Atomic Resolution Secondary Electron Imaging in Aberration Corrected STEM

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**BIOGRAPHY**
Hiromi Inada has a master’s degree in engineering from Nagaoa University of Technology. Joining Hitachi Ltd. in 1998 he focused on TEM and STEM development. From 2007 to 2009 he was stationed at The Center for Functional Nanomaterials in Brookhaven National Laboratory as a visiting researcher, working with Dr Yimei Zhu on aberration-corrected STEM studies. He currently works in the Science and Medical System Design Division of Hitachi High-Technologies with responsibility for TEM/STEM design.

**ABSTRACT**
Aberration-corrected electron microscopy has enabled sub-nanometre atomic scale characterisation to be applied to a wide range of materials although to date only transmitted electron signals have generally been studied. The very latest studies into atomic resolution secondary electron (SE) imaging using an aberration-corrected STEM are described. Following a series of experiments, we have succeeded in observing isolated single atoms using secondary electron signals. In addition, we have clearly observed atomic resolution not only for heavy elements like uranium but also for lighter elements such as silicon and carbon.

**INTRODUCTION**
Realising the Potential of Atomic-Resolution Imaging
In recent years the aberration-correction technique has brought a revolution in analytical microscopy by making atomic-resolution imaging and analysis routinely achievable in both transmission and scanning transmission electron microscopy (TEM and STEM).

Scanning electron microscopes (SEM), however, remain widely used in the observation of specimens at the micrometer and nanometer scales and are key to the development of many materials and devices. In the SEM, the secondary electron (SE) imaging mode is commonly used, enabling us to observe three-dimensional topographic images of the sample surface. One of the most advanced commercially available SEMs (Hitachi’s SU9000 In-lens SEM/STEM) achieves a spatial resolution of less than 0.4 nm. Whilst studies into even higher-resolution SE imaging have been approached, none of the research to date has achieved atomic resolution [1]. We have now succeeded in observing secondary electron images of single uranium atoms using an aberration-corrected 200 kV cold field-emission (CFE) STEM. We have observed atomic resolution images not only for heavy elements like uranium but also for lighter elements such as silicon and carbon [2, 3]. The observation of atomic resolution secondary electron imaging is unprecedented and may lead to new theories of signal generation, along with a host of new applications – some of which are highlighted here.

**Scanning Transmission Electron Microscopy**
This work was undertaken using the Hitachi HD-2700 dedicated STEM installed at the Brookhaven National Laboratory in NY. This STEM incorporates a probe aberration-corrector (CESCOR, from CEO S GmbH) (Figure 1) [4]. The microscope also features a cold field-emission gun with high brightness and a small energy spread, originally developed in consultation with Crewe in 1970s [5] and now fully optimised for stable observation and analysis in aberration-corrected microscopes. Coupled together these two technologies provide a small electron beam diameter (<0.1 nm) with high current for analysis and high signal-noise ratio imaging.

**Figure 1:** A Hitachi HD-2700 200 kV scanning transmission electron microscope as installed at the Brookhaven National Laboratory in New York, USA. This STEM incorporates a probe aberration corrector (CESCOR) from CEO S GmbH.
beneath the specimen, enabling the simultaneous observation of secondary and transmitted electron images.

**ATOMIC RESOLUTION IMAGING WITH SECONDARY ELECTRONS**

**Secondary Electron Imaging – A Complementary Imaging Signal**

Figures 2a to 2c show a low magnification comparison of SE, ADF-STEM and BF-STEM images of a core-shell catalyst specimen (palladium core and platinum shell) at 200 kV and with a 0.1 nm diameter electron probe [6]. We note that the SE image clearly demonstrates the topographical structure, enabling the distribution of particles with respect to the carbon support matrix to be studied. Thus, at low magnification the SE image provides useful structural information in itself. SE imaging capability, however, also plays an important role in locating a suitable region for electron energy-loss spectroscopy (EELS) and energy-dispersive X-ray (EDX) analysis enabling unwanted background material to be avoided.

Figures 2d to 2f show high resolution images of a gold nanoparticle on a carbon film. Au (111) lattice fringes with 0.24 nm spacing are marked, whilst particle morphology and faceting can be clearly observed in the SE image. Once again, the ability to simultaneously observe SE and TE signals is of paramount importance.

**Atomic Resolution Secondary Electron Imaging of Single Atoms**

Figure 3 shows simultaneously acquired SE and ADF-STEM images of isolated and clumped uranium atoms on a 2-nm thick carbon film (at 200 kV with a convergence angle of 28 mrad). The test specimen is a typical uranyl acetate negatively stained tobacco mosaic virus (TMV) on a carbon thin film, enabling the comparison of single atom imaging capability with...
conventional ADF-STEM Z-contrast imaging. Quantitative analysis of the images indicates the existence of uranium atom information in the SE image. Across 50 possible atom positions in the SE and ADF images, the averaged full-width and half-maximum (FWHM) intensity profile is less than 0.1 nm.

Atomic Resolution Secondary Electron Imaging of Light Elements
Our results indicate that SE atomic resolution is achievable for both heavy and light elements, although the spatial resolution for light elements may be reduced [2, 3]. We have successfully observed atomic resolution SE images on composite functional materials such as SrTiO$_3$ and YBa$_2$Cu$_3$O$_7$.

Figure 4 shows one example of one of these studies on graphitic carbon (Z=6) with Pt catalyst particles. The carbon graphite lattice (shown by arrows) corresponding to 0.34 nm is observed in Figure 4a. In comparison with STEM imaging, the SE imaging technique appears more sensitive to light elements, yielding higher intensity and contrast as well as topographical information. The observation of atomic images of light elements was unexpected, and certainly challenges the traditional understanding of the mechanism of SE image formation.

Signal Generation: Are the Images really formed by Secondary Electrons?
An important aspect of this study was to understand whether the electrons emerging from the surface were indeed secondary or whether they were backscattered electrons. By applying a positive bias voltage to the specimen, the low energy SEs (<50 eV) are suppressed from escaping from the surface and only higher energy backscattered electrons can reach the detector.

Figure 5 is an image comparison of SE intensity as a function of specimen positive bias. The minimum grey level is set to zero and the maximum grey level to 35,000 for the purposes of image comparison. The specimen was biased at 0, 2, 5, 10, 15 and 25 eV, respectively. The signal intensity decreases rapidly with increasing positive bias and the signal is saturated at biases above 20 eV. Studies show that for 200 keV primary electrons the SEs represent 90% of the total SE+BSE signal. Additionally, using electron trajectory simulations...
of the optical configuration used in this experiment, it can be shown that only 0.002% of the SE3 generated by BSEs are passed through the upper pole-piece hole. Thus the signal content is shown to be predominantly SE.

In respect of the atomic resolution observed it is also necessary to consider the beam-sample interaction volume. Monte-Carlo simulations suggest that the electron interaction volume at 200 keV is 1/100th of that at 20 keV. The traditional mechanism of secondary electron imaging is attributed to the inelastic scattering and delocalised decay of collective electron excitation. An atomic-scale SE image mechanism is proposed in which contrast depends on the width of the point spread function, arising from inelastic scattering with large momentum transfer.

**Atomically Resolved Imaging even on Bulk Specimens: Example Applications**

In the industrial field, SEM is widely used for routine inspection and measurement as well as materials development. The routine metrology of semiconductors for process control has become increasingly challenging as the design-size of devices has reduced. Conventionally the routine metrology of semiconductor devices has been performed using high resolution SEM, whilst the very latest semiconductor devices at the 45-nm node require TEM or STEM imaging on thin specimens in order to provide sufficient resolution for reliable assessment of gate oxide thickness and other key parameters. The atomic resolution SE imaging technique may prove invaluable to semiconductor metrologists by ensuring atomic resolution is available without the complexity of thin specimen preparation.

Figure 6a shows high-resolution images of a single crystal Si thin specimen (50 nm) [7]. In the thin specimen, silicon dumbbells (0.136 nm) and the corresponding power spectrum spot (004) can be observed. Most significantly, even with a 1-µm thick specimen, high-resolution SE images still present lattice fringe information in the Si(111) plane with a resolution of 0.314 nm (Figure 6b). This result suggests that: 1. measurement can be calibrated ‘on-the-fly’ using the lattice fringe in the same region of interest with high accuracy; and 2. the specimen for atomically resolved SE imaging can be much thicker than that for ADF and BF-STEM. We have confirmed that automatic metrology software based on the contrast measurement can be applied with this SE image.

We believe the atomic-resolution SE imaging technique can be applied to a variety of materials and systems where thin specimen preparation is either challenging due to material properties or impractical due to throughput requirements.

**Conclusions**

High-resolution SE imaging enables the surface characteristics of specimens to be observed simultaneously with the acquisition of transmitted electron signals. We have demonstrated atomic resolution secondary electron imaging for low and high-Z materials using the Hitachi HD-2700.

We have shown the results of atomic resolution SE imaging and we can conclude that there are three key parameters to be satisfied: 1. An ultrahigh efficiency through the lens SE detector is required. 2. An aberration-corrected electron beam with a diameter of around 0.1 nm used simultaneously with high current is required. 3. A high-energy primary electron beam results in a small interaction volume in the specimen.

The atomically resolved SE imaging technique is already being applied to a wide variety of materials, although the full potential of the technique is yet to be realised. The ability to correlate surface and structural properties is key to understanding many material systems. The ability to perform atomic resolution imaging on bulk materials may revolutionise high-throughput electron microscopy, particularly in semiconductor metrology or in applications where thin specimen preparation is difficult or impractical.

**Acknowledgements**

The author is grateful to Dr Yimei Zhu, Dr Joseph Wall, Dr Lijun Wu and Dr Dong Su of The Brookhaven National Laboratory, Dr Ray Egerton of The University of Alberta and Dr Jim Ciston of The Lawrence Berkeley National Laboratory for collaborative work on the experiments and for fruitful discussions on the contrast mechanism. The author also thanks Dr Michael Dixon of Hitachi High-Technologies Europe for support.

**References**