LIMITS TO SPATIAL RESOLUTION IN THE HRTEM

Michael A. O'Keefe

National Center for Electron Microscopy, University of California, LBL B72, Berkeley, CA 94720

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof, or The Regents of the University of California.

Ernest Orlando Lawrence Berkeley National Laboratory is an equal opportunity employer.

LIMITS TO SPATIAL RESOLUTION IN THE HRTEM

Michael A. O'Keefe

National Center for Electron Microscopy, University of California, LBL B72, Berkeley, CA 94720

The resolution of the high-resolution transmission electron microscope is limited by the specimen as well as by the HRTEM.[1] For specimens that beam-damage, image resolution depends upon electron energy and electron dose. For small-cell crystalline specimens, Bragg's law quantifies allowable resolutions, preventing image resolution from reaching instrumental resolution. Specimen thickness becomes increasingly important at higher resolutions. For a resolution of d(Å), we need the specimen to diffract at u=1/d. Consideration of the Ewald sphere intersection with the specimen shape transform (fig.1) shows that thickness must be less than $t_{max}=2/(\lambda u^2)=2d^2/\lambda$. For a resolution of 1.0Å at 300keV, thickness must be less than 100Å; to achieve 0.7Å requires halving this value to 50Å (fig.1).

Resolution in an image depends on the spatial frequencies of the information (diffracted waves) transferred from the amplitude spectrum (specimen exit-surface wavefunction) into the image intensity spectrum (Fourier transform of the image intensity). The microscope imaging system changes the phases of the diffracted waves contributing to the image intensity spectrum; these changes affect not only the phases of terms in the image intensity spectrum but also their magnitudes. Changes in magnitude impose an amplitude limit on linear information transfer called the "information limit" of the microscope. Changes in phase generate a limit called the "resolution" of the microscope.[2] Broadly, the information limit is the highest spatial frequency that can be linearly transferred from the exit-surface wave into the image; the Scherzer resolution is the highest spatial frequency that can be transferred to the image with the same phase as all lower frequencies.[3] For specimens thin enough to be considered weak phase objects, imaged at the Scherzer optimum defocus of $-\sqrt{(1.5C_S\lambda)}$, the Scherzer microscope resolution is $d_S=0.67C_S^{1/4}\lambda^{3/4}$. A Scherzer image shows the projected potential of the specimen at the microscope resolution. Because resolution is strongly dependent on wavelength, d_S can be improved by increasing the voltage of the microscope. Currently, high-voltage HREMs have resolutions approaching 1.0Å.[3]

Information at frequencies beyond d_S can be transferred by underfocusing from Scherzer defocus (fig.2a) to form higher passbands (fig.2b). This approach is bounded by the information limit imposed by focus spread (fig.2c). For thicker crystals, the focus-spread effect is described in terms of the transmission cross-coefficient.[4] For thin crystals, focus spread is described by quasi-coherent imaging theory[5] in terms of envelope functions[6,7]. Focus-spread envelope is $E_\Delta(u)=\exp\{-\frac{1}{2}\pi^2\lambda^2\Delta^2u^4\}$, and falls to exp(-2) at $u_\Delta=(\pi\lambda\Delta/2)^{-1/2}$ giving an information limit of $d_\Delta=v(\pi\lambda\Delta/2)$. Focus-spread, $\Delta=C_C[(\delta V/V)^2+(2\delta I/I)^2+(\delta E/E)^2]^{1/2}$, arises from electron beam energy spread (δE) and variations in microscope voltage (δV) and objective current (δI); C_C is the chromatic aberration coefficient of the objective lens. d_Δ can be improved by decreasing C_C, δV or δI. A factor under user control is energy spread; reducing the beam current lowers δE and Δ.[8] A large decrease in δE is obtained with a field-emission source; mid-voltage FEG-TEMs approach the information limits of high-voltage HRTEMs.[3]

Information in focal series of images is applied by comparing experimental images with simulated ones (fig.3), or by using focal-series reconstruction to combine them into one image. Methods of focal-series reconstruction include the linear-image Schiske algorithm[9], the non-linear procedures of Kirkland,[10] the paraboloid method,[11] and maximum likelihood.[12] With these methods, a microscope's resolution is no longer determined by its Scherzer limit, but approaches its information limit, once phase errors from 2- and 3-fold astigmatisms are corrected and vibration eliminated.[13]

1. F.A Ponce and M.A. O'Keefe, *Proc. 44th Ann. Meeting EMSA*, Albuquerque, N.M. (1986) 522-525.
2. O. Scherzer, J. Appl. Phys. **20** (1949) 20.
3. M.A. O'Keefe, *Ultramicroscopy* **47** (1992) 282-297.
4. M.A. O'Keefe, *Proc. 37th Ann. Meeting EMSA*, San Antonio, Texas (1979) 556-557.
5. M.A. O'Keefe and W.O. Saxton, *Proc. 41st Ann. Meeting EMSA*, Phx, Arizona (1983) 288-289.
6. J. Frank, *Optik* **38** (1973) 519-536.
7. R.H. Wade and J. Frank, *Optik* **49** (1977) 81-92.
8. O.L. Krivanek, *Optik* **43** (1975) 361-372.
9. P. Schiske, in Image Proc. and CAD, Ed. P.W. Hawkes, Acad. Press, London, (1973) 82-90.
10. E.J. Kirkland, *Proc. 42nd Ann. Meeting EMSA*, Detroit, Michigan, (1984) 432-433.
11. D. Van Dyck and M. Op de Beeck, *Proc. 12th Int. Congr. Electr. Mic.*, Seattle, Wash. (1990) 26-27.
12. W. Coene *Proc. 50th Ann. Meeting EMSA*, Boston, Massachusetts, pp. 986-987, 1992.
13. Work supported by Director, Office of Energy Research, Office of Basic Energy Sciences, Material Sciences Division of the U.S. Department of Energy, under contract No. DE-AC03-76SF00098.

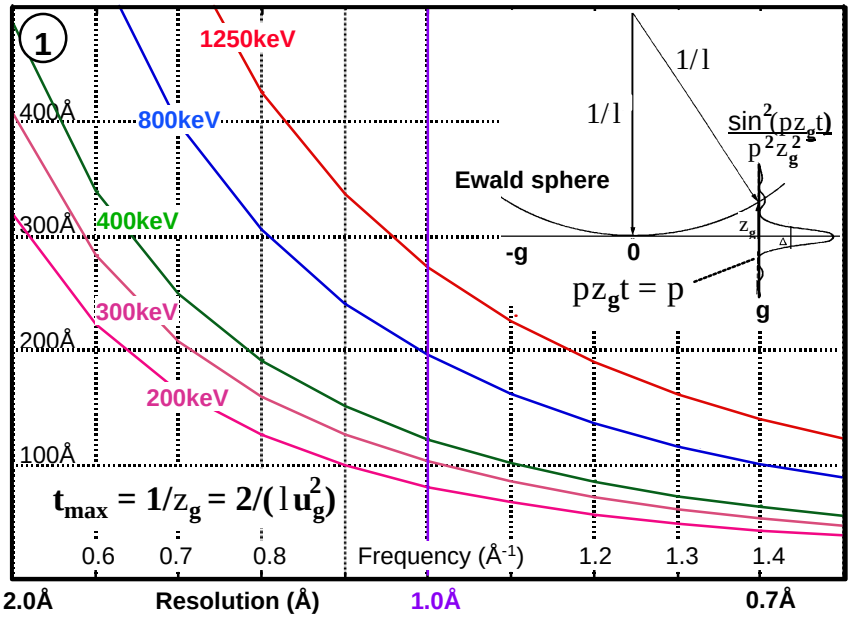

Fig. 1. Image resolution is limited by specimen thickness. Plots at several microscope accelerating energies indicate the specimen thickness at the first zero in the specimen shape-function, $t_{max} = 1/z_g = 2/(l u_g)$.

For significant diffraction at any resolution, sample thickness must lie well below the appropriate curve. For 1Å resolution, maximum thickness should be below 260Å at 1250keV, 180Å at 800keV, 120Å at 400keV, 100Å at 300keV, and 80Å at 200keV.

To reach 0.7Å resolution, maximum values halve: 130Å at 1250keV, 90Å at 800keV, 60Å at 400keV, 50Å at 300keV, and 40Å at 200keV.

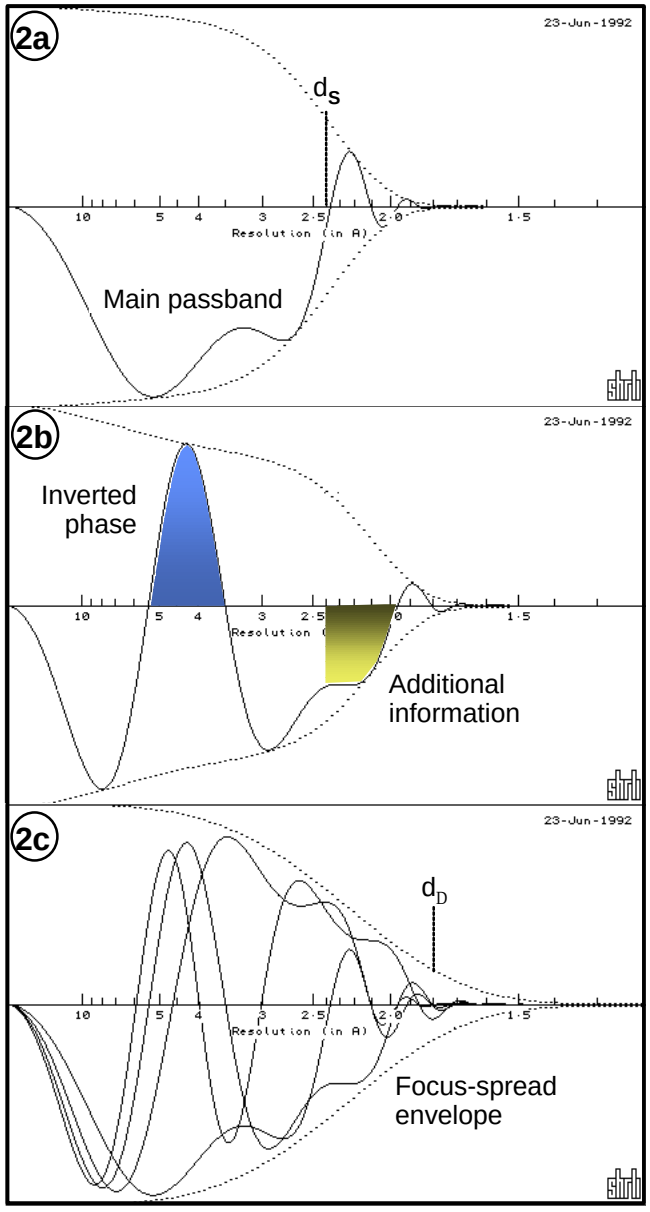

Fig. 2. Contrast transfer functions (CTFs): (a) at Scherzer defocus the main passband passes spatial frequencies to d_S with the same phase. (b) at second-passband defocus, additional information is passed at higher frequencies but mid-range frequencies suffer phase inversion. (c) at all defocus values, focus-spread envelopes all the possible linear CTFs and imposes an information limit of d_Δ.
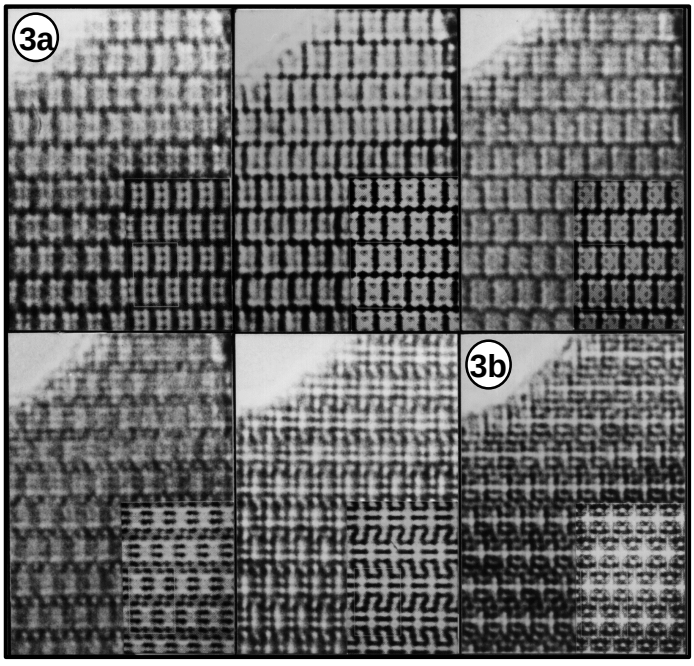
Fig. 3. Experimental focal series of $Nb_{12}O_{29}$ with inset simulations (lower right): (a) at Scherzer defocus, groups of 2x3 large tunnels are visible as 2x3 white dots. (b) at second-passband defocus, smaller tunnels are visible as small white dots lying between the groups of large tunnels, but the large tunnels are misphased and appear as a mixture of black and white contrast.