Achieving atomic resolution in secondary electron imaging using aberration corrected STEM

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Secondary electron imaging in a STEM

Imaging surface U atoms

Nature Materials, 8, 808 - 812 (2009)

Imaging single atoms using secondary electrons with an aberration-corrected electron microscope

Y. Zhu1*, H. Inada2, K. Nakamura3 and J. Wolf1

Aberration correction has embanked on a new frontier in electron microscopy by overcoming the limitations of conventional round lenses, providing sub-angstrom-sized probes1. However, improvement of spatial resolution using aberration correction so far has been limited to the use of transmitted electrons both in scanning and stationary mode, with an improvement of 20-40% (refs 3-5). In contrast, advances in the spatial resolution of scanning electron microscopes (SEMs), which are by far the most widely used instrument for surface imaging at the micrometre-nanometre scale6, have been about the particles’ locations, much of which is lacking in the transmission image. In the past decade or so, high-resolution STEM has proven an indispensable critical-dimension metrology tool for the semiconductor industry. The semiconductor roadmap identifies the need for ultrahigh-resolution STEM in the quest for ever-decreasing device sizes7.

We attempted to achieve the highest possible SEM resolution and to determine whether it is limited by the basic physics of secondary production or by the instrumentation. We explored well-defined samples (single uranium atoms) in an instrument.
Is there any contrast delocalization?

- SEM & ADF images are equally sharp, with FWHM 1Å for SEM and 0.8Å for STEM.
- there is NO detectable delocalization signal in atom images.

Localized contrast must be due to high-angle momentum transfer single-electron excitation.
Study of single-electron excitations by electron microscopy

II. Cathodoluminescence image contrast from localized energy transfers

By S. J. Pennycook and A. Howie
Cavendish Laboratory, Madingley Road, Cambridge, CB3 0HE, England

[Received 1 October 1979 and accepted 10 November 1979]

ABSTRACT

Observations of cathodoluminescence in ZnS single crystals using scanning transmission electron microscopy exhibit a dependence on crystal orientation indicating that about 20% of the signal originates in localized excitations. This figure can be explained in terms of energy transfer to valence excitations from energetic secondary electrons ejected from inner shells. Similar effects may operate in electron-beam-induced-conductivity images, X-ray production and other processes.


**Study of single-electron excitations by electron microscopy**

**II. Cathodoluminescence image contrast from localized energy transfers**

By S. J. PENNYCOOK and A. HOWIE

Cavendish Laboratory, Madingley Road, Cambridge, CB3 0HE, England

[Received 1 October 1979 and accepted 10 November 1979]

<table>
<thead>
<tr>
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<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
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The table above provides a detailed analysis of the stopping power and localization of energy transfers in various electron shells and bands, with specific values for each parameter across different materials.
Imaging cathodoluminescence in a STEM

Incoherently generated signals from ZnS

Cartridge to allow simultaneous detection of cathodoluminescence and transmitted electron signals.


Comparison between experimental intensities (solid curves) and 11-beam theory for (a) transmitted bright-field intensity, and (b) cathodoluminescence intensity; dashed curve in (b) is for a square-well situated on the Zn atoms, dashed/dotted curve is for a square-well situated on the S atoms.
Imaging cathodoluminescence in a STEM


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Pb on Carbon substrate

Pt/graphite core/shell

SE

ADF

SE image

ADF image

BF image

10kVdc

sample

10kVdc

sample

BSE

SE suppressor

SE detector

preamplifier

SE detector

preamplifier

BF detector

Hitachi HD2700C for HR-SE/ADF

Cs corrector

scan coil

metal cover

Hitachi in-lens design pole piece

BF detector

SE suppressor

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Imaging surface and bulk simultaneously (ADF/SE pairs)

CaO particles on MgO

SrTiO$_3$

SrTiO$_3$ >1um thick
Atomic imaging of surface and bulk

$YBa_2Cu_3O_7$

$t/\lambda = 0.45, \ t = 52 \ nm$

SE imaging appears to be more sensitive to light elements than ADF imaging

SE

raw data

ADF

average of 55 unit-cells

SE

ADF

SE
Separating SE from BES - sample biasing
BSE contributes 2% of total signal, but 4% of atomic signal.
Bias experiment: U stained TMV clumps and carbon film

SE: 85-90%, BES/FSE: 10-15%

Normalized SE/ADF intensity (U+C)

contrast level: Gatan display

contrast level: Min: 0; Max: 30,000 e−

Normalized SEM/ADF ratio vs bias

Normalized Intensity ratio (SEM/ADF) vs bias (V)

Normalized SE/ADF intensity (U+C)

+10 eV

+15 eV

+25 eV

+50 eV
Can we image surface structure?

\[ p \sim \exp\left(-\frac{z}{\lambda}\right) \]

\(p\): the escape probability
\(z\): depth below the surface
\(\lambda\): SE mean free-path.

SrO terminated

TiO\(_2\) terminated

SrTiO\(_3\) annealed in Oxy @ 1050°C

collection angle 46~104 mrad

\(\lambda \approx 115\)nm for SrTiO\(_3\)
Why we can achieve atomic resolution in SE imaging?

Atomic imaging using secondary electrons in a scanning transmission electron microscope: Experimental observations and possible mechanisms

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b Hitachi High Technologies Corp., Ibaraki, Japan
c University of Alberta, Edmonton, Canada

The SE generation & scattering processes

 specimen surface

Emission into vacuum

SE propagation

~2nm

Special issue for John Spence’s 65th birthday
A unique approach to calculate atomic resolution SE images

1. Wave function in (n+1)th slice:
   \( \psi_{n+1}(x, y) = p_n(x, y, \Delta z) \otimes [t_n(x, y, z) \psi_n(x, y)] \)

2. Intensity of PR and SE electrons:
   \( I_{n}^{PR}(x, y) = \psi_n(x, y) \psi_n^*(x, y) \),
   \( I_{n}^{SE}(x, y) = I_{n}^{PR}(x, y) L_n(x, y) \)

3. The object function for SE generation:
   \( L(r) = A^2(r) \)

4. The SE generation function:
   \( G_n(p) = \sum_{x,y} I_{n}^{SE}(x, y) \)

5. The SE transmission function:
   \( T_n(p) = \exp \left( -\frac{z_n}{MFP} \right) \)

6. Total SE intensity:
   \( S(p) = \sum_n G_n(p) T_n(p) \).

p : probe position
x,y : multislice coordinates
r : atomic inelastic scattering coordinates

Wu, Egerton, Zhu, Ultramicroscopy (2012)
Crewe special issue
SE image simulation using an object function model

SE images, top surface: TiO

SE images, top surface: SrO
SE image simulation using an object function model

Simulation: thickness=15nm, MFP=2nm, convoluted with a Gaussian function with FWHM=0.1nm)

Wu, Egerton, Zhu, Ultramicroscopy (2012)
The simulation: \( \text{MFP}=2 \text{nm} \), convoluted by a Gaussian spread function with FWHM=0.16 nm, \((C_s=9.6 \mu \text{m}, \Delta f=1.4 \text{ nm}, A_1=0.16 \text{ nm}, A_2=5.12 \text{ nm}, B_2=1.76 \text{ nm}, A_3=206.9 \text{ nm}, S_3=110.6 \text{ nm}, A_4=4.3 \mu \text{m})\) based on measured parameters of Hitaci 2700C.
Secondary electron imaging at atomic resolution using a focused coherent electron probe


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(Received 3 December 2012; published 5 February 2013)

Atomic resolution imaging using secondary electrons (emitted as a result of the interaction of incident fast electrons with a specimen) was only achieved recently. There has been considerable speculation as to the physical process behind the imaging. In this paper we use a quantum mechanical model to show that the image contrast is due to variations in the probability of electron ejection in inner-shell ionization events initiated by the primary beam, an atomic event that provides the imaging contrast. The angular probability distribution of the ejected electrons is key to this contrast: different atomic species within the specimen. For a given species, the angular probability distribution is predominantly determined by the angular momentum quantum number of the core electron prior to ionization. The model is compared to experiment and reproduces the intensity distribution.
The challenge of SE imaging: surface quality!

SE is sensitive to low-z carbon contamination.

Extension into vacuum is not due to edge enhancement.

A clean sample surface is extremely important!
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