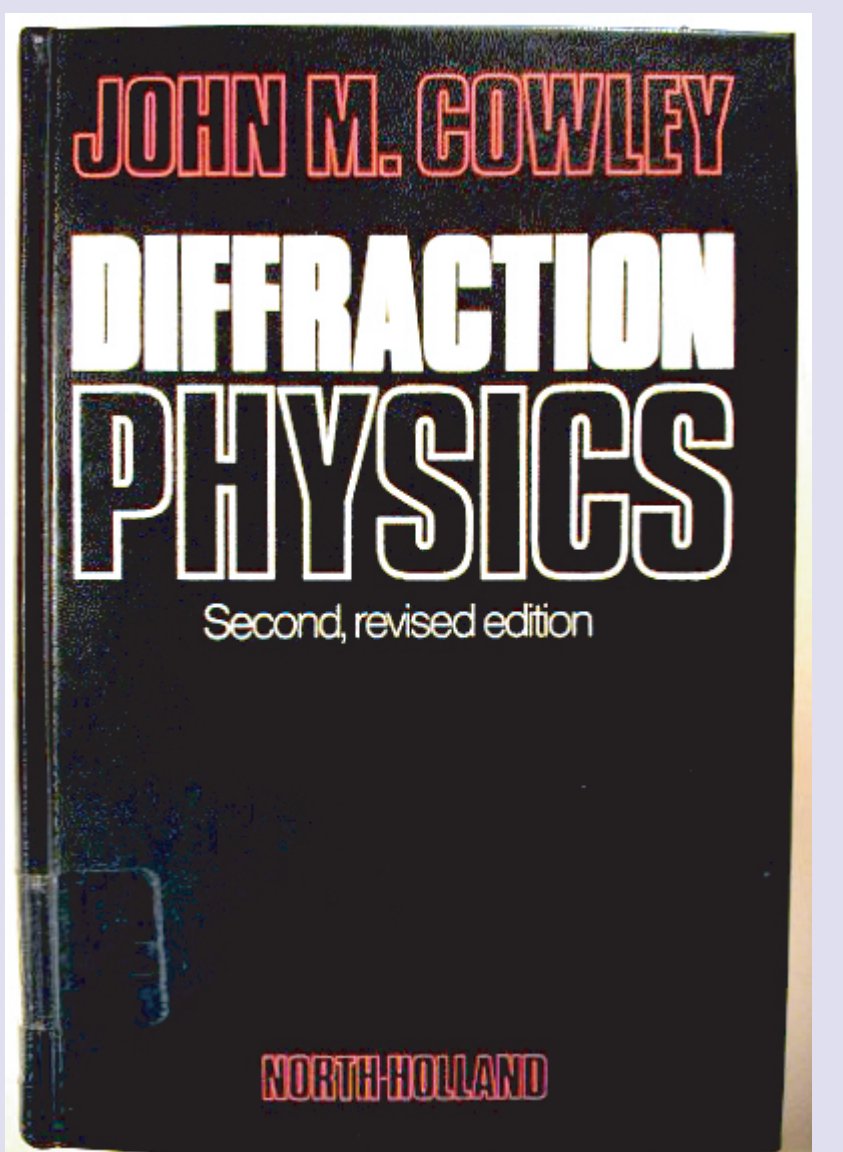




# John M. Cowley

1923-2004



## The early work in Australia

### ELECTRON DIFFRACTION AND RECTIFICATION FROM SILICON AND PYRITE SURFACES

By J. M. COWLEY AND J. L. SYMONDS.  
Received 3rd October, 1946.  
Summary.  
Electron diffraction and rectification investigations have been conducted on surfaces of pyrite and silicon for different surface conditions. There is evidence that, for the best rectification, the crystal lattice should be almost perfect and free from fracture or mosaic structure. A general picture of the pyrite layer on pyrite is built up from electron diffraction evidence.

Physics Department, University of Adelaide, South Australia.  
Trans. Faraday Soc., London 44:53-60 (1948).

### ELECTRON DIFFRACTION BY FATTY ACID LAYERS ON METAL SURFACES.

By J. M. COWLEY.\*  
Received 3rd October, 1946.  
Summary.  
The structure and effect of heating of layers of palmitic acid on various metals have been investigated. The temperature at which the orientation of the molecules is lost is below the bulk melting point for crystalline layers, and is 103°C for monolayers, being independent of the metal used as base. A new orientable layer formed from fatty acids on metals is described, corresponding to an orthorhombic structure and giving a characteristic pattern. Such layers are not soluble in the usual solvents, do not lose their orientation until heated to over 400°C, and remain on the surface in crystalline form to about 500°C. It is suggested that these properties arise from a change taking place under the influence of the electron beam.

Physics Department, University of Adelaide, South Australia. Trans. Faraday Soc., London 44:60-68 (1948).

John Maxwell Cowley was born February 18, 1923 in Adelaide, South Australia. He received his B.S. in 1942, and M.Sc. in 1945 at the University of Adelaide. Electron diffraction, especially diffraction from disordered or imperfect crystals, was central to Prof. Cowley's scientific interests, and it seems fitting that he started his career at the University of Adelaide where Lawrence Bragg had worked. However, Bragg had left in 1910, and at the time Cowley started his Masters work, under Prof. Roy Burdon, there was little work being done there. One of the very few instruments available was a previously unused Finch-type electron diffraction camera, and Cowley's Masters work with that instrument resulted in the two publications at the left.

### NATURE No. 4016 October 19, 1946

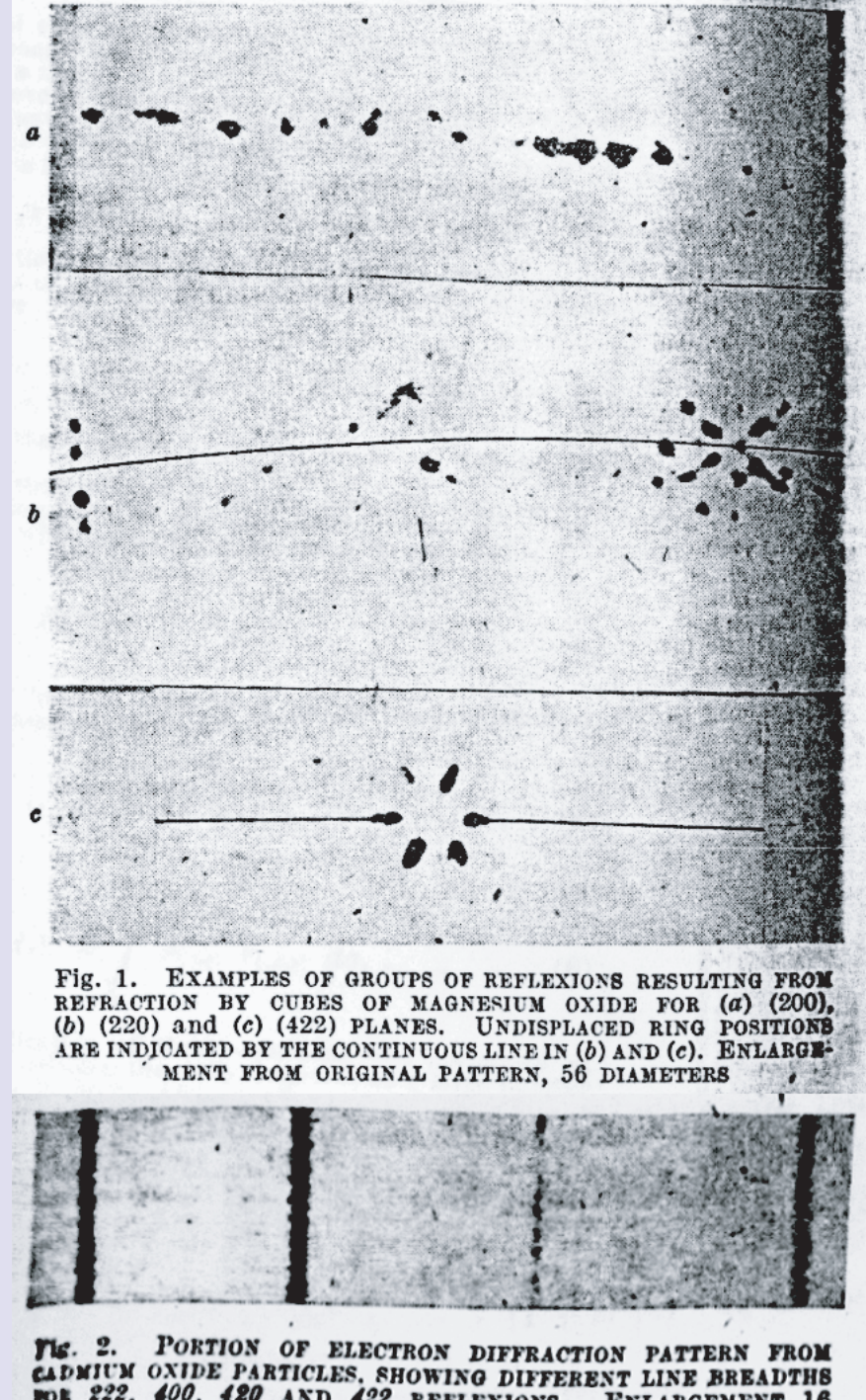


FIG. 1. EXAMPLES OF GROUPS OF REFLECTIONS RESULTING FROM REFRACTION BY GROUPS OF ALUMINIUM OXIDE FOR (a) 60° AND (b) 120° INCIDENCE. (c) 120° BEAM. (d) 120° BEAM. (e) 120° BEAM. (f) 120° BEAM. (g) 120° BEAM. (h) 120° BEAM. (i) 120° BEAM. (j) 120° BEAM. (k) 120° BEAM. (l) 120° BEAM. (m) 120° BEAM. (n) 120° BEAM. (o) 120° BEAM. (p) 120° BEAM. (q) 120° BEAM. (r) 120° BEAM. (s) 120° BEAM. (t) 120° BEAM. (u) 120° BEAM. (v) 120° BEAM. (w) 120° BEAM. (x) 120° BEAM. (y) 120° BEAM. (z) 120° BEAM.

### Structure Analysis of Single Crystals by Electron Diffraction. II. Disordered Boric Acid Structure.

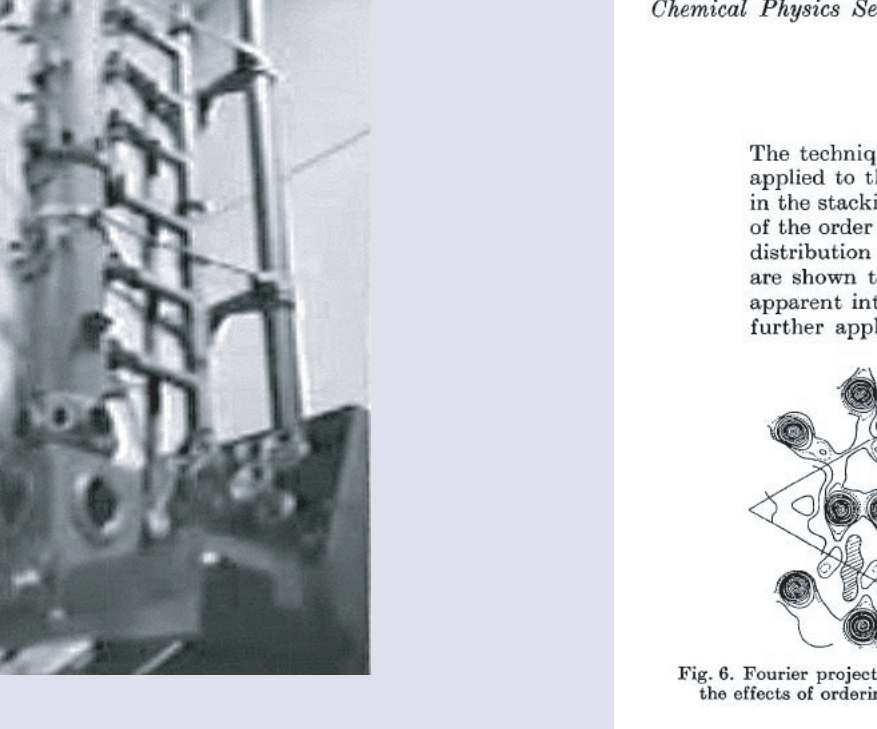
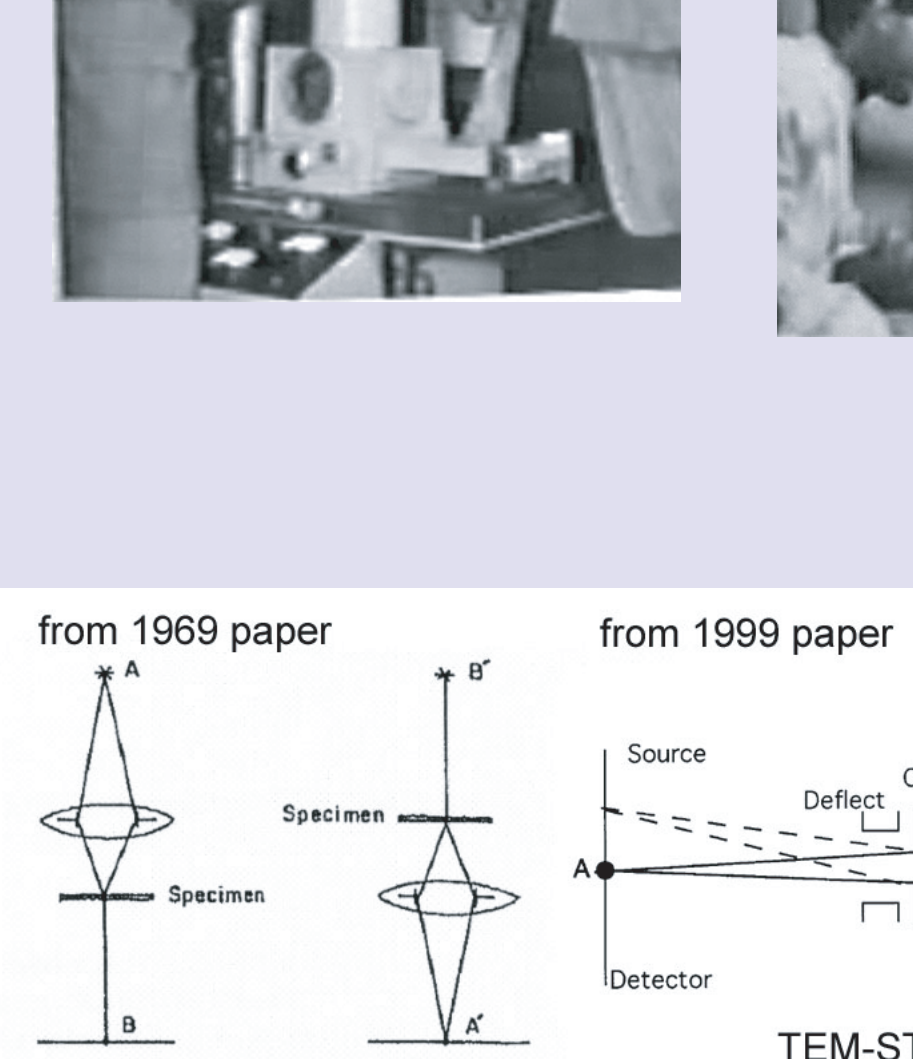


FIG. 4. Fourier synthesis of a boric acid layer, removed for the effect of ordering by the distribution function method.



After returning to Melbourne, Cowley and Alex Moodie completed their first ED camera, which was designed by Lloyd Rees. Cowley talks about his important long-time collaboration with Alex Moodie in a video interview conducted by Sterling Newberry at the 1991 EMSA meeting in San Jose, and he shows a pictures of the first and second ED cameras built at Melbourne. In the first picture, taken in 1948, Moodie is on the left and Cowley on the right. In the preface to the second edition of his classic book Diffraction Physics, Cowley credits Moodie as his "mentor in matters of diffraction theory". The second camera, with six lenses, was used by Peter Goodman to do the first work in symmetry determination from convergent-beam ED.



The new instruments could be used to record ED patterns from small areas of thin crystals, and in 1953, Cowley published the first crystal structure solution derived from single crystal electron diffraction data. This work was well ahead of its time; the technique did not become well established for decades.

From 1963 to 1969 Cowley was Professor of Physics at the University of Melbourne. At the University, he did his first work on electron microscopy, using a JEM-7, with students Peter Turner and Andrew Pogany, while continuing to work with the CSIRO group including Alex Moodie, Peter Goodman, Andrew Johnson, and David Cockayne. At the suggestion of David Wadsley, who was having difficulty sorting out the complexities of niobium oxides by x-ray diffraction, initial attempts were made at determination of the atomic structure of crystals by TEM. This was done using a Philips 200, and some suggestions of characteristic

### X-Ray Measurement of Order in Single Crystals of Cu<sub>3</sub>Au<sup>8,9</sup>

J. M. COWLEY<sup>1</sup>  
Massachusetts Institute of Technology, Cambridge, Massachusetts  
(Received August 4, 1949)  
X-ray diffraction methods have been used to measure long- and short-range order parameters for single crystals of the alloy Cu<sub>3</sub>Au held at elevated temperatures. It is shown that short-range order parameters may be obtained by a three-dimensional Fourier analysis of the "scattering power" for the diffuse background scattering of x-rays, expressed as a function of reciprocal lattice coordinates. Appropriate experimental and computational procedures are outlined. Intensity measurements have been made with a special arrangement of a bent-crystal monochromator and a Geiger-counter spectrometer. Short-range parameters are given for the first ten shells of atoms surrounding a given atom for three temperatures above the critical temperature of Cu<sub>3</sub>Au. These results, and those for the long-range order parameter agree well with theoretical predictions.

Between 1947 and 1949 Cowley completed his PhD work at MIT under the renowned Bertram E. Warren, working in x-ray diffraction, as reflected in the above paper in *J. Appl. Phys.* (1950) 21:24-30.

### The Scattering of Electrons by Atoms and Crystals. I. A New Theoretical Approach

By J. M. COWLEY AND A. P. MOORE  
Chemical Physics Section, Division of Industrial Chemistry, C.I.B.I.O., Melbourne, Australia  
The scattering of electrons by atoms and crystals is treated in a new theoretical approach. The scattering of electrons by atoms is treated in terms of the scattering of electrons by a potential well. The scattering of electrons by crystals is treated in terms of the scattering of electrons by a periodic array of potential wells. The scattering of electrons by atoms and crystals is treated in terms of the scattering of electrons by a periodic array of potential wells.

### The Scattering of Electrons by Atoms and Crystals. II. The Effects of Finite Source Size

By J. M. COWLEY AND A. P. MOORE  
The scattering of electrons by atoms and crystals is treated in a new theoretical approach. The scattering of electrons by atoms is treated in terms of the scattering of electrons by a potential well. The scattering of electrons by crystals is treated in terms of the scattering of electrons by a periodic array of potential wells. The scattering of electrons by atoms and crystals is treated in terms of the scattering of electrons by a periodic array of potential wells.

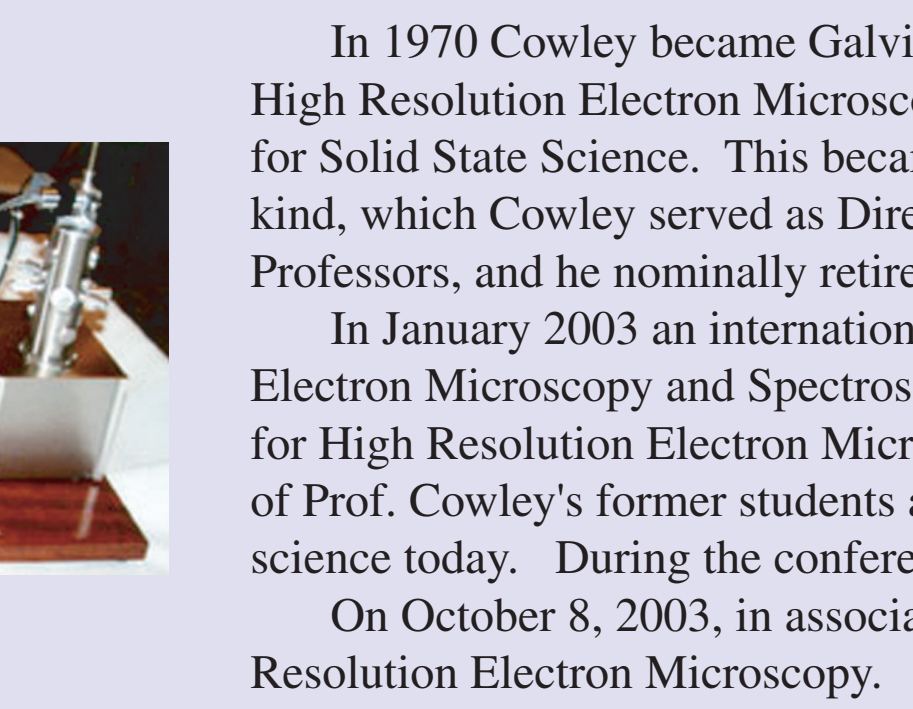
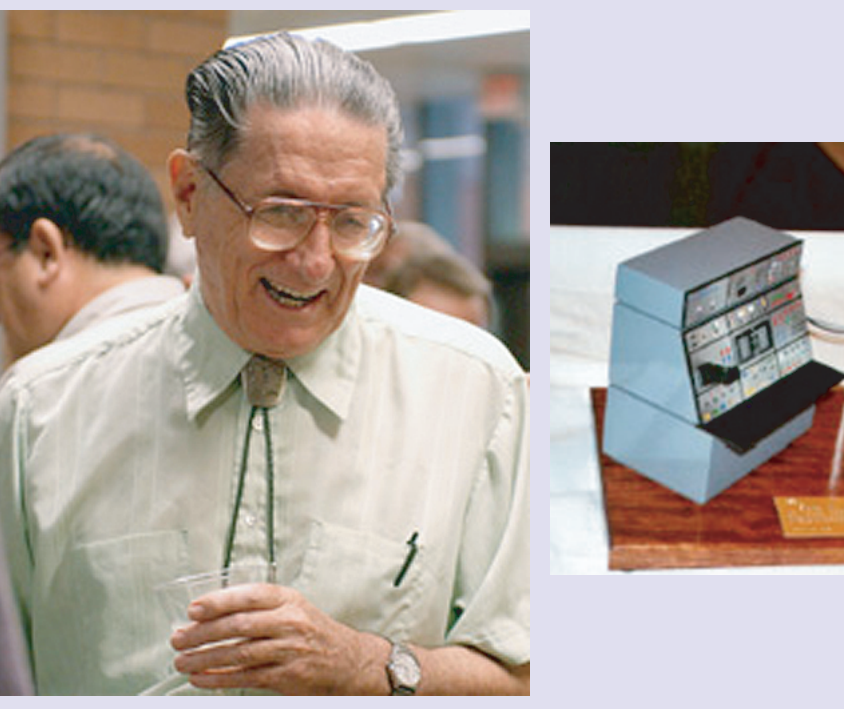
### The Electron-Optical Imaging of Crystal Lattices

By J. M. COWLEY  
The need to account for the observations on thin crystals led to the development of the many-beam theory with Alex Moodie. In 1957, Cowley was awarded a D.S. from the University of Adelaide, and Cowley and Moodie published a well-known series of papers on diffraction theory, starting with the famous multiple method (over 750 cites to date), which provides the basis for much of the diffraction and imaging simulation software in use today. During this time Cowley was Chief Research Officer and Head of the Crystallography Section, CSIRO Division of Chemical Physics, Melbourne. From the abstract to the 1957 paper, they write "Particular cases considered are the wave function at the exit surface of a crystal, corresponding to the image produced by an ideal electron microscope, and the diffraction pattern, or angular scattering function, of a crystal. The present theory is particularly suited to the study of the diffraction of electrons by very thin crystals and crystals containing imperfections. Its applications to matters of practical importance in this field will be considered in a future publication."

atom grouping could be seen, from the work of John Sanders and John Allpress. 1969 also saw the consideration of high-resolution STEM imaging, recently developed by Crewe, for atomic imaging, and the concept of reciprocity between TEM and STEM imaging (after far) was described. In 1969, Cowley attended his first EMSA meeting, at St. Paul, MN, and he was soon to be a permanent resident in the US.

Cowley, J.M. (1969) Image contrast in the transmission scanning electron microscope. *Appl. Phys. Lett.* 15:58-59.  
Cowley, J.M. (1999) Electron Nanodiffraction. *Microsc. Res. Tech.* 46:75-97.

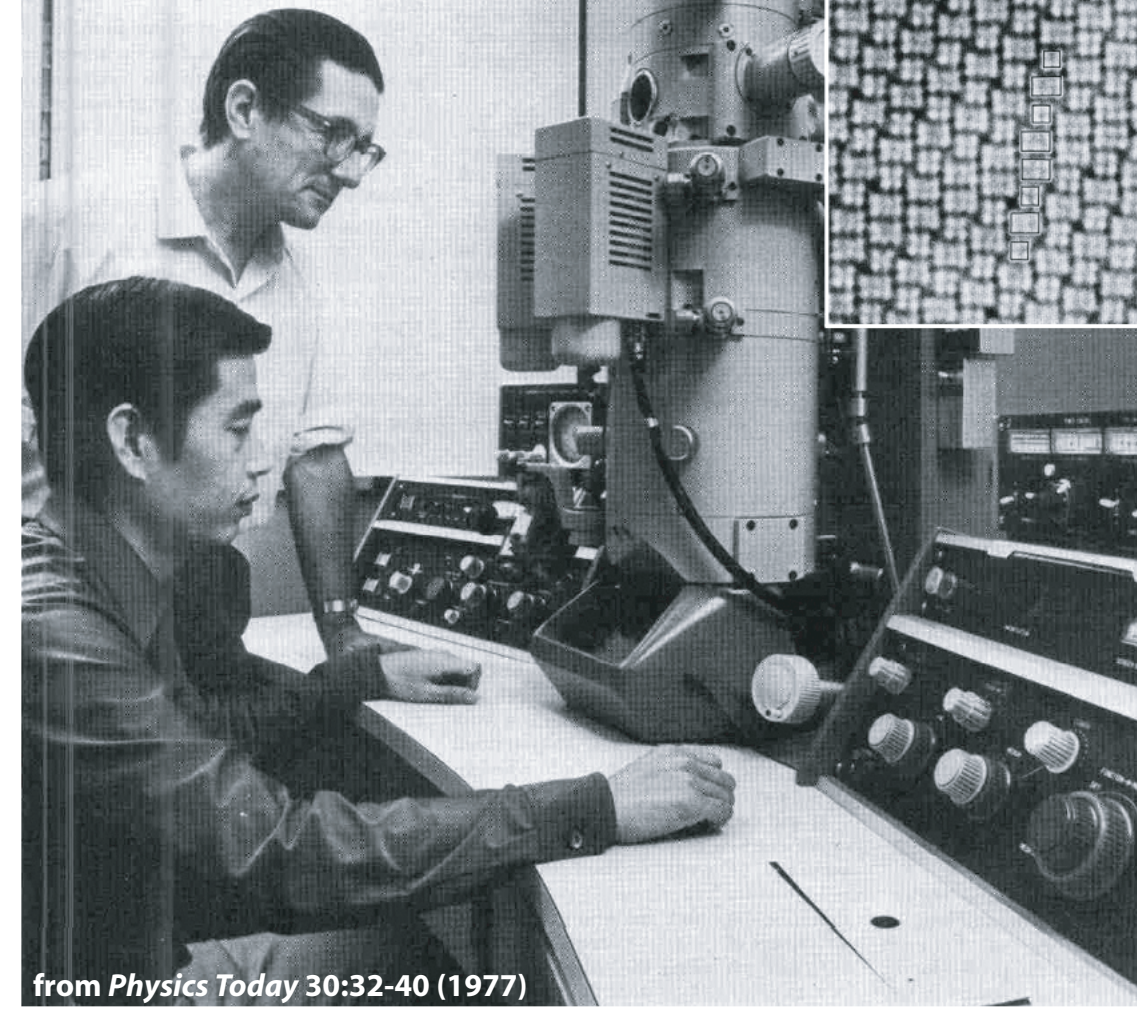
## The Center for High Resolution Electron Microscopy at Arizona State University



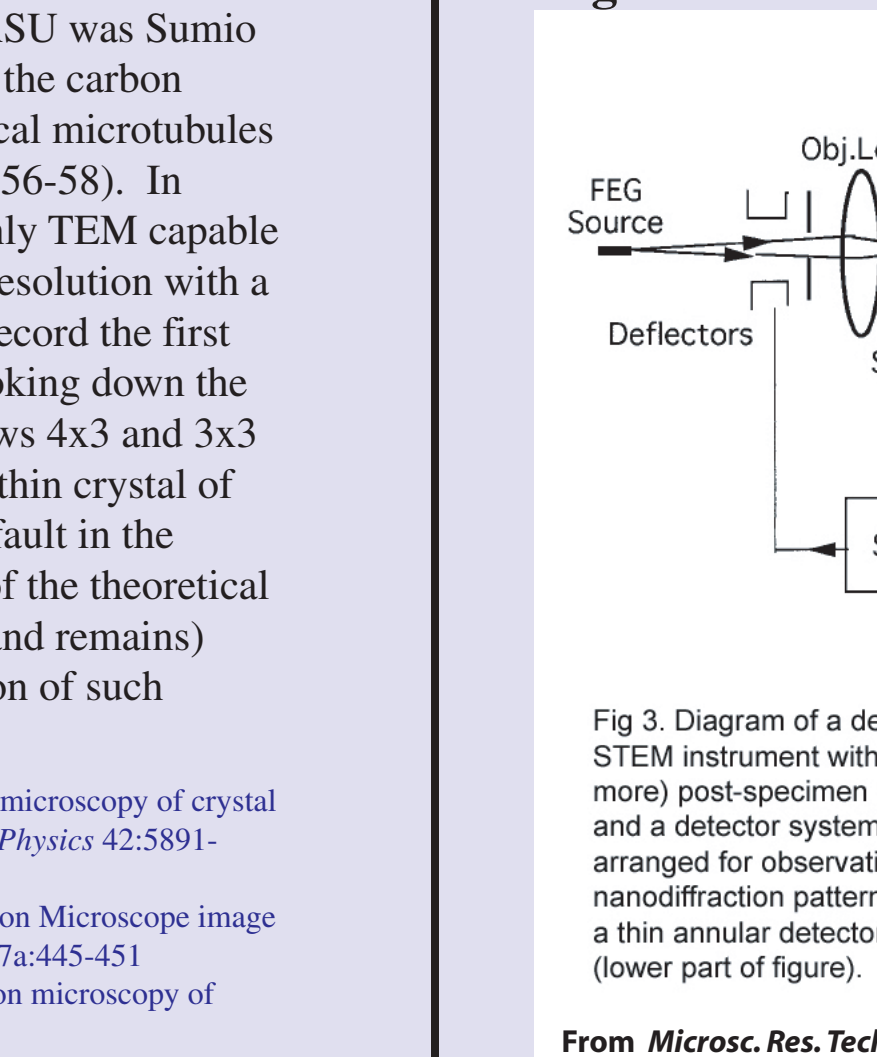
In 1970 Cowley became Galvin Professor of Physics at Arizona State University. In 1978 he established the Center for High Resolution Electron Microscopy as a National Science Foundation regional instrumentation facility, within the Center for Solid State Science. This became the NSF National Facility for High Resolution Electron Microscopy, the best of its kind, which Cowley served as Director from 1983 to 1990. In 1988, Cowley became one of the ASU's first Regents' Professors, and he nominally retired in 1994, although he continued his very productive research at ASU. In January 2003 an international workshop entitled "Recent Developments and Applications of Atomic Resolution Electron Microscopy and Spectroscopy-A Silver Jubilee" was held at ASU, celebrating the 25th anniversary of the Center for High Resolution Electron Microscopy, and honoring Cowley on his 80th birthday. The meeting was attended by many of Prof. Cowley's former students and associates, which represent many of the most prominent individuals in materials science today. During the conference dinner, Cowley was presented with a scale model of his VG HB-5 STEM. On October 8, 2003, in association with his 80th birthday, the facility was named The John M. Cowley Center for High Resolution Electron Microscopy.

Cowley edited or served on the editorial boards of several journals, and occupied official positions in various professional societies. He received the highest awards of the American Crystallographic Society (the Bertram Eugene Warren Award in 1976 with Sumio Iijima), the Electron Microscopy Society of America (Distinguished Scientist Award, 1979), and the International Union of Crystallography (Ewald Prize in 1987 with A.F. Moodie). He was a fellow of the Australian Academy of Science (1961), the Royal Society of London (1979), and the American Physical Society (1984). Cowley is well-known for his book *Diffraction Physics*, the standard reference in the field, with over 950 cites to date (first edition in 1975, second in 1981, and third edition in 1995 by Elsevier). In the first edition preface, Cowley writes that the work "Reflects my own interest particular interests in electron diffraction and diffraction from disordered or imperfect crystals and employs an approach which is particularly suited to the treatment of these topics." Cowley also contributed to and edited *Electron Diffraction Techniques* (Vols. 1-4), Oxford Univ. Press 1992, and *High-Resolution Transmission Electron Microscopy and Associated Techniques*, (Buseck, Cowley, and Eyring, Oxford 1988). At the time of his death, he was working on five papers, had outlines for another book and two book chapters, and was co-PI on two grant proposals.

### High-resolution TEM of crystals, image formation theory



Cowley's first post-doc at ASU was Sumio Iijima, who went on to discover the carbon nanotube (Iijima, S., 1991, Helical microtubules of graphitic carbon. *Nature* 354:56-58). In 1971, using a JEM-100B, the only TEM capable at the time of achieving 0.4nm resolution with a goniometer, Iijima was able to record the first images of atoms in a crystal, looking down the zone axis. The inset at left shows 4x3 and 3x3 octahedra of oxygen atoms in a thin crystal of niobium oxide, with a stacking fault in the center. Cowley's development of the theoretical bases of imaging crystals was (and remains) essential for correct interpretation of such images.

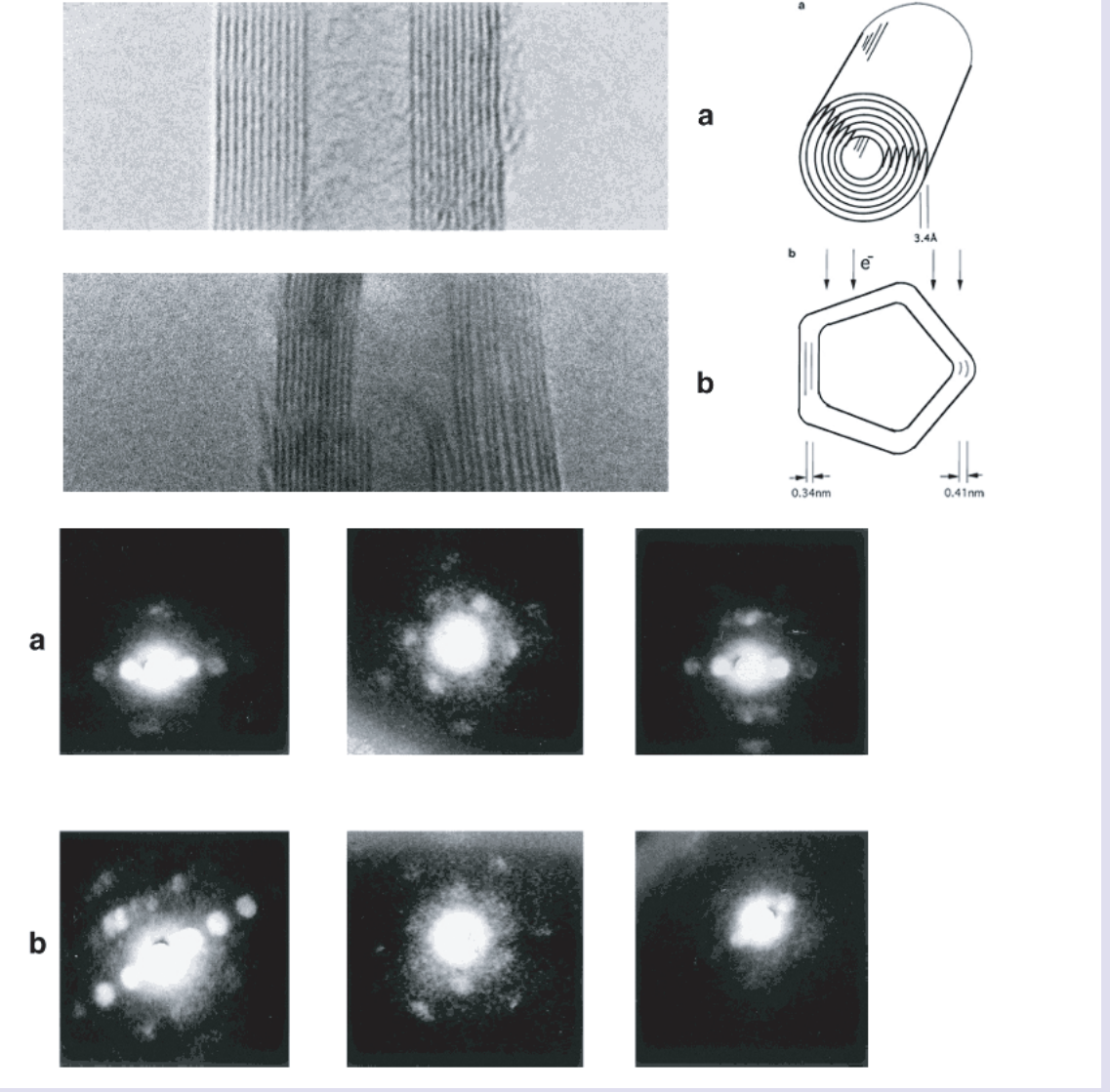


Iijima, S. (1971) High-resolution electron microscopy of crystal lattice of titanium-niobium oxide. *J. Appl. Phys.* 42:5891-5893.  
Cowley, J.M. and Iijima, S. (1972) Electron Microscope image contrast for thin crystals. *Z. Naturforsch.* 27a:445-451  
Cowley, J.M. and Iijima, S. (1977) Electron microscopy of atoms in crystals. *Physics Today* 30:32-40 (1977)

In 1978, ASU acquired a VG HB-5 cold field-emission gun STEM. This was to remain Cowley's instrument of choice for 25 years. The versatility of the instrument was ideally suited to Cowley's inventiveness, and the HB-5 was much-adapted over the years. Special-purpose detectors were designed, based on detailed theory, to optimize imaging in various modes (bright-field, dark-field, thin and thick specimens, crystalline and amorphous specimens, coherent and incoherent imaging). The theoretical work enabled calculation of simulated images, which ensures correct interpretation of the recorded images. These included darkfield imaging with the margin of a high-angle annular detector, and use of a thin annular detector that records the signal at the intersection of convergent-beam diffraction spots. The latter improves the resolution of bright-field images by a factor of 1.7 (Cowley et al., 1995). The HB-5 was fitted with additional detectors for imaging and microanalysis of many kinds, as seen at the left. Until his death, Cowley used the HB-5 weekly.



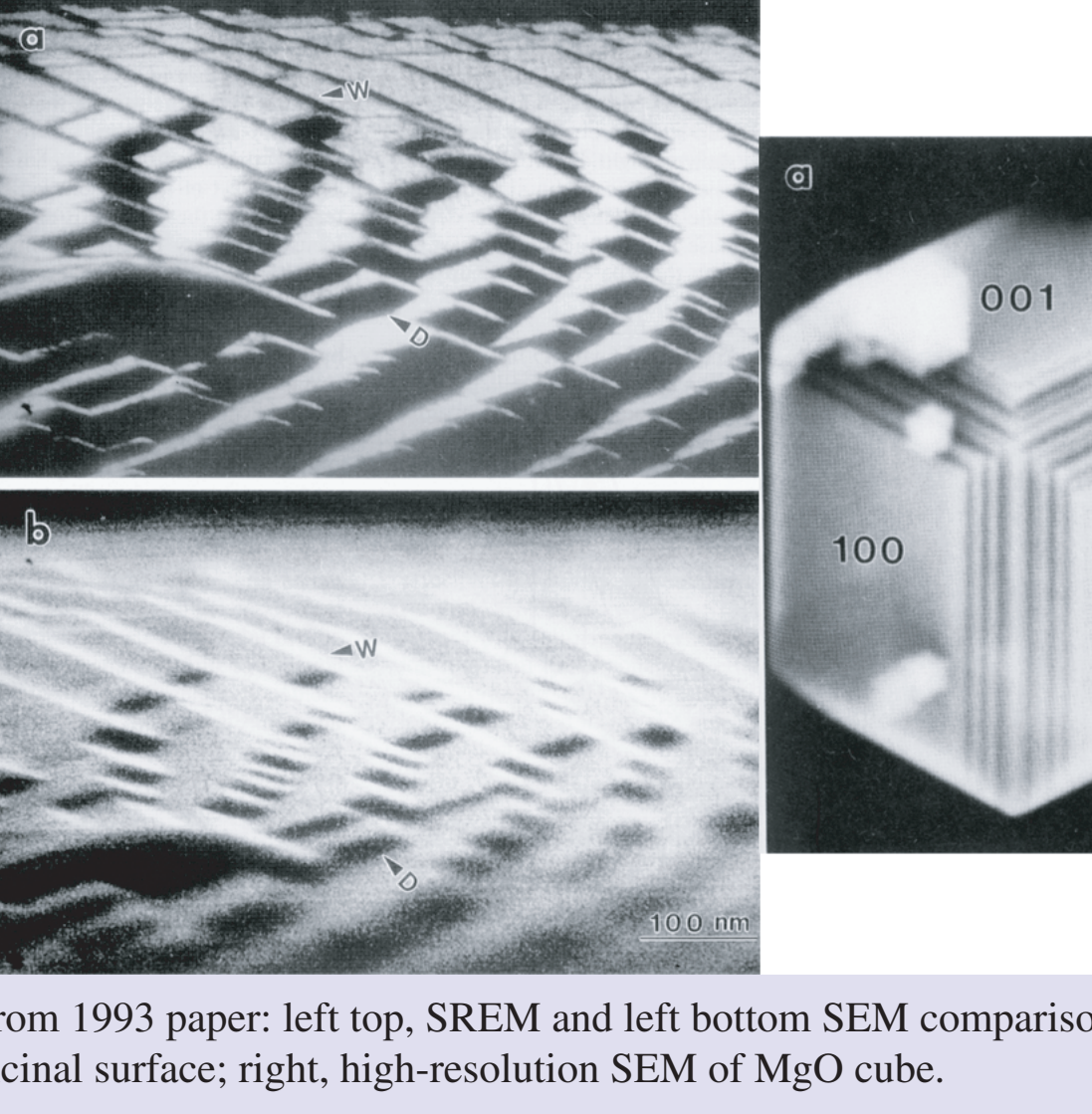
### Nanodiffraction for determination of local structure at high resolution



Coherent nanodiffraction offers structural information to about 0.03 nm, from very small regions of the specimen. When the probe is smaller than the unit cell of a crystal, and a large objective aperture is used in STEM mode, the disks of the convergent beam diffraction pattern overlap. The intensity distribution in the diffraction pattern then depends on the relative positions of the atoms, and is very sensitive to movements of the probe. Therefore, scanning the probe over the specimen while recording diffraction patterns leads to higher-resolution information than can be obtained by direct imaging. This technique required the implementation of a TV system capable of recording nanodiffraction patterns at 30 frames per second. By recording both an image and a nanodiffraction pattern at each point in the specimen, an image with resolution better than 0.1 nm may be calculated, even though the probe size is typically 0.3-0.4nm. Because of the large amount of data that would need to be analyzed, the technique was proposed for investigation of details in specific areas in a normal STEM image. The picture at the left, from the 1999 paper, shows how recording the diffraction pattern as the probe is moved across the sample led to the discovery of a polygonal form of carbon nanotubes. The *Micron* paper cited below was his last published.

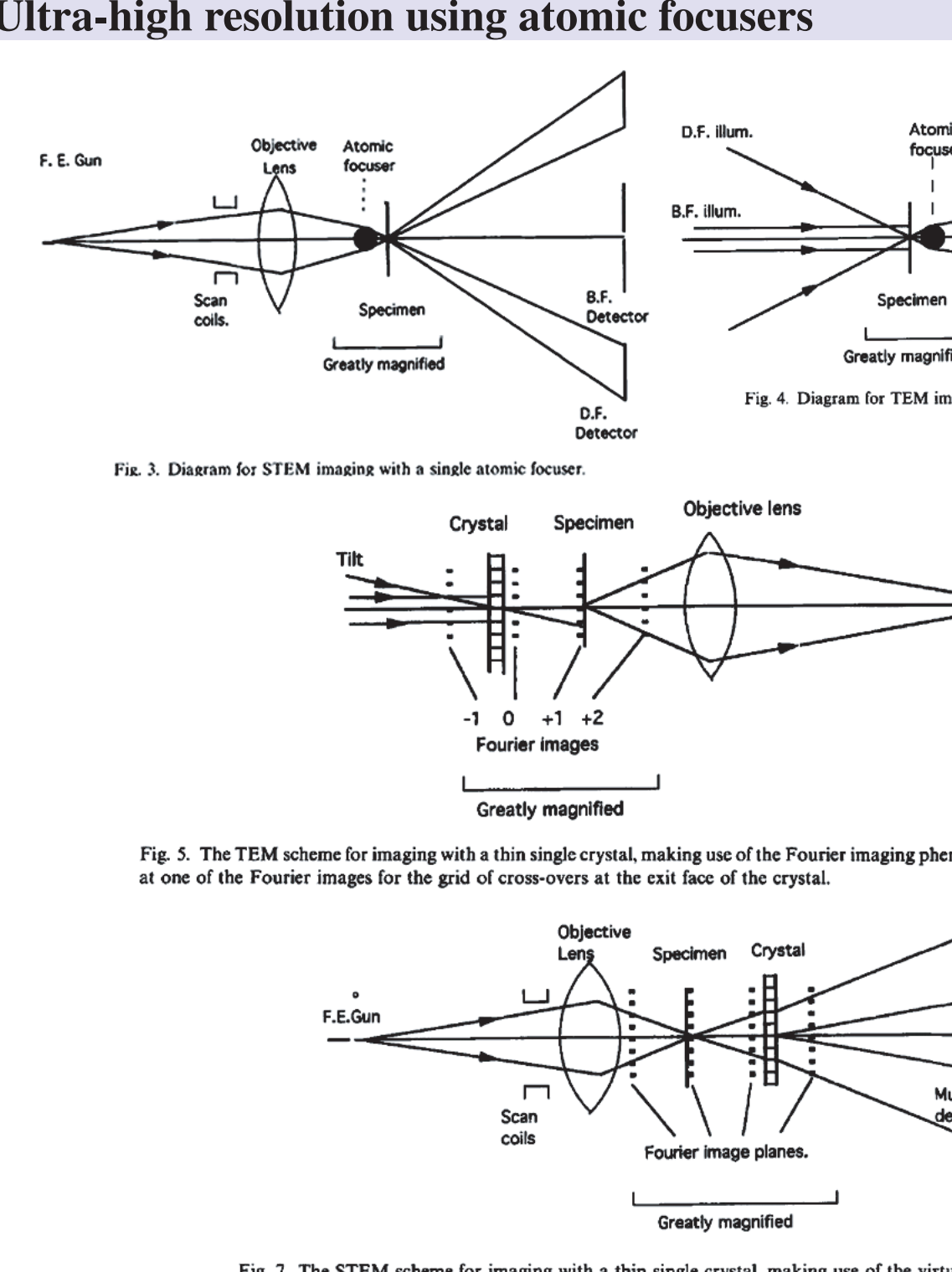
Cowley, J.M. and Spence, J.C.H. (1979) Innovative imaging and microdiffraction in STEM. *Ultramicroscopy* 3:433-438.  
Cowley, J.M., Osman, M.A., Humble, P. (1984) Nanodiffraction from platelet defects in diamond. *Ultramicroscopy* 15:311-318.  
Cowley, J.M. (1999) Electron Nanodiffraction. *Microsc. Res. Tech.* 46:75-97.  
Cowley, J.M. (2001) Comments on ultra-high resolution STEM. *Ultramicroscopy* 87:1-4.  
Cowley, J.M. (2004) Applications of electron nanodiffraction. *Micron* 35:345-360. (Review)

### Imaging and analysis of surfaces (SEM, SREM, RHEED, REM)



Excellent efficiency for detection of secondary electrons (SEM) was achieved in the HB-5 in the high-resolution mode, by detecting the electrons from the opposite side from the source. Resolution on the order of 1 nm was achieved. This was compared with scanning reflection (SREM) images using the high-energy electrons reflected off a highly tilted specimen. SREM was found useful for observing monoatomic surface steps and surface dislocations on bulk crystals. Images could also be formed with the Auger electrons. Images and microdiffraction patterns from reflected, low-loss high-energy electrons (RHEED) were also obtained. For this work, the HB-5 became "MIDAS": Microscope for Imaging, Diffraction and Analysis of Surfaces. Keeping in mind the principle of reciprocity between TEM and STEM, parallel experiments in techniques related to reflection TEM (REM) were also carried out.

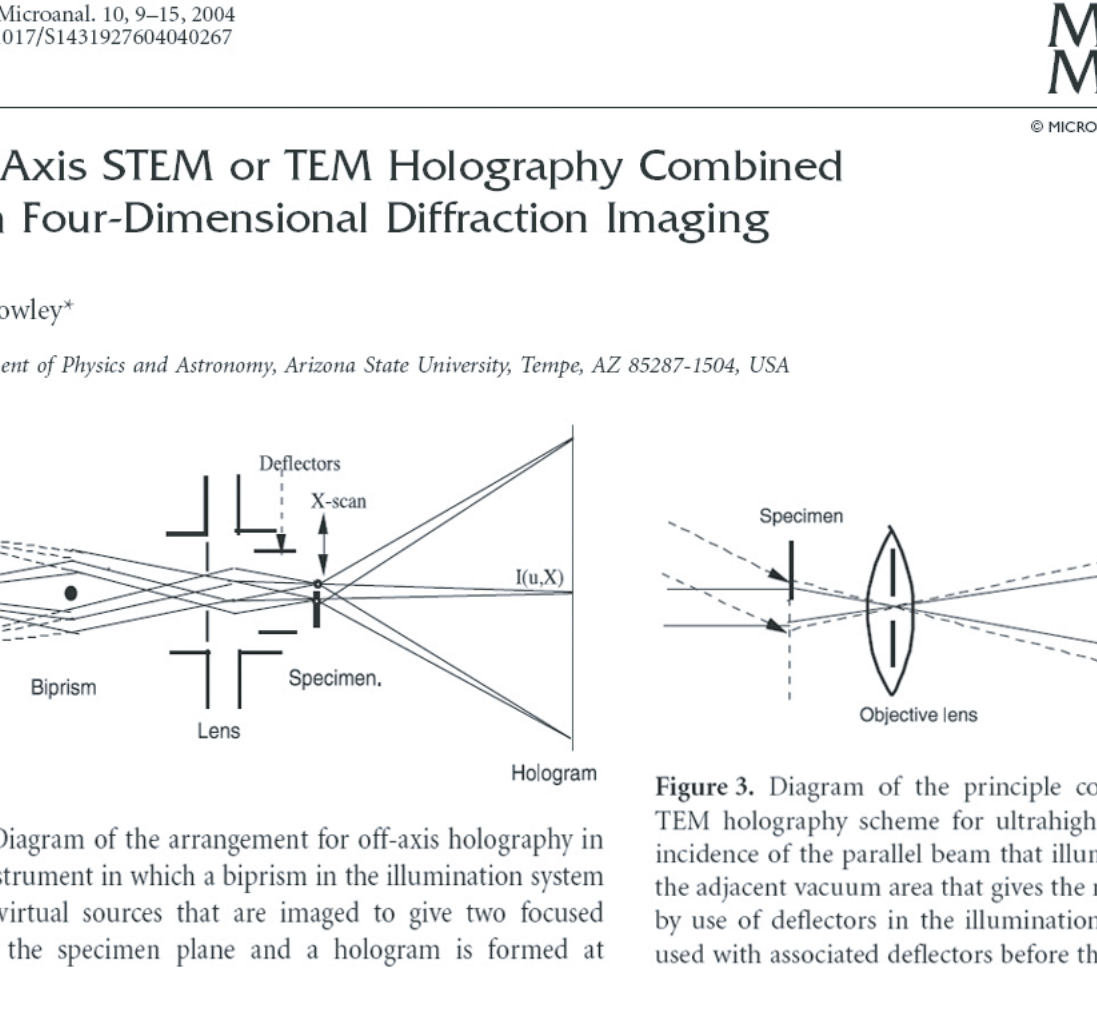
### Ultra-high resolution using atomic focusers



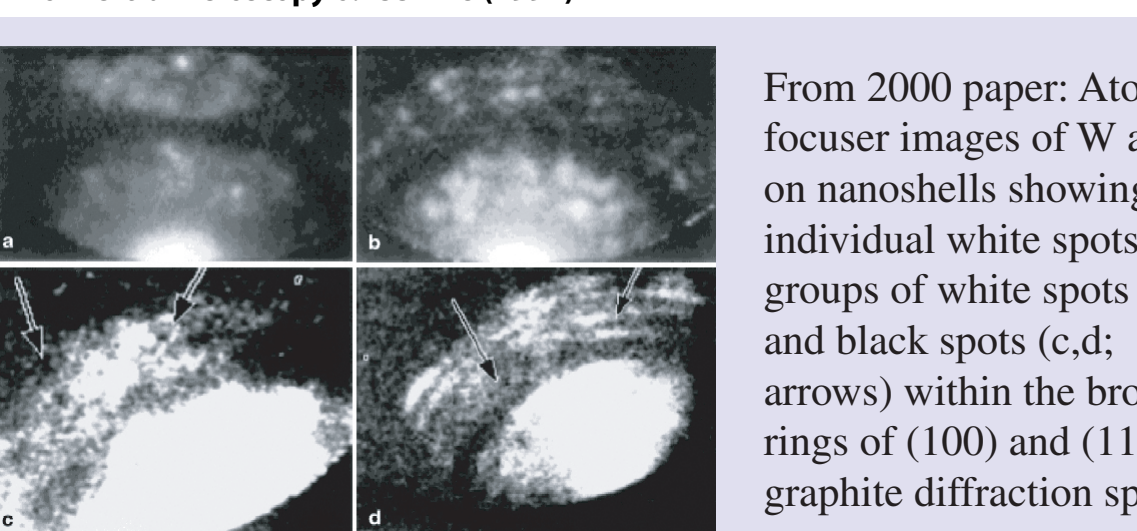
Ultra-high resolution may be achieved in STEM mode by placing the specimen within a few nanometers from an atom or thin crystal array of atoms. The atom acts as a lens to focus the probe as small as 0.05 nm. Because the focal length of this "lens" is so short, the spherical aberration is very low, and the resolution obtained may be higher than the microscope could attain in normal operation. In the corresponding TEM mode, the atoms highly magnify a small area of the specimen. To acquire a larger image area, piezoelectric translation, as is used with scanned-probe microscopies, could be used for scanning the specimen relative to the atom. Using an array of atoms in the form of a thin crystal, a larger-area image could be formed by selecting different focusing atoms using beam tilt in TEM mode, or by using an array of detectors at different angles in STEM mode. Since the depth of focus is very shallow because of the high convergence angle, slice-by-slice imaging of a relatively thick specimen may be possible. High-spatial resolution microanalysis, by EELS and x-rays, as well as Auger may be possible, with comparable beam intensity. The concept was validated experimentally using graphitic carbon as the focuser.

Lin, J., and Cowley, J.M. (1987) High-resolution scanning electron microscopy of surface reactions. *Ultramicroscopy* 23:463-472.  
Lu, P., Lin, J., and Cowley, J.M. (1989) Electron inelastic plasmon scattering and its resonance propagation at crystal surfaces in RHEED. *Acta Cryst. A* 45:325-333.  
Cowley, J.M. (1992) Resolution limitation in the electron microscopy of surfaces. *Ultramicroscopy* 47:187-198.  
Lin, J. and Cowley, J.M. (1993) Scanning reflection electron microscopy and associated techniques for surface studies. *Ultramicroscopy* 48:381-416.

### Electron holography



Gabor's scheme for overcoming the problem of aberration correction in electron lenses was to record an image of the diffraction plane, then use light optics to correct the spherical aberration and distortions (*Proc. Roy. Soc. London*, A197:454 (1949); B64:449 (1951)). The method failed at the time because the aberration of the EM lens could not be determined with sufficient accuracy. Cowley and Walker (1981) pointed out that for a weak phase object, using the high coherence available with a field-emission gun, the information inside the central disk of a nanodiffraction pattern is an in-line hologram. A Ronchigram may be used to accurately measure the aberration and defocus, and computation, rather than light optics, may be used to calculate the corrected image. However, an in-line hologram also contains a defocused, aberrated "conjugate image" that must be removed. Cowley did this by correlation of holograms obtained from series of overlapping regions as the incident beam was scanned over the specimen. Alternatively, an electron biprism may be used in TEM or STEM to form two beams, one through vacuum and one through the specimen, a technique known as off-axis holography. Interference between the two beams forms a hologram that does not include the conjugate image. Making use of the TEM/STEM reciprocity principle, Cowley described twenty forms of holography. In addition to Gabor's original scheme (Lin and Cowley, 1986), several other forms have been realized in practice by various researchers.



From 2000 paper: Atomic-focuser images of W atoms on nanoshells showing: individual white spots (a); groups of white spots (b); and black spots (c,d; arrows) within the broad rings of (100) and (110) graphitic diffraction spots.

Cowley, J.M., Spence, J.C.H. and Smirnov, V.V. (1997) The enhancement of electron microscope resolution by the use of atomic focusers. *Ultramicroscopy* 68:135-148.  
Sanchez, M., and Cowley, J.M. (1998) The imaging properties of atomic focusers. *Ultramicroscopy* 72:213-222.  
Cowley, J.M., Dunin-Borkowski, R.E., and Hayward, M. (1998) The contrast of images formed by atomic focusers. *Ultramicroscopy* 72:223-232.  
Smirnov, V.V. (1998) Atomic focusers. *J. Phys. D: Appl. Phys.* 31:1548-1555.  
Cowley, J.M. and Hudis, J.B. (2000) Atomic focuser imaging by graphite crystals in carbon nanotubes. *Microsc. Microanal.* 6:242-246.  
Cowley, J.M. and Winterton, J. (2001) Ultra-high resolution electron microscopy of carbon nanotube walls. *Phys. Rev. Lett.* 87:016101-1-4.

Among his last contributions, was a paper in his special Festschrift issue of *Microscopy and Microanalysis* edited by David Smith (February 2004). In the paper, Cowley brought together much of his earlier work to propose new methods of ultra-high resolution electron microscopy, combining nanodiffraction, atomic focusers and holography.

Since the image formed by an atomic focuser has very low aberration, the difficulty of characterizing aberrations for an in-line hologram is alleviated. Off-axis STEM holography also benefits from the use of atomic focusers. If an atomic focuser is used to reduce the diameter of the reference beam only, the area of the specimen that is illuminated is imaged with a resolution equal to the diameter of the reference beam. Thus, an image up to about 0.5nm in diameter may be imaged at about 0.05nm resolution. The probe could then be

Cowley, J.M. and Walker, D.J. (1981) Reconstruction from in-line holograms by digital processing. *Ultramicroscopy* 6:71-76.  
Lin, J.A. and Cowley, J.M. (1986) Reconstruction from in-line electron holograms by digital processing. *Ultramicroscopy* 19:179-190.  
Cowley, J.M. (1992) Twenty forms of electron holography. *Ultramicroscopy* 41:335-348.

translated, as described above, to form a larger image. Four-dimensional diffraction imaging, in which both an image and a nanodiffraction pattern are recorded at each point in the image (Rodenburg, 1993; also described above), can also be combined with off-axis holography. To avoid the problem of collecting and calculating a large number of diffraction patterns and small images, appropriate scanning voltages are applied to crossed biprisms in the STEM, forming four probes on the specimen. The Fourier transform of the projected hologram then yields sidebands that contain the corrected image. In TEM, the angle of incidence of the illumination is scanned to produce two beams and a scanned, biased crossed biprism is inserted before the image plane. Alternatively, a scanned and biased crossed biprism is placed before the specimen and a fixed biprism after.

Rodenburg, J.D., McCallum, B.B., Nellist, P.D. (1993) Experimental tests on double-resolution coherent imaging via STEM. *Ultramicroscopy* 48:304-314.  
Smirnov, V.V. and Cowley, J.M. (2002) In-line electron holography with an atomic focuser source. *Phys. Rev. B*, 65:041101.  
Cowley, J.M. (2003) Ultra-high resolution with off-axis STEM holography. *Ultramicroscopy* 96:163-166.