Dark-Field Scanning Transmission Ion Microscopy via Detection of Forward Scattered Helium Ions with a Microchannel Plate[[1]](#footnote-1)

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**Keywords**: helium ion microscopy (HIM), scanning transmission ion microscopy (STIM), Monte Carlo simulation, Stopping and range of ions in matter (SRIM), Z-contrast imaging

**Abstract**

A microchannel plate was used as an ion sensitive detector in a commercial helium ion microscope for dark-field transmission imaging of nanomaterials, i.e. scanning transmission ion microscopy. In contrast to previous transmission helium ion microscopy approaches that used secondary electron conversion holders, our new approach detects forward scattered helium ions on a dedicated annular shaped ion sensitive detector. Minimum collection angles between 125 mrad and 325 mrad were obtained by varying the distance of the sample from the microchannel plate detector during imaging. Monte Carlo simulations were used to predict detector angular ranges at which dark-field images with atomic number contrast could be obtained. We demonstrate atomic number contrast imaging *via* scanning transmission ion imaging of silica-coated gold nanoparticles and magnetite nanoparticles. While the resolution of scanning transmission ion microscopy is known to be degraded by beam broadening in the substrate, we imaged magnetite nanoparticles with high contrast on a relatively thick silicon nitride substrate. We expect this new approach to annular dark-field scanning transmission ion microscopy will open avenues for more quantitative ion imaging techniques and advance fundamental understanding of underlying ion scattering mechanisms leading to image formation.

**Introduction**

Helium ion microscopy (HIM) is conventionally a surface imaging technique known for offering high spatial resolution (), large depth of field, and the ability to image non-conductive samples ([Ward, et al., 2006](#_ENREF_12)). The technique is similar in operation to a scanning electron microscope (SEM), except it utilizes a focused probe of helium ions that is rastered over the sample ([Scipioni, et al., 2009](#_ENREF_11)). Helium ions generate primarily low-energy secondary electrons (SEs) that only escape from the top nanometer of the sample surface, making SE images formed in the HIM extremely sensitive to surface topology ([Bell, 2009](#_ENREF_1)). However, for thin samples a majority of incident helium ions transmit through the sample, either forward scattering or transmitting unscattered. Ion-sample interactions are more complex compared to electron-sample interactions due to the possibility for incident helium ions to change ionization state due to scattering, e.g. from singly to doubly ionized or from singly ionized to a neutral state. Like electrons, ions may also sputter the sample material, resulting in forward scattering of sample atoms. Using Monte Carlo simulations, Notte et al. showed that 50% of 40 kV helium ions transmitted through 100 nm cubic magnesium oxide (MgO) nanoparticles, allowing scanning transmission ion microscopy (STIM) with an expected resolution of 3 nm due to beam broadening ([Notte, et al., 2010](#_ENREF_10)). Due to the reduced interaction volume, scanning transmission microscopy modes, such as STEM and STEM-in-SEM ([Klein, et al., 2012](#_ENREF_7)), often provide higher resolution and additional information about nanomaterial samples compared to surface sensitive techniques ([D'Alfonso, et al., 2013](#_ENREF_3)). Scipioni et al. reported various STIM modes including bright-field (BF) and annular dark-field (ADF) imaging, as well as exit-surface SE imaging that is similar to BF imaging ([Scipioni, et al., 2009](#_ENREF_11)). Thickness fringes and dislocations were observed in MgO nanocrystals imaged with 40 kV BF STIM ([Notte, et al., 2010](#_ENREF_10)). Hall et al. determined the thickness of freestanding silicon nitride membranes using pixel intensities from BF STIM images ([Hall, 2013](#_ENREF_4)). To date, STIM imaging has relied on conversion sample holders that utilize a polished metal surface coated with gold that converts transmitted helium ions into SEs ([Hall, 2013](#_ENREF_4)). The polished surface is angled towards an Everhart-Thornley detector where the SEs are collected to form an image ([Hall, 2013](#_ENREF_4)). A hole can be placed in the polished surface on the optic axis to allow the unscattered ions to pass through to enable ADF imaging ([Notte, et al., 2010](#_ENREF_10)). While not detailed in the publications, the sample holder used by Scipioni et al. ([Scipioni, et al., 2009](#_ENREF_11" \o "Scipioni, 2009 #11)) and Notte et al. ([Notte, et al., 2010](#_ENREF_10" \o "Notte, 2010 #8)) was a custom built multisurface SE conversion sample holder capable of BF and ADF imaging by adjusting a voltage bias on a mesh cage surrounding the sample holder. However, the indirect nature of the STIM image formation process when using an SE conversion holder limits fundamental understanding of the ion scattering processes that govern contrast mechanisms. While this approach has the advantage of not requiring a separate detector below the sample, many assumptions must be made about the conversion of transmitted ions into SEs ([Hall, 2013](#_ENREF_4)). For instance, one must assume that the SE emission per transmitted ion on the polished metal surface does not vary with position ([Hall, 2013](#_ENREF_4)). While SE conversion holders have been shown to provide useful STIM image contrast, detection of transmitted helium ions with a dedicated detector would allow for more faithful interpretation of STIM images, which could open new avenues for quantitative STIM imaging, such as standard-less atom counting ([LeBeau, et al., 2010](#_ENREF_8)) and mass-thickness determination ([Loferer-Krossbacher, et al., 1998](#_ENREF_9)) that have been demonstrated for high angle annular dark field (HAADF) STEM and TEM imaging. Helium ions offer an advantage over electrons for quantitative imaging because diffraction contrast is not significant in ADF STIM images due to their small wavelength compared to electrons.

Here we demonstrate ADF STIM imaging of nanoparticles on thin supports *via* detection of forward scattered helium ions using an ion sensitive microchannel plate (MCP) detector. This approach differs from previous STIM experiments in several ways: (1) Image formation does not rely on conversion of transmitted helium ions into SEs, (2) dedicated detection eliminates the need for assumptions to be made about the image formation process, and (3) the detector is retractable and separate from the sample, which allows for control over the ADF collection angles during imaging. Compared to backscattered HIM imaging, ADF STIM is expected to show similar atomic number contrast but with higher spatial resolution. ADF STIM represents a superior imaging technique for thin samples, such as nanoparticles and 2D materials, where forward scattering of helium ions is strongly preferred compared to backscattering. Our simulations indicate that for the nanoparticle samples imaged in this article, > 90% of the helium ions forward scatter, while only ~5 % or less backscatter. Due to this propensity for forward scattering, ADF STIM enables imaging of thin samples with less ion dosage compared to backscatter imaging, which will reduce the amount of sample damage. Higher spatial resolution is expected for ADF STIM compared to backscatter imaging due to the reduced interaction volume.

**Materials and Methods**

Imaging was performed on a Zeiss Orion Plus HIM operated at 25 kV or 30 kV accelerating voltage; the beam current was typically 3 pA to 10 pA.[[2]](#footnote-2) SE conversion ADF STIM imaging wasSE The HIM is equipped with an annular shaped MCP that is mounted as a backscattered detector a few millimeters below the pole piece (Figure 1a). We inverted the MCP and placed it ~10 mm below the sample to enable STIM imaging. The detector has a 2 mm diameter hole in the center to allow unscattered transmitted ions to pass through. The active area of the MCP detector has an inner annulus diameter of 4 mm and outer annulus diameter of 15 mm. A custom-built sample holder was used to cantilever a thin sample between the pole piece and MCP ([Holm & Keller, 2015](#_ENREF_5)). The sample-to-detector distance and the radii of the inner and outer annulus of the active detector area determined the minimum and maximum collection angles of the annular detector (Figures 1b, and 1c). In this configuration, ions forward scattered to angles smaller than the minimum collection angle or larger than the maximum were undetected, while ions scattered to collection angles defined by the active detector area were integrated serially to form the ADF STIM image. The accessible minimum collection angles () for this detector configuration ranged from approximately 125 mrad to 325 mrad, with corresponding maximum collection angles () between 440 mrad and 900 mrad (Figure 1c). The size of the angular range () increases as a function of minimum collection angle. The accessible minimum and maximum collection angles can be extended to larger angles in the future by decreasing the total height of the cantilever sample holder or decreasing the distance between the MCP and the pole piece. The current MCP detector configuration does not allow for BF imaging as there is no way to define this collection angle range on the annular shaped detector. Images were acquired with a size of 512 x 512 pixels, line integration of 4 lines, and a pixel dwell time of 100 μs. Imaging was performed on a field of view larger than 1.2 μm to avoid ion beam damage to the sample. For a smaller field of view, increased ion doses etched the sample during imaging. Larger beam currents than used for conventional SE imaging in the HIM (typically sub-pA) were necessary to provide adequate signal for ADF STIM imaging. Future design of a more sensitive MCP detector with higher gain will enable imaging of a smaller field of view with lower beam current. The sample-to-detector distance and reported collection angles were determined by subtracting the working distance of an in-focus image of the sample from an in-focus image of the detector surface. The error associated with this collection angle measurement due to the uncertainty in the working distance of the focal plane (stemming from the relatively large depth of field (single to tens of microns)) is only on the order of hundredths of a percent. However, as the working distance is only reported to tenths of a millimeter by the software, this introduces an error of approximately 5% in the reported collection angles.

Nanomaterial samples were deposited on transmission electron microscopy (TEM) grids by drop casting aqueous suspensions. Magnetite nanoparticles (Ocean Nanotech, USA) with a nominal diameter of 30 nm ± 2.5 nm were deposited onto 30 nm thick silicon nitride (SiN) membranes (Norcada, Canada). Silica-coated gold nanoparticles (Nanocomposix, USA) with a gold core diameter of 20 nm ± 3 nm and silica shell thickness of 20 nm ± 5 nm were prepared on lacy carbon supported ultrathin carbon TEM grids (01824, Ted Pella, USA).

Monte Carlo simulations of helium ion trajectories were performed using the freely available software SRIM (available at www.srim.org). The software is based on calculations of the stopping and range of ions in matter (SRIM) ([Zeigler, et al., 1985](#_ENREF_14)). The simulations assumed a point source of helium ions incident on the surface of a uniform thin film of material. The “ion distribution and quick damage calculation” option was used to compute the trajectories of 10000 helium ions per simulation. The angular trajectories of forward scattered ions were calculated from the exit-surface position vectors of the ions. Detected ion yields (Figure 2) were calculated by integrating the total number of ions forward scattered onto a virtual ADF detector with an angular range defined by the minimum and maximum detector collection angles, and then dividing this by the incident number of helium ions on the material.

**Results and Discussion**

Monte Carlo simulations were performed to determine the ADF minimum collection angles at which we could obtain atomic number contrast ADF STIM images. We simulated the trajectories of 10000 helium ions with initial energies of 30 keV (Figure 2a) and 25 keV (Figure 2c) through uniform films of materials with atomic numbers and mass-thicknesses corresponding to those of the two nanoparticles and substrates imaged. The simulated film thicknesses for the spherical gold and magnetite nanoparticles were taken to be the nominal particle diameters. The simulation for the 20 nm gold nanoparticles also included two 20 nm silica films sandwiching the gold layer to more accurately reproduce the core-shell configuration. The ion beam interacts with a cross-section of the silica shell that is almost twice as thick as the nominal shell thickness of 20 nm, so we simulated the silica layer to be , which is equal to the path length the helium ions traverse through the silica shell where the core and shell meet.

Figures 2a and 2c show the detected ion yields as a function of the ADF detector angular range for the samples in Figures 3 and 4. In these plots, the y-axis corresponds to the integrated number of ions forward scattered to angles encompassed by a virtual ADF detector with an angular range defined by a minimum (, lower x-axis label) and maximum collection angle (, upper x-axis label). The maximum collection angles are only shown in Figure 2a for clarity, but the other plots in Figure 2 have the same corresponding maximum collection angles. The total angular range for each virtual ADF detector was set by the inner and out radii of the experimental MCP detector (See Materials and Methods) and the sample-to-detector distance, which was varied to change the angular range of the virtual ADF detector. For example in Figure 2a, for a virtual ADF detector with an angular range of to approximately half (yield = 0.5) of the incident helium ions were forward scattered onto the virtual ADF detector. The yield is proportional to the signal detected by the ADF detector and can therefore be used to predict image contrast as a function of angular range.

Figure 2a shows the resulting detected ion yields for 3 nm carbon, 20 nm gold, and 20 nm silica, which correspond to the substrate, core, and shell materials of the silica-coated gold nanoparticles shown in Figure 3. Ion scattering through the carbon layer, which had the lowest mass-thickness and atomic number, resulted in minimal ion yields for ADF detectors with and an ion yield that peaked for a virtual ADF detector with . Ion scattering in the silica layer produced a larger detected yield over a broader range of detector collection angles with a maximum yield for an ADF detector with , while scattering in the gold layer resulted in an ion yield maximum for a detector with . This trend suggests that scattering in the Monte Carlo simulations is similar to Rutherford elastic scattering, i.e. mass-thickness contrast, where elements with larger densities and higher atomic numbers forward scatter ions to larger angles.

Atomic number contrast images are formed at ADF detector minimum collection angles where the detected ion yield increases proportional to the material’s atomic number. The dashed line in Figure 2a shows that this regime corresponds to minimum detector collection angles of . T, so we calculated the contrast for several silica shell thicknesses with a constant gold core size In general the contrast increased as a function of and decreased with increasing silica shell thickness (Figure 2b). The minimum collection angle above which the contrast was positive corresponds to the regime where atomic number contrast imaging is expected (inset Figure 2b). The inset shows that the above which atomic number contrast is predicted increases approximately linearly with silica shell thickness.

Figure 2c shows the detected ion yield for 25 keV helium ions incident on 30 nm SiN and 30 nm Fe3O4 (cf. images in Figure 4). The ion yield of the Fe3O4 includes the underlying SiN substrate because the materials are similar in mass-thickness. Due to the higher density and average atomic number of Fe3O4 compared to SiN, the simulations predicted that atomic number contrast images can be obtained for virtual ADF detectors with minimum detector collection angles of . Figure 2d again shows that the simulated contrast increases as a function of .

For comparison, an ADF STIM image of silica-coated gold nanoparticles on a TEM grid acquired using the SE conversion holder is shown in Figure 3a. The TEM sample was mounted ~15 mm away from a polished gold-coated metal surface that was angled towards the Everhart-Thornley detector. We removed the BF angle limiting aperture cone from the holder and placed a 2 mm diameter metal disk on the optic axis 3 mm below the sample to block the unscattered helium ion beam from the conversion surface, allowing for ADF imaging. The metal disk blocked all the unscattered and forward scattered helium ions for angles of . A stage bias of +200 V was applied to eliminate contrast due to SE and higher energy Auger electron emission from the sample surface. The ADF STIM image acquired with the conversion holder showed atomic number contrast, as the gold cores were more intense than the silica shells. The intensities of the silica shells and lacy carbon were approximately the same, but the edges of the lacy carbon had higher intensity compared to the center. There were two disadvantages to using this conversion holder. Applying a positive bias to the sample was necessary to limit the contribution of sample surface SE and Auger emission to the image contrast, but this also reduced SE emission on the conversion surface, which created an inherently noisy ADF STIM image. Secondly, the sample holder and conversion plate are a single piece, so the ADF collection angles could only be changed by removing the sample and changing the size of the beam blocking metal disk.

ADF STIM imaging *via* a dedicated MCP ion detector yielded images with no SE contribution and allowed for imaging with variable detector collection angles. Imaging with the MCP only required inverting the backscattered detector that already had the desired annular shape, while ADF imaging with the SE conversion holder required custom fabrication of millimeter sized beam blocks. Figure 3b shows a representative ADF STIM image of silica-coated gold nanoparticles acquired using the MCP detector with a minimum collection angle of (. The image clearly demonstrates atomic number contrast, as the highest intensity portions of the image are the gold cores, followed by the silica shells, and finally the lacy carbon and the thin carbon had the lowest intensity. Figure 3c shows a higher magnification image of the core shell nanoparticles showing the high-intensity gold cores surrounded by lower-intensity silica shells. The intensity of the gold core was approximately twice that of the silica shell, corresponding to a contrast of , in agreement with the simulated contrast shown in Figure 2b. Figure 3d is an image of the same nanoparticles acquired at a lower minimum collection angle of . The digital brightness and contrast settings were the same for Figures 3c and 3d, allowing for direct comparison of image intensities. The silica shells had higher intensity in Figure 3d, while the gold and carbon had similar intensity compared to Figure 3c. Comparison to the Monte Carlo simulations reveals that the silica intensity changes in the images were due to the large change in the yield of silica over the range of to (Figure 2a). On the other hand, the changes of the gold and carbon yields over the range of to were relatively small, consistent with their similar intensities between the two experimental images. Additionally, in Figure 3d the silica shell thickness had a distinct effect on the contrast between the silica and gold. The gold cores in particles with relatively thin silica shells (to) were resolved with atomic number contrast; however, there was minimal contrast between the gold core and silica shell for slightly thicker silica shells (to, green arrow). Particles with larger silica shells () showed reverse contrast, meaning the silica shell had a higher intensity than the gold core (red arrow). This imaging artifact was recently identified for low energy (20 keV to 30 keV) ADF STEM imaging, and was attributed to a competition between the mass-thickness contrast of the two materials ([Brodusch, et al., 2013](#_ENREF_2); [Holm & Keller, 2015](#_ENREF_5); [Woehl & Keller, 2015](#_ENREF_13)). In this case, the smallest nanoparticles (to silica shell) showed atomic number contrast because the gold core scattered more ions to the ADF detector than the silica shell, as indicated by the detected ion yields and simulated contrast in Figures 2a and 2b. However, the slightly larger nanoparticles (to silica shell) showed minimal contrast between gold and silica because the larger silica shell and small gold core scattered a similar number of ions to the ADF detector (cf. Figure 2b). The largest nanoparticles ( silica shell) showed reverse contrast because the thick silica shell scattered more ions to the detector than the gold core. The Monte Carlo simulations of the gold core and silica shell clearly shows that for 40 nm thick silica shells, the image contrast reverses for virtual ADF detectors with (Figure 2b and inset). While there was not complete quantitative agreement between the simulated and experimental contrast for the various thicknesses of the silica shells, the overall trend of decreasing contrast and eventual contrast reversal with increasing shell thickness predicted by the simulations was consistent with the experimental images. The quantitative disagreement was likely due to the simplified 1D simulation geometries compared to the 3D geometry of the nanoparticles. Contrast reversal was the most prevalent artifact observed during ADF STIM imaging and was eliminated by increasing the minimum collection angle of the ADF detector, as exemplified by Figures 3c and 3d. Monte Carlo simulations were instrumental in identifying the atomic number contrast collection angle regimes to avoid this artifact.

To test the suitability of STIM for obtaining ADF images on thicker substrates, we imaged 30 nm magnetite (Fe3O4) nanoparticles supported on a 30 nm amorphous SiN membrane (Figure 4). Individual nanoparticles were easily resolved with sufficient contrast compared to the substrate. The images in Figures 4a and 4b were acquired with ADF minimum collection angles of ( and (. The STIM image acquired with a lower minimum collection angle (Figure 4a) had a higher background signal resulting from the thick SiN substrate, and lower image contrast compared to the image with the higher minimum collection angle (Figure 4b). Both of these ADF images were obtained with minimum collection angles larger than that required for atomic number contrast as predicted by Monte Carlo simulations (Figures 2c and 2d). The intensity of the magnetite nanoparticles was approximately twice that of the SiN substrate in the image acquired with (), while increasing the minimum collection angle to increased the intensity ratio in the image to . The approximate intensity ratios and increase in contrast with increasing minimum collection angle were consistent with the Monte Carlo simulations in Figure 2c and 2d. Together, the simulations and experiments indicate that increasing minimum collection angle can be used to decrease the background signal and increase contrast in ADF STIM images of samples on thick substrates.

Preliminary measurements of the spatial resolution for ADF STIM imaging using the SMART macro ([Joy, et al., 2000](#_ENREF_6" \o "Joy, 2000 #16)) showed an edge resolution of for a sputtered gold on carbon TEM sample. The magnetite nanoparticles on SiN were resolved with an edge resolution of , with the decrease in resolution likely due to beam broadening in the thick substrate. We do not expect the spatial resolution to change significantly with the sample-to-detector distance and minimum collection angle of the ADF detector because the convergence angle of the helium ion beam is approximately 0.5 mrad, so beam divergence and beam broadening will not be significantly affected by the working distance. The measured edge resolution is consistent with previous Monte Carlo predictions for BF STIM imaging that predicted a resolution of 3 nm for a 100 nm MgO nanocube ([Notte, et al., 2010](#_ENREF_10" \o "Notte, 2010 #8)). Future work will focus on systematically investigating the effects of sample thickness, atomic number, and substrate thickness on image resolution.

**Conclusions**

An ion sensitive MCP was utilized as a dedicated transmission detector in a commercial helium ion microscope to perform ADF STIM imaging. Our new approach differs from previous STIM imaging efforts in that (1) we detect forward scattered helium ions using a dedicated ion sensitive detector and (2) the ADF collection angle can be continuously varied during imaging. We demonstrated atomic number contrast by imaging silica-coated gold nanoparticles, where the core-shell structure was clearly resolved. Though STIM image resolution is limited by beam broadening, imaging of magnetite nanoparticles on a thick SiN membrane demonstrated adequate resolution and atomic number contrast. Contrast changes in both samples were observed with changes in minimum detector collection angle, which correlated well with SRIM simulations. This detector modification could enable STIM imaging on any HIM equipped with an MCP in the backscatter configuration; future applications of this technique include end point detection for ion milling, sub-nanometer resolution imaging, and diffraction-based (i.e. ion blocking) measurements. Sub-nanometer resolution imaging could be realized for 2D materials where beam broadening does not reduce spatial resolution, as the helium ion beam has a size on the order of several angstroms. This first demonstration of ADF STIM imaging *via* ion detection with a dedicated ion detector is a step towards realizing a dedicated scanning transmission helium ion microscope.

**Acknowledgments**

We thank Aric Sanders for useful discussions on microchannel plates and detector design in the helium ion microscope, John Notte for useful discussions on forward scattered ion detection in the helium ion microscope, and Jason Holm for the sample holder. T.J.W. and R.M.W. thank the National Research Council Research Associate Program for funding.

**References**

Bell, D.C. (2009). Contrast Mechanisms and Image Formation in Helium Ion Microscopy. Microscopy and Microanalysis **15**(2), 147-153.

Brodusch, N., Demers, H. & Gauvin, R. (2013). Dark-Field Imaging of Thin Specimens with a Forescatter Electron Detector at Low Accelerating Voltage. Microscopy and Microanalysis **19**(6), 1688-1697.

D'Alfonso, A.J., Forbes, B.D. & Allen, L.J. (2013). The interaction of a nanoscale coherent helium-ion probe with a crystal. Ultramicroscopy **134**, 18-22.

Hall, A.R. (2013). In Situ Thickness Assessment During Ion Milling of a Free-Standing Membrane Using Transmission Helium Ion Microscopy. Microscopy and Microanalysis **19**(3), 740-744.

Holm, J. & Keller, R.R. (2015). Analytical Transmission Scanning Electron Microscopy: Extending the Capabilities of a Conventional SEM Using an Off-the-Shelf Transmission Detector. Microscopy and Microanalysis **Supp S2**.

Joy, D., Ko, Y. & Hwu, J. (2000). Metrics of resolution and performance for CD-SEMs. Proceedings of SPIE **3998**.

Klein, T., Buhr, E. & Frase, C.G. (2012). TSEM: A Review of Scanning Electron Microscopy in Transmission Mode and Its Applications. In *Advances in Imaging and Electron Physics, Vol 171*, Hawkes, P. W. (Ed.), pp. 297-356.

LeBeau, J.M., Findlay, S.D., Allen, L.J. & Stemmer, S. (2010). Standardless Atom Counting in Scanning Transmission Electron Microscopy. Nano Letters **10**(11), 4405-4408.

Loferer-Krossbacher, M., Klima, J. & Psenner, R. (1998). Determination of bacterial cell dry mass by transmission electron microscopy and densitometric image analysis. Applied and Environmental Microbiology **64**(2), 688-694.

Notte, J., Hill, R., McVey, S.M., Ramachandra, R., Griffin, B. & Joy, D. (2010). Diffraction Imaging in a He+ Ion Beam Scanning Transmission Microscope. Microscopy and Microanalysis **16**(5), 599-603.

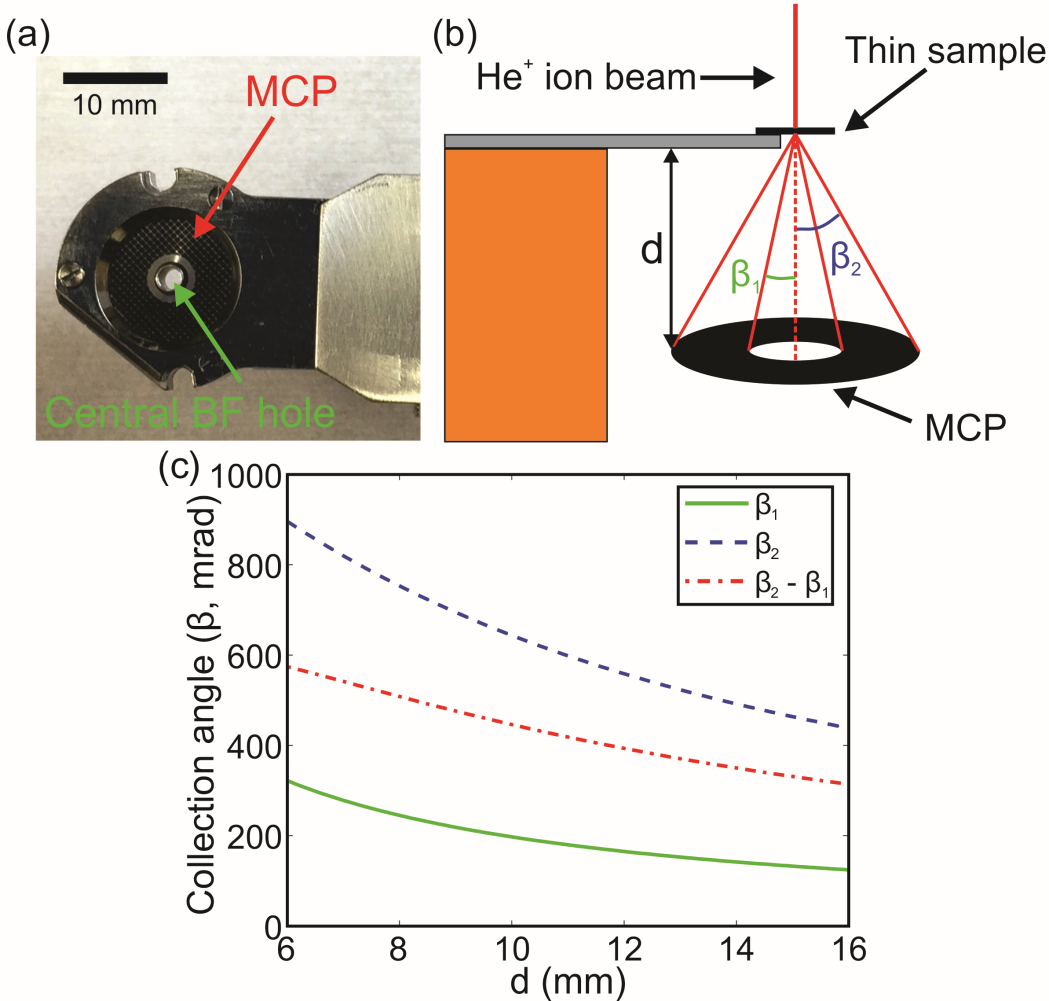
Scipioni, L., Sanford, C.A., Notte, J., Thompson, B. & McVey, S. (2009). Understanding imaging modes in the helium ion microscope. Journal of Vacuum Science & Technology B **27**(6), 3250-3255.

Ward, B.W., Notte, J.A. & Economou, N.P. (2006). Helium ion microscope: A new tool for nanoscale microscopy and metrology. Journal of Vacuum Science & Technology B **24**(6), 2871-2874.

Woehl, T.J. & Keller, R.R. (2015). Dark-Field Image Contrast in Transmission Scanning Electron Microscopy: Effects of Substrate Thickness and Detector Collection Angle. Ultramicroscopy **In preparation**.

Zeigler, J.F., Biersack, J.P. & Littmark, U. (1985). *The Stopping and Range of Ions in Solids*. New York: Pergamon.

**Figures and Figure captions**

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**Figure 1.** (a) Optical image of the MCP with the active detector area denoted in red and the center hole for the unscattered beam to pass through denoted in green. (b) Schematic representation of the ADF STIM imaging geometry showing a thin sample cantilevered between the incident ion beam and the annular MCP detector. The distance, , determined the minimum and maximum collection angles of the detector. (c) Accessible minimum and maximum collection angles and their difference for the annular MCP detector.

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**Figure 2.** (a) Monte Carlo simulated detected ion yield as a function of the minimum and maximum ADF detector collection angle (ADF angular range) for the silica coated gold nanoparticles in Figure 3. Each data point represents the fraction of 10000 simulated helium ions incident on the sample that are forward scattered into a virtual ADF detector with an angular collection range between and . (b) Simulated contrast between the gold core and silica shell as a function of ADF angular range for various silica shell thicknesses. The inset shows the minimum above which atomic number contrast is expected, as a function of silica shell thickness. (c) Simulated detected ion yield as a function of the ADF angular range for the magnetite nanoparticles on a silicon nitride substrate shown in Figure 4. (d) Simulated contrast between the magnetite and silicon nitride as a function of ADF angular range.

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**Figure 3.** ADF STIM images of silica coated gold nanoparticles supported on ultrathin carbon. (a) Cropped ADF STIM image of gold nanoparticles acquired on the Everhart-Thornley detector using the SE conversion transmission holder. The minimum collection angle for the image was . (b-d) Cropped ADF STIM images acquired *via* detection of forward scattered helium ions using an ion sensitive MCP. Images were acquired with minimum collection angles of (b,c) ( and (d) . The scale bars in each image are 200 nm. The red arrow in (d) denotes contrast reversal in the larger core-shell nanoparticles while the green arrow denotes nanoparticles with no core-shell contrast.

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**Figure 4.** ADFSTIM images of 30 nm diameter magnetite nanoparticles supported on 30 nm SiN. The images were acquired at 25 kV accelerating voltage with detector minimum collection angles of (a) () and (b) ). The scale bars in each image are 200 nm.

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