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Reflective Small Angle Electron Scattering to Characterize Nanostructures on Opaque Substrates

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Abstract

Features sizes in integrated circuits (ICs) are often at the scale of 10 nm and are ever shrinking. ICs appearing in today's computers and hand held devices are perhaps the most prominent examples. These smaller feature sizes demand equivalent advances in fast and accurate dimensional metrology for both development and manufacturing. Techniques in use and continuing to be developed include X-ray based techniques, optical scattering and of course the electron and scanning probe microscopy techniques. Each of these techniques have their advantages and limitations. Here the use of small angle electron beam scattering measurements in a reflection mode (RSAES) to characterize the dimensions and the shape of nanostructures on flat and opaque substrates is demonstrated using both experimental and theoretical evidence. In RSAES, focused electrons are scattered at angles smaller than 1° with the assistance of electron optics typically used in transmission electron microscopy. A proof-of-concept experiment is combined with rigorous electron reflection simulations to demonstrate the efficiency and accuracy of RSAES as a method of non-destructive measurement of shapes of features less than 10 nm in size on flat and opaque substrates.

Both incremental improvements in electronics fabrication and disruptive technologies have in common improved performance through shrinking dimensions. Integrated circuits (IC) such as dynamic random-access memory (DRAM) that appear in most of our hand-held and desktop devices and flash memory and 3D transistors such as FinFETs have features that already breach the 10 nm mark.¹ Equivalent advances in fast and accurate dimensional metrology are needed for both development and manufacturing.²⁻⁶ So far, this challenge has been met with techniques that include X-ray scattering, optical scatterometry (OCD), electron microscopy and atomic force microscopy, each with its advantages and limitations. Here, we introduce small angle electron scattering in reflection mode (RSAES) as a non-

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destructive measurement of dimensions and shapes of features less than 10 nm in size on flat and opaque substrates. In RSAES, focused electrons are scattered at angles smaller than 1° with the assistance of electron optics typically used in transmission electron microscopy (TEM).⁷ We combine a proof-of-concept experiment with rigorous electron reflection simulations to demonstrate the efficiency and accuracy of RSAES.

Efficient measurement of features smaller than 10 nm requires: (1) high dimensional precision including 3D capability, (2) a measurement footprint small enough to measure micron-size test arrays of nanodevices, yet large enough to get a good statistical sampling, (3) little to no sample preparation and (4) a strong output signal so that samples can be measured in seconds, not hours.⁴ OCD involves scattering light of various wavelengths. On its own, scatterometry encounters severe difficulties in terms of resolution when measuring length-scales below 30 nm, much less 10 nm (requirement 1).^{8–11} OCD also hits theoretical limits from electron confinement effects at 7 nm.⁴ Electron microscopy and atomic force microscopy work well to determine dimensions to high precision except those with high aspect ratio.⁴ Also, they are unable to provide good statistical sampling of an entire array of nanodevices (requirement 2), often require laborious and time consuming sample preparation (requirement 3) and can even charge the sample with electrons leading to systematic measurement errors.³ A complementary scattering technique is needed for its statistical accuracy and ease of sample preparation. X-ray scattering^{2,6,11} has been initiated by one of the authors (W.-L. W.) as a viable dimensional and shape metrology where the used wavelength is about a tenth of a nanometer; however, once an X-ray source has been collimated to a spot size of a few tens of micrometers or below (requirement 2), the net scattered X-ray flux is too small to produce a viable measurement in a few minutes, or samples must be taken to a high-brightness X-ray source such as the Advanced Photon Source¹¹ (requirement 4). Hybrid Metrology combines methods to mitigate existing limitations but not resolve them.⁵

RSAES leverages existing TEM technology to probe structures with high dimensional precision and a small footprint. Electron scattering cross-sections that are typically 10^8 times larger than those of X-rays.^{12,13} Also, RSAES will be less affected by the charging issues affecting CD-SEM as these high scattering cross sections combine with high reflection efficiencies at small angles to allow lower electron beam currents.

RSAES uses two sets of electron optics (Figure 1a). Collimating optics focus an electron beam through a sample surface to achieve high resolution and high scattering intensity simultaneously. Projection optics focus the reflected electrons onto a detector to generate a high-intensity diffraction pattern (RSAEP). As an example, consider an etched Si structure with a height of 10 nm. A reflected electron beam with a wavelength of 10 pm will exhibit constructive/destructive interference first at an elevation angle of about 0.03° , and the first five interference fringes will occur below 0.15° . To measure the height with an error less than 5 % will require an angular resolution better than $\theta < 0.005^\circ$. Similar angular resolutions are needed in the transverse direction to measure pitch and cross-section geometry. At these very small angles, the signal strength of RSAES is also very good. For a typical mean inner potential of 10 eV^{14,15} for silicon, and an electron wavelength of 10 pm, at $\theta = 0.1^\circ$, the Fresnel reflectance is 75%, and at $\theta = 1.0^\circ$, it is 8%.

RSAES differs from Reflection Electron Microscopy (REM)^{16,17} and Reflection High-Energy Electron Diffraction (RHEED).^{18–20} REM is an imaging technique, and RHEED lacks projection optics. RHEED measures features that are atomic length-scale. The Laue diffraction patterns generated by RHEED range over a few degrees while Reflective Small-Angle Electron Diffraction Patterns (RSAEPs) are ten to 100 times smaller.

The necessary configuration (Figure 1a) cannot be fully realized in existing TEMs, but a proof-of-concept (POC) experiment was performed with a commercial TEM. The experiment shows that the diffraction pattern is sensitive to the height of three-dimensional nanostructures.

High angular resolution small angle electron scattering via TEM has three basic protocols.²¹ The three lens method (3LM) of Mahl *et al.* was used here because it allows the sample to be illuminated by a convergent electron beam, hence, a high scattering intensity is expected.²² The high dispersive (HD) diffraction mode common to commercial TEMs provides sufficient resolution but insufficient scattering intensity. A JEM2100F microscope²³ was operated at 100 keV which is above the desirable voltage of 8–20 keV commonly used in RHEED.¹⁸ The 3LM settings for the last condenser lens and intermediate lenses differ from HD presets (Table I). The last condenser lens was excited near its maximum in HD mode to place the incident beam cross over near the exit of the condenser lens, thus, to provide a large distance between the specimen and the cross over. This large distance multiplied by the magnification factor of the intermediate lens and the projection lens combination is the effective sample to detector distance, L_{eff} , which is set at 40 meters for this work. Since the cross over is located above the specimen and the resulted illumination is a divergent type, the scattering intensity was rather weak. In the 3LM mode used in this work the last condenser lens was under-focused and the cross over was located beneath the specimen such that the specimen illumination beam was convergent and hence enhanced the e-beam flux on the sample. The first intermediate lens was adjusted to enable its focal point to reach the object plane of the second intermediate lens. The rest of the intermediate lenses were set to bring the scattering image at the back focal plane of the first intermediate lens to the object plane of the projection lens (PL) with a desirable magnification factor. A condenser aperture of 100 μm was the only aperture used.

In the HD mode, electrons were diffracted off a carbon film with periodic 463 nm spaced square pattern grooves in a transmission geometry for a L_{eff} value of 40 meters. This sample was provided by JEOL for its HD mode calibration,²³ it is also commonly available from other sources including suppliers of electron microscope accessories. The TEM lens settings of the HD mode were listed in Table 1. With the carbon film specimen in place we switched the operation mode to a 3LM mode to achieve a L_{eff} of 40 meters in transmission geometry (Figure 1b). The distance between the diffraction spots was the metric to assure that the desired L_{eff} of 40 meters was achieved in 3LM. The lens settings of the 3LM mode was also listed in Table 1 and the diffraction patterns of the carbon film with 463 nm square grids obtained via 3LM mode was given in Fig. 2a. Projection optics allow diffraction patterns to be measured with high angular resolution even using electrons with wavelengths around 3.7 pm (100 keV). The distance between the adjacent diffraction spots corresponds to an angular range of 2.3×10^{-4} degrees.

In the POC experiment, we performed RSAES on an electro-deposited Cu line grating on Si substrate with a pitch of 200 nm a height of 50 nm and line width of 70 nm. Using an alignment procedure similar to that used previously for REM¹⁷, the sample was placed on a modified grid which was in turn placed on a double-tilt sample holder tilted a fraction of a degree away from the TEM column while keeping the copper lines parallel to the incident electron beam (Figure 1 c). The lens settings used for the 3LM mode with a L_{eff} of 40 meters were used for the POC experiment. However, the exact value of L_{eff} in the POC experiment is not known since the sample position of the HD experiments was not exactly maintained for the POC experiment. A modified double tilt sample holder was used for the POC work. In addition, the reflected electrons did not pass through the center of the intermediate/projection lenses, severe distortions are expected in the observed RSAES patterns given in Fig. 2b. Bright reflection striping was consistent with diffraction from the Cu-line grating. Stripe intensities oscillated along their length demonstrating sensitivity to 3D structure. The bending of the diffraction stripes results from the oblique angle of the sample normal and the detector plane with perhaps additional distortion coming from the unorthodox use of the projecting optics without any apertures after the specimen.

The incident beam footprint was analyzed using a minor change of the intermediate lens setting to obtain an image of the of the sample area that contributed to the RSAES patterns. Ten gratings were clearly visible indicating that the incident electron beam footprint had a width of 2.0 μm . Using 0.1° as representative of the glancing angle, the corresponding length of the footprint would be 1200 μm . For smaller gratings, a smaller condenser aperture could be used, for example one with a diameter of 5 μm instead of 100 μm would give an estimated footprint of dimensions of 0.10 μm by 60 μm .

After the proof-of-concept experiment, simulations of the desired RSAES configuration were performed but without the complication of electron optics (Figure 1d). To simulate the strong electron-material interactions, the time-independent Schrödinger's equation was solved numerically using a complex potential for Si, $U_{si} = (-12.1 - 1.9i)$ eV, with the real part equal to the Si mean inner potential,¹⁵ and the imaginary part introducing finite quantum coherence due to inelastic collisions. To simplify this demonstration, and remain consistent with the experiment, a source focused well below the scattering region, 5 cm, was simulated.

The wave equation was solved taking advantage of the grating periodicity and using finite differences,

$$\psi(x, y, z) = \sum_n c_n(z) e^{i(k_x + g_n)x}. \quad (1)$$

A little different from the horizontal multislice method,^{18,24} the Fourier coefficients, $c_n(z)$, were found directly instead of using wave source terms. k_x is the transverse component of the incident wave-vector, and g_n are the scattering wavenumbers. Radiative boundary conditions were applied at the top and bottom of the simulation cell.

$$(\partial_z - i\Gamma_n)c_n(z)|_{z=z_{\text{top}}} = -2i\Gamma_n c_n^I, \text{ and} \quad (2)$$

$$\left(\partial_z + i\sqrt{\Gamma_n^2 - V_{Si}}\right)c_n(z)|_{z=z_{\text{bottom}}} = 0$$

where $\Gamma_n = \sqrt{k_z^2 - g_n^2 - 2k_x g_n}$, k_z is the vertical component of the incident wave, $V_{Si} = (2m_e/\hbar^2)U_{Si}$, $c_n^I = 1$ if $n = 0$ and zero otherwise, and $g_0 = 0$ rad/Å. The incident probability current density normal to the $x - y$ plane is $(\hbar/m_e)\Gamma_0|c_0^I|^2$. The scattered probability current is $(\hbar/m_e)\Gamma_n|c_n(z_{\text{top}}) - c_n^I|^2$ when Γ_n is real. The scattering from any incident wave function was constructed using superposition and reciprocity²⁵. Numerical calculations were performed using Mathematica, v.10.²³ with 65 total scattering vectors and $z = 0.0700$ nm.

Figure 3a shows the reflection diffraction pattern (RSAEP) resulting from a convergent spherical wave. Palm frond-like patterns reveal the pitch of the simulated grating. These intensity “ridges” have the shape of Kikuchi bands similar to what has been observed in RHEED¹⁸. Finer intensity undulations reveal more information about the grating geometry but are difficult to interpret by inspection. Collimating the electron beam produces a different RSAEP (Figure 3b). Creating a composite RSAEP from different elevations of the collimated source or using a vertical slit aperture produces the RSAEP in Figure 3c that can be more easily interpreted. Vertical stripe spacing is inversely proportional to the grating pitch, while vertical intensity oscillations along the stripes are inversely related to the grating height. The RSAEP is similar to the structure factors used for RHEED oscillations to estimate monolayer completion during molecular beam epitaxy¹⁸. All RSAEPs are shown in linear-scale revealing very strong reflectivities with peak intensities, greater than 10 % and usually much higher. The multimedia view demonstrates how RSAEPs are sensitive to changes in grating shape, height, pitch and line-width.

The proof-of-concept experiment and simulations show that RSAES can access a wealth of geometrical information available when incident and scattered angles are less than 1° enabling precise dimensional measurement of sub – 10 nm three-dimensional features. Scattered intensities are strong enough to be measured on a linear-scale, and sampling areas can be easily tuned using electron optics. The simulations not only show RSAES’ potential, but also demonstrate that the tools necessary to quantitatively analyze diffraction patterns are available now, using either the multiple scattering (dynamic) simulation method used here or other methods.^{18,24,26–28}. The use of electron optics in focusing incident beams and defocusing scattered beams combined with the high scattering cross-sections of electrons allows for an unprecedented combination of measurement signal, angular resolution and statistical measures of sample geometry, thereby, meeting a great many requirements of future nanotechnology.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

Schematics of RSAES and related experiments and simulations, including electron gun (red), incident electron beam (dark-blue), transmitted or reflected/diffracted electron beam (light-blue), electron optical elements (green), electron detector (white) and sample grating (orange) with opaque substrate (light orange). (a) RSAES apparatus. (b) 3LM mode experiment with carbon grating (black grid). (c) proof-of-concept experiment. (d) simulated geometry.

Figure 2.
Diffraction pattern from (a) 3LM mode in a transmission geometry and (b) POC experiments illustrated in Figures 1b and c.

Figure 3. Simulated diffraction patterns (RSAEPs) (a) from spherical wave-section, (b) from a highly-collimated beam incident at elevation angle, $\theta=0.20^\circ$ and (c) from a composite pattern from vertically scanned collimated beam. Intensity scale is linear. (d) Simulated electron density corresponding to collimated beam in Figure b. Yellow indicates boundary of etched Si grating. The multimedia view shows results from various gratings. (Multimedia View)

Table I

HD presets and 3LM operating current settings.

	HD: 40 m	3LM
CL1	1.92	1.93
CL2	X	X
CL3	2.39	1.43
CAP	100 μm	100 μm
CM	X	X
OB	X	X
OBA	X	X
OBM	X	X
FLAP	X	X
IL1	1.04	0.91
IL2	2.29	3.04
IL3	1.18	1.15
PL	3.36	3.36

X denotes where lens or aperture were not activated or engaged