An Order of Magnitude Improvement in STEM Resolution: Wavelength High-Energy Electron Localization

Walter Gordon Morris¹, Squatter Madras², and Alwyn Eades³*

- ¹Hardye's School, Dorchester, Dorset, DT1 2ET, UK
- ²H.H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, UK
- ³Department of Materials Science and Engineering, Lehigh University, Bethlehem, PA 18015

Introduction

There have been many attempts to improve the resolution of electron microscopes. Transmission electron microscopes are normally limited in resolution by a balance between the diffraction limit (which could be overcome by the use of a large objective aperture) and the spherical aberration of the lenses used (which could be overcome by the use of a small objective aperture). In recent years successful attempts have been made to achieve resolutions beyond this limit by holography [1, 2] and also by the development of aberration correctors [3].

In this paper we bring together two concepts (each of them well-known) to propose a new method of achieving image resolution in scanning transmission electron microscopy (STEM) that goes beyond the resolution of these existing methods. The two elements that combine to make the new technique possible are the appearance of singularities in the caustics, which are formed in waves that are subject to random phase shifts, and the imaging of detail well below the size of the image-forming probe when the probe contains fine-scale structure.

Resolution in a Scanned Image

It is well known that in a scanned image, the resolution in the image can be much better than the overall size of the probe. In a scanning electron microscope (SEM), for example, the signal leaves the sample from an area corresponding to the interaction volume, but the detail seen in the image can be

orders of magnitude smaller than the interaction volume [4]. In more general terms, information may be seen in an image on a scale that is related to the maximum slope of the spatial variation of the intensity of the probe, not the overall size of the probe [5]. Therefore to achieve high-resolution images, we do not need a probe with a size equal to the resolution sought. We need only a probe that contains a strong local intensity fluctuation on the scale of the resolution.

Caustics in Randomly Scattered Waves

When waves are focused to form a small probe, they are subject to fields that are deliberately made very symmetrical. However, if waves are scattered by a more random scattering potential, the result is the production of caustic surfaces [6]. These caustic surfaces are described by catastrophe theory [7]. The conclusion of studies in this field is that in the fairly-near field of the waves scattered from a random potential, there will be caustic maxima (see figure 1). These maxima have a sharpness that would be infinite in geometric optics but in a wave theory approach can be shown to be broadened by the wavelength [7].

The Principle of the Technique

The principle of the proposed technique is that a coherent electron beam illuminates a very thin amorphous film of a light element. This acts as a scattering potential to produce a randomly phase shifted wave. The exit wave from this phase-shifting film is a wavefront with random variations in phase on the scale of atomic diameters and below.

As this wavefront propagates, caustics will form, including places where a caustic peak is locally much smaller than the atomic size, limited only by the wavelength of the electrons. This caustic peak will be surrounded by a much larger area of electron wave but with essentially no significant structure. We imagine that the beam falling on the amorphous phase-shifting film is limited in diameter so that the likelihood of more than one caustic in a given plane is negligible. In this plane below the phase-shifting film, the sample to be studied is placed and scanned under the probe by a piezoelectric drive similar to those used for atomic force microscopes. The image of the sample under investigation is thus as sharp as the caustic peak

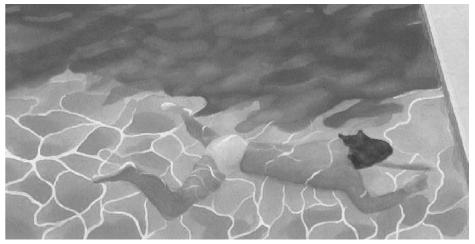


Figure 1: Detail from a painting by David Hockney: "Portrait of an Artist (Pool with Two Figures)" 1972. Acrylic on canvas. 84×120 inches. ©David Hockney. The figure shows caustics produced on the bottom of a swimming pool by the scattering of sunlight by the random pattern of waves on the surface.

^{*} jae5@lehigh.edu

DIATOME diamond knives

Development, Manufacturing, and Customer Service since 1970

What have we achieved in this period?

ultra 45° the first diamond knife with an absolutely score-free, hydrophilic cutting edge.

semi the first diamond knife for alternating sectioning ultrathin/semithin.

cryo the diamond knife for sectioning at low temperature.

histo the first diamond knife for semithin sections for light microscopy.

ultra 35° the diamond knife for optimized sectioning results in almost all applications.

STATIC LINE II the ionizer for eliminating electrostatic charging in ultramicrotomy.

cryo-P a cryo knife with a patented platform for section pick up.

cryo immuno the optimized cryo diamond knife for the Tokuyasu technique.

ultra sonic the oscillating diamond knife for room temperature sectioning.

cryotrim 45 and 25 optimizing trimming with diamond blades.

ultra AFM & cryo AFM the first diamond knives for AFM at room and low temperatures.

cryo 25° for sectioning frozen hydrated specimens.

What services can we offer you?

- Technical assistance in all fields of ultramicrotomy.
- Free sectioning tests for all types of samples.
- Make use of our many years of experience in perfecting our knives.
- Custom knives, tools, and boats.
- Special purchase programs.
- Workshops and training.



For more information, please call or write us today, or visit us online at:

www.emsdiasum.com

DIATOME

for all your sectioning requirements

P.O. Box 410 • 1560 Industry Rd. Hatfield, Pa 19440 (215) 412-8390 • Toll Free: 1-(800) 523-5874 Fax: (215) 412-8450 or 8452

email: sgkcck@aol.com • stacie@ems-secure.com www.emsdiasum.com

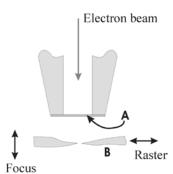


Figure 2: Schematic diagram to show the principle of the method. A is a thin amorphous film that serves to introduce a suitable scale of random phase shifts on the electron wave. B is the sample to be studied. It is moved vertically to find "focus" that is a position where the electron wave field contains a singular caustic. The sample is rastered under the beam to form the image. Note: Rastering the electron beam does not serve, in this case.

in the probe but with an added diffuse background formed by the skirt surrounding the caustic peak. We coin the term Wavelength High-Energy Electron Localization for this new technique.

Figure 2 shows a schematic diagram of the apparatus. A thin amorphous film (A) imposes random phase shifts, as a function of lateral position, on the electron incident The wave field below A contains caustics produced by these phase shifts. Thus the electron wave field that is incident on the sample (B) is a large diffuse patch crossed by bright caustics. The transmitted beam then

contains information on the sample, B, down to a resolution corresponding to the size of the caustic in the illumination.

At any height of the sample, the electron wave field will contain caustics that are basically lines similar to those in Figure 1. However in certain planes below A, the wave field will contain singularities [7]. In such planes the intensity will be extremely bright in a local region, rather than moderately bright along a line. If the sample, B, is located in such a plane, the image will resolve detail down to the size of the singularity.

There is no way (that we are aware of) that would allow us to examine the wave field formed by the phase-shifting sample to see directly the caustics formed. We anticipate that the sample to be studied would be moved along the beam direction, looking to see fine detail in the images produced until a caustic singularity is found through observation of fine detail in the image.

There are two apparent difficulties. On the one hand the imaging probe will have the complex shape of a caustic singularity, rather than the Gaussian shape of an "ideal" probe. On the other hand there will be no way to check that there is just one singularity. Both of these problems have straightforward solutions, but, for reasons of space, it is not appropriate to give them here.

Comparison with an Earlier Method

When a lens is used to focus a beam, the process may be thought of as producing a caustic, but one in which, because of the symmetries so painstakingly imposed on the fields, the caustic is highly degenerate, rather than one of the structurally stable singularities that result from unsymmetric scattering. Thus, when Cowley and others suggested that resolution could be enhanced by putting an atom (one atom) into the electron beam, they were proposing one special case of the present method, albeit one that seems impossible to realize [8].

Conclusion

We propose a new method for obtaining much higher resolutions than can be obtained with current instrumentation.

It seems plausible that, if the requisite stabilities can be achieved, direct imaging should be readily obtained that is an order of magnitude better than present results.

See page 68 for a note added in proof.

References

[1] D Gabor, Nature 161 (1948) 777-78.

[2] E Volkl, LF Allard and DC Joy, eds., *Introduction to Electron Holography*, New York, Kluwer Academic/Plenum Publishers, 1999.

[3] An excellent source of up-to-date information on aberration-corrected microscopy is given in a special issue of *Phil. Trans.*: *Philos T Roy Soc A* 367 (2009) 3631–3870.
[4] JI Goldstein, DE Newbury, P Echlin, DC Joy, CE Lyman, E Lifshin, L Sawyer, and JR Michael, *Scanning Electron Microscopy and X-ray Microanalysis* (Third edition 2003), New York, Kluwer Academic/Plenum Publishers, 197ff.

[5] PE Batson, DW Johnson, and JCH Spence, *Ultramicroscopy* 41 (1992) 137–45.

[6] MV Berry, JF Nye, Nature 267 (1977) 34-36.

[7] MV Berry, C Upstill, Catastrophe optics: morphologies of caustics and their diffraction patterns Progress in optics Vol. XVIII (1980) 257–346.

[8] JM Cowley, JCH Spence, and VV Smirnov, *Ultramicroscopy* 68 (1997) 135–48.

Μт

Research Engineer in Advanced Analytical Electron Microscopy

Department of Materials Science and Engineering Lehigh University, Bethlehem, PA

The Nanocharacterization Laboratory at Lehigh University houses an impressive array of electron-optical instruments including two state-ofthe-art aberration-corrected analytical electron microscopes. It is also the home of the world-renowned Lehigh Microscopy School. The Research Engineer position entails managing all aspects of TEM/STEM operations within this user facility. Major responsibilities of the successful candidate include: training users in the operation of the laboratory's TEM/STEM microscopes and related specimen preparation equipment, including a dual-beam FIB; performing routine maintenance and overseeing essential repairs of the instrumentation; managing routine operation, including logs, bookkeeping, billing, maintenance records, and safety; making recommendations and assisting in the purchase, installation, and commissioning of new equipment; liaison with external users from other universities, government laboratories, and local industry; providing outreach and tours for the general public; and participating in the annual Lehigh Microscopy School (http://www.lehigh.edu/microscopy). Further details of the Nanocharacterization Laboratory and position requirements can be found at http://www.lehigh.edu/~inmicro/.

Applicants should submit a complete CV and the names of three referees to Sharon Coe, Department of Materials Science and Engineering, Lehigh University, 5 East Packer Avenue, Bethlehem, PA 18015-3195, USA. We will start reviewing applications after March 15, 2010, and the search will continue until the position is filled. *Lehigh University is committed to recruiting and retaining women and members of minority groups*.

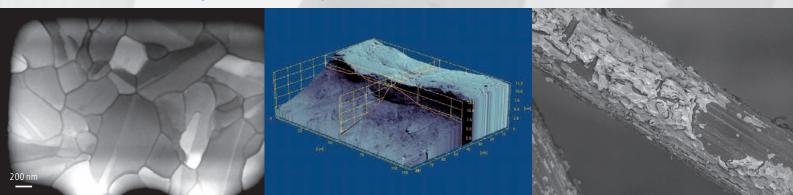
MERLINTM

Analytical Power for the Sub-Nanometer World

Nano Analytics | Total Information | Ease of Use | Future Assured



Field Emission - Scanning Electron Microscopes (FE-SEM) of Carl Zeiss



Carl Zeiss SMT Inc.

One Corporation Way Peabody MA 01960

USA

Enabling the Nano-Age World®

Tel. +1 978/8267909 Fax +1 978/532 5696 info-usa@smt.zeiss.com www.zeiss.com/nts



We make it visible.