitigate residual effects due to C_5 .
3. Peak intensity along with total integrated intensity of the incident electron beam are significantly increased.
4. Because of the reduced influence of lateral coherence, resolution is less sensitive to incident beam tilt in TEM so that orientation of the specimen with respect to the beam can be precisely fine tuned by beam tilt without resolution degradation.
5. Delocalization of object information which is proportional to C_S is strongly reduced; this is especially important in high resolution interface studies; image interpretation is thus simplified.
6. For simulation of high resolution images, there is improved measurement of imaging parameters.

Dr. Haider concluded that the most realistic way to achieve sub-Ångstrom resolution will be either to combine a C_S corrector with a 300 kV instrument or a C_S corrector and monochromator with a 200 kV instrument.

Aberration Correction in STEM: Ondrej Krivanek

For STEM imaging at high resolution, high-angle annular dark field (HAADF) has become a standard technique. The importance of spherical aberration correction in STEM can be demonstrated by comparing theoretical annular dark field resolution with and without C_s (=C_3) correction. The C_3 - limited resolution is given by

$$d_3 = 0.4\, C_3^{1/4}\, \lambda^{3/4}$$

which for 100 kV and C_3 = 1.0 mm is 0.19 nm and for 200 kV and C_3 = 0.5 mm is 0.12 nm. For the same instrument with C_3-corrector, the annular dark field resolution is limited by C_5 which gives

$$d_5 = 0.4\, C_5^{1/6}\, \lambda^{5/6}$$

which for 100 kV and C_5 = 100 mm is 0.08 nm and for 200kV and C_5 = 1 mm is 0.03 nm. Thus the resolution is improved by a factor of about 2 for present generation of aberration correctors. Once C_5 and other higher order aberrations are also brought under control, a resolution improvement of about 4x can be expected relative to uncorrected microscopes.

There are two other relevant reasons for pursuing C_3-correction in STEM, which are related to chromatic aberration and to probe current. In lattice imaging in TEM the effect of chromatic aberration may be partially mitigated by tilting the incident beam so that it is half way between 000 and the principal operating reflection g, thus taking advantage of the achromatic circle. In STEM annular dark field images, for every spatial frequency q there is interference between rays at $\pm$ q which also takes advantage of the achromatic circle. To put it another way, the phase difference due to chromatic aberration for these rays $\pm$ q is independent of defocus changes. Thus, inherently in annular dark field STEM the effects of chromatic aberration are sharply diminished.

Another important demonstration of the effect of C_3-correction, especially for EELS microanalysis, lies in the relationship of probe current to probe size for an uncorrected and a corrected STEM. For example, for a 100 kV STEM with source brightness of 10^9 A / cm^2 str and uncorrected C_3 = 1 mm, the probe size is about 0.4 nm at a probe current of 1 nA.

With the corrector on and C_5 < 50 mm, the probe size is reduced to ~0.13 nm, which means that single atom "nanoanalysis" would then be possible, even for 100 kV.

Aberration correction is a subject with a more than 60 year history [P.W. Hawkes, E. Kasper, Principles of Electron Optics, vol.2, Academic Press, New York, 1996, Chap. 41]. Table 4 is a brief summary of this history, which Dr. Krivanek has assembled and which he reviewed in some detail in his presentation. There have been a number of partial successes, such as Deltrap's quadrupole-octupole corrector which nulled spherical aberration in a probe-forming system more than 35 years ago by means of 4 combined quadrupole/octupoles [J.H.M. Deltrap, PhD Thesis, University of Cambridge, 1964; Proc. 3rd EUREM Congress, Prague, vol. 1 (1964) 45]. He had no interest, however, in applying his development to microscopy. As indicated in Table 4., twenty years later Krivanek and Dellby developed a variation of the Deltrap corrector which successfully improved the resolution of a VG HB5 at Cambridge, clearly demonstrating that the principle of aberration correction for improvement of resolution in STEM was sound. This development has been reviewed by Krivanek, Dellby and Lupini, including improvements for the next generation design for the VG HB501 at IBM Watson [O.L. Krivanek, N. Dellby, A.R. Lupini, Ultramicroscopy 78 (1999) 1–11]. A comparison of the principal electron optical elements and first order electron trajectories for the Cambridge and IBM correctors are shown schematically in the following Fig. 2.

In the original HB5 corrector design there are six combined quadrupole-octupole elements (identical elements with twelve poles each) , whereas in the design for the HB501 there are four quadrupoles and three octupoles which are spatially separate elements. The latter design makes it possible to operate the octupoles in (higher) moderate saturation without having to deal with changing first order trajectories due to quadrupole strengths, as was necessitated by the combined quadrupole-octupole elements. In both designs the C_s correctors are situated between the condenser lens system and the scan coil assembly. In the case of the HB5, this results in a lengthening of the column; for the HB501, however, the scan coil assembly is redesigned and moved into the objective lens housing of the STEM, the corrector replacing the original scan coil/alignment assembly. Such a corrector behaves as a rotation-free weak round lens which imparts adjustable negative spherical aberration to the wavefront; in addition it compensates for all parasitic *axial* aberrations up to fourth order. Several key parameters for the C_s corrected HB5 and the HB501 designs are reproduced in Table 5, extracted from the reference by Krivanek, Dellby and Lupini, which contains many more interesting details, as did Dr. Krivanek's presentation.

As indicated in Table 5, all of the significant parameters have been refined in the case of the HB501, the major application of which will be EELS and high angle dark field imaging at IBM Watson so that the reduced focal length of the objective is not a serious limitation; of course, it would be unacceptable in the case of an NTEAM-type As indicated in Table 5, all of the significant parameters have been refined in the case of the HB501, the major application of which will be EELS and high angle dark field imaging at IBM Watson so that the reduced focal length of the objective is not a serious limitation; of course, it would be.

Table 4. Brief history of aberration correction

Corrector Type	*First Proposed*	*Subsequent Versions*	*Proof-of-Principle*	*Improves Resolution of Its Microscope*	*Improves Resolution of Any Microscope (at its kV)*
Cs only: 2 cylindrical lenses / 3 octupoles	Scherzer 1947		Seeliger 1951–54	Mollenstedt 1954–56	
Cs only: 4 quads / 3 octupoles (combined)	Archard 1955		Deltrap 1964	Krivanek+Dellby 1997	
Cs only: 4 quads / 3 octupoles (separate)	Thomson 1967	Beck+Crewe 1972–75	Krivanek+Dellby 1999	Dellby + Krivanek 2000	Dellby + Krivanek 2000
Cs only: 2 sextupoles / 2 (4) round lenses	Beck 1979	Crewe 1980 Rose 1981 Shao 1988 Rose 1990	Chen and Mu 1990	Haider 1997	Haider 1998
Cs + Cc: 4 mag. quads / 2 el. quads 3 octs	Hardy 1967	Rose 1971	Hardy 1967	Zach + Haider 1995	Zach 1997
Cs + Cc: 5 el. quads / 5 mag. quads 3 el. octs	Hardy 1967 Rose 1971	Pohner 1976 Koops 1978 Bernhard 1980 Hely 1981 Haider 1984	Koops 1978	Hely 1981	

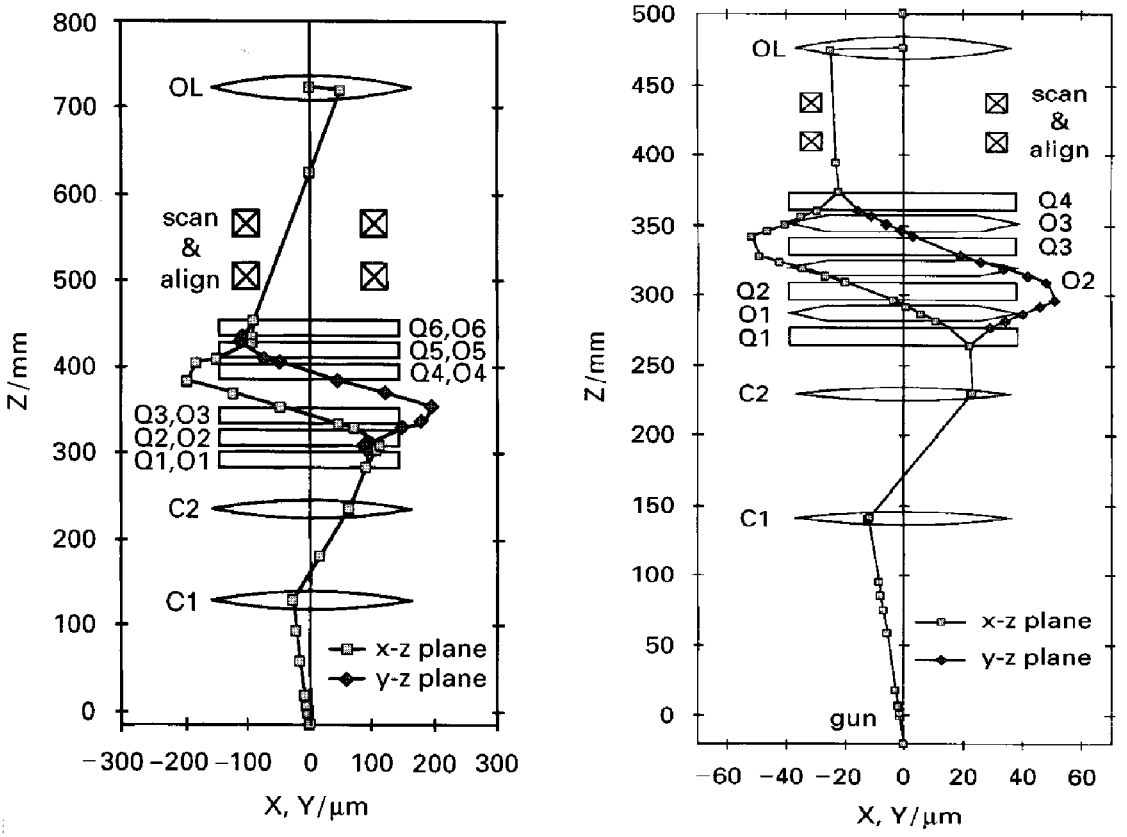


Fig. 2. Principal electron optical elements and first order electron trajectories for the C_s correctors for the VG HB5 and the HB501. (Reprinted from Ultramicroscopy, vol. 78, Krivanek *et al.*, "Towards sub-Å electron beams", pp. 1–11 (1999) with permission from Elsevier Science.)

unacceptable in the case of an NTEAM-type, however. Dr. Krivanek showed very recent high angle dark field images of Si from the HB501 operated at

Table 5. Several key parameters for two C_s-corrected STEMs

Parameter	VG HB5 (Cambridge)	VG HB501 (IBM Watson)
Primary energy (keV)	100	100
Obj. focal length (mm)	4	1.5
Intrinsic obj. C_s (mm)	3.5	1.3
Intrinsic obj. C_c (mm)	3.5	1.3
C_c of corrector (mm)	7	0.2
C_c of system (mm)	10.5	1.5
Corr. pow. supp. stab. (ppm)	1	0.5
Lateral drift of probe (nm)	0.2	0.02

120 KV, demonstrating 2.5 Å resolution with the Nion corrector off (i.e., the octupoles off and the quadrupoles on) and 1.36 Å, the dumbell spacing, with the corrector operating.

Dr. Krivanek, looking further ahead, presented information on the precision in various aberration coefficients required for correction of a 200 kV instrument to achieve probe sizes of 0.2, 0.1 and 0.05 nm. These are summarized in Table 6. [Note: regarding use of symbols for various aberration coefficients in Haider's presentation, the following correspondences exist for the two conventions:

For defocus C_1 is the same; regular astigmatism $C_{1,2} = A_1$; axial coma $C_{2,1} = B_2$; 3-fold astigmatism $C_{2,3} = A_3$; (3rd order) spherical aberration $C_3 = C_3$; (4th order) axial coma $C_{4,1} = B_4$; (5th order)) spherical aberration $C_5 = C_5$.]

The following conclusions were drawn and predictions made by Dr. Krivanek:

1. Spherical aberration in STEM is now a solved problem.
2. C_s correction will improve DF STEM.
3. Beam current in a given probe will increase by more than the resolution improvement squared.
4. Improved resolution, increased beam current STEM will find many new and interesting applications in materials science and biology.
5. C_c correction in STEM is not necessary at the moment and probably too difficult.
6. Aberration correctors correct aberrations, not instabilities. Stability requirements increase in aberration-corrected systems. This is a solvable problem, but it will require particularly stringent precautions in microscopes using objective lenses with large polepiece gaps.

Table 6. Precision needed for microscope set-up (200kV)

Aberration		Precision needed for probe size of		
Name	Krivanek Symbol*	0.2nm	0.1nm	0.05nm
beam drift (pm)	$C_{0,1}$	40	20	10
defocus, 2- fold astig. (nm)	C_1, $C_{1,2}$	20	5	1.2
axial coma (□m)	$C_{2,1}$	16	2	0.2
3-fold astig. (□m)	$C_{2,3}$	5	0.7	0.09
3rd order aberrations (□m)	C_3, $C_{3,2}$, $C_{3,4}$	1000	80	5
4th order aberrations (mm)	$C_{4,1}$, $C_{4,3}$, $C_{4,5}$	300	10	0.3
5th order aberrations (m)	C_5, $C_{5,2}$, $C_{5,4}$, $C_{5,6}$	80	1.3	0.02

* See Krivanek, Dellby and Lupin, Ultramicroscopy 78 (1999) 1-11, for definition of aberration coefficients.

The Problem of Chromatic Aberration

The subject of chromatic aberration was touched on by many speakers during the Workshop, a brief introduction to which is presented here. Chromatic aberration produces a smearing of an image which arises from several sources. This smearing or defocus spread D for instrumental factors is given as

$$D = C_c\left[(\Delta V/V)^2 + (\Delta I/I)^2 + (\Delta E/E)^2\right]^{1/2}$$

where C_c is the chromatic aberration coefficient, ΔV is the amplitude of short-term acceleration potential fluctuations, ΔI is the amplitude of short-term lens current fluctuations and ΔE is spread in energy of the electron source, usually taken as the FWHM or FW at one-tenth maximum. The first two contributions reflect short term instabilities in the high voltage and lens current power supplies; the last, the intrinsic characteristics of the electron source and subsequent prespecimen energy filtering (monochromator). The solution to these problems rests with the degree of perfection of the engineering. In addition, there is what one may call specimen-specific chromatic aberration associated with various energy loss mechanisms as electrons interact with the specimen, which is the basis for EELS and which can be mitigated in large measure for imaging and diffraction purposes by post-specimen energy filtering, either in-column or post-column. Thus in principle, each of these factors is manageable, increasingly so with time.

Monochromator Development: Frank Kahl and Peter Tiemeijer

The energy spread of a cold field emission electron gun is typically 0.2–0.4 eV and of a Schottky FEG, 0.5–0.7 eV. Current practice for decreasing chromatic aberration associated with this energy spread of the electron source involves installation of a monochromator preceding the electron accelerator section of a TEM or STEM. A monochromator is a precision energy filter which typically reduces the energy spread of the beam by energy-dispersing the beam to a defining slit which excludes electrons of energies differing from the peak by some predetermined limits. The net result for TEM, STEM and SEM is improved spatial resolution for imaging and spatial and energy resolution for microanalysis; however, the total beam current incident on the specimen is reduced relative to that emerging from the electron gun. Several such monochromators have been proposed including the retarding Wien filter [M. Terauchi et al., Microsc. Microanal. Microstruct.2 (1991) 351–], the electrostatic omega filter [H. Rose, Optik 85 (1990) 95] and the fringe-field Wien filter [T.T. Tang, Optik 74 (1986) 51; H.W. Mook, P. Kruit, Ultramicroscopy 81 (2000) 129–139]. The subject of monochromator development was reviewed in some detail during the Workshop by Frank Kahl who had worked with Harald Rose at Darmstadt University of Technology in development of the SÅTEM monochromator and by Peter Tiemeijer who is involved in monochromator development at FEI, Eindhoven.

Frank Kahl's talk dealt with the development of an omega-type monochromator for the analytical SÅTEM for Stuttgart, a microscope which should be delivered in about 2002. While the general concept is the same, requirements for such a "gun monochromator" are different from those of the well known in-column omega filter because of its extreme sensitivity to relatively very small desired energy spread ΔE. For example, a large dispersion is required so that the average electron energy entering the monochromator should not exceed a few keV, rather than a hundred or more. As in an in-column omega filter the energy spread exiting the monochromator is defined by a selection slit on the optic

axis at the plane of mirror symmetry of the filter where an image of the electron source must be formed. In the instrument design process, analytical solutions for the paraxial rays are computed, starting with three conditions and six system parameters. From the solutions three free parameters remain, allowing fast scanning of the three dimensional solution manifold for feasible solutions which can then be explored in further detail including the effects of fringe fields within the filter. The Boersch effect which effectively broadens the source is especially important at crossovers because of the low energy of the electrons; this effective broadening, however, is less than 0.1 eV. Regarding the loss of brightness in the case of the SÅTEM monochromator, for large beam currents (100 nA) brightness is decreased by a factor of 7 (astigmatic ray path); for small currents (10 nA), by a factor of 2. The conclusion is that a useable monochromator with $\Delta E = 0.2$ eV is feasible.

Question: How close is the SÅTEM monochromator design to realization?

Response: Dr. Kahl referred the question to Max Haider who responded that construction should be complete by CEOS in August (2000) and the system should be operating at low kV on the SEM test bench in October. It should be shipped to Stuttgart early in 2001.

Question: (Ondrej) How many voltages can be employed without realignment of the monochromator? Are there additional quadrupoles for this purpose?

Response: In tests of the sensitivity of the energy resolution to misallignment of the source and monochromator, measurements showed that the energy resolution was not limited for a relative displacement up to 100 mm. (A lengthy discussion ensued relating to problems of changing the accelerating voltage.)

Question addressed to the audience: (Alwyn Eades) The monochromator for decreasing ΔE of the source is placed near the source potential; earlier we heard that C_c correction was presently not feasible at TEM voltages; would C_c correction be possible similarly by placing the corrector near the gun potential, just as one puts the C_s corrector before the element producing the aberration?

Response: Max Haider responded that a C_c-corrector at the illumination side makes sense only for a STEM. For TEM, the C_c-corrector has to be placed after the object plane. The C_c correction for STEM is possible at a place near the gun, however one has to consider the demagnification of the objective lens and the very different electrical potential at the gun area compared with the potential at the object plane. Therefore, the generation of a chromatic aberration with negative sign and a length of meters or even kilometers for the corrector (in order to compensate for the C_c of the objective lens) is necessary. This is theoretically possible, but it seems to me that it is not experimentally feasible. In addition, it is doubtful, if such a C_c-corrector would be useful because one has to consider the difficulties to realize such a system and, in contrary, what can be gained with a C_c-corrector for a STEM in comparison with an already existing monochromator, for example.

Question: How does the $\Delta E = 0.2$ eV for this monochromator with a Schottky emitter compare with that for a cold FEG?

Response: A cold FEG can provide a sufficient current with $\Delta E = 0.2 - 0.3$ eV without a monochromator, but calculations suggest that you have to distinguish between results for Z-contrast STEM and EELS.

Peter Tiemeijer presented the second talk on monochromator development. A major emphasis at FEI in monochromator development is for improved EELS energy resolution which is presently limited by high voltage supply instabilities (typically 0.2 eV), the spectrometer resolution (typically 0.7 eV) and DE of the electron source (typically 0.7 eV for a Schottky field emitter). The goal is 0.1 eV energy resolution, requiring both hardware and software approaches for a 200 kV TEM/STEM. Instabilities of the high voltage supply can be reduced significantly by thermal and acoustic isolation. In addition, Gatan is improving the GIF for sub-0.1 eV resolution, involving improved stability in the bending magnet and the addtion of octupoles to correct the important third order aberrations. The monochromator for reduction of DE of the source is a double-focussing Wien filter (crossed magnetic and electric fields normal to the beam direction) between the gun lens and the accelerator. Fig. 3 is a sketch of the optics of the test column employed, including the Wien filter. The filter is 50 mm long and operates between 0.5 and 3 kV beam potential, the relatively large beam potential reducing the Coulomb interactions. So far the filter has been tested on a 20 kV column; individual beam sweeps show 0.1 eV resolution EELS spectra. Images of the dispersed beam spot indicate that the aberrations due to the filter are small, limiting the energy resolution only by 0.02 eV.

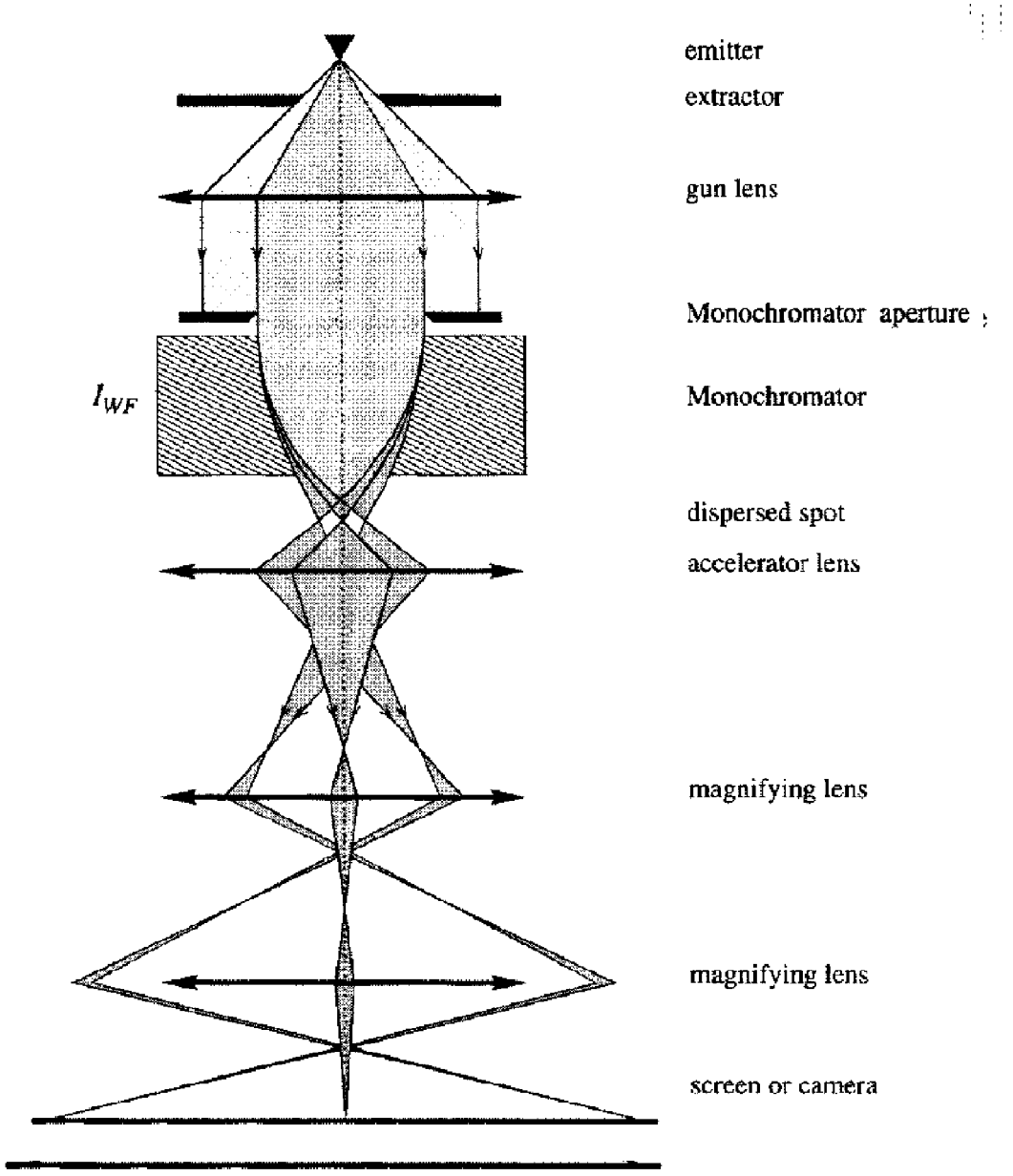


Fig. 3. Optics of the test column. (Reprinted from Ultramicroscopy, vol. 78, Tiemeijer, "Measurement of Coulomb interactions in an electron beam monochromator", pp. 53–62 (1999) with permission from Elsevier Science.)

The gun is an important element in the monochromator design; the gun lens may be used as a decelerating (then accelerating to the monochromator) or accelerating gun lens (then decelerating to the monochromator). This allows flexibility in the various compromises of initial aberration, dispersion, beam intensity, resolution etc. The final question is, what can the monochromator do for the NTEAM microscope? When you consider the various instabilities, you may conclude that for a 10 mm gap objective all of the instabilities taken together cannot exceed 0.25 ppm which is quite small. The major problem rests with the

objective lens current instabiliy which must be dealt with if C_c correction with a monochromator is to be worthwhile. There has also been discussion about loss of current with a monochromator; we believe you do not necessarily have to loose current by monochromizing. Dr Tiemeijer briefly described a technique for operation of the system with the dispersed source imaged on the specimen. In this way all the beam current passes and one obtains a line illumination with the slower electrons on one side and the faster ones on the other of the specimen image. Using the stigmator inside the monochromator, the line of illumination can be elongated in the non-dispersive direction to form a square illuminated spot.

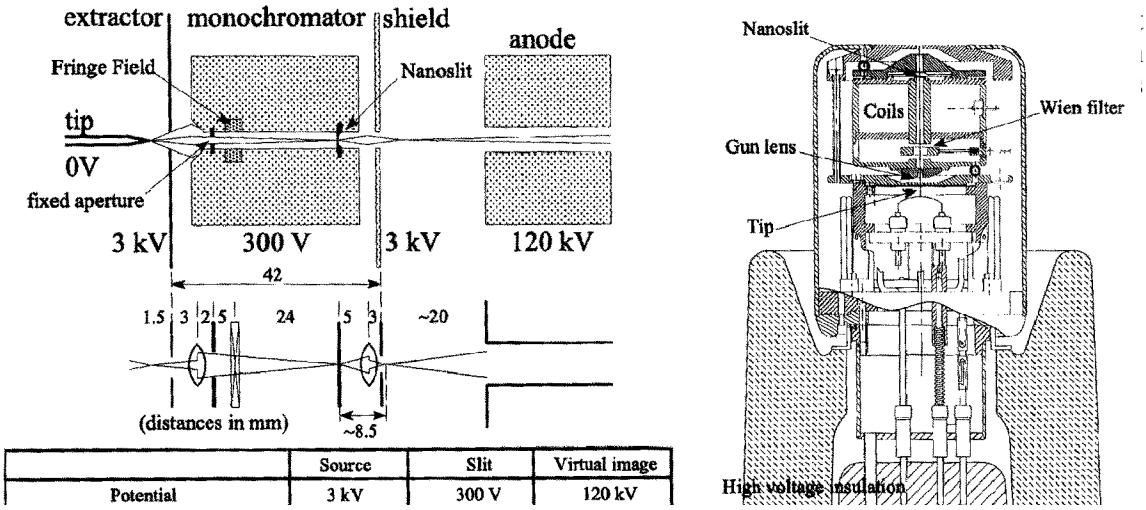
He also addressed briefly Kohler illumination. In the electrostatic omega filter Kohler illumination is obtainable simply, since the dispersion of the first half of the omega filter is corrected in the second half of the filter. In the monochromator, this correction is absent. However, he explained that the effect is sufficiciently small that it does not impede high resolution imaging.

[See also P.C. Tiemeijer, M.H.F.Overwijk and A.F. de Jong, Microsc. Microanal. 6 Suppl 2: Proceedings, (2000) 170-171, which briefly discusses some aspects of this topic including the software approach which aims at improving the resolution of the EELS spectra by maximum entropy deconvolution. This can reduce the original 0.8-0.9 eV resolution to better than 0.3 eV, the resolution improvement being proportional to the logarithm of the signal-to-noise ratio. The combination of present hardware improvements and deconvolution offers the prospect of EELS with resolutions well below 0.1 eV.]

Question: More important than the current is the brightness, since you can always increase the current by increasing the spot size. Have you ever measured the loss in brightness in your experimental setup? The problem is we cannot determine the spot size because of instabilities so we have not been able to measure this. But we do plan to do so in the coming months. Calculations indicate that the accelerating gun lens will increase the brightness by a factor of 2 (compared to the standard decelerating gun lens).

[Note: While it was not discussed during the Workshop, a monochromator of the fringe-field Wien filter type (FFM) has been constructed and successfully tested on the VG 5 STEM with W filament gun at IBM Watson, the results of which are reported by Mook and Kruit [Ultramicroscopy 81 (2000) 129–139; see also H.W. Mook, P.E. Batson, P. Kruit, Inst. Phys. Conf. Ser. 161, IOP, 1999, 223–226]. This monochromator is rather compact as illustrated in Figure 12 in the reference by Mook and Kruit and is included below as Fig. 4.

For any monochromator design there is a fundamental limitation imposed by the Coulomb interaction of electrons at focal points along the optical path. Longitudinal repulsive components give rise to additional energy spread (the Boersch effect) and lateral repulsive



	Source	Slit	Virtual image
Potential	3 kV	300 V	120 kV

Fig. 4. Basic optics and construction of FFM (fringe field monochromator) on field emission source. (Reprinted from Ultramicroscopy, vol. 81, Mook and Kruit, "Construction and characterization of the fringe field monochromator for a field emission gun", pp. 129–139 (1999) with permission from Elsevier Science.)

Pole Piece Gap and Chromatic Aberration and Comments on the SÅTEM and SESAMe Projects in Europe: Bernd Kabius

In a microscope corrected for spherical aberration, the major factor limiting resolution, aside from electrical and environmental instabilities, is the product $C_c\Delta E$, where ΔE is the energy spread of the beam. Dr. Kabius' main objective was to put these limitations in the context of in situ microscopy for which a considerably larger pole piece gap than is customary in dedicated high resolution instruments is usually required. But first he discussed a series of broader issues. Because of the practical difficulty of correcting C_c, at least in the near term, the immediate goal is to reduce ΔE so far as possible. While it would be very desirable to reduce ΔE to 0.1 eV, this would require stability in high voltage power supply of 0.2 ppm which is presently not likely. Thus for the SESAMe project, consisting of three microscopes, the more realistic assumption of ΔE = 0.2 eV has been made, requiring stabilities in the 0.5 ppm range; this is sufficient for achieving a resolution of 0.09 nm, due to the limiting envelope for temporal coherence.

The SESAMe Project is a collaborative project involving Max-Planck-Institute—Stuttgart, the University of Tübingen, CEOS and LEO. SESAMe 1 is a LaB_6 instrument which incorporates a post-specimen 90° energy filter and was delivered to Tübingen during 2000. SESAMe 2, which will be delivered probably in 2001, will be equipped with a FEG which is compatible with the monochromator. SESAMe 3, which will be delivered to MPI—Stuttgart, probably in 2003, will include an improved in-column energy filter (the MANDOLIN) with an isochromicity ΔE = 0.2 eV for a 2048^2 CCD. The STEM probe size will be 0.18 nm with an acceptance angle >100 mrad with an energy width of 10 eV for HAADF and CBED.

The 200 kV SÅTEM design includes a Schottky emitter with monochromator, a C_s corrector and a post-specimen 90° energy filter for analytical work. The 90° filter has two basic advantages over earlier omega filter designs, improved isochromicity with a ΔE = 1.0 eV over a 50 mm diameter area and a large acceptance angle for convergent beam electron diffraction (better than $\pm$100 mrad) with a ΔE = 10 eV. Another obvious advantage is that the SÅTEM is suitable for both high resolution imaging and for analytical work. The SÅTEM microscope will be delivered to Jülich probably during 2002.

When these several projects are completed, we will have a series of components which can be combined to obtain new microscopes. One of these would use the C_s corrector not only to achieve ultimate resolution but also to move the pole pieces and open the gap, so that for a gap of 1 cm it will still be possible to achieve a resolution of 0.1 nm and if the gap is opened to 2 cm, the resolution will still be similar to that of the current Jülich microscope with the prototype C_s-corrector (0.14 nm), assuming ΔE = 0.2 eV and electrical stabilities in the 0.5 ppm range. Another possible solution might be to go to a smaller ΔE; if in future years we could attain ΔE = 0.1 eV, a resolution of 0.07 nm could be achieved in a 1 cm gap. The stabilities required to do this are beyond the realm of possibility today, however.

For a TEM devoted primarily to dynamic in situ studies, the size of the gap becomes an important issue because C_c increases with objective focal length, as demonstrated in Fig. 5.

In addition to the situation in which the specimen sits within the lens gap, it can just as well sit above the lens. [Note: while most in situ experiments are performed with side entry holders, some of the most versatile, robust holders

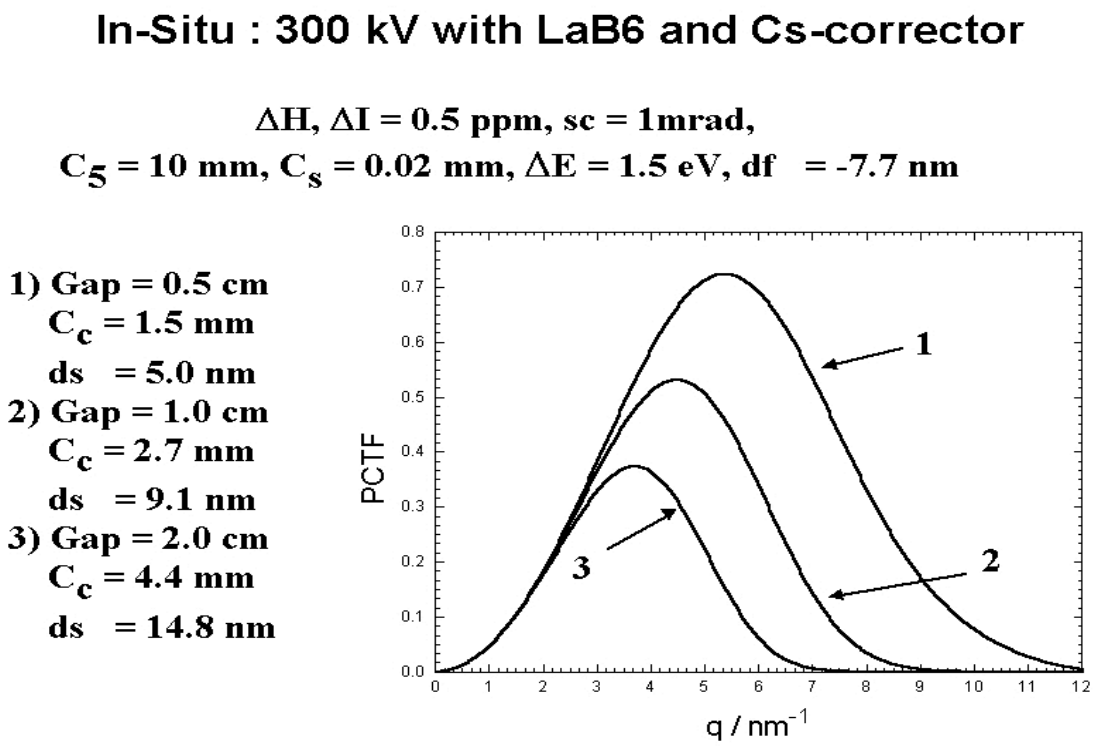


Fig. 5. Effect of objective gap dimension on C_c and on the associated phase contrast transfer function for a C_s-corrected TEM with LaB_6 source. (Unpublished. Used with permission of author.)

are those which have been employed by Messerschmidt for mechanical property studies at the MPI—Haale HVEM, which are all top entry. Also the ultrahigh temperature holder (2200 K) along with a number of other experimentally useful holders at the Ultra High Voltage Electron Miroscope Facility at the University of Osaka are top entry.] A C_s corrector would still be required, of course; an energy filter can be added for analytical studies and for observation of specimens of larger thickness.

In response to a question regarding exposure times for beams with very narrow DE, Dr. Kabius replied that by eliminating coherence and thereby obviating the spatial coherence damping envelope one may use a larger convergence angle, thereby reducing exposure time. So for DE = 0.3 eV approximately 75 percent of image intensity will be lost (relative to no filtering); a factor of four in intensity can easily be obtained by converging the beam.

Question: Earlier speakers were rather more pessimistic about going to large gaps because of increased sensitivity to instabilities and stray fields. For the Jülich microscope, compensation of stray fields was necessary in the microscope room, reducing the field from 0.4 mG by a factor of ten. Improving power supply stabilities is a progressive technology. Dr. Kabius indicated that he was not so concerned about these limitations.

Question: Can one turn off the monochromators described in this talk without upsetting the alignment, for example, if one wanted to perform a high intensity irradiaition before

switching to a higher resolution condition for analysis? The CEOS monochromators mentioned by Dr. Kabius are electrostatic and therefore can be switched on and off.

Toward an Ideal SEM: Peter Tiemeijer

One of the reasons for this second talk is that SEM presents another possible application for the Wien filter (monochromator) to improve resolution. The talk is not exactly about ideal SEM but rather about aberration correction in SEM, employing no magnetic field or one magnetic field. Two correctors are being studied at Philips Research: one is a purely electrostatic corrector and the other a Wien filter corrector. Both of these can correct both chromatic and spherical aberration for low voltage SEM. The choice of minimizing magnetic fields is due to the relatively small effect of magnetic fields on ions which can also be produced by the gun and rather to concentrate on electric fields for which ion optics are employed. In the case of the electrostatic corrector, lens elements are chosen having positive and negative C_c values so that the net C_c of the corrector is negative. Of course, there can be no negative electromagnetic lens so that quadrupoles are employed as we have seen in earlier talks; to increase the dispersion one may modulate the beam potential. (He then explained

The second design is the Wien filter with only one magnetic field, which also has a negative C_c. The system consists of electric, magnetic and quadrupole fields. Both designs also correct for spherical aberration by employing octupole lenses.

Question (Alwyn): Would you compare the performance or the difficulty of producing these designs with that which has been implemented in Haider's group?

Response: The implementation of the Wien filter is fairly simple, we think, with only two power supplies, and the stabilities are in the range of 10 ppm, not 0.5 ???ppm.

Question (Alwyn): Do you agree with him, Max?

Response: In theory, yes.

Question: Can you make a Wien filter for energy filter too so you have an imaging Cc, Cs corrector plus energy filter?

Response: You use this in the 2p mode and there's no dispersion left at the end because it is so long.

Comment (Alwyn Eades): If I had money to buy an aberration-corrected system, it would not be any of the systems presently being built. It would have to be a standard 30–40 kV SEM with long working distance and a large beam current into a small probe. I still have not heard anyone say that they are building that. If I am wrong and someone is building such an instrument, please let me know because for a lot of applications, such as electron backscattering diffraction and microanalysis, that would make a huge difference and I think a more important difference than pushing the envelope in TEM.

Murray Gibson: We discussed earlier the effects of stray fields on long working distances. Have you given that any thought?

Alwyn Eades: No, I didn't know that stray fields was the big issue until yesterday.

Reports on Existing Projects and Pending Proposals

The Jülich Experience: Knut Urban:

The aberration-corrected instrument in the Institute for Microstructure Research of Jülich Research Center is a Philips CM 200 with C_s corrector described in the earlier talk by Max Haider. Professor Urban indicated that small Au clusters had been employed in order to determine the imaging properties of the microscope, but that the main research emphasis for the instrument was in defect studies in general, with a strong emphasis on interfaces in particular. In his presentation, he commented briefly on seven points relating to spherical aberration corrected TEM: resolution, contrast, contrast delocalization, image reconstruction and correction, Bragg contrast, microdiffraction and instrumentation. Due to coherence problems, the resolution of the Jülich instrument with corrector (C_s = 0.05 mm) does not by principle achieve the information limit of the instrument (Philips specification C_s = 1.2 mm). In addition the corrector itself increases the chromatic aberration of the instrument (C_c = 1.7 mm compared to 1.3 mm measured), associated with the transfer lenses of the corrector. To compensate for this the heater current of the Schottky emitter is reduced by about 10 percent to reduce the thermal spread and the influence of the Boersch effect on chromatic aberration at reduced intensity. The energy spread DE is thereby reduced from about 1 eV for the basic instrument to 0.7 eV. The demonstrated resolution of the corrected instrument, as Max Haider mentioned also, is improved to 0.13 nm at 200 kV.

Professor Urban then reviewed the theory of image contrast delocalization in the presence of spherical aberration and showed several examples of this troublesome effect and of its cure. One observation is that in the corrected microscope for a defocus of 50 mm Scherzer defocus and Lichte defocus practically coincide, giving optimum imaging conditions with respect to delocalization. In an uncorrected microscope the phase contrast transfer function is very complicated for Lichte defocus, making image interpretation very difficult. In imaging under phase contrast conditions it is necessary to choose a value of C_s other than zero in order to convert the phase information into amplitude information. In the presentation tomorrow by Markus Lenzen there will be more detailed discussion about the optimum defocus and C_s values which should be chosen for phase contrast imaging. Professor Urban, however, made several related comments. First, in choosing these values one should ensure that the slope of the contrast transfer function is kept as steep as possible at small spatial frequencies. So the C_s = 50 mm was chosen for imaging of GaAs , for example, showing 0.14 nm resolution for the 111 dumbbells, on this basis. The value of C_s is still sufficiently small that contrast delocalization is small. Delocalization of selected area diffraction information is similarly reduced to near zero. He also discussed briefly the case of C_s = 0 and the contribution of electron channeling to image formation in relation to defocus and specimen thickness variations. A final comment dealt with the problem of depth of focus in high resolution phase contrast images of specimens which are not normal to the incident beam and when C_s is small.

Professor Urban next turned to the question of whether exit wave function reconstruction is necessary when corrected microscopes are employed for high resolution imaging (the software approach in relation to the hardware approach). The most important point, however, in wave function reconstruction involves its elimination of artifacts arising from the difference between image and object frequencies, the former extending to higher values than the latter, and the artifacts arising from non-linear interactions between beams. To eliminate

these artifacts for quantitative high resolution, wave function reconstruction must be performed. In addition, because the contrast transfer function is at best far from ideal, i. e., far from unity and dependent on spatial frequency, focal series reconstruction is still essential for quantitative high resolution microscopy.

Of course, not all microscopy is done under high resolution conditions. Thus, for Bragg contrast imaging in a corrected microscope, the conditions on microscope adjustment for good imaging are considerably relaxed. For example, one may tilt the beam off the optic axis by up to 10 mrad in lieu of precise specimen tilting which is often impossible. Similarly in the case of dark field, precise centering of the desired reflection is also unnecessary; in fact, displacing the objective aperture is often adequate for acceptable quality images.

Electrical stability, elimination of time-varying fields and mechanical stability of the site and of specimen stages are increasingly more important in achieving the potential of aberration-corrected microscopes. In addition, because it is critical for high resolution applications to have an amorphous segment (not carbon contamination) preferably in an edge of the specimen in order to accurately determine the imaging parameters for subsequent reconstruction. As resolution continues to improve, it may turn out that the range of spatial frequencies available in typical amorphous thin films is insufficient for accurate determination of these instrumental parameters.

The Oxford Project: John Hutchison

John Hutchison, Department of Materials Science, University of Oxford, began his remarks with a brief overview of two projects in high resolution imaging using their recently acquired JEOL 3000F FEGTEM. Although the UK was relatively slower in acquiring FEGTEM instruments, there are now about 10 in the country. The first experimental project John described was the image resolution of the oxide sublattice structure in complex oxide materials. Using through-tilt and focal series to reconstruct phase and amplitude image contrast, a resolution of better than 1.4Å was demonstrated, a significant improvement on the instrument's nominal Scherzer resolution of 1.6Å.

A second project consisted of high resolution imaging of RbI (Rubidium Iodide) and KI (Potassium Iodide) filled single walled carbon nanotubes. John described perhaps "the world's smallest crystals" of 2 atoms wide and 2 atoms deep of KI contained in a nanotube. Using 20 image focal series, phase and amplitude maps were constructed that imaged and distinguished single K and I atoms separately in a 3 x 3 atom crystal. By accurately measuring the atomic positions in these small crystals, the structure was found to be tetragonally distorted from the bulk crystal cubic symmetry.

Following these illustrations of recent high resolution work, John described the current state of the next generation microscope project at Oxford. Funding for this instrument was obtained from the Joint Infrastructure Fund (JIF), one of about three avenues of possible funding for this sort of project in the UK. The other two include the usual Engineering and Physical Sciences Research Council (EPSRC) and the Joint Research Equipment Initiative (JREI). The location for this microscope project, and that of other recently acquired or projected equipment will be in the recently renovated buildings at the University's Begbroke Science and Business Park, about 8 km north of Oxford. This site is ideal for low surface vibrations, as it is located well away from highways or industry.

The Oxford One Å Project, as it is called, is funded and contracted with JEOL, with final specifications nearly finished at the time of this talk. As currently envisaged, the microscope will most likely be based on the JEOL 2010FEF instrument with variable

voltage from 60 to 200 kV and Schottky field emitter source. As this will also be fully STEM capable (for John Titchmarsh's analytical interests), three-fold astigmatism correction will be possible on both condenser and objective lens systems. C_s correctors will also be incorporated in illumination and imaging lenses and provision will be made for spectroscopy and energy-filtered imaging. The objective lens pole piece gap should be about 5 mm (John's guess), allowing about 35° sample tilt. Detectors will include a high angle annular dark field (HAADF), TV rate, CCD and photographic image recording. The target probe size will be one Å, with a similar point resolution. Sample holder stages will be compatible with the existing JEOL3000F, allowing testing and cost saving. Finally, John had "no comment" on the price of this project.

[Note: A second project in the UK is the "SuperSTEM" Project initially proposed by Professor Mick Brown (Cambridge University), and now funded in 2001**.** The facility which will be based on aberration-corrected VG STEM instruments will be built at the Daresbury Laboratory.]

The Oak Ridge High Temperature Materials Laboratory Acquisition: Larry Allard

The High Temperature Materials Laboratory (HTML) at ORNL intends to acquire a monochromator-equipped, spherical aberration corrected FEG-TEM/STEM or FEG-STEM in the coming months. After commenting on the politics of instrumentation funding, Larry Allard summarized a number of specifications which have been written in conjunction with this acquisition which has been funded by DOE—Environmental Engineering. It is expected that an order will be placed before the end of summer.

[Note: shortly after the Workshop, the contract for this instrument was awarded to JEOL; the instrument will be a TEM/STEM with spherical aberration corrector for the STEM probe to be supplied by CEOS GmbH; the monochromator will likely be an omega filter type of JEOL design. The design of the instrument will also allow for addition of TEM aberration corrector at a later date.]

Questions: (Knut) Was there any indication in your discussions with the manufacturers of moving to a corrected 300 kV instrument? And can this be combined with a stage having 0.3 nm per sec drift or better?

Response: Yes. The HF-2000 hyperstage is better than that.

General Discussions

During the part of the Workshop dealing with theory and development of hardware for aberration correction two general discussion sessions were held in which several comments and short presentations were made, which are briefly summarized here.

Comment by Markus Lentzen Some comments on focal series reconstruction and the need for C_s correction. There are various choices in doing high resolution including minimizing delocalization, maximizing phase contrast, maximizing amplitude contrast or even minimizing either of these latter two. If you choose $C_s = 0$ and $Df = 0$, as Urban pointed out earlier, you get amplitude contrast, you minimize phase contrast and you have no delocalization. But if you want to play around with phase contrast, you have to choose

either C_s or Df not equal zero. He described briefly for given C_s the difference among Scherzer focus, the focus for minimum contrast and Lichte focus of minimum delocalization (which is linear in C_s). In general the strategy is to maximize phase contrast to produce a zero at the information limit of the microscope and at the same time this maximizes the area under the phase contrast transfer function. With $C_s = 0$ you can still have phase contrast by defocusing to achieve smaller phase plate in comparison with the Scherzer defocus. Examples were shown of several imaging conditions from the Jülich microscope which Urban described earlier. For zero C_s, there is phase contrast for defocus of about -7 nm, and amplitude contrast at half the extinction distance. C_s ~54 mm optimizes delocalization (~1 Å) with phase contrast for defocus around -13 nm. The optimized Scherzer focus is -20 nm. For real experimental situations, although you may have a reduction of C_s, you still have nonlinear imaging and even if you consider small amplitudes for diffracted beams for linear imaging you still have a mixing of amplitude and phase contrast mechanisms. If you want to work in an aberration-free environment, the specimen may not admit it; for instance, if you have a wedge shaped specimen, you still have different focus at different locations, or if you have a particle embedded in an amorphous matrix, you also may have the situation in which the periphery of the particle may be in focus while the center is not. A focus series will get around this, of course, and utilize reconstruction to obtain the wave function, allowing "refocusing' after the experiment. Similarly one can perform the aberration correction after the experiment, obtaining the tableau very quickly. So the question is, if we can do focal series reconstruction, do we need C_s correction anymore? Clearly, yes! because C_s correction allows immediate view of the structure in the microscope. In addition the useable CCD area in the corrected microscope is significantly larger which is also valuable for subsequent focal series reconstruction; one can optimize the global tilt with the beam tilt controls.

Comment by Murray Gibson: The effect of AC magnetic fields which are the primary cause of blurring depends on distance. For an electron drifting some distance L, the electron experiences a deflection in distance proportional to L^2 and inversely proportional to energy $E^{0.5}$. So for L = 10 mm compared to 1 mm, the blurring is increased by a factor of 100. This is easily seen in the SEM with increasing working distance, where this is the major source of blurring (not the increased aberration coefficient as usually stated in the manual). This is a problem in any lens of increased focal length, as Ondrej mentioned. One may address this in several ways. Reducing the magnetic fields by shielding or by active compensation as mentioned by Bernd Kabius. Another idea, originally suggested to me by Arnold Bleaker, is to employ a pancake lens, a snorkel lens. As you move the specimen far away, there is unlimited space, but up close, the focal length is short. This gives a lot of flexibility. Perhaps one could design some hybrid of a snorkel and normal pole piece configurations. (A many-sided discussion ensued.)

Comment by Ondrej Krivanek: We all agree that the shorter the focal length of the lens, the better the performance. So the question deduces to how much resolution are you willing to sacrifice in favor of increased space. There are of course already a number of 1 Å resolution microscopes with 10 mm gaps in the world (the ARM II series). Radiation damage and capital cost for these HVEMs are significant issues, of course. We need to focus on the kinds of experiments which need to be done in situ. For example, experiments which might capitalize on MEMS technology. Phil Chang at Etec has built an entire SEM including the gun in a 10 mm space. These same techniques may permit doing experiments on a very small scale, not only manipulating the specimen but making it as well.

Comment by Murray Gibson: I agree but there a number of types of experiments where space is essential, for instance, for given conductance in a UHV microscope or where the

specimen or apparatus must be heated to high temperature and kept some distance from other surfaces.

Comment by Ondrej Krivanek: You gain some space with a corrector in the system, because you typically reproduce the back focal plane elsewhere in the system which removes the necessity for placing an objective aperture very near the specimen.

Question (Alwyn): Is stray magnetic field interference the only problem one has to worry about in going to larger gaps?

Response: Max Haider responded that stray electric fields may also show up. For example, he observed interference from a local TV station at 100 MHz.
Comment by Nestor Zaluzec: A comment to focus on a roadmap for NTEAM. Ultimately we are after information about our samples from some signal. We have seen today two roads, one concentrating on probe-forming or illumination systems and the other on the road of the imaging and post-specimen systems. Both require intelligent engines to do the corrections and enhancements because the procedures are too complicated to be done manually. The next level up are a series of barriers to doing experiments which we need to identify and to evaluate. These include environmental barriers, such as magnetic, electric fields, and acoustic fields; stage drift and the issue of proper and stable positioning of the sample; and electrical stabilities. And so on, a progression of barriers, what are the ones which face us at this point and how do we overcome them? At some point we will agree that now it is not economically feasible to build a commercial scheme like that. But NTEAM is not limited at this point in time by commercial barriers. I would prefer to see, this is the limit and here's what it takes to get around it.

Comment by Steve Donnelly: As someone interested in in situ experiments, I would be delighted with 2 Å resolution and a relatively large gap to work in. I don't see the point of trying to combine that into an instrument with sub-Å resolution.

Comment by Shigeto Isakozawa: I want to introduce you to another Cs-correction system. The method of Defocus-Image Modulation Processing was originated by Dr. Ikuta in 1989 as a reconstruction method using through-focal series, and we have been developing this method for electron microscopy since that time. (near end of Tape III B) We record 256 images taken at 4 nm focus step intervals; the integration to attain the DIMP image is performed combining the focal series images and a focus-dependent weighting function. In this manner we retrieve the aberration-free image, requiring typically 2–3 hours. Our goal has been to develop a microscope using this concept which allows aberration-free imaging in realtime. Dr. Isakozawa briefly described such a realtime system involving active modulation of the high voltage to produce an image pair (+Df, -Df) every 1/15 second which is displayed as a difference image at half NTSC video frame rate. He also displayed high quality high resolution dynamc image grabs to demonstrate the success of the technique even for very short atom columns.

Question : What is the effect of the stability of the cold FEG on the frame rate time scale?

The high voltage stability is 2×10^{-6} ; the measured ripple is about 50 mV. The intensity fluctuation normally ranges from 5–8 percent over a 2 or 3 minute interval.

Question: Does the high voltage modulation have a significant effect on the field of view?

Response: High voltage modulation amplitude is typically 200–300 V at a frequency of 1–2 kHz.

Question: The negative part of the linear contrast function to the positive part and superimposing all the images. How does the method treat non-linear contrast features? Even in thicker crystals, one should have a lot of non-linear contrast features.

Response: Weak phase approximation only, very thin objects.

Question: This looks like it would be applicable to any microscope; there are no changes to the electron optical column. Could this be retrofitted to an HF 2000 in the field or even to other manufacturers instruments? And how about the CCD camera?

Response: The CCD camera was developed by Osaka University and high voltage system, by Hitachi.

Question: Have you actually checked the value accuracy for a weak phase object; the Au specimen you showed is a critical specimen in that respect? This approximation is the basis of such an enormous technology, one should think about it. Isn't that the diplomatic expression?

Response: The theory has been developed by others; I'm afraid I don't know the answer.

Comment by Kai Xiu: We considered a quadruplet of quadrupole-octopoles aimed at the C_c-correction of a high resolution TEM with large-gap pole piece and low beam energy. The strategy of adding appropriate numbers of octopoles to balance the third order aperture aberrations and correct coma is studied in depth. We conclude that for a moderate electrode potential of the electrostatic quadrupole, its power stability poses the most severe limit on the achievable resolution. It is expected that as long as the power instability of its electrostatic quadrupoles can be maintained below 0.1 ppm, a quadrupole-octopole quadruplet C_c corrector featuring thick field distribution can provide a correction of C_c up to 6.5 mm without introducing any other aberration effects.

Comment by Alwyn Eades: I would like to return to the NTEAM design and to think about how the core of it might be configured. The first question is what kind of objective lens system will be used, an immersion lens or one for which the specimen sits outside the magnetic field. If the latter, probably we will want both a probe-forming and an image-forming lens with the specimen between them, outside their fields. We want a reasonably large working space here as well. I would like to suggest several relevant points to be factored into the eventual decision-making process in the context of aberration correction; others will surely add to these factors. The first advantage of immersion optics is that it is well known. The disadvantage is that the sample remains in a strong magnetic field which is disadvantageous for some in situ experiments and techniques (angle-resolved Auger spectroscopy, electron backscattered diffraction, or microscopy of many magnetic materials). Conversely with separate lenses, the sample is in a magnetic field-free region and the probe- and image-forming lenses are decoupled and thus controlled separately. But there are also disadvantages; one is the uncorrected C_s and C_c will be larger than otherwise and C_c becomes then more important, assuming C_s is correctable. Further there is the possibility for worse magnetic screening depending on the placement of the outer cores of these lenses. In addition it is unclear whether the same angular range of scattered electrons can be collected as in the immersion lens configuration because of the field distribution within the latter. Such decisions are critical to the NTEAM design.

Comment by Murray Gibson: I wanted to add to this that third idea which I mentioned yesterday regarding lenses with the concentration of field quite far from the sample. This is both good and bad as you pointed out. Another possible approach is use is the snorkel

design of Mulvey, for which Bleeker did calculations for 100 kV in situ application and published it about 10 years ago. There is, of course, no second pole piece and the field is concentrated outside the lens, so that with the specimen close to the lens (a couple of mm) and the field after it where TEM mode is realized or with the specimen following the field (maybe 4 mm from the lens) in which case the configuration would act like a STEM. And, of course, the specimen may be moved much further away. The aberration coefficients are within a factor of two or so of those of an immersion lens so they're not unmanageable. So there are a number of possibilities for designing lenses which offer more space within which to work. It is clear from the various comments which have been made that decreasing the focal length you can reduce the effects of stray fields, so for a given experiment there may be an optimum gap and the one-size-fits-all approach does not optimize performance. These performance requirements should become much more clear beginning this afternoon; some experiments will require sub-Å resolution and others 2 Å resolution or worse.

Max Haider: Regarding the snorkel lens, as the specimen is moved further from the pole piece, the aberration coefficients increase tremendously, not a factor of two but maybe 10 or 20, as I recall. A special corrector would have to be designed for this pole piece.

Comment: The importance of stray fields is being overemphasized because there are several corrective measures available which were discussed earlier.

Nestor Zaluzec: Many layers of mu-metal don't cure these problems, at least in the VG (603) because we are still piercing the shielding with aperture drives and so on.

Murray Gibson: If the volume to be compensated is small enough, active compensation can be very effective. We don't know the answer to all these questions; clearly it is an issue which needs to be looked at in detail.

Comment by John Spence: Just before VG's bankruptcy, Mike Sheinfein did very detailed calculations for that kind of lens. I want to make a very quick point about image interpretation to send you off to lunch (not to change the subject). To interpret images without spherical aberration, called the projected charge density approximation, the image is slightly out of focus with $C_s = 0$ and is proportional to the charge density projected, not the potential projected. As in classical light optics case of hundred years ago, out-of-focus phase objects are sort of differentiated, they are second derivatives. It's the same thing here in the absence of spherical aberration; we get the second derivative of the potential which by Poisson's equation is the charge density; positive defocus gives bright atoms, negative defocus gives dark atoms. The images on the two sides of focus are complementary. The important point is that, while you must not forget about multiple scattering as pointed out earlier, this projected charge density term is not part of a mathematical expansion, and so it does a better job of dealing with multiple scattering and in fact it is exactly correct if there are no excitation errors for the flat Ewald sphere. So here's an approximation which includes all multiple scattering within the approximation and may prove useful. I go into more detail in my book!

Comment: The remark about the flat Ewald sphere is a pretty big stickler there.

Materials Research in an Aberration-Free Environment

The second half of the Workshop was devoted at discussions of scientific needs for and anticipated impact of aberration-corrected instrumentation, based on a large number of short presentations by participants. To this end it had been suggested to a number of participants before the Workshop was convened that in these brief presentations they might describe their current research by way of introduction and then address the question of how aberration correction might impact their own future research and its direction. This part of the Workshop was divided Into three sessions, each organized and conducted by personnel from one or more of the participating National Laboratories.

- Session 1 was devoted mainly to HREM and was conducted by U. Dahmen (NCEM/LBNL).
- Session 2 focussed on in situ studies and was conducted by C. Allen (EMC/ANL) in cooperation with I. Petrov (CMM/FSMRL—UIUC).
- Session 3 was devoted to chemical and elemental microanalysis and was conducted by I. Anderson (SHaRE/ORNL).

For the sequence of speakers in these three sessions, please refer to the program in Appendix A. Particularly in the case of the HREM and In Situ Sessions, issues relating to all three session areas were often intermixed. Table 7 illustrates the spectrum of research interests and some instrumental requirements reflected by the participants speaking in these sessions. For reference, participants are listed alphabetically in Table 7. For the sake of logical flow of the Report, the strict chronological order of the agenda (Appendix A) is generally observed, but not in every detail.

Session 1: HREM-Related Studies and Aberration-Free Microscopy
Session Chair: Ulrich Dahmen

Introductory Overview: Ulrich Dahmen

While emphasizing various aspects of high resolution imaging, Uli Dahmen (NCEM/LBNL) presented a comprehensive overview to introduce the second half of the Workshop program, addressing a wide range of issues involving the role of very high spatial resolution microcharacterization techniques in general in the study and analysis of materials phenomena, including in situ studies, with emphasis on full analytical quantitation. The key areas of materials research addressed in his presentation were (1) interface science, (2) defect science, (3) phase transformations, (4) nanostructured materials, and (5) microelectronics.

For each of these, he established a perspective which was followed by a list of typical challenges which serve as a broad outline for Part 2 of the Workshop.

Interface Science: Internal interfaces, such as grain boundaries and interphase interfaces, are far less well understood than are surfaces, yet they play often times a decisive role in limiting mechanical, electrical and magnetic behavior of materials. While most surface phenomena have interface analogs, interfaces are under solid constraints which give rise to entirely unique phenomena associated with elastic compatibility, bicrystallography and abrupt changes in-chemical composition. Challenges in this area include the following:

- Mapping interface segregation with sub-monolayer accuracy

- Probing electronic structure with atomic column resolution
- Determining quantitatively non-periodic atomic structure and local relaxation
- Observation of atomic mechanisms an dynamics of interfaces in situ during phase transfirmations or deformation
- 3-D reconstruction of interfacial defect structures

Table 7. Summary of User Requirements for Aberration-Corrected Instrumentation

Participant	Affiliation	Requirement	Comment
I. Anderson	Oak Ridge National Laboratory		Spectrum imaging, multivariant techniques
J. Bentley	Oak Ridge National Laboratory	More stable stages; better detectors; x-ray focussing optics; CTEM/STEM 0.1 eV EELS to look at core losses on single atom columns	
R. Birtcher	Argonne National Laboratory	Large sample tilt, ion beams, fast video	
N. Browning	University of Illinois - Chicago	Cold FEG STEM; 10 pA probe; 5-10 s EELS collection	
U. Dahmen	Lawrence Berkeley Laboratory	HREM on technologically important materials –better resolution needed.; valence state with single column EELS	3-D reconstruction of interfaces; to see single point defects-dislocation cores
M. De Graef	Carnegie Mellon University		Phase reconstruction in Lorentz microscopy
K. Downing	Lawrence Berkeley Laboratory	Ability to tune C_S over a large range	Has radiation-sensitive light element samples and wants better CCD detectors. Now use 400kV beam with 4 microns under focus for contrast and decellerate electrons before hitting CCD camera.
V. Dravid	Northwestern University		In-situ oxidation/reduction, measurement of dopant profiles
M. Haider	CEOS GmbH	Cc, Cs corrected SEM	Better contrast and sharper images, Cc affects the tails of the probe
R. Hull	University of Virginia	FIB in microscope, make electrical contact patterns; fast video, control temperature (100mK), strain sample, make electrical and optical measurements Interest in ion stimulated	

		deposition and imaging of hexagonal lattice of electrons in compound semiconductors	
J. Howard	Noran Instruments		WDS w/x-ray optics, microcalorimenter
J. Hutchison	Oxford University	Sub 2 Å resolution with gases for in situ environmental studies	
S. Isakozawa	Hitachi Ltd.		Defocus image modulation processing for HREM
M. Kirk	Argonne National Laboratory		Interstitial/vacancy study
C. Kisielowski	Lawrence Berkeley Laboratory NCEM	50 meV energy resolution and 0.5–0.6 Å pt-to-pt res.; automatic sample alignment to zone axis	HREM and light element detection, exit wave reconstruction from as recorded images seeing oxygen
O. Krivanek	Nion Company		Nanoanalysis: 1.5 Å probe with 1 nA current
M. Lentzen	Inst. of Solid State Physics, Jülich	Cs correction; "direct" view is convenient giving gain in useable CCD area	
M. Libera	Stevens Institute of Technology	Good spatial resolution and better energy resolved EELS to resolve differences in polymers	
M. McCartney	Arizona State University	Improved minilenses (C_c limit); scan/descann coils; stage stability; space for coils to apply oscillating magnetic field on specimen Wants to bias transistors, stimulate emission from lasers in situ	
L. Marks	Northwestern University	Large tilt for 3D diffraction patterns; pulse biasing of ceramic samples	
A. Meldrum	Univ. of Alberta	STEM (sub Å); EDS and EELS for surfaces and interior atom column resolution for nanoparticles; e.g., map Co in a CoPt nanoparticle	
I. Robertson	University of Illinois - Urbana	25 mm gap with 2-3 Å resolution; sample tilt, with temperature and strain control; better detectors; faster video	Study structure during growth and then irradiate and observe and control structure.
F. Ross	IBM Research Division	Larger beam current and filter images, larger gap (1	Wet cells in situ growth of Cu

		cm), tilt beam—no image degradation	
R. Sinclair	Stanford University	Atomic column composition analysis for defects and interfaces; 300kV; computer alignment and adjustments; low magnification capabilities; high temperatures and gas pressures; data storage and faster video recording	Follow chemical changes with time
J. Spence	Arizona State University		Nano precipitates with no aperture for a large convergence, if no Cs then have greater intensity. Tygrography (?)hybrid of diffraction and imaging
E. Stach	Lawrence Berkeley Laboratory NCEM	Large gap pole piece to fit MEMS microtensile tester	
E. van Cappellen	FEI Company		Discribed the FEI dual beam FEGSEM?HR-SEM and FIB
N. Zaluzec	Argonne National Laboratory	Optimize detectors, better stages, still need to optimize cold FEG	
Y. Zhu	Brookhaven National Laboratory		Electronic and magnetic structure studies
J. M. Zuo	University of Illinois—Urbana	0.01 mrad beam convergence, stable sample with temperature control; automated software to acquire and analyze diffraction patterns like x-ray interpretation	

Defect Science: Defects in crystalline solids control much of their physical behavior; point defects in diffusion and many irradiation-related phenomena; line defects in deformation, certain phase transformations and crystal growth; planar defects in deformation and intergrowth phenomena. Challenges in this area include the following:

- Atomic resolution imaging of dislocation core structures in metals, superalloys, semiconductors and ceramics
- Imaging of individual point defects or small defect clusters
- High resolution, high precision mapping of local strains in materials
- Measurement of local electronic structure

Phase Transformations: The study of solid state phase transformations is the foundation of modern materials science, promoting the design of new materials through characterization of structure, composition and bonding. Challenges related to this area include the following:

- Nanocrystallography
- Nanoscale composition analysis
- Quantitative HREM: Errors bars on local atom positions and chemistry and phase identification

- Integration of theory and computation with instrument development
- Atomic scale mechanisms and dynamics of phase transformations
- Atomic structure of glasses

Nanostructured Materials: Nanoscale materials are of increasing scientific and technological importance and are dominated by effects of surfaces and interfaces. They are critical materials for catalysis, quantum confinement structures and nanotube applications. Many physical properties are strongly size dependent in the nanometer size range including electronic, optical, magnetic, mechanical, thermodynamic, kinetic and chemical properties. Challenges in this area include the following:

- Nanocrystallography
- Phase identification
- Relating nanoscale theory and simulation to experiment
- In situ measurement of local electrical and mechanical properties
- Structure of interfaces involving nanocrystalline solids
- Size and shape dependence of phase transformations

Microelectronic Materials: As device structures move to progressively smaller sizes, electron optical imaging and diffraction techniques are the only techniques capable of resolving the structures produced. Challenges in this area include the following:

- Structure of gate oxide, and crystalline/non-crystalline interfaces
- Imaging core structures of interfaces and dislocations with atomic size discrimination
- Mapping strain with high precision and sub-nanometer spatial resolution

Discussed at nearly every microscopy-related meeting, it is generally agreed that a major effort in specimen preparation is needed to push electron beam microcharacterization forward to take full advantage of the capabilities of forthcoming instrumentation. Uli Dahmen suggested that the National Lab User Facilities are uniquely positioned to accept this challenge to develop both specimen preparation techniques and instrumentation, including the use of MEMS and STM technologies for in situ preparation and manipulation.

A summary of specific issues, which depend on further development of instrumentation, concluded Dr. Dahmen's talk. They include the following:

- Atomic level spectroscopy: single column microanalysis;
- Electron nano-crystallography at the 0.5 Å level;
- Determination of core structures of line defects,
- Atomic structure of interfaces in three dimensions
- Iterative model/experiment refinement of defect structures in three dimensions (atomic resolution tomography);
- Optimized, artifact-free specimen preparation (thinning, growth, MEMS);
- Structure of glasses—fluctuation microscopy;
- In situ microscopy of atomic scale mechanisms and kinetics, including environmental cell studies, in real time, with in situ manipulation methods (MEMS) and
- In situ microscopy of atomic scale mechanisms and kinetics: real materials, model materials, nanostructures, devices;
- Measurement of local physical as well as structural properties: functional materials, model materials, nanostructures and devices;
- Imaging of magnetic and electric fields.

In Session 1 dealing with HREM-related studies. a number of these topics are addressed in greater detail by other participants.

Comment: Murray Gibson You mentioned in your summary of specific issues the structure of glasses. Diffraction techniques have not been successful in determining the structure of glasses which generally exhibit medium as well as short range order. Medium range order is, however, addressed well by variable coherence microscopy.

Question: Robert Hull I am trying to get a sense of what extra advantages or opportunities are gained by having a 1Å or 0.5Å resolution instrument as opposed to current instrumentation. To play devil's advocate, the question really is, what has high resolution microscopy really achieved that should merit its being considered essential to nanostructural science and the nanotechnology initiative? I'm convinced that the answer is out there but it has not been clearly articulated by the community.

Response: Uli Dahmen We may be too hard on ourselves. If you look around at other communities, for example the neutron scattering community, they are very good at stretching their case but when you look closely, I believe that the microscopy community is better able to point to successes such as the nanotube discovery by electron microscopy. In addition I cited a number of contributions which, while incremental in nature, nevertheless were essential in contributing to understanding of, for example, the structure of interfaces. I doubt that the neutron scattering community could do better.

Comment: Knut Urban It was mentioned that the structure of quasicrystals was solved by electron diffraction. In fact, the initial "structure" was demonstrated by HREM and subsequently the detailed structure established by a combination of X-ray and neutron diffraction. The electron microscopy of quasicrystals is a good example of something which has been oversold under the aegis that we have achieved atomic resolution. So we are asked today why do we need better and better instruments to achieve atomic resolution when atomic resolution has already been achieved. Of course, the answer is, if it has been achieved at all, "atomic" resolution has been realized only in a few very favorable orientations yielding limited information. The fact is that technically relevant materials require significantly higher resolution for this technique to be at all useful. So far we have done our work by selecting the material consistent with the instrument we have had available; now it is time for us to position ourselves so that we can choose the material in relation to its relevance. We want to be able to do our microscopy under high resolution conditions in real and interesting materials when that is appropriate.

Response: Uli Dahmen Yes, it was just the point I tried to make in one of my first viewgraphs which showed the number of achievable and meaningful orientations which could be utilized for various materials as progressively better resolutions became available.

Even with the current generation of JEOL ARM series of HVEMs, we are able to study only rather simple grain boundaries and interphase interface structures and then only in low index orientations. More general boundaries require much higher resolution as illustrated by Table 8. The ability to achieve a wide variety of orientations is certainly essential in defect analysis in complex materials. One of the most exciting prospects from aberration correction, however, is the possibility for atomic resolution tomography when such a variety of orientations is available for structure imaging.

Table 8. Image resolution and structure imaging.

Material	Number of Orientations Available for Structure Imaging for a Given Pt-to-Pt Resolution			
	2Å	1.6 Å	1.0 Å	0.6 Å
Diamond	1	1	2	9
Aluminum	2	2	6	10
Silicon	1	2	7	13
Iron (bcc)	1	1	4	12
Iron (fcc)	1	2	5	12
Tungsten	1	1	5	13

Source: NSF Report on Atomic Resolution Imaging

Comment: Howard Birnbaum Let me address the situation in the USA. I wouldn't bet on the inability of the neutron scattering community to construct a strong case for its successes and breakthroughs. But that is not the real point we should address. Yesterday, Murray Gibson presented an estimate that, I don't know, 45 percent of all materials physics and 65 percent of all materials chemistry papers use electron microscopy. I've done this exercise also and come to roughly the same conclusion. But when you look critically at a large fraction of these publications, you discover that the token electron micrograph is presented as window dressing. The real problem in this country is that, as microscopists, we haven't really effectively penetrated the physics and chemistry literature, the consequence being that they regard microscopy as an essential tool in the same sense that they regard STM now. The Si 7x7 reconstruction is a good example of a phenomenon discovered by TEM, but there are many fewer papers in which this is studied by TEM since then than by STM. We simply have not penetrated the physics and chemistry communities and that is where our sights should be focussed.

Response: Uli Dahmen I agree, and that is exactly why we need to make electron microscopy much more quantitative, because that should be the effect it would have.

Comment by Bob Sinclair I was asked to comment on image resolution in metals and semiconductors. I believe that 1Å or 0.5 Å instrument development is extremely important, and since hopefully this is to be associated with in situ microscopy I would like to show a video segment to illustrate why that goal is superior to what we have generally available to us today. (An in situ HREM movie taken at 200 °C of the growth of Ge crystal using Ag as a diffusion medium through which Ge diffuses from amorphous Ge on one side to crystallize on the other. Instrument resolution is 1.8–1.9 Å. The lattice of Ge is well resolved at about 2.0 Å but, when you look at the Ag, the in situ conditions of heating, translating, refocusing and so on are sufficient to wipe out the lattice resolution at the 1.9 Å level.) We have done such in situ experiments with a wide variety of metals and alloys, and getting high quality and informative images is extremely difficult in our instrument. So the point is, if one is to perform in situ studies on a host of materials, especially those which have technological importance, something like a 1 Å machine will be needed to get good, consistent in situ results at the 1.5–2.0 Å level even.

I was also asked to comment on in situ microscopy in the broader context. I certainly support what Uli has already said. As has been pointed out, to get resolution of particular defects, it is necessary to have a range of zone axes available, and to do this 1 Å even may not be enough. Where so much these days involves high tech materials, nanomaterials are an everyday subject of study, multilayers of different components for read/write heads, for example. In addition to high resolution structure, often one needs good compositional

information, atom column by atom column in the structure; the interfaces, are they rough, what are the interdiffusional effects across the interfaces. Compositional analysis is every bit as important as the structural analysis. Especially for this kind of material system, specimen preparation kills you every time, so we really do need vast improvements in advanced specimen preparation methods as well. Resolution requirements are more exacting, for instance, in nanostructures involving tetrahedral boundaries in semiconductors to separate the two species, especially if one wishes to do in situ experiments, 1.4 Å for Si and smaller spacings for compound semiconductors, and again in such systems the interface technology and its evaluation are critical.

The last point I want to make is in regard to synchrotron facilities which have discovered how to garner huge amounts of funding for their operations, which is certainly one of the problems ahead of us in regard to NTEAM. The mode of operation at such user facilities is for a group to work around the clock for two weeks accumulating mountains of data which are analyzed elsewhere, and then the process repeats itself. Of course, as we all know a typical electron microscopy experiment is quite different from a synchrotron experiment because EM is generally highly interactive and labor intensive for which the next step may be based, at least in part, on the results of what you have just seen happen, especially in in situ studies. How we incorporate this type of flexibility into an NTEAM type of instrument which might attract the sychrotron mindset is problematic. We also need small facilities which will feed into the big instruments like NTEAM to prepare the various aspects of the experiment ahead of time, the holder with the specimen, the specialized analytical tools and so on. I am very enthusiastic about this whole concept, and I feel if we can work together as a community we can push it forward.

Comment: Uli Dahmen I like the idea of feeder facilities. To carry that concept one step further, it would be tremendously advantageous if the specimen holders (modules) were compatible with a number of instruments to permit various aspects of a given experiment to be carried out at multiple sites.

Comment by Christian Kisielowski I want to address two issues: the needs of materials science regarding spatial and energy resolutions and regarding advanced specimen preparation, particularly in the context of high resolution imaging. Closely related to the issue of resolution is that of precision in atom position determination, for which we would like 1 pm. The detection of light elements, such as oxygen, nitrogen, boron and carbon, is of equal concern, not only for microanalysis but also for high resolution imaging. Table 9 summarizes spatial resolutions obtained over the past ten years or less by various TEM and STEM techniques. When we examine the spectrum of materials available to us today we may conclude the following with respect to imaging: (1) a spatial resolution for imaging of 50 pm or better is very desireable, (2) the precision of atom column location should be ~1 pm, and (3) for modeling of defects energy differences of ~50 meV should be achievable.

Specimen preparation is a major problem for high resolution because of sensitivity to factors such as thickness (image contrast can increase by a factor of four for 2 nm thick Si compared to 8 nm, with a corresponding improvement in atom position definition) and surface roughness (e.g., radiation damage due to ion milling). There are periodic advances in this area, recent examples of which include FIB and low angle/low voltage ion milling with specimen cooling which avoids most of the artifactual problems associated with conventional ion milling and allows much closer thickness control. Also remilling of FIB-prepared specimens using low voltage milling will restore the base material for high resolution observation. The final point I would like to make has to do with radiation damage, particularly of non-metals, by coating the backside of the specimen before observation with a thin layer of carbon (which evidently mitigates differential sputtering resulting in the introduction of defects).

Comment: Murray Gibson Another use for the beam tilts is their automated computer control to look for symmetry or intensity in the image so you can find precisely in real time the zone axis, for instance.

Response: Yes, everyone would like to have an automated procedure of this sort.

Question: You mentioned wanting an image resolution of 0.5 Å and energy resolution of 50 meV. Is this at one time or not?

Response: I would like to be able to discuss this because they are mutually contradictory objectives.

Comment by John Spence**:** Post-specimen energy filters make electron diffraction more accurate than X-ray diffraction. In fact, electron diffraction now allows us to see chemical bonds even. X-ray diffraction suffers from extinction errors ranging from 2–50 percent, whereas one requires an accuracy of better

Table 9. Best resolutions obtained by various TEM and STEM methods.

METHOD	VOLTAGE	EMITTER	BEST RESOLUTION
HVEM	1.25 MV	LaB_6	95 – 89 pm Moebus, Phillipp et al., 1998
	1.25 MV	Schottky	49.8 pm (info. limit) Kawasaki et al., 2000
Cs-corrector	200kV	Schottky	140 pm Haider et al. 1998
Z-Contrast (STEM)	300kV	Cold FEG	78 pm Nellist & Pennycook, 1998
Other IVEM	200kV	Schottky	150pm Coene et al., 1992
	300kV	Schottky (sftwr. corr. astig.)	140pm Thust, Coene et al. 1998
	300kV	Schottky (hrdwr corr. astig.)	80 $\pm$ 3 pm NCEM
Holography	300kV	Schottky	104 pm Orchowski et al. 1995

than 1 percent to see a bond. This is now achievable by quantitative CBED [J.M. Zuo, M. Kim, M. O'Keefe, J.C.H. Spence, Nature 401 (1999) 49–52]. This was demonstrated by combining QCBED and X-ray diffraction in a study of Cu_2O from which high quality

charge distribution maps were obtained, allowing the direct imaging of *d* holes on the Cu atoms and also demonstrating the existence of Cu-Cu bonds in this compound.

We have also used multiple scattering calculations to demonstrate that the structural periodicity along a dislocation core may be measured from coherent CBED patterns obtained with a sub-nanometer probe parallel to the dislocation core. The resulting "half order" HOLZ ring resulting from a double-period core reconstruction model proposed for Si by Bennetto *et al*. in 1997 is detectable (in the simulation) above the background of thermal diffuse scattering which is included in the calculations. Multiple scattering calculations are used to show the temperature and thickness dependence of this ring. Using experimental coherent CBED, coupled with such calculations, it may be possible to deduce activation energies for core processes. Such experiments are well suited for aberration-corrected STEM.

Question: How large is the computational supercell you use, and what restrictions are there on its contents ?

Response: Once the supercell is larger than the probe, you can put anything in you like, it could be a glass.

Comment: Knut Urban This comment concerns quantification in general; we have talked a bit about why electron microscopy is not so appreciated as STM. In STM we always say we measure the images. I think what we must do is make real measurements in the microscope also.

Comment: J.M. Zuo Regarding the opportunity to combine diffraction with high resolution imaging, the diffraction pattern gives a correlation function while imaging gives phase information for starting information regarding defect structures. So progress can be made in that direction by combining the two.

Comment: Uli Dahmen Yes, basically doing nanocrystallography. If that were developed into a generally available quantitative technique that would be very valuable.

Comment by Nigel Browning: I don't want to sound too negative about aberration correction but I would like to think first about what we can do with the microscopes which we already have. (He then described the equipment at UIC, mainly a 2010F with 1.2–1.3 Å resolution.) My point here is to point out some of the problems one may encounter in doing aberration correction. We spent at least six months trying to understand the source of various instabilities, for instance from video monitor interference (price: 0.5 Å resolution). To begin with, there was sound damping on the walls and an active anti-vibration system under the microscope and a dynamic field compensation system, and still there were residual problems. In the case of the field compensation system which sits about 20 cm from the specimen position, compensation is very effective at the 5Å level but as you press the resolution further you begin to see the dynamic compensation field deteriorating the image. The microscope senses the field much more sensitively than the compensation system does. So the key to doing very high spatial resolution studies is, one must be extremely meticulous about quality of the environment in every detail.

Finally a major advantage to STEM is that the same probe is employed for all the functions, imaging, EDS, EELS and so on, all of the signals simultaneously. So from the point view of aberration correction, the resolution for each of these modes is improved by a single corrector system. We are pursuing aberration correction, but not for the 2010F which appears to still have some residual instability, but rather with a cold FEG dedicated STEM. (He then showed a number of examples demonstrating the integrated functionality of

STEM and what can be done with a standard state-of-the-art microscope even without aberration correction.)

Comment by Yimei Zhu: The major TEM-related efforts at BNL include the electronic structure of superconductor interfaces and the magnetic structure of hard magnetic materials employing particularly techniques of quantitative diffraction and imaging (with J. Tafto), quantitative EELS of light elements (with R. Egerton), electron holography in hard magnets (with M. McCartney) and phase reconstruction (with M. DeGraef, who presented this topic in more detail later in the Workshop). With off-axis electron holography of B-2212 the Bi-O double layer is clearly revealed in the phase image. He then described a diffraction technique developed some time ago known as parallel recording of diffraction intensity, a modified CBED technique with the FEG probe focussed slightly before the specimen. He applied this also in Bi-2212 to determination of the thickness and charge density (from 000) and the displacements (from various reflections 00l) across a stacking fault and tilt and twist boundaries with an accuracy of 0.5 Å. Aberration corrected instrumentation will greatly improve quantitative HREM structure and interface characterization. In addition the ability to determine the imaging parameters accurately will be very beneficial for image simulations and phase reconstruction work. It is probably less crucial in the cases of quantitative diffraction and Lorenz microscopy, however.

Comment by Molly McCartney: We employ a Philips EM300 for imaging of electrostatic and magnetic fields, in both cases with the objective lens turned off. To provide some magnification, however, there is a minilens in the lower bore of the objective, the chromatic aberration of which limits the information limit to 1 nm. We wish we had a lens system with a variable magnification on the objective lens. Aside from the resolution issues, we are mostly limited in what we can do by the very limited space available within the objective pole. At present to vary the applied field seen by the specimen, we tilt the specimen with the objective on, the magnetization of our thin specimens generally being in the plane of the film. As an example, she described a study of a trilayer specimen of Co and Ni with a Au spacer layer to study the coupling between the Co and Ni layers. What we would like is to be able to arrange a system of coils in the gap for application of small magnetic fields, perhaps in plane with the specimen. We have tried this with a single coil on the objective aperture strip to study the effects of field frequency. There are relaxation effects at low frequencies, 10–100 Hz, that mimic spin glass behavior in ferromagnets. To do such experiments well requires more space; this is also the case in studies involving electric fields. Several other examples were presented involving electron holography. There are many interesting and important studies waiting to be performed which require a significantly higher level of sophistication than can presently be brought to bear.

Comment by Kenneth Downing: This presentation dealt with the special problems facing the biologist in regard to electron microscopy, including the fact that for the most part the materials involved contain only light elements (C, O, N) guaranteeing low contrast imaging but allowing work in the kinematical regime and that they are very susceptible to radiation damage, requiring low exposure. The result is at best a low S/N ratio in nearly all images, which means if we are to successfully do high resolution imaging, we must average many hopefully equivalent units in the process of getting enough information. In the case of crystals this is not a big problem. For single particles (molecules), he described the problem of solving the structure of hollow microtubules, employing a 400 kV microscope and a large defocus, sometimes several microns, enough to make almost any materials scientist shudder. The reason for the large defocus is to produce good contrast on the scale of the detail desired, perhaps 50 Å spacing. of course this is unacceptable for high resolution. What we want though is a medium or high resolution image of the microtubule, a much finer scale, say 7 Å, and much closer to focus.

Image processing may involve taking a number of segments of a microtubule from a given image and averaging them all together to produce a refined structure image of a segment. This may require averaging information from 10–100 microtubules. This amounts to a rather labor intensive process. So it would be of great benefit to have much greater contrast, of course. He then described the contrast transfer function (CTF) under various circumstances, the conclusion being that for biologists it could be tremendously advantageous not necessarily to eliminate spherical aberration but to have a variable C_s which could be tuned to values up to several hundred mm or more. Biologists are used to working in regimes where there are many oscillations in the CTF anyway and not particularly near Scherzer defocus and so this is a way of significantly extending the effective resolution as the calculated CTFs show (resolution to 4Å, for example, for C_s = 100 mm, a defocus of 15,000 Å in a 300 kV instrument).

Another major problem is the recording of information. To achieve really high resolution we would like to have up to a million particles to average (the particles are diluted in solution to achieve adequate separation). The limited performance of CCDs is a real problem as illustrated by a Monte Carlo calculation of 400 kV electron trajectories in a 40 □m thick scintilator, resulting in a large point spread function for the camera. Also the thermal noise due to electron energy losses becomes worse the higher the voltage. What we are doing is to float the camera at a high voltage (-250 kV) to slow the electrons down before they reach the CCD. (Laughter) We are just short of having experimental results from such a system.

Comment: Ondrej Kivanek I don't think you need a variable C_s but rather a phase plate in the back focal plane which does not charge up, a working version having recently been demonstrated in the PRC. Also in regard to the deceleration of the electrons, it would be better to put the specimen at 400 kV.

Comment by Matthew Libera: This presentation addressed a related family of materials, namely polymers, dose-limited resolution and new sources of contrast. Polymers are rich in structure at the 1–100 nm length scales, including interfaces, nano/microemulsions and other phase separated structures. Many of the comments that Ken Downing made apply equally well to some of the materials we encounter, including those comments relating to C_s.

As in imaging of biological materials, the well-established practices of differential staining and Au labeling have a place and a number of examples are presented. On a good day achieving a resolution of 50 nm is extraordinary. More interesting, however, is the application of spatially resolved energy loss associated with the valence electron structure of C, N, O etc. for imaging (low loss spectrum imaging). Matt again showed a number of examples of this such as distinguishing polystryrene from polyethylene without staining, the mapping of □□*-bonds in polystyrene at 35 nm/pixel (the technique requires adequate separation of pixels to minimize the effects of in-plane secondary electrons which create significant disks of confusion and wipe out the low loss peak at about 6.5 eV) and the imaging of water in a frozen hydrated copolymer.

Comment by Marc DeGraef: This presentation described a non-interferometric phase reconstruction procedure (i.e., an alternative to electron holography) and is a collaboration mentioned earlier by Yimei Zhu. Derived from the transfer function formalism and applied to Lorentz imaging, solution of basic transport of intensity equation (TIE) in the small angle approximation (mrad) allows direct reconstruction of phase of the exit wave. The gradient of phase corresponds to in-plane magnetic induction times specimen thickness. The longitudinal derivative is proportional to two images at a given defocus interval. Thus analysis requires meticulous alignment of the images which is the most difficult step in the process. In addition image pixel size and defocus step must be well calibrated. Several

examples of such reconstruction analyses of magnetic structures were presented. The method also shows potential for application to Cs-corrected microscopes, where deconvolution of the microscope transfer function becomes a trivial exercise.

Discussion: There was some discussion of this method in relation a similar one described about thirty years ago.

Session 2: In Situ Studies and Aberration-Free Microscopy
Session Chairs: Charles Allen and Ivan Petrov

Introductory in situ study presentations were made by Bob Sinclair, emphasizing the general guidelines and requirements, and by Eric Stach. emphasizing the scope of such studies and some of their related difficulties.

Introductory Overview I (Session 2)—Guidelines and Requirements
Robert Sinclair

In situ TEM may be defined as TEM studies of dynamic events in a specimen under some externally applied and presumably controlled stimulation. Stimulation methods have included heating to simulate thermal processing, cooling, mechanical deformation, irradiation (damage, sputtering, metastable phase formation), application of electric or magnetic fields, or deposition. The general requirements include a stable microscope (medium voltage instruments are most versatile but HVEM continues to be very useful), specialized specimen holders, excellent image recording systems (video tapes may degrade significantly in a week following recording) and appropriate specimen configuration and preparation. If an experiment is to be performed under high resolution conditions, all parameters must be more precisely controlled. In situ experiments are advantageous because (1) of the direct and continuous view of dynamic events, (2) the events may be recorded continuously, (3) the method allows a rapid survey of a range of dynamic behavior, (4) the information is multifaceted, (5) the information may be unique and unanticipated. The in situ method, however, is experimentally difficult and may require constant intervention by the operator(s). In addition experiments must be carefully designed ahead of time, the results which may be voluminous carefully analyzed and checked for reliability and possible artifacts. The advantages of TEM for in situ experiments include (1) the wide range of magnifications over which observations may be made, depending on the experiment, (2) simultaneous diffraction information and elemental and chemical analysis and (3) the direct observation of defects. But there are disadvantages also: (1) specimens are relative small (but they may be much smaller in the future), (2) specimen preparation is often the rate limiting step, (3) because the specimens are thin, phenomena may be surface dominated (great for thin films but not when bulk behavior is desired), and (4) presence of surface artifacts (often another specimen preparation problem).

Because in TEM we are often dealing with thin foils (as opposed to thin films), from the behavior of which we hope to infer bulk behavior, we must ensure that this is in fact the case for the results. This may be done reliably by (1) comparing the final in situ observations in thin vs. thick areas (if necessary remilling the specimen slightly at the conclusion of the experiment so that what was too thick can now be observed to see if the microstructure is the same as in the origin area of obseravtion) and with the electron beam on and off (except

for intermittent observation), (2) comparing microstructures developed in situ vs. ex situ for identical conditions (annealing, for example) and finally (3) comparing in situ vs. ex situ results (activation energies, for instance, to make sure what you saw was a bulk phenomenon, not a surface phenomenon). Sometimes you can be clever about specimen preparation; for example, when you are preparing a cross-section specimen for an experiment, you can glue as the other half of the specimen a material whose behavior is well known. Another important consideration is the quantitative reliability of the results, that is, that the magnifications and camera constants, and other experimental parameters such as specimen temperature, strain, magnetic field and so on are well calibrated. It may be necessary to check these against known standards in situ. Finally in so far as possible, the experimental phenomena should be modeled theoretically for comparison.

To summarize, in situ TEM

- Provides direct structural information of dynamic behavior which can be continuously recorded
- Increasingly emphasizes the importance of experimental control and quantitative analysis
- Can yield unique information which may lead to unforeseen advances.

As for the future, we can expect to see more refined in situ studies and analyses (many at high resolution), under more extreme conditions of temperature and perhaps environmental pressure, under combined stimuli such as temperature, stress and irradiation (that is, more closely simulating the model of reality), and with much more concomitant chemical and elemental micro(nano)analysis. Finally, the new generation of aberration-corrected microscopes will spawn increased innovation in the design and conduct of in situ experiments.

Related Comment: Murray Gibson A comment on the efficiency of in situ experiments for identifying the conditions under which a phenomenon occurs. Combinatorial materials is a big buzz word; the basic idea, for instance, is to examine in real space a range of compositions. But in real materials science there are other parameters in time and space, not just composition; temperature, rates and so forth. In situ procedures are an aspect of combinatorial materials and is a very efficient way of searching parameter space compared to conventional techniques.

Related Comment: Knut Urban So much depends on stage development. To realize a good in situ holder may typically take two years to complete. What company will be involved in producing such specialty specimen holders?

Several Responses: We do it ourselves.

Introductory Overview I I (Session 2)—Scope and Difficulties
Eric Stach

(This presentation was actually made during Session 1.) The main goals of in situ TEM include the following:

- Quantitative observation of dynamics and mechanisms of material phenomena
- Direct, real time measurement of property alteration caused by changes in microstructure
- Correlation of observations with quantitative models.

In this regard, we are able to understand the mechanisms involved in deformation, phase transformations, crystal growth, magnetic and ferroelectric domain wall motion, to name a

sampling of phenomena. But, indeed, there are pitfalls associated with this experimental method in electron microscopy. First and foremost are thin foil effects (which are largely overcome by HVEM which is rapidly becoming extinct in this country). We require carefully designed and prepared specimen geometries to compare with theoretical models and to address possible thin foil effects. And last but not least we must devise novel ways to apply external stimuli and to measure properties of small control samples within the confined space of the TEM objective lens. Aberration corrected instrumentation should be a significant help in this regard by providing addition space. Thin foil effects strongly limit our ability to obtain quantitative information about mechanical properties, diffusional, massive and martensitic transformations and real time quantitative HREM of material dynamics.

Emphsizing the practical significance and relation to modeling of phenomena associated with in situ studies, Dr. Stach then presented a number of examples illustrating a variety of types of such studies involving SiGe heterostructures, GaN, carbon nanotubes, and in situ nanoindentation, the last two employing a specimen holder in which the specimen could be manipulated piezoelectrically, based on the technology of scanning probe microscopy.

He then turned to a topic which Murray Gibson had referred to in his overview of the Workshop and the possibilities for NTEAM instrumentation, namely, the utilization of MEMS (micro-electro-mechanical systems) technology, in this case for the fabrication of a 4.2 mm x 2.4 mm micro-tensile tester for incorporation in an electron microscope. This was manufactured by C. Keller of MEMS PI at Cal Berkeley and represents the kind of new thinking which Professor Sinclair called "increased innovation". He also showed SEM micrographs of several MEMS devices including a pair of microtweezers for grasping a thin fiber or nanotube during a microtensile test, one of a number of highly specialized techniques which would be profitably pursued. Other examples which he proposed include in situ measurement of load-displacement characteristics correlated with simultaneous observation of deformation phenomena and more elaborate micromachines to bias samples and to measure their physical properties. For UHV instruments, it is possible to incorporate many types of surface science techniques with simultaneous TEM observation for crystal growth, catalysis, electronic and magnetic materials fab studies and the like.

Comment: John Hutchison A number of in situ studies which have been active at Oxford University were briefly reviewed by Professor Hutchison, including a number of environmental cell studies, particularly of non-metals such as $Nb_{12}O_{29}$, performed at high resolution**.** In general a 5 mm polepiece gap is adequate and in fact desirable for such environmental studies in order to diminish scattering by the environmental gas over the electron path length. For the most part 2 Å resolution is adequate for these studies, but , of course, better resolution makes any experiment easier.

Question: What if the resolution were adequate for imaging of the anions as well as the cations in such systems?

Response: That would open up a whole new area for study.

Comment: Uli Dahmen: There is a new 300 kV FEG instrument in Denmark with environmental cell in which an information limit of 1.4 Å has been achieved.

Related Comment: Howard Birnbaum. A comment about the effect of the electron beam in environmental cell studies. In one case we had 13 kPa of hydrogen and an aluminum specimen. From the rate at which bubbles formed on the surface voids, we estimated that the actual fugacity of the hydrogen is 40 MPa because of the influence of the electron beam on

molecular dissociation and ionization reactions. So the pressure you measure in the cell may be very different from the effective fugacity.

Question (Murray Gibson to John Hutchison): Would there be any advantage of STEM for such experiments in terms of the total current, the kinds of things you talk about in the plasma in which the total current is important. If that were minimized, does that mitigate the problem which Howard describes to some degree?

Response: We have not thought about that.

The Materials Science Laboratory in a TEM
Ian Robertson

In preparing for this presentation we adopted the approach of specifying the requirements of a microscope specifically for the purpose of performing dynamic studies of materials processes. Therefore, we did not restrict ourselves to current and near future advancements but rather to a futuristic approach, proposing that the design philosophy be altered to allow experimental laboratories to be readily incorporated into the electron microscope column. In the following we describe laboratories for conducting critical experiments and suggest a modular objective section with a redesign of specimen holders to use new technologies. It is realized that not all of this is currently possible but this is the direction we would propose for future generations of microscopes.

For many in situ experiments TEM is required, rather than STEM, because of the necessity for essentially instantaneous image recording capability to capture real time-varying events. And while the emphasis is largely on pushing the envelope of high resolution imaging in conjunction with aberration correction, few in situ experiments require 1 Å resolution; to achieve the goal of a materials science laboratory within the microscope, many of us would be delighted to have instrumentation with ~25 mm gap and a resolution of 0.2–0.3 nm at a fraction of the capital and maintenance cost of a modern HVEM. For an NTEAM-type of instrument we envision that various modular designs for in situ control and analysis could be employed. For such instrumentation, the emphasis at the Frederick Seitz Materials Research Laboratory at the U. of I—Urbana, as we see it now, would be in the following areas involving such modules:

- Surface and interface structures during growth of films
- Morphological instabilities during strained-layer crystal growth
- Nanomaterials: ion implantation and ion beam assisted deposition
- Gas-solid interactions: effects of gases in metals; nucleation and growth of hydrides
- Mechanical property studies
- Laser irradiation studies

In order to achieve such a range of objectives, we propose an NTEAM instrument which is a TEM/STEM In an ideal system, we would like both LaB_6 and FEG electron sources (not practical at present, but this would be very valuable), aberration correction system for probe formation (STEM) as well as post specimen aberration correction (TEM), in-column energy filter and multiple image capture modes (CCD, IP, film, ADF detector) and microanalysis capabilities. We envision a wide range of studies which would be handled by multiple objective sections (modules), each of which would be devoted to a particular type of study. Thus some aspects of design of the microscope in this proposal start with the materials science in mind. We realize that all this poses great challenges.

Thus a partial summary of general specifications might include the following:

- Experimental modules for the objective region
- Reasonable turnaround time for changing of objective modules
- Wide pole piece gap (≥25 mm)
- Spatial resolution ~0.2–0.3 nm
- Probe size ~1 nm
- Electron energy: 20–200 keV
- Capability for low to medium energy ion beams (tens of eV to 30 or 40 keV)
- Precise control of illumination area and orientation
- Adaptation of MEMS technology for innovative specimen holder design and control
- Large dynamical range detectors and detector efficiencies
- Energy filter with large field of view
- Tunable C_c correction
- Large field of view for dynamic observations
- Enhanced data handling and storage; faster video recording.

He described a number of specific examples of contemporary in situ studies at the U. of I. MRL ranging from in situ vapor deposition to reasonable pressure gas reaction cell experiments (40 Torr rather than 2 Torr as discussed by John Hutchison for the high resolution studies in the Oxford cell; the effects of hydrogen on dislocation velocities). He concluded with a number of specific things which cannot presently be done but which would be well suited to an NTEAM instrument.

Question (Max Haider): One of the requirements that you mentioned was a tunable C_c. Why do you need that?

Response: The chromatic aberration I referred to is actually associated with the specimen and in the case of the gas reaction cell, with the atmosphere. (This problem is dealt with by a post-specimen energy filter.)

Related Comment: Eric Stach: I would just like to echo the need for improved CCDs and parallel recording for in situ studies, which was a part of Ian's first slide. The present state of affairs is surely one of the key limitation for today's in situ microscopy.

Related Comment: Howard Birnbaum You said something that struck me as being worth emphasizing again. Anyone who has done in situ experiments knows that you spend a lot of time before you get things right and before things work. As you make more complex holders, it becomes progressively more difficult to get things to work in the microscope, particularly if you have to operate within the instrumental constraints imposed by the manufacturers, space, feed-throughs etc. I'm going to argue against the modular concept in which you exchange various objective lens parts in setting up your experiment. Rather you should do the experiment outside the microscope first to make sure the various aspects of the holder work, the inputs and outputs are in the right places and so on.

Related Comment: Nestor Zaluzec Howard, although I agree with you in principle, I think it's a big mistake to have so many different modules that you can plug into one machine. You need to focus in on a certain number, decide what you want to do, pick a set. I really didn't like the Illinois idea of the instrument that is going to do everything for everybody. I tried that; it doesn't work.

Response: Howard Birnbaum I know your effort very well and you did not try that. You made one machine that had all these additional things added to it and that does not work, I agree. What I'm talking about is, you build a microscope and if you want to do high resolution, you have a high resolution objective section; if you want to do gas reactions, etc.

The NTEAM doesn't have to start out with a dozen different modules but it has to start out with a design for which various modules can be made over time. If you contract with a manufacturer to deliver a microscope, he will deliver it in 3–5 years, and what you want then may be very different from what you wanted originally. The modular concept gives a flexibility that extends the microscope capability to new areas of science as they develop.

Related Comment: Karl Merkle I believe the different "modules" should be different variations of the NTEAM instrument itself. If you think you can exchange the pole piece in a state-of-the-art instrument, you can forget about it. It won't work.

Response: Howard Birnbaum We are not exchanging pole pieces but exchanging the whole objective lens section (Editorial comment: as MPI—Stuttgart did when the side entry objective section replaced the original top entry section. However, such an exchange in the case of corrected electron optics would require complete realignment of the entire system, a difficult and very time-consuming process).

NTEAM as an In Situ Materials Irradiation Facility
Robert Birtcher

The emphasis for NTEAM within Argonne's Materials Science Division, like that of the MRL at U. of I—Urbana, is in in situ studies. Historically, the specific emphasis over the past twenty years or so largely has been in conjunction with irradiation effects studies, ranging from analysis of fundamental irradiation-induced defect clusters to effects of ion and/or electron irradiation on electrical and mechanical properties and phase equilibria. These were often in the context of nuclear electric power generation and the development of host materials for long term nuclear waste storage. We see ourselves moving into other areas as well, including semiconductor devices, problems encountered in processing, other nanomaterial areas, and low earth orbit and space applications where radiation fields effect both performance of devices and of materials.

Within this DOE User Facility the staff has worked closely with a number of groups from various institutions, including the U. of I—Urbana with which it has had an especially close relationship over the years. The experimental capability of in situ ion irradiation is a core competency in the Materials Science Division which we would include in the Argonne version of NTEAM, but many of the other features outlined by Ian Robertson are quite generic and would also be incorporated. Especially with the possibility of moving the objective aperture to a conjugate focal plane as suggested by one of the speakers, we believe that we could tolerate a somewhat smaller pole piece gap and thereby hopefully achieve better resolution when that is required. The concept of modular experimental modules which Ian Robertson discussed and which was introduced in another workshop here in 1989 in conjunction with possible replacement of the HVEM still seems like a viable approach to increasing the versatility of the instrument without interfering significantly with the design of the microscope or in its ease of operation . Employing such modules is a way of greatly expanding the instrument's capabilities without a proportionate increase in its complexity.

A partial summary of general specifications for the Argonne NTEAM might thus include the following:

- Electron energies 80–300 keV
- Dual electron sources preferred; TEM for most in situ studies
- Ion energies 0.1–1,000 keV or more
- Ion beam interfaces including FIB and other low energy sources such as a cluster beam source (ion beam assisted deposition studies, e.g.)
- Excellent ion and electron dosimetry

- Pole piece gap 11–15 mm
- Objective aperture in a conjugate plane
- Wide variety of specimen holders, preferably compatible with one or more other microscopes
- Microanalysis capabilities
- Remote access, especially for collaboration
- Modules allowing optical, electrical and mechanical probes.

He also described the existng HVEM-Tandem Facility and its instrumentation: the HVEM and 300 kV H-9000 and their ion beam interfaces. Examples of several types of in situ ion irradiation studies of materials were presented.

Comment by Mark Kirk Relying heavily on Ian Robertson's and Bob Birtcher's talks, he describes a specific displacement damage study of a rather fundamental nature involving ion irradiation. The general objective is to do quantitative image and diffraction measurements. An important aspect of this study is the development of a very weak beam imaging technique (e.g., 6g, g) for the imaging at low temperature (20 K) of individual defect clusters which allows discrimination between vacancy and interstitial type clusters from the nature of the contrast. This is work with Mike Jenkins (Oxford) and Hiroshi Fukashima (Hiroshima) which has been published recently. This illustrates a range of in situ processes involving the Hitachi H-9000 in the HVEM-Tandem Facility including very low dose in situ ion irradiation (good dosimetry), use of the Oxford double-tilt He-cooled holder, of 100 kV to avoid annealing of the defects due to sub-threshold events, and of the Gatan 622 camera for precise focusing and stigmation. Resolution requirements for this type of experiment correspond to that of the instrument. He described the experimental and analysis procedures for results in some detail. The very weak beam technique is now being employed in an attempt to identify the embrittling defect in ferrous alloys subject to radiation-induced embrittlement.

He strongly supports the idea proposed by Ian Robertson of having two interchangeable electron sources because in situ studies often require a large field of view (low magnifications, thousands or a few tens of thousands of times) or high magnifications to resolve detail on the nanometer scale or both in a given study. This would be a great advantage.

Related Comment: Bob Birtcher One thing I forgot to mention which Mark's talk reminded me of is the ability to change electron energy without a lot of hastle, because there are many studies in which changing the energy to control or create displacement events or to avoid even subthreshold events is essential.

Comment by Robert Hull This presentation deals with in situ studies in semiconductor materials and devices. Semiconductor materials and systems offer opportunities and motivation for understanding the behavior of materials under extreme conditions: local mechanical stresses (to >1 GPa), "electrical stresses" (to $>10^3$ A/cm^2) and "optical stresses" (optical emission currents from a semiconductor laser to $>10^6$ W/cm^2). The potential technological ramifications of such studies are enormous, the electronics and telecommunications industries being likely to approach a trillion dollars per year by 2010. In this context, the goal is to apply as many "stresses', singly and in combination, and to measure as many "signals" (optical, electrical, microscopical...) as possible in situ, simultaneously. To give an indication of where we are today, Table 10 lists the various specimen holders for in situ experiments in the speaker's laboratory. While these holders have been very useful, more sophisticated versions should certainly be possible which

would be much more useful, versatile and user friendly. One major problem is in making electrical connections. The ability to do micro-machining with in situ FIB would open vast new territory for these studies. Another opportunity might to build wave guide structures into the specimen to get optical signals in and out.

For example, to build wave guides to get optical signals in and out. We have heard a lot about the extraordinary opportunities in electron optics design which are coming to pass. There are parallel opportunities in ion optics design, the present spatial limit today is about 10 nm but 1 nm is conceivable and of great

Table 10. Specimen holders for in situ studies.

Specimen Holders	Temperature Range (°C)	Other Capabilities
Heating: ST and DT	30–1400; 30–1000	
Heating, Electrical, ST	30–1400	Electrical Current >1 A
Heating, Straining, ST	30–1000 [a]	Elongation Rate >0.04–0.4 mm/s
Heating+Electrical+Optical ST	30–500 [b]	Electrical current >1A Optical Flux >102 W-cm^2
Heating+Indentation [c]	30–600	Indentation; xy positioning ~ 10 nm, z ~1 nm

[a] Furnace temperature; heat transfer to semiconductor specimen severely limited. [b] No water cooling. [c] Under construction. ST=single tilt. DT=double tilt.

application possibility. One could consider a broader initiative in "charged particle" microscopy.

He described a number of specific in situ studies including in situ film growth in an Hitachi H-9000 UHV with digermane, disilane and oxygen inputs and a Ti evaporator. The resulting TEM specimen may be transferred in UHV for SEM and AES and for other film growth methods including MBE and e-beam evaporation (in collaboration with R. Tromp and F. Ross at IBM Watson). To illustrate an in situ growth experiment, he showed results of TEM measurements of dislocation velocities during and following growth of SiGe/Si heterostructures in which the time resolution of the camera becomes a problem at relatively low dislocation velocities. More generally, much faster time resolution would allow access to a much broader range of materials problems. Sufficient signal is available if we trade spatial resolution (i.e., number of pixels) for time resolution.

He concluded with two proposals for challenging in situ experiments, one for the electron imaging of Wigner lattices (electron lattices) which form in semiconductor heterostructures at very low temperatures (~100 mK) in fields of order 10 T, and the other for in situ electron microscope observations of ion beam-induced deposition which is less demanding experimentally.

Related Comment: John Spence. I am reminded of the work of Furuya in Japan (in situ FIB in a 200 kV TEM) and also of Petrov at Santa Barbara, the FIB completely destroyed all the electrical properties of the semiconductors. There was a lot of damage and Ga everywhere. In fact Petrov sent a FIB back to the manufacturer for that very reason.

Response: To avoid those extreme levels of damage, low energy ion optics would be well suited. We have also looked at ways of passivating the surfaces during milling to mitigate

these sorts of problems. But you're right, there are problems, but there are also solutions if you take the time to work through them, I believe.

Related Comment: John Spence. Regarding the Wigner lattice proposal, we also looked into this, as others have too, and concluded that the degree of long range order is not all that large in the presence of defects because of the weak binding energy (<1 meV).

Comment by Frances Ross This presentation deals with the UHV TEM at IBM–Watson for in situ studies and with Phil Batson's 120 kV VG STEM with C_s corrector by Nion. So far Phil has completed tests comparing performance before and after installing the aberration corrector. Without the corrector, the resolution is 2.0-2.5 Å and it is not possible, for example, to resolve the dimer pairs at the core of a misfit dislocation in a Si/SiGe quantum well structure. This makes it impossible to distinguish clearly between several models for the dislocation core structure. With the corrector installed, the instrument resolution is improved to about 1.3 Å and dumbbell images have been obtained. Among other experiments, this resolution will remove the ambiguity in images of dislocation cores, allowing analysis of the core structure. A monochromator has also been installed on this microscope and a spectroscopic resolution of 70 meV has been demonstrated. Experiments involving high resolution imaging and spectroscopy of single atomic columns are planned.

The Hitachi H-9000 UHV TEM at IBM's TJ Watson Research Center has been optimized for in situ studies. It is a side entry microscope with a base pressure of 2x10-10 Torr, C_s = 1.2 mm, C_c = 1.5 mm and a 5 mm gap. A Gatan Imaging Filter has also been installed. A variety of in situ experiments can be performed on this microscope: capillary tubes extend into the specimen area so that gases can be introduced for oxidation and chemical vapor deposition, and an electron beam evaporator is located above the pole piece for metal deposition. We have previously grown nanostructures such as Ge islands on Si (100) and studied the effects of oxygen on the C49 to C54 transformation in $TiSi_2$. We have also designed a closed liquid cell for a variety of electrochemical studies. A corrected instrument would significantly improve our ability to do further in situ experiments; in most of our work, a resolution of 3 Å is acceptable, so that a larger polepiece gap, up to one centimeter, may be possible. This will allow more elaborate evaporators, gas dosing and liquid cells to be designed. Furthermore, real time energy filtered imaging would greatly benefit from increased illumination intensity.

Question: How well do you have to allign the beam with respect to the optic axis in a C_s corrected instrument?

Portion of the Response (Max Haider): The beam can be precisely alligned with respect to the optic axis, and then beam tilts of 20–25 mrad are acceptable.

Comment by Eric Van Cappellan This talk is on the important subject of specimen preparation. Everyone recognizes that without a good specimen, the best, most expensive microscope is impotent. Yet there is an incredible discrepancy between budgets for the microscope and for specimen preparation equipment. This may have to change as the requirements for specimens become more refined and complex. He then reviewed the status of FIB for specimen preparation, which is fast, accurate in defining the position of thin area and is compatible for dealing with difficult material systems such as metal/non-metal multilayers. The disadvantages are that there are thin amorphous or at least heavily damaged layers on surfaces, and the capital investment is large. The FIB has been combined with SEM to permit observation of the specimen during thinning. Alternative sources in addition to a Ga, such as In, are being aggressively investigated. The possibility of adding Ar ion milling as a final step for cleaning the surfaces is also under investigation. As Bob Sinclair

indicated you can actually fabricate portions of the specimen, electrical pads or small amorphous areas for focusing and stigmation and so on and thus have a high degree of freedom in designing the specimen. The FIB must be widely accepted in order to reach its full development potential.

Related Comment: Robert Hull Rather than focussed ion beams, if you can get columated beams of Ar, Ne or Xe, that's what we really need for specimen preparation. Could you comment on that please?

Response; If you don't use a liquid metal ion source, then you end up with gigantic pieces of equipment compared to today's FIB, the column of which is quite compact. As I said, only when people accept FIB widely can we expect to see the full potential realized, including what you suggest.

Comment by Vinayak Dravid Various in situ experiments and in situ analyses were described for the Hitachi HF 2000FEG with electron holography and EDS capability at Northwestern. Specimen holders include electrical biasing of a specimen, which, for instance, have been employed to show in a bicrystal specimen an increase by four orders of magnitude the electrical resistivity across the grain boundary. Holography can be employed to map out the electrical potential across such interfaces, which cuts to a very fundamental level. Particularly in non-metals cation impurities at interfaces become very important in determining the properties of the material as a whole. With the bright prospect of aberration corrected electron microscopy, we can have now the expectation of controlled doping of such interfaces (nanotitration) in systematic studies which would involve a variety of analytical techniques including high resolution imaging, elemental and chemical nanoanalysis and electron holography as well as electrical property measurements and the like. He presented other interesting examples including PZT dot array studies and other experiments involving ferroelectric domain wall motion and the design of such a specimen with the aid of FIB.

He also addressed the question of enhanced capability for in situ experiments by incorporation of piezoelectric positioning for specimen manipulation with nanometer precision. In fact, one might consider putting an AFM or STM in the microscope column as an additional analytical tool. An example of the potential power of combining STM and TEM is in studies of the initiation of surface cracks in materials under in situ loading conditions. He also described in situ nanolithography using this technology. The final area he suggested was the imaging of fluids containing nanoparticles in the TEM.

Discussion: Regarding the last suggestion which would be of interest particularly to the biological community, the question was raised by Robert Hull of trying to interest NIH in sponsoring such development and subsequent research, including the application of high resolution spectroscopy to such solid/fluid systems. Vinayak responded that indeed most of the users interested in this area are life science users funded by NIH. John Spence added that they had submitted such a proposal and quickly discovered that for NIH to even consider a proposal requires a tremendous amount of preparatory development. Vinayak added though that he had had a recent conversation with someone at NIH regarding this policy, and he was assured that this was not categorically the case.

Comment by Laurie Marks He described the modular UHV analytical systems interfaced to a Hitachi UHV H-9000 with LaB_6 (known as Mark II) at Northwestern, which include an MBE deposition chamber and an analytical chamber incorporating a dual anode X-ray source, field emission electron gun and a spherical capacitance electron energy analyzer. He described a number of studies for which this complex array of capability has been

employed for electron crystallography, in situ thin film growth and studies of surface structures and dynamics. He then addressed the question of the potential value of aberration corrected instrumentation for surface imaging and electron crystallography. In surface imaging, the major issue is signal because the intensity of surface reflections are typically three or four orders of magnitude weaker than the transmitted beam; that is, in studies of surfaces, when it comes to S/N, diffraction is always going to beat imaging hands down. He pointed out that there is no UHV TEM with FEG in this country (several in Japan though) and only one UHV TEM with a GIF in the world (at IBM). Spherical aberration correction may be helpful in plan view surface imaging in conjunction with FEG because of increased current density, but the system must be bakeable (in profile imaging of surfaces, a LaB_6 source in an uncorrected microscope is superior to FEG because of the latter's image delocalization problem). In the case of electron crystallography, he suggested that aberration correction would allow a precession camera (involving automated fully eucentric tilting, beam tilting for HOLZ) to be a powerful new tool for studies of small areas (~10 nm; a quasi-kinematical diffraction pattern which can directly interpreted as in the X-ray case). Probably a STEM-type approach. For electron crystallography, this could be a very significant development. In these cases, a larger pole piece gap is not required.

Related Comment: Murray Gibson. We have been interested in thin film epitaxial growth and the measurement of strain associated with the several aspects of that. In dark field imaging which is very useful, for such situations the strains are not associated with quantized sources like Burgers vectors of dislocations, for example, and as a consequence more precise measurements have to be made. The issue is control of deviation parameters to permit such higher precision and we don't do this very well as a rule. He then presented recently published results showing strains associated with individual quantum dots as a function of their size, from which it was possible to draw some conclusions regarding the growth processes. It would be so much easier and superior if one could observe a single area of specimen and change the incident beam orientation (to control the deviation parameters), which is one of the things you get with aberration correction. He disagreed, of course, with Laurie regarding the general utility of C_s correction. He also disagreed regarding the space issue and quoted the Cu film growth experiments in a TEM equipped with an Auger spectrometer. When you have access to more space you certainly can do more things to characterize the material which makes the experiment more relevant to the real world. The more tools of characterization, the better.

Related Comment: Knut Urban Regarding the delocalization issue, that all depends on the resolution you are after. If you use an aperture to exclude the rapid oscillations in the transfer function in an FEG instrument, there is essentially no delocalization. So if you are satisfied with 2.5 Å in the basic CM 200, say, delocalization is not an issue for you in a FEG instrument either. And the latter has a lot more possibilities to offer.

Laurie Marks responded that his point was that for surface microscopy, aberration correction buys you very little additional and probably only non-essential information. But John Spence expressed a different view. In the quantification of selected area diffraction, the area contributing to the pattern is determined by C_s. In corrected instruments, you can get patterns from smaller, better defined areas where each part of the pattern represents exactly the same area. Alwyn Eades wondered who in his right mind even uses selected area diffraction anymore. John Spence responded that it is the only way to study diffuse scattering. The discussion continued for some time thereafter.

Comment by Al Meldrum This presentation deals with two different types of problems: first, in situ studies of geologic materials in conjunction with radiogenic age dating and second, the characterization of nanocrystalline systems, for instance, in the context of

optoelectronic properties of interfaces of semiconductor nanocrystals. He approached these subjects from the point of view of a regular user of both the HVEM-Tandem Facility at Argonne and the Metals and Ceramics Microscopy Facility (through SHaRE) at Oak Ridge, beginning with his days as a graduate student at the University of New Mexico. In the studies of geochronology, in situ ion beam techniques are employed to study radiation damage in materials in order to establish a time-temperature model for the behavior of radioisotope-bearing minerals over their lifetime. For these studies current instrumentation poses primarily two important limitations for more accurate modeling purposes, inadequate in situ high resolution imaging capability and insufficiently precise EELS analysis of individual damage cascades. The variations of NTEAM which have been described including in situ ion irradiation capability should readily mitigate such limitations.

The second area is the characterization of nanocrystalline materials, especially semiconductors and ferromagnetic fine particles, which is now recognized as a fundamentally as well as technologically important research area, and for which we are certainly pushing the limits of current generation instrumentation. Several examples are described, such as the apparent ion beam modification of nanocrystal interfaces using not only high resolution imaging but also X-ray mapping and EELS analysis for chemical bonding information at interfaces such as for Si nanoparticles in silica. From present state-of-the-art, "we think we are modifying the interfaces, but we do not have the TEM technology to adequately characterize them." In fact, significant TEM is remarkably absent from current nanoparticle symposia which, however, routinely include large numbers of synchrotron studies. Somehow we are missing the boat. Again fine, intense electron probes associated with aberration corrected instrumentation should largely mitigate these limitations. Especially in the in situ versions it should also be possible to study stimulated light emission or other appropriate signals in parallel with the other more traditional analysis techniques.

Related Comment: Howard Birnbaum Let me comment on the lack of TEM in nanoscience research, an area which attracts scientists from a wide range of diverse disciplines and backgrounds. Electron microscopy is never going to be there if we depend on these people to become electron microscopists. The challenge for the microscopy community is to create liason so that joint work is done.

Related Comment: Christian Kisielowski We are all swept up by the growing interest in nanotechnology and nanoscience; we see this in the users of the NCEM, for instance. From your talk I thought particularly well presented was the need for many kinds of analysis on the atomic scale. It struck me that this was closely connected with one of the points Uli Dahmen has made regarding the need for being able to tilt to very different zone axes to "see" high resolution details from the sea of nanoparticles. I completely disagree with those who claim they need only 2 or 3 Å resolution; in nanoscience every fraction of an Ångstrom improvement will turn out to be valuable.

The speaker agreed wholeheartedly with both of these comments.

Session 3: Analytical Studies and Aberration-Free Microscopy and Microanalysis
Session Chair: Ian Anderson

The fine, intense electron probe provided by the combination of FEG and C_s-correction present exciting prospects for elemental nanoanalysis and electronic structure information at the level of single atom columns and atomically sharp interfaces.

Preliminary Remarks: Ian Anderson

As we have heard, aberration correction promises a host of advantages for imaging of electron transparent crystals, both in CTEM and STEM, including higher spatial resolution, reduced contrast delocalization and wider pole piece gaps. Concurrent developments in analytical detectors will enhance the versatility of these next generation instruments for materials characterization, including EELS detectors with higher spectral resolution and EDX detectors with higher spectral resolution or count rates. For in situ microscopy studies, improved (faster) video recording has been identified as a key enabling technology.

Analytical microscopy offers special challenges for aberration correction for at least two reasons: first, characteristic signals have significantly lower yields than corresponding image signals and second, analytical signals may suffer from large tails on otherwise sharp probes. Lower signal yields mean longer acquisition times (resolution may be limited by specimen drift) or higher probe currents (resolution may be limited due to opening of apertures). To compensate we may need to trade spatial resolution for probe current. (Nigel Browning has suggested that probe currents for imaging are sufficient for collection of EELS signals and presumably EDX signals also; noisy spectrum images may provide sufficient integrated signal.) The existence of large tails on sharp probes is a problem because the electron distribution from the entire probe contributes to the effective resolution, not FWHM or just the sharp probe.

Analytical studies have already figured prominently in Workshop presentations. For example, Uli Dahmen and Christian Kisielowski made reference to single column EELS as a natural complement to aberration-corrected HRTEM; Matt Libera, to EELS spectrum imaging of polymers (low loss and core loss); Nigel Browning, to EELS concurrent with Z-contrast imaging; and Frances Ross, to Phil Batson's EELS of dislocations in semiconductors. The message here seems clear: as we approach true atomic resolution, there will be more and more mixing among the traditional disciplines of high resolution, in situ and analytical.

Comment by Ondrej Krivanek

I would like to expand on several ideas I mentioned in my talk the other day. For STEM microanalysis and imaging, both the probe current and the probe size are important. With full correction of spherical aberration (C_3), the probe size is mainly limited by C_5, i.e.:

$$d_5 \sim 0.4\, C_5^{1/6}\, I^{5/6}.$$

In the next generation of aberration correction we will worry about correcting C_5. We will then probably begin to see the effects of C_7, namely,

$$d_7 \sim a_7\, C_7^{1/6}\, I^{7/8}.$$

Hence for higher order aberrations, the probe size will be roughly proportional to l and thus the resolution will improve with increased primary energy. In going from 100 keV to 200 keV, for instance, 1 Å probe should be reduced to about 0.7 Å.

Similar considerations apply to the resolution limit due to chromatic aberration, which may well become an important factor for NTEAM. It is:

$$d_{cc} \sim a_{cc} (C_c \, l \, DE/E_o)^{1/2}.$$

Because l is proportional to $Eo^{-0.5}$ (and to $1/E_o$ at relativistic energies) d_{cc} also is roughly proportional to l/l. Hence once more the resolution will improve by going to higher primary energies. In other words, attaining close to 1 Å resolution at 100 keV, as we have already done, shows that sub-Ångstrom resolution should be readily reachable at 200 keV and higher.

Another point I would like to emphasize is that using a STEM probe that is no bigger than the resolution limit due to aberrations is O.K. for high resolution imaging, but not a good practice for microanalysis. This is because for rapid and noise-free microanalysis you need a probe current of 0.1 – 1 nA, which can only be obtained if you don't demagnify the image of the source too much. Taking the effects of finite source demagnification into account, the STEM probe size is approximately:

$$d_{total} \sim (d_{aber}^2 + d_{geom}^2)^{0.5}$$

where d_{aber} is the probe size due to the aberrations and d_{geom} is the ideal size of the demagnified source, i.e. the probe size that we would get due to the finite source size in the absence of any aberrations. The probe current is proportional to d_{geom}^2, and hence for microanalysis it crucial not to reduce d_{total} to be close to d_{aber}, as this means reducing d_{geom} so much that almost no current remains in the probe. As a practical example, with a 100 kV aberration-corrected STEM with an aberration-limited probe size of 0.7 Å, to attain a total probe size of 0.8 Å, you must demagnify the source to $d_{geom} \sim 0.4$ Å, whereas for a 2 Å probe size, demagnifying the source to $d_{geom} \sim 1.9$ Å is sufficient. Hence the 2 Å probe will contain about 20 times more current than the 0.8 Å one, even though it is only 2.5x as large. With a brightness of 2×10^9 A $(cm^2 \, str)^{-1}$, which may be attainable with CFEG, the 2 Å probe will have nearly 1 nA of current in it. At this level of current and resolution, single atom nanoanalysis (forget microanalysis now) should be possible in a wide variety of materials. In a doped semiconductor system, you might be able to map out just where the individual dopant atoms are. I can't think of any other generally applicable technique which can analyze single atoms in situ and determine their chemical species.

Question: How about atom probe techniques?

Response: The specimen is miniscule, hard to prepare, and destroyed in the mapping process. At any time only surface atoms are analyzed.

Question: How about the effect of specimen thickness on beam spreading?

Response: Electron channeling largely mitigates through-thickness beam spreading when precise atomic column orientations can be chosen with respect to the incident electron.

Another point I would like to address is the addition of aberration contributions from different lenses in the microscope column. Each element in the column contributes to the net spherical aberration in a way which strongly depends on the width of the beam in that element. The conventional way of expressing this is to consider the magnification of the intermediate crossovers following each lens (in the STEM) or the magnification of the intermediate image preceding the lens (in the CTEM) when adding up the aberrations. For N lenses, each with an intermediate magnification M_n, the total spherical aberration is:

$$C_{s\ total} = \sum_{n=1}^{N} C_{sn} / M_n^4$$

Hence the parts of the probe-forming system that make the largest contribution to $C_{s\ total}$ are the ones that produce the smallest intermediate crossovers. Normally there is only one lens in the column that produces a very small crossover - the objective lens. But in a cold field emission STEM being used to produce a large beam current, the size of the (virtual) crossover produced by the gun (typically a few nm) can become comparable to the size of the probe on the sample. At this point the contribution of the gun and of the first condenser to $C_{s\ total}$ will begin to dominate, and $C_{s\ total}$ will go through the roof. For instance, we have experimentally measured $C_{s\ total}$ of an uncorrected VG HB 5 for a final probe size of around 2 nm, and found it to be greater than 100 mm! This is precisely the reason why cold field emission normally does not do well in producing large currents into moderate size probes. However, with an aberration corrector, even a $C_{s\ total}$ of several meters will be correctable, and hence this limit will largely disappear, as long as the corrector's optics is flexible enough to correct the wide range of $C_{s\ total}$ values that may be encountered depending on what source demagnification one is working with.

That concludes my comments directed at microanalysis. Now I would like to turn for a moment to a topic that seems to be rather controversial, and that is how to bring together the different parties that need to speak with one voice if the NTEAM proposal is to be successful.

A unifying idea for the NTEAM, it seems to me, would be to essentially Lego-ize the instrument, i.e. to define the basic modules and leave the specific configuration up to the users. With Lego, a kid can use her imagination and creativity to build a castle or a missile. This kind of approach might be what's needed to unite the various constituencies at the National Labs to stand behind one proposal and at the same time appeal to the broader community. The NTEAM initiative then might come down to a microscope which is reconfigurable. If this were the case, a lot of effort should be devoted to defining the basic building blocks. Standardize the interface including the various aberration correctors and let the user decide what objective lens assembly to put in. At any given time the instrument will not be able to do everything, but over time by reconfiguring, it may. This mitigates all of the arguments about large gap or narrow gap, corrector before the objective or after the objective, microanalysis or not and so on. Perhaps NTEAM should therefore stand for National Transformable Electron Analysis Machine.

Comment by Max Haider As an introduction to comments on aberration correction for STEM, he briefly reviewed C_s and C_c correction for a low voltage SEM which had been designed and constructed in the European Molecular Biology Laboratory (EMBL) in Heidelberg in the 1990s [J. Zach, M. Haider, Nucl. Instr. and Mth. A 363 (1995) 316]. This consisted of a field emitter and corrector with four quadrupoles; this system has the intrinsic

advantage of allowing compensation of axial chromatic aberration C_c, provided at least the two inner elements are compound electromagnetic elements. If such a quadrupole corrector is employed only for correction of C_s (= C_3), this system is more complicated than the hexapole corrector (the number of non-rotationally symmetric elements is doubled). But if both C_c and C_s are to be compensated in an SEM or a high resolution STEM, a quadrupole corrector is required, consisting of at least four electrostatic or magnetic quadrupole fields, of which at least two have electrostatic and magnetic elements combined for simultaneous excitation. A larger number of multipole elements increases flexibility, but at the expense of additional power supplies and alignment complexity. [For a more detailed discussion and comparison of corrector configurations for STEM and TEM and the problem of C_c correction and ultimate STEM resolution, see M. Haider, S. Uhlemann, J. Zach, Ultramicroscopy 81 (2000) 163–175.]

In the EMBL corrected low voltage SEM, in addition to the improved resolution at 1 kV, for example, there was significant improvement of image contrast relative to that for the uncorrected instrument. In STEM, as in SEM, the subject of contrast is often as important as that of resolution. This is directly connected with the sharp intensity of the C_c-corrected electron probe; for example, for a 17 mrad probe of a 100 kV STEM, the peak intensity of the corrected probe is about three times that of the uncorrected probe, normalized to the same total current. The effect of C_c-correction is to suppress the long energy tail of the probe. Another way to look at this is to consider the fraction of electrons in a given probe confined within a certain diameter at the specimen: again for a 17 mrad probe, 59% of electrons are confined within 1.4 Å in the corrected case and 3.1 Å in the uncorrected case.

Analytical Electron Microscopy in an Aberration-Free Environment
Jim Bentley

This presentation was prepared by the speaker in collaboration with Ian Anderson, Ed Kenik and Neal Evans. It will include three main topics:

- Signal strength and instrument configuration for analytical electron microscopy (AEM)
- Cc correction for energy filtered TEM analyses
- Aberration correction for analyses in STEM modes (EELS, EDS), electron diffraction and ALCHEMI.

Low signal strength in analytical electron microscopy has several important implications. We must realize that the factors limiting AEM are different from those for HREM and thus the implications for spherical and chromatic aberration correction are also somewhat different. While less of a problem in EDS than in EELS, element-specific spectroscopies involve signals which are many orders of magnitude lower than those required for imaging techniques. For example, following is a list of relative signal strengths involved in various analysis methods for a typical 20 nm thick ferrous alloy:

Analysis Method	Signal/Incident Intensity
CBED	1
HREM	0.5
BF TEM	0.3
Weak Beam DF	10^{-2}
HAADF STEM	10^{-3}
SAD	10^{-3}

EFTEM Fe-L23	10^{-5}
Fe K- or L-Shell X-rays	10^{-8}

To compensate for weak signals larger probe currents (size) and/or acquisition times must be employed.

To take advantage of improvements in analytical instrumentation which can complement those for aberration corrected instrumentation instrument configuration will involve mechanically more stable specimen stages permitting longer exposure times in the collection of spectrum images and larger space for advanced detectors, detector geometries and non-conventional spectrometers, for example, detector arrays and improved collection efficiency, X-ray focussing optics, and lower systems background for higher peak to background ratios. For maximum analytical flexibility, the microscope should be configured for both CTEM and STEM, permitting hybrid operational modes such as rocking beam for ALCHEMI (1 mrad CTEM-like illumination plus beam scanning to raster the incident orientation). Of course, spectrum imaging (a data set allowing any series of energy-filtered images or series of spectra to be accessed. A number of specific examples were discussed illustrating some of the variety of chemical and elemental information which already can be employed and the current limitations and prospects for these with vastly improved instrumentation.

Jim Bentley then turned to the topic of the importance of C_c correction for EFTEM analyses. A rather wide variety of aspects related to this may be summarized as follows:

- Core-loss EFTEM yields composition and sometimes bonding information, which involves many pixels but relatively short acquisition times; it may be especially advantageous for damage-rate limited processes.
- Spatial resolution for EFTEM analyses are more limited by a combination of S/N and objective lens chromatic aberration than by spherical aberration.
- For good resolution pixel size (mag), binning, probe current and exposure time must be appropriately matched.
- Post-specimen C_c correction has an immediate impact on resolution.
- Atomic resolution should be possible routinely for some elements.
- Improved detectability of low concentrations at more modest resolutions.
- Equivalent considerations for TEM spectrum lines will allow high spatial resolution over an extended energy-loss range.

For analytical STEM modes, the implications of aberration correction were summarized as follows:

- STEM-EELS is more efficient than EFTEM; maximum spectral information for a given dose; the better choice for process limited by total dose.
- Required are ~0.3 nm probes with ~1 nA, not <0.1 nm with tens of pA.
- Spectroscopy at today's routine HREM level is not an inconsiderable advance; at or beyond the practical limit for EDS (beam broadening); at EELS limit (interaction delocalization).
- 0.1 eV EELS with monochromator, pre-specimen aberration corrector and high resolution electron spectrometer seem realistically achievable.
- "Synchrotron spectral resolution at atomic spatial resolution".
- Bonding and chemical effects at defects through near-edge fine structure, especially in combination with first-principles theoretical calculations of electronic structure. For example, segregation and bonding effects at interfaces (alloy embrittlement, electroceramics), catalysts and other nanostructured materials.

Several of the points above were illustrated with information gleaned from the Key West Workshop in January 2000.

For electron diffraction the implications of aberration correction are summarized in the following:

- Extend ongoing work to measure accurately structure factors by analyzing zero-loss filtered (elastic scattering) convergent beam electron diffraction patterns with dynamical theory calculations.
- Really-wide-angle CBED becomes compatible with small probes for the first time.
- Possibly useful for orientation imaging microscopy (OIM) at a resolution of ~1 nm, e.g., for nanocrystalline materials; perhaps other modes of electron diffraction will evolve as a consequence.
- Energy-filtered CBED patterns to map lattice parameter or strain variations at high spatial resolution.

Aberration correction will also have a major impact on X-ray spectrometry. EDS is a very broadly applicable tool for compositional analysis and will surely remain so. The topic of microcalorimetry (bolometry) will be discussed in more detail by Jim Howard, but a brief summary of relevant points are as follows:

- The bolometer achieves more than an order of magnitude improvement in resolution as well as sensitivity because of improved peak to background for equivalent collection geometries.
- Diminished spectral overlaps.
- Single atom detectability across the periodic table; complements EELS.
- For specimen thickness of 20 nm and probe size <0.5 nm, 1 nA is adequate.
- Bolometer count rates (1000 cps) are compatible with high spatial resolution (a small specimen thickness limits beam broadening).
- Extend interfacial segregation sensitivity to 0.01 ML. E.g., interfacial segregation in Ni-based superalloys, there is reduced spectral overlap among lines from 5d transition elements such as Hf, Ta, W, and Re and K lines of Ni and Al. Possible to measure low concentrations of dopants in semiconductors.

A number of additional examples were presented of various techniques and materials applications.

The possibilities for significantly improved Atom Location by Channeling-Enhanced Microanalysis (ALCHEMI) were summarized as follows:

- Pre-specimen C_s correction will allow beam orientation to be varied without significant translation of the beam with respect to the specimen.
- "Automated" conventional ALCHEMI at 10 nm resolution.
- "Real-space" ALCHEMI with small probes correlating characteristic X-ray variations as probe is translated within the unit cell to yield site occupancies.
- Extend applications to ordered alloys exhibiting poor site discrimination because elastic scattering amplitudes of adjacent planes or columns are similar (L12 ordered Ni_3Fe).
- Extend applications to selected ordered alloys with multiple sublattices occupied by a single host element (ordered $Mg_{12}La$ intermetallic).

Several examples of ALCHEMI results were described.

As Ian Anderson pointed out in his introductory comments, it is very likely that significant improvements in resolution, probe size and space will stimulate much more integration of the methods of high resolution, in situ and analytical studies. We can now think about combining HREM structural information with high resolution elemental and chemical information for understanding applications such as segregation at interfaces. Or to explore

the possibility of obtaining AEM data fast enough to follow compositional evolution during in situ experiments, such as diffusional phase transformations.

For a number of systems, EFTEM composition mapping methods may become feasible but a number of factors come into play. For the specimen, the energy loss in EELS must be sufficiently large to be localized, but not too large in order to retain sufficient signal from a reasonable thickness. For the collection of data and acquisition of information content at the target resolution, collection angle, window width and magnification (pixel size) must be optimized. For EFTEM composition mapping, the limiting instrumental factor appears to be incident current density. For example, we know from work in an uncorrected 200 kV FEG (Schottky) instrument with GIF the necessary defocus of the intense but highly non-uniform illumination results in insignificant gain in current density over LaB_6. Finally, even if the experimental challenges can be met, elastic-inelastic multiple scattering will complicate direct image interpretation.

The challenges are considerable, but meeting them will be exciting.

Question (Alwyn Eades): I don't think you need C_s correction to do ALCHEMI mapping with better spatial resolution. Just use look-up tables as you drive the deflectors. And a second comment on your really wide angle CBED; I presume that has the bright and dark field superimposed in which case that is what is called a Kossel pattern. In the application you suggested for orientation mapping of small grains, at the moment the TSL method of conical scanning looks more promising.

Response: Absolutely. I don't know why someone hasn't done that. And on the second remark, I agree.

Comment (Bob Sinclair): For thermally activated reactions, all you have to do is reduce the temperature so the reaction is slower so you have sufficient time to collect the data. So microanalysis during the experiment seems very feasible.

Response: Do you have to worry about relative rates of surface and volume diffusion then.

Reply: Not usually. At least ion milled specimens have a thin layer which prevents short circuits to the surface.

Question: The bigger gap means a larger solid angle for detection, doesn't it?

Response: I'm not sure the larger gap allows the collection angle to be larger necessarily. It does make interfacing the detector easier though. The net result of a 30 mm^2 Si crystal coupled with the distance to the specimen and columation for today's detectors will probably not change significantly.

Comment: A bigger detector is not a good approach. What you want is to put in multiple detectors that you can control. A bigger gap will let you do that.

Comment by James Howard: For many years elemental microanalysis has been performed in SEM, TEM/STEM and dedicated STEM by X-ray energy dispersive spectroscopy (EDS) commonly employing Li-drifted Si detectors (Si[Li]). The purpose of this presentation is to describe current developments of other analytical X-ray detectors as alternatives to common present day practice. These include the following:

- Wavelength dispersive spectrometers (WDS) employing hydrid X-ray optics (improved light element sensitivity)

- Microcalorimeter EDS (WDS resolution with most of the benefits of semiconductor EDS)
- Silicon drift chamber detectors (very high throughput but with energy and resolution tradeoffs).

In the first case, the hybrid X-ray optic element between the specimen and the WDS spectrometer is a capillary which partially parallelizes the X-ray beam to compensate for the combination of distance to the spectrometer and the relatively low detection efficiency. The optics are particularly well suited for low energies (0–10 keV). Intensity gains of a factor of more than 10 are realized for energies below 1 keV with typical WDS peak to background ratios but the data collection is serial.

The microcalorimenter, based on developments at NIST, employs resistance thermometry at ~100 mK, just above the superconducting transition temperature of the thermometer material, to detect photons of different energies from the heat they deposit. The energy resolution is outstanding compared to semiconductor EDS (at 10 times the price though) and 2–3 times worse than WDS for energies in the 1 keV range. Data collection is parallel.

Silicon drift chamber detectors count electron-hole pairs with 150 eV resolution at 250 K, 5 mm^2 detector and 1,000 cps; at 30,000 cps, the resolution is still 170 eV. They can be operated routinely at 300 K and have high count rate capability. They are an especially good survey tool for X-ray mapping.

In summary, refinement of semiconductor EDS is reaching its limits, but will surely be the best value for all around microanalysis for some time to come. Si drift chambers could be the best high count rate detectors. WDS with hybrid X-ray optics bridges the gap between current EDS and microcalorimeter EDS. And finally, microcalorimeters have great potential for microanalysis but have a long way to go to compete in solid angle and count rate with Si[Li] detectors.

Question: With the cryogenic system in place why don't you have an array of detectors instead of just one?

Response: It's a major problem because of all the thermal shielding for each detector to get them in advantageous positions. We don't see being able to do that, at least for the next few years.

Spectrum Imaging and Multivariant Techniques
Ian Anderson

The technique of spectrum imaging provides a new paradigm that combines the strengths of imaging and microanalysis. In contrast to conventional energy dispersive spectroscopy, for example, which has provided mainly spot and line analyses of particular elements, spectrum imaging will acquire the entire spectrum in parallel creating comprehensive, correlated imaging and microanalysis of a microstructure. Of course, the method is applicable in both SEM and STEM. A full spectrum is acquired for each pixel in a two dimensional array (the "spectrum image"). Large raw data sets result. For example, such an array with today's technology might involve a two hour acquisition time including a 1 s dwell time for each of 100x75 pixels and 1024 channels, yielding a 15 MByte file. In the near future this will expand to 1024x768 pixels and 2048 channels, for instance, which will yield a 3 GByte file. Methods for mining such large data sets are required to take full advantage of spectrum imaging (for example, the commercial spectrum imaging package by EMiSPEC allows elemental mapping of a selected area, accessing only a small part of the potential

information). Such a method is multivariant statistical analysis which satisfies the following key design criteria for such analysis procedures:

- Works for all data sets, not just for special cases
- Analyses are stable (convergent)
- Require no *a priori* input on the part of the operator
- There are no adjustable parameters
- All spectrally distinct features are identified
- There is strong potential for automated analysis.

Large data sets are not peculiar to microanalysis as has been pointed out in several other talks previously. As CCD arrays become larger, for example, such data sets for images and diffraction patterns will become more and more unwieldy and also will require development of robust ways for analyzing these very large data sets.

(Ian presented several examples of ways in which multivariant statistical analysis may be applied to spectrum imaging data from a computer chip cross section.)

What do people think about collecting very noisy images in which you can't see what you want to see in a given image?

Comment: Murray Gibson Ken Downing mentioned an example of that involving the addition of a large number of very noisy images to produce one with the necessary information. Also another example of imaging is the technique of variable coherence microscopy. Your point is well taken. If you are going to take some average property of the image, say, the information per pixel could go practically to zero and there is still a lot you will be able to do if you recognize that fact, not only in microanalysis but in high resolution imaging and so on.

Comment by Jian Min Zuo Many comparisons have been made for electrons, X-rays and neutrons in many scientific and non-scientific areas and I would like to add one more comparison here. We want to take advantage of the basic differences for these three by combining information which they provide to extract accurate structural information. The structure factors for electrons and X-rays behave very differently for ions while neutrons interact with nuclei only, and the magnitudes of the three scattering factors are very different, especially electrons scatter 10^4 times stronger than X-rays and even more compared to neutrons. For low angle scattering angles electrons are unusually sensitive to valence electron distribution of the target. For materials with large lattice parameters such as high Tc oxides, low order elastic scattering of electrons approaches the low angle regime where the valence electron scattering is large. At high angles electron scattering factors taper off and become fairly constant, reflecting the target nuclear positions. For X-ray scattering, sensitivity is to electron density. We would be able to communicate effectively with chemists and physicists if we could show how the three probes can be combined to reveal very fundamental details of material structure: bonding, magnetic spin arrangements, as well as atomic positions. This is what we have done for copper oxide, which John Spence mentioned very briefly. By combining electron diffraction measurement of low order structure factors with high-order structure factors from X-ray diffraction, we were able to obtain a highly accurate charge density difference map of cuprite revealing d-orbital holes in the copper oxide.

He also described application of small electron probe for studying local structures and the advantage of the Cs-corrector and energy-filtering for in-situ study of phase transitions using electron diffuse scattering technique. The advantage here is that electron can probe length scales from sub-nanometer to microns, which is very useful for characterizing

complex materials with many length scales. Electron diffraction differs from X-ray and neutron scattering in that electrons can form images at atomic resolution. Diffraction gives quantitative structural information. Our challenge is how to combine the real-space high resolution imaging with localized quantitative electron diffraction to obtain critical structural information about complex materials.

Comment by Nestor Zaluzec The purpose of what we are after here is more signal to improve the data collection process. What is the limitation of the signal, where does it come from? The system must be stable; mechanically and electrically stable. If we are to generate a 1 Å probe, it doesn't do us any good if the specimen drift is 5 Å/min. We need to spend time on perfecting that. We need to spend time on perfecting the electron sources. We have heard a lot about illumination correctors but not much about the illumination source. I still prefer cold FEG as the better way to go, but to use it we have to solve the problem of the cold FEG, the source being too close to all manner of surfaces and becoming contaminated. We need to think about the specific scattering event we are trying to measure, for imaging, for spectroscopy of this or that sort and so on. We need to optimize the process involved in that scattering event. And optimize the detectors. I see aberration correctors at two ends of the spectrum; one with the STEM and its probe-forming system and the other, the image corrector. We are trying in both cases to collect the signals more appropriately and more efficiently. The aberration corrector should optimize things for the particular scattering process.

Summary Remarks: Ian Anderson. We heard from both Ondrej and Max what aberration correction buys us in the analytical world, not so much by pushing the envelope of spatial resolution but by so significantly increasing the probe current at a given spatial resolution. In particular it was gratifying to see the viewgraph which Ondrej showed of resolution vs probe current; we operate on the flat portion of the curve so that we can worry about the envelope on the detector side instead of the probe current side of the problem. We expect to see detector developments to capitalize on the improved probe current. The statistical methods which are available today will allow us to take full advantage of deriving good statistics from noisy spectra.

Closing Comments: Murray Gibson

Scientific Challenges

A number of specific scientific challenges which have not been met satisfactorily by current technology emerged from the presentations. These challenges are presented here according to the organization suggested in Uli Dahmen's overview on the second day of the Workshop. Five key areas are emphasized: interface science, defect science, phase transformations, nanostructured materials, and microelectronics.

Interface Science

Internal interfaces are far less understood than are surfaces. In addition to the fact that most surface phenomena have interface analogs, interfaces are under solid constraints associated with elasticity, plasticity and bicrystallography, resulting in entirely new phenomena.

- Mapping interfacial segregation with sub-monolayer accuracy
- Probing electronic structure with atomic column resolution
- Quantitative determination of non-periodic atomic structure and local relaxation

- Observing atomic mechanisms and dynamics of deformation at interfaces

Defect Science

Defects control much of the behavior of crystalline solids; e.g., point defects (diffusion, irradiation effects), line defects (deformation, crystal growth, some phase transformations) and planar defects (deformation, intergrowths, point defect reactions).

- 3D reconstruction of bulk and interfacial defect structures
- Atomic resolution imaging of dislocation core structures in metals, superalloys, semiconductors and ceramics
- Imaging of point defects and small clusters of defects
- High resolution mapping of localized strains with high precision
- Determination of local electronic structure around defects in semiconductors and insulators

Phase Transformations

The study of solid state phase transformations is the foundation of modern materials science. Design of new materials through characterization of processing-related structures, compositions and bonding.

- Observing atomic mechanisms and dynamics of phase transformations at interfaces and second phase precipitates
- Quantitative error bars on local atomic positions or identity of chemical species
- Phase identification
- Nanoscale compositional analysis
- Atomic scale mechanisms and dynamics of transformations involving crystalline phases
- Atomic structure of glasses and the crystal <-> glass transition

Nanostructured Materials

Nanoscale materials are of increasing scientific and technological importance; e.g., in catalysis and quantum confinement. In general, electronic, optical, magnetic, mechanical thermodynamic, chemical and kinetic properties are size dependent.

- Nanocrystallography and phase identification
- Correlation of nanoscale theory and experiment
- In situ measuement of electrical, magnetic and mechanical properties
- Interface structures in nano crystalline systems
- Size and shape dependence of phase transformations in nanostructural systems

MIcroelectronics

Device structures are constantly decreasing in size. Electron optical imaging is the only technique capable of resolving many such structures

- Structure of gate oxide and amorphous/crystalline interfaces
- Imaging core structures of interfaces and dislocations with atomic site discrimination
- Mapping residual strains with high precision and sub nanometer spatial resolution

Some Specific Frontiers in Materials Research

It is possible to identify several outstanding breakthroughs in materials research which can be expected from the availability of spherical and chromatic aberration corrected TEM/STEM.

A Frontier of High Resolution Imaging and Elemental Nanoanalysis

For the most part at present, only one or two projections of crystal structure are achievable in HREM because limited spatial resolution allows usually only those crystal orientations with maximum or near maximum atomic column spacings to be usefully accessed. This is especially true for oxides, nitrides and other non-metallic materials. With the ability to achieve direct sub-Ångstrom image resolution on a routine basis, structural tomography (structure image reconstruction in 3D) can become a reality. For this technique, high angle annular dark field STEM imaging (HAADF) which is dominated by incoherent scattering may be better suited than TEM because of the diminished importance of changing projected thickness with specimen tilt on image contrast and the somewhat more direct interpretation of high resolution images.

In the case of chemical and elemental nanoanalysis, electron channeling largely mitigates through-thickness beam spreading when precise atomic column orientations can be chosen with respect to the incident probe. Thus for a 1–1.5 Å probe, single column analysis should be realizable. This is one of the principal objectives of the spherical aberration corrected STEMs which are currently being engineered and tested (the IBM, Cornell and Oak Ridge VG STEMs, for example, in collaboration with Nion, Inc.). In the STEM mode, the chemical analog of structural tomography may also become a reality, utilizing already well established spectrum imaging procedures.

The combination of structural and chemical tomography would raise the level of sophistication and quantitation of interface science to that of surface science—a truly exciting breakthrough in materials research. For realization, both tomographies strongly depend on instrumental aberration correction—certainly spherical aberration and, for best results, chromatic aberration as well. To this end, current efforts underway in Europe and the USA should be encouraged, supported and extended.

A Frontier of In Situ Materials Research

By their very nature, as clearly indicated by a number of Workshop presentations, ideally in situ materials experiments involve manipulation and probing of the specimen in one or more ways *simultaneously* during observation, often combining heating or cooling, straining, irradiating with ions, photons and/or electrons, controlling magnetic or electric fields, energizing devices and making a variety of physical property measurements, environmental assessments and chemical analyses. The introduction of spherical and chromatic aberration correction will allow significantly increased space within which such a microlaboratory is confined, at the same time achieving or surpassing spatial resolutions with 100–300 kV instrumentation, which today is achievable only in much more costly state-of-the-art HVEMs which provide less available space (cost of a JEOL ARM-1000 or ARM-1250, for example, is roughly $25M for delivery in North America; 1 cm gap). Such space is critical to incorporating manipulators and detectors associated with the ideal stated above, an ideal which can become a reality.

Outcomes and Strategies

Among the anticipated outcomes of such instrumentation development are

- Standardized, modular instrumentation

- Specialized stages, holders and detectors for each Center
- Incorporation into user programs of the Centers
- Promotion of future commercial availability
- Stimulation of US optical science, important to lithography, microscopy and semiconductor inspection, for example.
- Interface science being raised to the level of sophistication of surface science
- Characterization of defects on anion sublattice of simple and complex ceramics and in multicomponent semiconductors
- Subsurface atomic level spectroscopy.

Achievement of these outcomes involves a long and arduous path which includes

- Preparation and dissemination of the report of this Workshop
- Coalescence of working partnerships
- Work to clarify the complementary roles of electron, neutron and photon scattering techniques in materials research and engineering
- Preparation of a proposal to DOE with other possible cooperating sponsors
- Ensure that the proposal is high on the agenda of the participating laboratories, especially the participating National Laboratories
- In order to enroll the wider community, hold additional workshops and sessions at meetings such as the Symposium on Problem Solving in the Electron Microscope at the 2001 Spring MRS Meeting.

APPENDIX A.

WORKSHOP ON ABERRATION CORRECTION IN ELECTRON MICROSCOPY
Materials Research in an Aberration-Free Environment

TUESDAY (July 18, 2000)

1:00 pm Workshop Overview. Murray Gibson

Theoretical and Practical Approaches to Aberration Correction I

1:45 Correction Schemes for TEM. Max Haider
2:30 Aberration Correction in the STEM. Ondrej Krivanek
3:15 Break
3:45 The Jülich Experience—A Report. Knut Urban
4:15 Objective Pole Piece Gap—How Big Is Too Big for $C_S = 0$? Bernd Kabius
4:30 General Discussion

WEDNESDAY (July 19, 2000)

Theoretical and Practical Approaches to Aberration Correction I I

8:30 am The Oxford Project—A Report. John Hutchison
8:50 The Oak Ridge HTML Project—A Report. Larry Allard
9:10 Development of the SÅTEM Monochromator. Frank Kahl
9:30 Monochromator Development at FEI. Peter Tiemeijer
9:50 Toward an Ideal SEM. Peter Tiemeijer
10:10 Break
10:40 General Discussion
11:30 Group Photograph

Materials Research and Aberration-Free Microscopy I

1:00–3:45 Session 1. HREM Studies. (Session Chair: Uli Dahmen)
Uli Dahmen: Outlook and Future Challenges
Bob Sinclair: Comments on Resolution Requirements
John Hutchison: Environmental Cell for In Situ Studies
Christian Kisielowski: Beyond Lens Aberrations—Desireable Precision, Spatial and Energy Resolutions and Sample Preparation
Eric Stach: In Situ TEM, MEMS and a Variety of Problems
John Spence: Tygrography**?**—Hybrid of Diffraction and Imaging
Nigel Browning: STEM Applications
Yimei Zhu: Electronic and Magnetic Structures
Molly McCartney: Magnetic and Electric Field Imaging
Ken Downing: Very Special Problems for the Life Scientist
Matt Libera: Dose-Limited Resolution and New Sources of Contrast for Imaging Polymers
3:45 Break
4:00–5:30 Session 2A. In Situ Studies. (Session Chairs: Ivan Petrov and Charlie Allen)
Frances Ross: Present and Future Studies at IBM
Marc DeGraef with Yimei Zhu: Non-Interferometric Phase Reconstruction

Ian Robertson: In Situ Studies in the NTEAM—CMM at UIUC
Bob Birtcher: In Situ Studies in the NTEAM—EMC at ANL
Mark Kirk: Displacement Damage Studies with Ion Irradiation
Robert Hull: In Situ Experimentation in Semiconductor Materials and Devices

THURSDAY (July 20, 2000)

Materials Research and Aberration-Free Microscopy I I

8:30–9:45 Session 2B In Situ Studies. (Session Chair: Charlie Allen)
Bob Sinclair: In Situ Electron Microscopy—Guidelines, Prospectives and Opportunities
Eric Van Cappellan: The FEI HRSEM and FIB
Vinayak Dravid: Various In Situ Experiments and Measurements
Laurie Marks: 3D Reconstruction of Diffraction Data
Al Meldrum: In Situ Studies of Geologic Materials and the Characterization of Nanocrystalline Materials
Markus Lentzen: Focal Series Reconstruction and Need for C_s Correction

9:45–12:40 Session 3. Analytical Studies. (Session Chair: Ian Anderson)
Ondrej Krivanek: Requirements for Nanoanalysis
Max Haider: Chromatic and Spherical Aberration Corrected SEM
Jim Bentley: Synchrotron Spectral Resolution at Atomic Spatial Resolution
Jim Howard: X-ray Optics and Microcalorimetry: EDS with WDS Resolution
12:00N Closing Comments: Murray Gibson
Ian Anderson: Spectrum Imaging and Multivariant Techniques
Jian Min Zuo: Electron Diffraction Analysis
Nestor Zaluzec: Detectors, Stages and Cold Field Emission

APPENDIX B. WORKSHOP PARTICIPANTS

Summer Workshop on Aberration Correction in Electron Microscopy
July 18-20, 2000
Argonne National Laboratory
Argonne, IL

Lawrence Allard
High Temperature Materials Laboratory
Building 4515 MS 6064
Oak Ridge National Laboratory
1 Bethel Valley Road
P.O. Box 2008
Oak Ridge, TN 37831-6064
Ph: 865-574-4981
Fax: 865-576-5413
E:mail: allardlfjr@ornl.gov

Howard Birnbaum
Materials Research Laboratory
Univ. of Illinois - Urbana
1304 W. Green St.
Urbana, IL 61801
Ph: 217-333-2940
Fax 217-244-2278
E:mail: birnbaum@uimrl7.mrl.uiuc.edu

Charles Allen
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-4157
Fax: 630-252-4289
E:mail: allen@aaem.amc.anl.gov

Robert Birtcher
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-4996
Fax: 630-252-4289
E:mail: birtcher@anl.gov

Ian Anderson
Metals and Ceramics Division
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831
Ph: 865 574 0632
Fax: 865 574 0641
E:mail: andersonim@ornl.gov

Nigel Browning
Department of Physics (M/C273)
University of Illinois at Chicago
845 W. Taylor Street
Chicago, IL 60607-7059
Ph: 312-413-8164
Fax: 312-996-9016
E:mail: browning@uic.edu

James Bentley
Metals and Ceramics Division
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831
Ph: 865-574-5067
Fax: 865-574-0641
E:mail: bentleyj@ornl.gov

Xidong Chen
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-6968
Fax: 630-252-4798
E:mail: xchen@anl.gov

Russell Cook
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-7194
Fax: 630-252-4289
E:mail: cook@horus.et.anl.gov

Roseann Csencsits
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-4977
Fax: 630-252-4289
E:mail: csencsits@anl.gov

Brian Cunningham
Nissei Sangyo America
755 Ravendale Drive
Mountain View, CA 94043
Ph: 650-210-3442
Fax: 650-213-9381
E:mail:
dr.brian.cunningham@nissei.com

Ulrich Dahmen
National Center for Electron Microscopy
Building 72; 1 Cyclotron Rd.
Lawrence Berkeley National Laboratory
Berkeley, CA 94720
Ph: 510-486-4627
Fax: 510-486-5888
E:mail: udahmen@lbl.gov

Marc DeGraef
Department of Mater. Sci. and
Engineering
Carnegie Mellon University, Wean Hall
4307
5000 Forbes Ave.
Pittsburgh, PA 15213-3890
Ph: 412-268-8527
Fax: 412-268-3113
E:mail: mdg@xenon.mems.cmu.edu

Steven Donnelly
Joule Physics Laboratory
University of Salford
Manchester M5 4WT UK
Ph: +44 (0) 161 295 5392
Fax: +44 (0) 161 295 5119
E:mail: s.e.donnelly@physics.salford.ac.uk

Kenneth Downing
Lawrence Berkeley National Lab
326 Donner Lab
Berkeley, CA 94720
Ph: 510-486-5941
Fax: 510-486-5342
E:mail: khdowning@lbl.gov

Vinayak Dravid
Northwestern University
Materials Science & Engineering
2225 N. Campus Drive, 3013A MLSF
Evanston, IL 60208
Ph: 847-467-1363
Fax: 847-491-7820
E:mail: v-dravid@northwestern.edu

Alwyn Eades
Department of Materials Science and Engineering
Lehigh University
5 East Packer Avenue
Bethlehem, Pennsylvania 18015-3195
Ph: 610-758-4231
Fax: 610-758-4244
E:mail: jae5@lehigh.edu

Peter Genovese
JEOL USA
11 Dearborn Rd.
Peabody, MA 01960
Ph: 508-535-5900
Fax: 508-536-2205
E:mail:

Murray Gibson
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-4987
Fax: 630-252-4789
E:mail: gibson@anl.gov

Robert Gottschall
Department of Energy
19901 Germantown Rd.
Germantown, MD 20874-1290
Ph: 301-903-3428
Fax: 301-903-9513
E:mail:
robert.gottschall@science.doe.gov

Maximilian Haider
CEOS GmbH.
Englerstr. 28, D-69126 Heidelberg
Germany
Ph: -49 (0)6221 451 450
Fax: -49 (0)6221 451 449
E:mail: haider@ceos-gmbh.de

Jon Hiller
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-9558
Fax: 630-252-4289
E:mail: hiller@anl.gov

James Howard
Noran Instruments, Inc.
2551 W Beltline Hwy.
Middleton, WI 53562
Ph: 608-831-6511
Fax: 608-831-2313
E:mail:

Robert Hull
Department of Materials Science
Thornton Hall, University of Virginia
Charlottesville, VA 22903
Ph: 804-982-5658
Fax: 804-982-5660
E:mail: rh5C@virginia.edu

John Hutchison
Department of Material
University of Oxford
Parks Road, OX1 3PH Oxford, UK
Ph: +44 (0)1865-273705
Fax: +44 (0)1865-283333
E:mail:
john.hutchison@materials.oxford.ac.uk

Shigeto Isakozawa
Hitachi Ltd. Instruments
882 Ichige; Hitachinaka-city
Ibaraki Japan
Ph: 81-29-276-6147
Fax: 81-29-274-6771
E:mail: isakozawa@cm.naka.hitachi.co.jp

Bernd Kabius
LEO Electron Microscopy
Carl-Zeiss-Strasse 56
73446 Oberkochen
Germany
Ph: +49-(0)7364-943434
Fax: +49-(0)7364-944670
E:mail: kabius@leo.de

Frank Kahl
Oak Ridge Institute of Science and Education
(ORISE)
P.O. Box 117
Oak Ridge, TN 37831
Ph: 865-241-6617
Fax: 865-574-4913
E:mail: kahlf@ornl.gov

Michael Kersker
JEOL USA
11 Dearborn Road
Peabody, MA 01960
Ph: 508-535-5900
Fax: 508-536-2205
E:mail: kersker@jeol.com

Ondrej Krivanek
Nion Co.
1102 8th St.
Kirkland, WA 98033
Ph: 425-576-9060
Fax: 425-739-0312
E:mail: krivanek@compuserve.com

Young-Woon Kim
Materials Research Laboratory
University of
Illinois—Urbana/Champaign
1304 W. Green St.
Urbana, IL 61801
Ph: 217-244-0582
Fax: 217-244-2278
E:mail: yw_kim@mrl.uiuc.edu

Markus Lentzen
Institute for Solid State Physics
Forschungszentrum Juelich GmbH
D-52425 Juelich, Germany
Ph: +49 2461 61 6644
Fax: +49 2461 61 6444
E:mail: m.lentzen@fz-juelich.de

Marquis Kirk
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-4998
Fax: 630-252-4289
E:mail: kirk@anl.gov

Matthew Libera
Stevens Institute of Technology
Dept. of Chemical, Biochemical,
and Materials Engineering
Hoboken, New Jersey 07030
Ph: 201-216-5259
Fax: 201-216-8306
E:mail: mlibera@stevens-tech.edu

Christian Kisielowski
National Center for Electron Microscopy
Building 72; 1 Cyclotron Rd.
Lawrence Berkeley National Laboratory
Berkeley, CA 94720
Ph: 510-486-4716
Fax: 510-486-5888
E:mail: cfkisielowski@lbl.gov

Laurence Marks
Dept. of Mater. Sci. & Engr.
Northwestern University
2225 N. Campus Dr. Rm 2036 MLSF
Evanston, IL
Ph: 847-491-3996
Fax: 847-491-7820
E:mail: ldm@apollo.numis.nwu.edu

Masahiro Kawasaki
JEOL
11 Dearborn Road
Peabody, MA 01960
Ph: 978-536-2230
Fax: 978-536-2264
E:mail: kawasaki@jeol.com

Molly McCartney
Arizona State University
Center for Solid State Science
Tempe, AZ 85287-1704
Ph: 480-965-4540
Fax: 480-965-9004
E:mail: mccartney@csss.la.asu.edu

Alkiviathes Meldrum
University of Alberta
Dept. of Physics
Edmonton, AB T6G 2J1
Canada
Ph: 780-492-5342
Fax780-492-0714
E:mail: ameldrum@ualberta.ca

Robb Mierzwa
JEOL USA
3906 Lisa Ave
Sheboygan WI
Ph: 920-803-8945
Fax: 920-803-8946
E:mail: mierzwa@jeol.com

Dean Miller
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-4108
Fax: 630-252-4289
E:mail: miller@anl.gov

Mikio Naruse
JEOL Ltd.
1-2 Musashiro 3-chome
Akishima, Tokyo 196-8558
Japan
Ph: 81-42-542-2227
Fax: 81-42-546-8063
E:mail: naruse@jeol.co.jp

Ivan Petrov
Materials Research Laboratory
University of
Illinois—Urbana/Champaign
1304 W. Green St.
Urbana, IL 61801
Ph: 217-333-8396
Fax: 217-244-2278
E:mail: petrov@uiuc.edu

Ian Robertson
Materials Research Laboratory
University of Illinois—Urbana/Champaign
1304 W. Green St.
Urbana, IL 61801
Ph: 217-333-6776
Fax: 217-333-2736
E:mail: ianr@ uiuc.edu

Frances Ross
IBM Research Division
TJ Watson Research Center
PO Box 218
Yorktown Heights, NY 10598
Ph: 914-945-1022
Fax: 914-945-2141
E:mail: fmross@us.ibm.com

Herbert Ruoff
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-7783
Fax: 630-252-4289
E:mail: ruoff@anl.gov

Robert Sinclair
Stanford University
Mater. Sci. & Engr.
Stanford, CA 94305-2205
Ph: 650-723-1102
Fax: 650-725-4034
E:mail: bobsinc@leland.stanford.edu

John Spence
Physics Department
Arizona State University
Tempe, AZ 85287
Ph: 480-965 6486
Fax: 480-965-7954
E:mail: spence@asu.edu

Eric Stach
National Center for Electron
Microscopy
Building 72
Lawrence Berkeley National Laboratory
Berkeley, CA 94720
Ph: 510-486-4634
Fax: 510-486-5888
E:mail: EAStach@lbl.gov

Ray Twesten
Materials Research Laboratory
University of Illinois—Urbana/Champaign
104 S. Goodwin Ave
Urbana, IL 61801
Ph: 217-244-6177
Fax: 217-244-2278
E:mail: twesten@uiuc.edu

Stephen Streiffer
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-5832
Fax: 630-252-4289
E:mail: streiffer@anl.gov

Knut Urban
Institute for Solid State Physics
Forschungszentrum Juelich GmbH
D-52425 Juelich, Germany
Ph: +49 (0)2461-61-3153
Fax: +49 (0)2461-61-6444
E:mail: k.urban@fz-juelich.de

Roger Teppert
Hitachi
Nissei Sangyo America, Ltd.
2850 Golf Road, Suite 200
Rolling Meadows, IL 60008
Ph: 847-545-2225
Fax: 847-981-1792
E:mail: roger.teppert@nissei.com

Eric Van Cappellen
FEI Company
7451 NW Evergreen Park
Hillsboro, OR 97124
Ph: 503-726-2708
Fax: 503-640-7663
E:mail: evc@feico.com

Peter Tiemeijer
FEI Electron Optics - Building AAEp
PO Box 218
5600 MD Eindhoven
The Netherlands
Ph: +31-40-2766005
Fax: +31-40-2766820
E:mail: peter.tiemeijer@nl.feico.com

Michael van der Stam
FEI Company
Building AAE, Achtseweg Noord 5
5651 GG Eindhoven
The Netherlands
Ph: +31 40 2765103
Fax: +31 40 2766820
E:mail: mvs@nl.feico.com

John Treadgold
LEO Electron Microscopy, Inc.
2626 Embers Lane
Arlington Heights, IL 60005
Ph: 847-290-9566
Fax: 847-290-0045
E:mail: 102624.2026@compuserve.com

Mark Van der Zande
Philips Research Laboratories
Building WY4-002 Mailbox WY-41
Prof. Holstlaan 4
5656 AA Eindhoven
The Netherlands
Ph: +31 40 2744816
Fax: +31 40 2742293
E:mail: mark.van.der.zande@philips.com

Kai Xiu
212 Loomis Lab of Physics
Department of Physics
University of Illinois
1110 W. Green Street
Urbana, IL 61801
Ph: 217-244-3265
Fax: 217-333-9819
E:mail: kaixiu@uiuc.edu

Yimei Zhu
Dept. of Applied Science
Brookhaven National Laboratory
Bldg. 480
Upton, NY 11973-5000
Ph: 631-344-3057
Fax: 631-344-4071
E:mail: zhu@bnl.gov

Nestor Zaluzec
Materials Science Division
Argonne National Laboratory
9700 S. Cass Ave.
Argonne, IL 60439
Ph: 630-252-7901
Fax: 630-252-4289
E:mail: zaluzec@aaem.amc.anl.gov

Jian-Min Zuo
Dept. of Mater. Sci. & Engr.
University of Illinois—Urbana/Champaign
1304 W. Green St.
Urbana, IL 61801
Ph: 217-244-6504
Fax: 217-333-2736
E:mail: jianzuo@uiuc.edu