



This open access document is posted as a preprint in the Beilstein Archives at <https://doi.org/10.3762/bxiv.2020.101.v1> and is considered to be an early communication for feedback before peer review. Before citing this document, please check if a final, peer-reviewed version has been published.

This document is not formatted, has not undergone copyediting or typesetting, and may contain errors, unsubstantiated scientific claims or preliminary data.

Preprint Title Scanning transmission imaging in the helium ion microscope using a microchannel plate with a delay line detector

Authors Eduardo Serralta, Nico Klingner, Olivier De Castro, Michael Mousley, Santhana Eswara, Serge Duarte Pinto, Tom Wirtz and Gregor Hlawacek

Publication Date 09 Sep 2020

Article Type Full Research Paper

ORCID® iDs Eduardo Serralta - <https://orcid.org/0000-0001-6254-022X>; Nico Klingner - <https://orcid.org/0000-0001-9539-5874>; Olivier De Castro - <https://orcid.org/0000-0001-9968-6695>; Santhana Eswara - <https://orcid.org/0000-0003-4151-2304>; Gregor Hlawacek - <https://orcid.org/0000-0001-7192-716X>

License and Terms: This document is copyright 2020 the Author(s); licensee Beilstein-Institut.

This is an open access publication under the terms of the Creative Commons Attribution License (<https://creativecommons.org/licenses/by/4.0>). Please note that the reuse, redistribution and reproduction in particular requires that the author(s) and source are credited.

The license is subject to the Beilstein Archives terms and conditions: <https://www.beilstein-archives.org/xiv/terms>.

The definitive version of this work can be found at <https://doi.org/10.3762/bxiv.2020.101.v1>

1 Scanning transmission imaging in the helium ion microscope using a 2 microchannel plate with a delay line detector

3 Eduardo Serralta*^{1,2}, Nico Klingner¹, Olivier De Castro³, Michael Mousley³, Santhana Eswara³,
4 Serge Duarte Pinto⁴, Tom Wirtz³ and Gregor Hlawacek¹

5 Address: ¹Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-
6 Rossendorf, Bautzner Landstr. 400, 01328 Dresden, Germany; ²Technische Universität Dresden,
7 Dresden 01069, Germany; ³Advanced Instrumentation for Nano-Analytics (AINA), MRT Depart-
8 ment, Luxembourg Institute of Science and Technology, 41, rue du Brill, L-4422, Belvaux, Luxem-
9 bourg and ⁴Photonis Netherlands B.V., Dwazziewegen 2, 9301 ZR Roden, The Netherlands

10 Email: Eduardo Serralta - e.serralta@hzdr.de

11 * Corresponding author

12 Abstract

13 A detection system based on a microchannel plate with a delay line readout structure has been de-
14 veloped to perform scanning transmission ion microscopy (STIM) in the helium ion microscope
15 (HIM). This system is an improvement over other existing approaches since it combines the infor-
16 mation of the scanning beam position on the sample with the position (scattering angle) and time
17 of the transmission events. Various imaging modes such as bright and dark field or the direct image
18 of the transmitted signal can be created by post-processing the collected STIM data. Furthermore,
19 the detector has high spatial and time resolution, is sensitive to both ions and neutral particles over
20 a wide energy range, and shows robustness against ion beam-induced damage. A special in-vacuum
21 movable support gives the possibility of moving the detector vertically, placing the detector closer
22 to the sample for the detection of high-angle scattering events, or moving it down to increase the
23 angular resolution and distance for time-of-flight measurements. With this new system, we show
24 composition-dependent contrast for amorphous materials and the contrast difference between small

25 and high angle scattering signals. We also detect channeling related contrast on polycrystalline
26 silicon, thallium chloride nanocrystals, and single crystalline silicon by comparing the signal trans-
27 mitted at different directions for the same data set.

28 **Keywords**

29 helium ion microscopy; scanning transmission ion microscopy; delay line detector; channeling;
30 bright field; dark field

31 **Introduction**

32 The helium ion microscope (HIM) is an instrument that has already proven its value for high-
33 resolution imaging, compositional analysis, nanofabrication, and materials modification [1,2]. It
34 generates a focused helium (or neon) ion beam with sub-nanometer spot size and rasters it across
35 the sample. The beam can be used for both imaging and modification of samples at the nanometer
36 scale. The standard and most widely used imaging mode in the HIM is using an Everhart-Thornley
37 detector (ET) [3] for collecting secondary electrons (SE) emitted from the top surface of the sample
38 which carry mainly topographic information [4].

39 Other detectors and signals have been used to expand the capabilities of the HIM. Imaging with
40 back-scattered particles [5,6] can add compositional information and reveal buried structures [7].
41 Ionoluminescence has been studied by detecting the light emitted from the sample during ion
42 bombardment [8-10]. Moreover, compositional analyses using secondary ion mass spectrome-
43 try (SIMS) can be performed in the HIM with lateral resolution in the order of 10 nm [11-14].

44 Transmission mode imaging can further improve the capabilities of the HIM since it is dependent
45 on different contrast mechanisms and gives information on sub-surface features as well. There are
46 several ways of using the transmitted signal to form an image. In bright field (BF) mode, the im-
47 age is produced by mapping the intensity of the beam that has suffered very little, or no, deflection.
48 In dark field (DF) mode, the intensity of the deflected beam is used as the signal. In annular dark
49 field (ADF) mode, the intensity of the transmitted beam at a particular polar angle interval is inte-

50 grated over a complete annulus. Alternatively, the image can be formed using the beam deflected in
51 a polar and azimuthal angular interval.

52 For amorphous materials under perpendicular incidence, the transmitted beam is expected to be
53 scattered symmetrically around the axis of incidence. The average polar angle of scattering de-
54 pends on both the material and the thickness of the sample. Different materials and thickness com-
55 binations create distinct polar angle distributions of scattering producing a contrast similar to the
56 mass-thickness contrast in transmission electron microscopy. In BF mode, the areas of the sample
57 with little, or no, scattering appear as high intensity in the image, and regions of the sample that
58 scatter more than the detector's collection angle will appear as low intensity. In a complementary
59 manner, in ADF mode, the areas of the sample that scatter to the considered angular interval will
60 appear bright in the image, and the areas of the sample with little scattering will appear dark. BF
61 imaging has the advantage of having higher count rates for the same beam current in thin samples.
62 On the other hand, by adjusting the collection angles to fit the maximum of this distribution for a
63 given material and thickness, ADF imaging can enhance the contrast of certain compositional fea-
64 tures of the sample.

65 Crystalline materials can also give rise to additional contrast mechanisms. In crystalline materi-
66 als, the stopping force depends on the orientation of the crystal [15]. In some orientations, the tar-
67 get atoms are aligned in rows or planes, thereby creating easier directions for the penetration of
68 the projectile atom. If the projectile atom reaches the crystal at an angle smaller than the critical
69 angle for such an axial or planar channeling direction, the projectile will be steered along this di-
70 rection and will experience a reduced probability of undergoing large-angle scattering. Hence, it
71 will have a smaller energy loss per distance compared to random directions. This phenomenon is
72 called the channeling effect and has been described for MeV ions in detail in [16]. When compared
73 to a random orientation, the channeling directions also have reduced secondary electron [17], back-
74 scattering and sputter yields. Conversely, the ions have increased range and transmission probabilit-
75 ity in these directions. Channeling contrast in the HIM was demonstrated using SE imaging [17,18]

76 and using the back-scattering signal [19]. The channeling effect in the HIM has also been studied
77 using Monte Carlo [20] and molecular dynamics [21] simulations.

78 Measuring the energy of the transmitted particles would be a novel technique that adds an informa-
79 tion channel to the previously discussed transmission imaging modes. It will provide information
80 on the phenomena occurring during the projectile-target interaction and can increase the signal-to-
81 noise ratio [22]. Since most likely a considerable fraction of the transmitted particles at this energy
82 range is neutral [23,24], magnetic or electrostatic spectrometers cannot be used. Therefore, ion
83 energy-loss spectrometry and energy-resolved imaging require an energy-sensitive detector or a
84 detection system in which time-of-flight (ToF) measurements can be implemented.

85 Likewise, the use of the transmission signal in the HIM for visualizing diffraction patterns is, in
86 theory, possible but has not been reported yet. Diffraction patterns can add information on the crys-
87 tal lattice and orientation. However, this application demands a detector with high spatial resolu-
88 tion taking into consideration the energy range and typical space restrictions in the HIM.

89 In the past, several attempts to utilize the transmission signal in the HIM have been made. One
90 approach is converting the transmitted particles into SEs by positioning a material with high SE
91 yield below the sample and using the ET detector to collect the SEs coming from this material.
92 This method has been used in BF mode for assessing the thickness of milled materials in the mi-
93 croscope [25,26] and has also been implemented for ADF imaging [27,28]. Mass-thickness and
94 thickness fringes contrast have been shown in transmission mode in the HIM using this approach
95 with a combined bright and dark field conversion detector [29]. Another approach using an annular
96 microchannel plate detector was used for investigating gold-silica core-shell nanoparticles in ADF
97 mode [30]. These approaches require a physical aperture to restrict the acceptance angle when per-
98 formed in BF, and physically changing the distance between the sample and the annular detector to
99 adjust the acceptance angle interval when performed in DF. Finally, a position-sensitive detector
100 consisting of a silicon diode array has also been adopted for use in the HIM [31]. Later the same
101 group also studied channeling effects on single crystalline silicon with this detector [32].

102 In this work, we present a new system for comprehensive scanning transmission ion microscopy

103 (STIM) analyses that gives more flexibility to the user than the earlier approaches. We adopted a
104 microchannel plate (MCP) and a delay line readout structure as a position-sensitive detector to be
105 used in the HIM. A special in-vacuum detector support allows mechanically controlling the accep-
106 tance angle during analysis. The resulting system has high spatial resolution and can be positioned
107 to detect polar angles of deflection of up to 19° . The selection of the transmission imaging mode
108 and further tuning of acceptance angles can be done in post-processing. Additionally, ToF resolved
109 recording of the transmission events can be integrated into this system. Here, we use this system
110 to study mass-thickness dependent contrast on amorphous materials and demonstrate transmission
111 channeling contrast using polycrystalline silicon, thallium chloride samples and beam steering in
112 single crystalline silicon.

113 **Experimental**

114 The new STIM detector comprises a stack of two MCPs and a resistive anode layer with a delay
115 line readout structure behind it, as represented in Figure 1c. The combination of MCPs with a de-
116 lay line readout structure as a position-sensitive detector was first implemented for the detection
117 of 1 keV to 15 keV electrons [33], since then, it has been used in many other applications such as
118 in astrophysics [34], transmission electron microscopy [35], and hard x-ray photoelectron spec-
119 troscopy [36].

120 The detector working principle can be summarized as follows: At the front side of the MCP, SEs
121 are produced by the impact of the impinging energetic particles (He ions and atoms). These SEs
122 are drawn into the microchannels due to the applied bias. The number of SEs is multiplied by nu-
123 merous collisions along the way within the channels creating an electron cloud. The electron cloud
124 hits the resistive anode layer in front of the delay lines, and by capacitive coupling, induces sig-
125 nals on the delay line meanders. These signals are collected at the endpoints of each delay line and
126 passed through separate constant fraction discriminators for each delay line. Finally, with a time-
127 to-digital converter, the position of the cascade is computed by comparing the time of arrival of the

128 pulses at the ends of each delay line. This can be performed with ps accuracy and thus makes the
129 detector ideally suited for future ToF applications.

130 In the present form of the detector, we use two 50 mm by 50 mm square active area MCPs stacked
131 and rotated 90° to each other with a gap of 100 μm between them. The first MCP has a magne-
132 sium oxide coating to increase the SE yield [37]. The MCP pores have a diameter of 25 μm and
133 center-to-center spacing of 32 μm. The bias angle of the pores is 16°. The combination of delay
134 line readout performance and MCP characteristics results in a spatial resolution of 47.2 μm in the
135 x-direction and 58.1 μm in the y-direction. The detector has approximately 4 virtual megapixels
136 over its entire area. The MCP front is biased to a potential of approximately -2 kV, while the MCP
137 back is kept at approximately -400 V, relative to the anode which is at ground potential. The delay
138 line readout has a specified maximum count rate of 5 Mcps for randomly distributed events.

139 However, in practice, the count rate is limited by the non-random nature of the transmitted parti-
140 cles. In thin light samples, most of the transmitted particles will hit the center of the detector. The
141 pores of the MCP have an individual recharging time in the order of 0.5 ms, estimated consider-
142 ing the MCP pores as parallel capacitors and resistors. This results in the fact that a single channel
143 can only correctly detect count rates lower than 2 kcps or currents smaller than 0.3 fA. In addition
144 to that, given that the position on the detector is calculated based on the time difference from the
145 signals, when dealing with multiple simultaneous events, there might be multiple solutions for the
146 position and time combinations. Therefore, when trying to compute events that are too close in
147 time *and* position, the delay line structure algorithm might produce imaging artifacts with shapes
148 reflecting these multiple solutions for multi-hit events.

149 The STIM data consists of, for each detected event, the position on the detector, the position of
150 the beam on the sample, and a time reference to an internal or external signal (used in ToF mode).
151 The beam position is controlled by an external scan generator that also provides the beam position
152 to the detection hardware. The acquisition of the STIM data and the external scan generator are
153 controlled by a LabView interface based on an earlier implementation used for ToF-SIMS in the
154 HIM [14]. The program allows the visualization of the total transmitted signal on the detector, and

155 the creation of user-defined STIM images by selecting areas or radii as the signal providing parts of
 156 the detector. In any case, all the data is stored, therefore, the user can generate BF, ADF, or other
 157 DF images post-processing the transmission data at will at any time.

158 The experiments were conducted using the npSCOPE prototype, which is a high-vacuum instru-
 159 ment based on the gas field ion source (GFIS) column technology. This instrument combines the
 160 helium ion microscopy techniques with SIMS using a magnetic sector spectrometer, STIM with
 161 this new detector, and cryo-microscopy capabilities in a single instrument [38]. In comparison to
 162 the commercial HIM, this microscope has a larger vacuum chamber that allows the installation of
 163 the STIM detector and its movable support. A schematic representation of the measuring geometry
 164 is displayed in Figure 1a together with an overview image of the STIM detector, the stage without
 165 the sidewall, the adapted sample holder, cryo-shields, and ion optical column (Figure 1b).

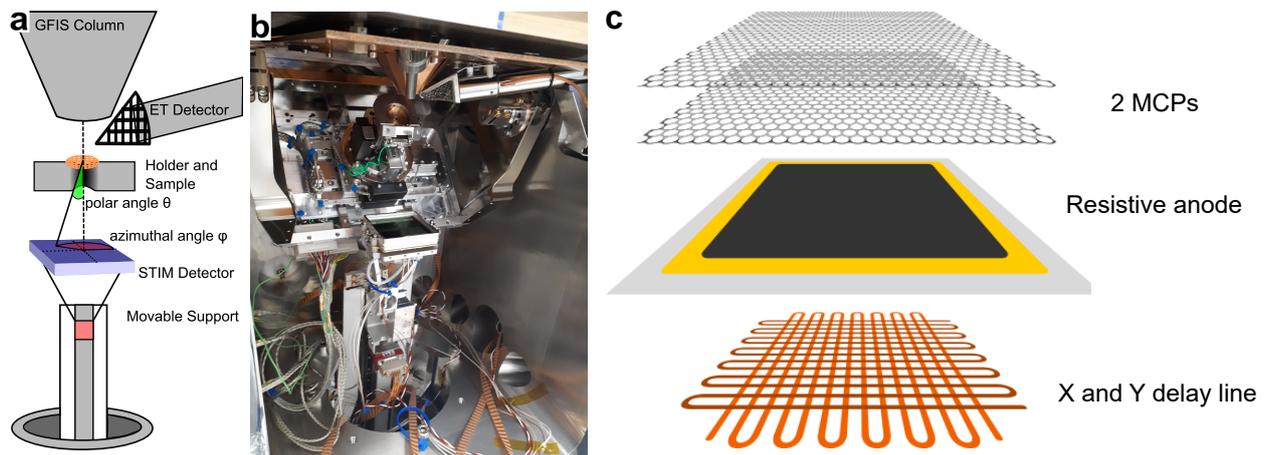


Figure 1: (a) Schematic representation of the STIM experiment. (b) picture of the inner part of the chamber. (c) Schematic representation of the STIM detector.

166 An in-vacuum movable linear support is used to control the detector distance to the sample. This
 167 means that the distance can be chosen to give the best compromise between maximum collection
 168 angle for high angle scattering events, and angular resolution with longer distance (and time-of-
 169 flight) for higher energy resolution. The support consists of a vertically mounted movable rail, on
 170 which a carriage supporting the detector can travel up and down. The rail is mounted on a flange
 171 attached around the pumping hole of the chamber. The motion is driven by piezo motors (Nanomotion
 172 HR-8), and controlled by a motion controller (Nanomotion XCDX) using a closed feedback

173 loop with optically encoded linear rails (Schneeberger Miniscale Plus). This construction is com-
174 patible with the high vacuum requirements, is self-locking and has no mechanical feedthroughs,
175 nor lubricants, and provides high accuracy in the detector's position (down to 100 nm). In the
176 npSCOPE prototype, the distance between the detector and the sample can be adjusted from
177 101 mm to 496 mm, with the closest position being limited by the current stage. This results in a
178 maximum acceptance angle of $\pm 19^\circ$.

179 The sample is currently mounted similarly to the one presented in [31]. A sample holder with an
180 extension arm with a hole on it is used to mount the sample. Since the extension arm is attached
181 at 45° , the stage has to be tilted so that the sample can be aligned with the column axis. In order
182 to allow the transmitted particles to reach the detector, we removed the sidewall of the *cradle* of
183 the current stage. With a new dedicated stage design (currently under construction), the detector
184 can reach a minimum distance to the sample of 50 mm, achieving a maximum polar angle of $\pm 34^\circ$.
185 The detector support is designed in a way that it can be adapted and installed into the commercially
186 available Orion NanoFab chamber, with a reduced travel range.

187 The images presented in this work in transmission mode were taken operating the microscope
188 at 30 kV acceleration voltage, with a $10\ \mu\text{m}$ aperture, in spot control 6 (crossover position of
189 $-247\ \text{mm}$) and a gas pressure of 5×10^{-7} mbar. These conditions provide an estimated beam cur-
190 rent of 50 fA.

191 **Results and Discussion**

192 **Mass-thickness contrast**

193 **Bright and Dark field contrast**

194 In Figure 2, we show images of a carbon film under lacey carbon using the SE imaging mode (Fig-
195 ure 2a), BF STIM (Figure 2b), and ADF STIM (Figure 2c). The two STIM images presented in
196 Figure 2b and Figure 2c were created after data acquisition by selecting different appropriate angles
197 from the same data.

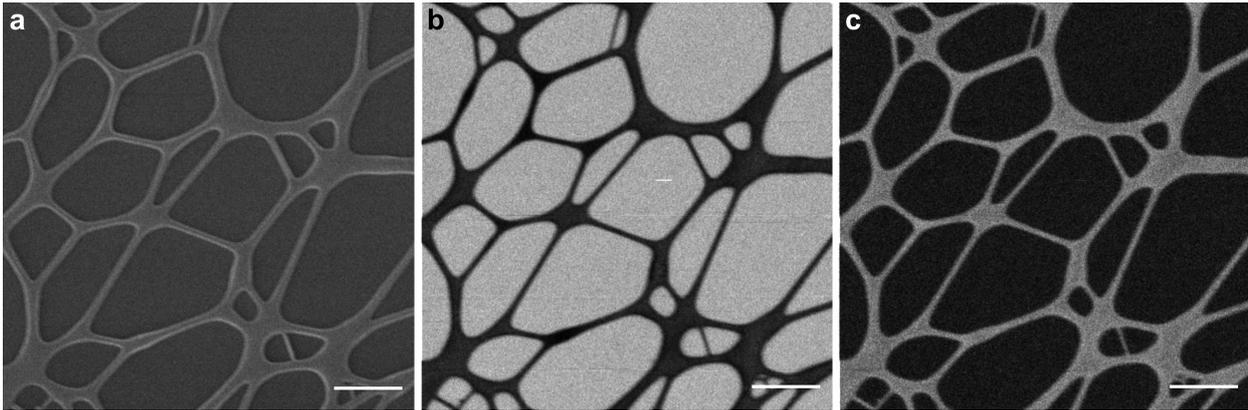


Figure 2: Micrographs of lacey carbon on carbon film in (a) secondary electron imaging mode, (b) bright field STIM image, with collection angle from 0° to 3° , and (c) annular dark field STIM image, with collection angle from 8° to 13.9° . The scale bars are $1\ \mu\text{m}$.

198 The areas of the sample where the carbon film has the lacey carbon on top show a different inten-
 199 sity compared to the areas where there is only a homogeneous film. In general, since the average
 200 polar angle of scattering increases with the thickness, correctly adjusting the cut-off angle for the
 201 BF image can effectively suppress the signal from thicker areas of the sample in the final image.
 202 On the other hand, in ADF, thin areas of the sample are suppressed, while thicker regions appear
 203 bright if an appropriate minimum angle is chosen.
 204 Here, we show two STIM images with different contrast using the same data set. During post-
 205 processing, the discrimination between ADF and BF has been done by choosing different minimum
 206 and maximum scattering angles for each image in order to maximize the contrast for each of them
 207 individually. Scattering angles between 0° to 3° have been used for BF while only scattered par-
 208 ticles with scattering angles between 8° to 13.9° have been used for the corresponding ADF. The
 209 contrast due to the difference in thickness of the material can be noticed in these images.

210 **Quantitative Analysis**

211 Figure 3a is a bright field image of a multilayer sample used to study STIM contrast using com-
 212 binations of light and heavy elements. In the BF image (Figure 3a BF angles: 0° to 4.5°), we can
 213 clearly differentiate all four regions based on their intensity levels.

214 The sample comprises a 20 nm thick silicon nitride membrane used as a support layer. A 20 nm

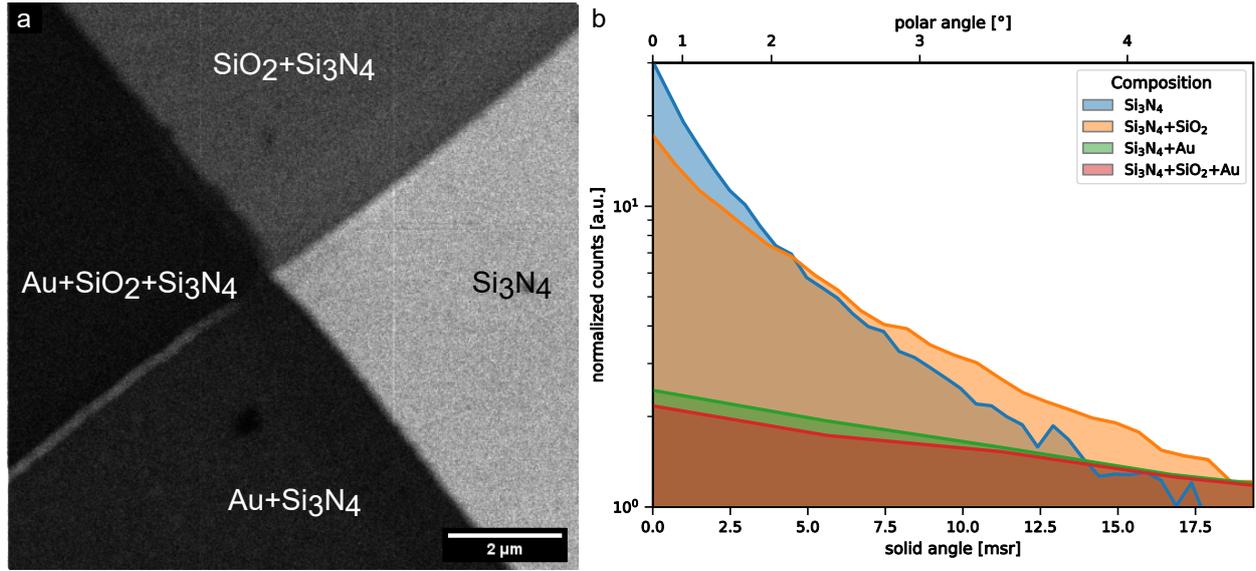


Figure 3: Bright field image showing contrast due to the exit angle dependence on the material and thickness of the layer. (a) Bright field STIM image with collection angle from 0° to 4.5° of a silicon nitride membrane with silicon dioxide deposited on the top left and gold deposited on the lower left. (b) TRIDYN simulation of the angular distribution of the transmitted beam.

215 thick layer of silicon dioxide was deposited on one half—visible on the top left half of the area
 216 in Figure 3a. Then, in the next step, a gold layer of 20 nm was deposited on the lower left half of
 217 the sample, creating four distinct areas on the window. The different material stacks are indicated
 218 in the STIM image. In Figure 3b, we show simulations on the exit angular distribution of 30 keV
 219 He for the different stacks of materials that are present in the sample, using TRIDYN [39] in static
 220 mode. The graph presented in Figure 3b shows the corresponding transmission angular distribution
 221 for the interval used in Figure 3a. The expected contrast between different areas of the sample for
 222 the detection range of 0° to 4.5° is calculated from these distributions. In Table 1, a comparison be-
 223 tween the contrast calculated in the simulations and the contrast obtained from Figure 3a is given.

Table 1: Bright field STIM contrast comparison: Intensity of the transmitted signal from 0 to 4.5°

| Material | Si_3N_4 | $SiO_2 + Si_3N_4$ | $Au + Si_3N_4$ | $Au + SiO_2 + Si_3N_4$ |
|--------------------------------|-----------|-------------------|----------------|------------------------|
| Average counts per pixel | 45.73 | 19.03 | 7.42 | 4.78 |
| Experimental signal normalized | 1 | 0.42 | 0.16 | 0.10 |
| Simulated signal normalized | 1 | 0.67 | 0.12 | 0.11 |

224 For this sample, the simulated and experimental contrast match qualitatively. A quantitative anal-

225 ysis shows relevant differences in the intensity levels of the regions. The relative intensity level of
226 the area with the layer of silicon dioxide on top of the silicon nitride differs considerably in the ex-
227 periment and simulation. The signal in the area on which only gold is deposited is stronger than
228 expected, meanwhile, the signal on the area on which only silicon dioxide is deposited is weaker.
229 A further study on the thickness of each layer using different techniques has not been performed,
230 although deviations of the layer thickness could be responsible for the observed mismatch.

231 **Beam steering and Channeling**

232 **Polycrystalline Silicon**

233 A 15 nm thick nanoporous polycrystalline silicon membrane (available from Electron Microscopy
234 Sciences, item number: 76042–79) has been investigated using STIM. In the SE image (Figure 4a),
235 one can note that the bigger pores are completely black, since they are totally open and no signal
236 comes from these areas. The smaller pores are possibly partially filled with carbon and have some
237 SE signal.

238 The bigger pores appear dark in dark field mode (Figures 4c-e) because there is no scattering. Un-
239 expectedly, the same pores appear dark in bright field mode (Figure 4b) as well. This behaviour
240 can be explained by the intensity of the full primary beam exceeding the local rate capability of
241 the detector. At a distance of 151 mm behind the sample, the beam diameter has only widened to
242 53 μm and quickly saturates the MCP pores with diameters of 25 μm , for a beam with 0.35 mrad
243 convergence. The high local current density temporarily discharges the irradiated pores prevent-
244 ing the creation of further electron cascades above the discriminator value, resulting in dark pix-
245 els. Grains that are thinner than the others, and smaller pores partially filled with residual carbon,
246 appear brighter than the average in BF and darker in DF as expected. This contrast is due to the re-
247 duced scattering which the ions undergo when passing through such an area. Figures 4c-e are dark
248 field images created using the same polar angle but different azimuthal directions on the detector
249 (different to annular dark field where all azimuthal angles are considered). The regions indicated
250 by the arrows show contrast variations in different azimuthal directions of detection, with the same

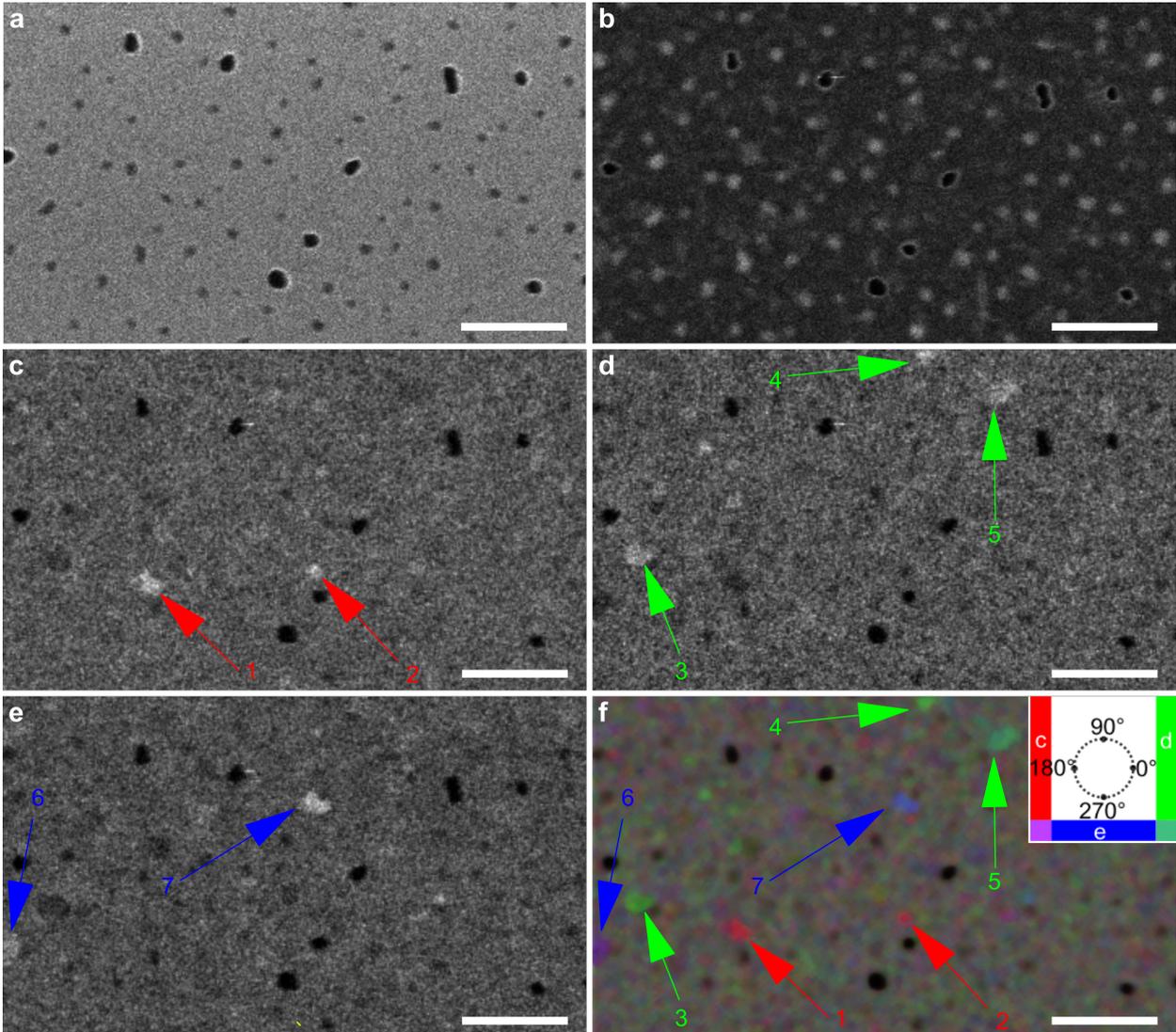


Figure 4: Helium ion microscopy images of the nanoporous polycrystalline silicon membrane. (a) SE image. (b) BF STIM image with polar angle $\theta < 3^\circ$ and ϕ from 0° to 360° . Post processed DF image with polar angles $\theta > 6^\circ$ and azimuthal angles ϕ from (c) 135° to 225° , (d) 315° to 45° , (e) 225° to 315° . (f) Composite colored image using (c), (d) and (e) as RGB color channels. The inset shows the color mapping used in Figure 6f for the areas on the detector. The scale bars are 250 nm.

251 polar angle. The size and shape of these regions are comparable to the size and shape of the grains
 252 of the sample. This contrast change can be explained by channeling and blocking effects. For a ran-
 253 dom orientation or amorphous material the polar angle of the scattering would depend only on the
 254 mass thickness product of the traversed material and no azimuthal pattern is expected. However, for
 255 crystalline materials, depending on the crystal orientation with respect to the beam, the ions can be
 256 channeled along a low index crystal axis or plane and, as a result, are steered into a particular direc-

257 tion. In a STIM image composed of the intensity in this particular polar and azimuthal direction,
258 the grain will appear brighter than the average. (e.g. grain 3 in Figure 4d). Conversely, the same
259 grain will show a lower intensity than a randomly oriented grain for other non-channeling direc-
260 tions, since the beam is not being scattered into random directions as much as it would be the case
261 for a randomly oriented grains (grain 3 in Figure 4c and Figure 4d). Figure 4f is an RGB image
262 created using the three different DF directions as color channels. Using appropriate azimuthal an-
263 gles for the channels, this composite image shows the grains that are steering the beam to directions
264 between two directions used for individual channels presented in Figures 4c-e. For instance, grain 5
265 appears as cyan (overlap between d and e) and grain 6 appears purple (overlap between c and e).
266 The exit angle distribution for the transmitted beam in a crystal depends on the blocking pattern
267 of the crystal for a given orientation. The best contrast for the grains is obtained at angles larger
268 than the largest critical angle for silicon. Therefore, we can infer that the ions are not following the
269 same channel from the beginning to the end. Since this effect would steer the beam to the angle
270 between the crystal axis and the beam, having an upper limit equal to the maximum critical angle
271 for channeling. In silicon, this value would be 3.51° , for the $\langle 110 \rangle$ directions, calculated using an
272 adaptation of [40]. This is also the direction where the minimum backscattering yield (maximum
273 transmission) is expected. Instead, the ions enter the crystal and, after some deviation due to ran-
274 dom scattering, they reach directions in which they are channeled. Holeňák et al. [22] showed the
275 blocking pattern of 50 keV helium through a 200 nm single crystalline silicon foil at a pseudo ran-
276 dom orientation. In their report, some high-intensity spots were present at angles higher than twice
277 the channeling critical angle.

278 **Thallium Chloride**

279 A transmission electron microscopy (TEM) grid coated with evaporated thallium chloride (avail-
280 able on <https://scienceservices.de/> with product code: Sku:E80045) was also analyzed using STIM.
281 This sample has several small crystallites randomly oriented and it is used as a diffraction standard
282 for TEM. Here, we perform a similar analysis to the one done with the polycrystalline sample.

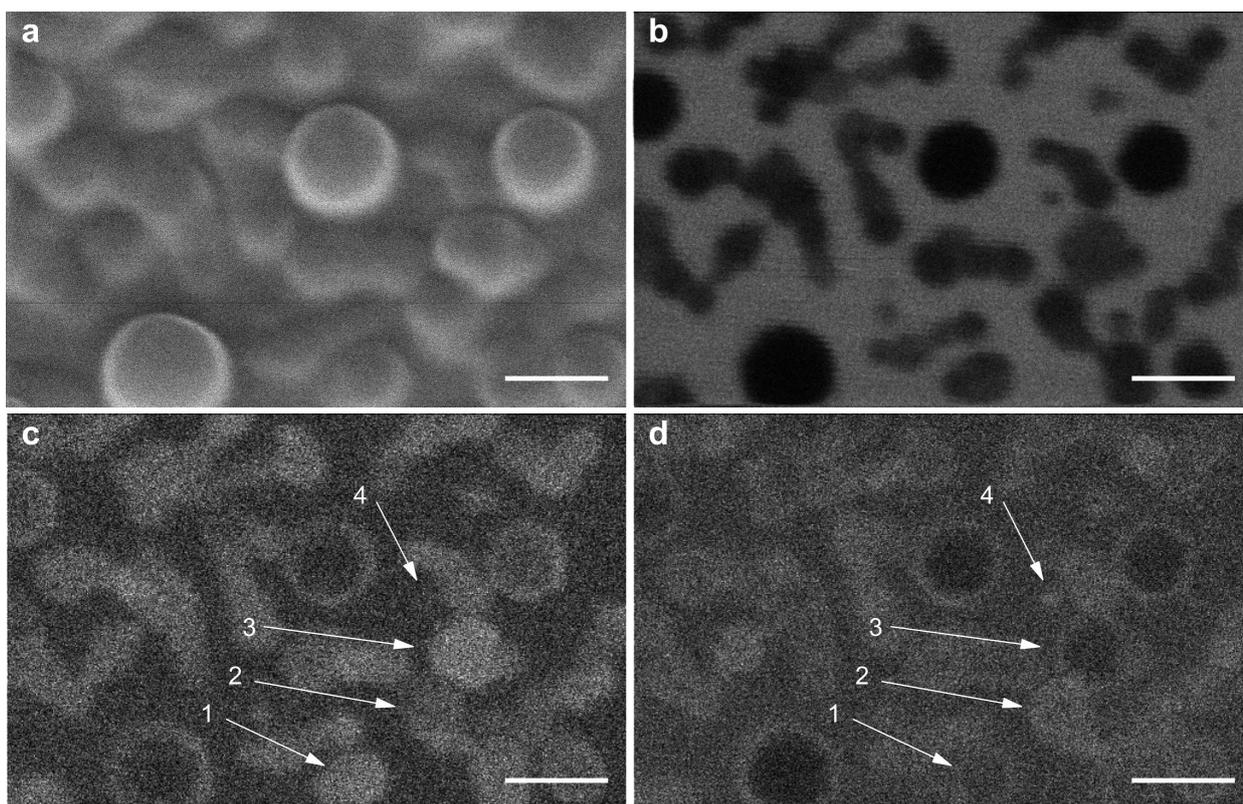


Figure 5: Thallium Chloride evaporated on a TEM grid. (a) Secondary electron image. (b) BF STIM image with acceptance angle of 0° to 4° . (c) DF STIM with polar angle from 6° to 19° and azimuthal angles ϕ from -45° to 45° . (d) DF STIM with polar angle from 6° to 19° and azimuthal angles ϕ from 135° to 225° . The scale bars are 100 nm.

283 The SE image presented in Figure 5a and the BF STIM image (Figure 5b) show crystallites with
284 different sizes. Additionally, the BF image (Figure 5b) gives information on the size of the crystal-
285 lite along the beam axis according to their intensity level. The DF images Figure 5c and Figure 5d
286 are made using same polar angle, but with opposite azimuthal directions.
287 A comparison between the size of the structures in the SE image (Figure 5a) and the BF image
288 (Figure 5b) shows larger structures in the SE image. Considering that a thin film of a light material

289 would show in the SE signal but would not increase significantly the scattering, hence, it would not
290 appear in the BF STIM mode, we assume that there is a film of approximately 10 nm over the crys-
291 tallites. As expected, most of the smaller crystallites that showed as dark in BF are bright in DF.
292 Some larger crystallites are dark both in bright field and dark field images because of their larger
293 size, which causes the beam to be scattered to angles higher than the maximum angle covered by
294 the detector. There are, however, crystallites marked by arrows that show different intensity lev-
295 els for different azimuthal directions. Crystallites 1 and 3 appear brighter in Figure 5c than in Fig-
296 ure 5d, while crystallites 2 and 4 behave in the opposite way. This difference would not happen for
297 amorphous samples and can be explained with preferential scattering along low index directions.
298 Since the crystallites are randomly oriented, the axis in which the transmission of ions is enhanced
299 points in different directions creating this variation of contrast for different azimuthal angles.

300 **Single Crystalline Silicon**

301 In Figure 6a-e, we show STIM, using different sections of the detector and the image of the trans-
302 mission signal (Figure 6). The sample was a 35 nm thick, $\langle 100 \rangle$ oriented silicon membrane win-
303 dow (available on <http://TEMwindows.com>, product code: US100-C35Q33). From Figure 6a,
304 one can see that the membrane has wrinkles that create different angles of incidence between the
305 sample and the incoming beam. The images shown in Figures 6b-e are DF STIM images created
306 using the same polar angles but different azimuthal angles. One can notice that the same areas of
307 the sample show different contrast at different DF directions. This means that different areas of the
308 sample scatter the beam in different preferential directions, depending on the local inclination of
309 the film. Since the membrane is oriented in the $\langle 100 \rangle$ direction, we assume that channeling will
310 predominantly occur along the same direction. The critical angle for 30 keV helium ions along this
311 direction is 1.16° , calculated using an adaptation of [40]. Therefore, areas that are bright in dark
312 field images (Figure 6b-e) can be interpreted as areas in which this channeling direction points to-
313 wards the corresponding dark field region on the detector due to the local inclination of the film. If,
314 for the given experiment, we assume that channeling will occur along the close by $\langle 100 \rangle$ direction,

315 we can obtain the local tilt angle from the measured polar angles. The images presented in Figure 6
 316 highlight the areas with a local tilt angle of 3.8° .

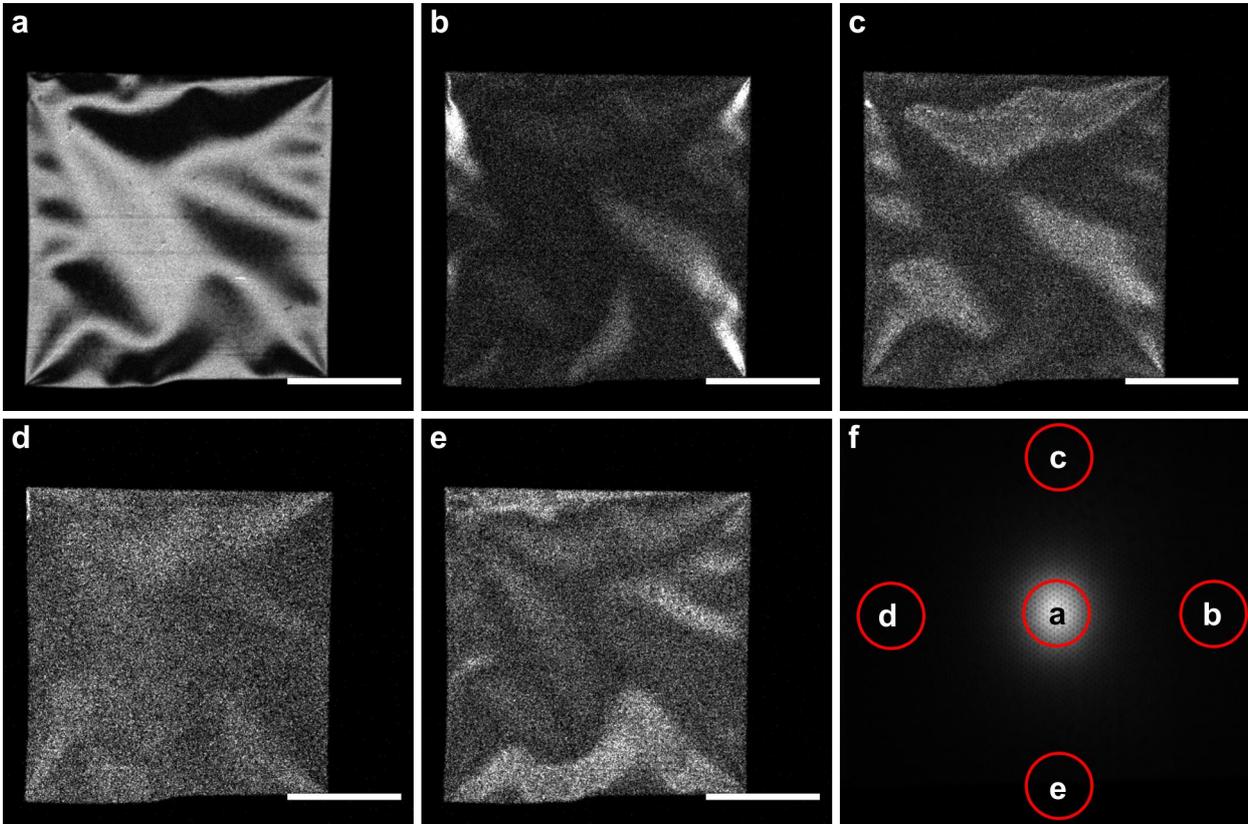


Figure 6: Single crystalline $\langle 100 \rangle$ silicon membrane STIM image in: (a) bright field with $\theta \leq 1.09^\circ$. Dark field centered at the polar scattering angle $\theta = 3.8^\circ$, and azimuthal angle center $\phi = 0^\circ$ in (b), $\phi = 90^\circ$ in (c), $\phi = 180^\circ$ in (d), and $\phi = 270^\circ$ in (e). (f) Detector image of the transmitted signal. The areas on the detector for the corresponding STIM image are marked in red. The scale bars are $50 \mu\text{m}$ for (a), (b), (c), (d), and (e). In (f), the distance from the center to edge corresponds to a 5.58° deflection in the polar angle.

317 Conclusions and Outlook

318 In this work, we presented the development of a detection system for STIM that adds new function-
 319 alities to instruments based on the GFIS ion column, as for example the helium ion microscope or
 320 other high lateral resolution light ion beam methods. The system is based on the combination of
 321 MCPs and a delay line detector mounted on a movable support so that the experiment geometry
 322 can be optimized. The used imaging detector is capable of a random count rate of up to 5 Mcps and

323 has a spatial resolution of approximately 50 μm . This detector has not shown performance degra-
324 dation due to energetic particle damage even when exposed to the primary beam directly. One ad-
325 vantage of this detector over earlier approaches is its flexibility and numerous supported imaging
326 modes. These include bright field, annular dark field, and dark field for channeling applications.
327 In the future the detector will also provide time-of-flight support for these modes with a time reso-
328 lution of 200 ps. In addition, the concept provides the possibility for post-processing the recorded
329 data into BF and DF according to the operator's needs.

330 Using this detection system, we show applications of STIM for amorphous, polycrystalline and
331 single crystalline materials. For the case of amorphous samples, we show the contrast change for
332 low and high scattering angles using BF and ADF detection. We also demonstrate the qualitative
333 match of the contrast in bright field mode with predictions from binary collision approximation
334 calculations using a test sample. In the case of polycrystalline silicon, we can see blocking pattern
335 related contrast in DF. Employing DF and post-processing, we see a contrast dependence on the
336 orientation of thallium chloride nanocrystals. Finally, beam steering effects were shown to occur
337 for a single crystal silicon sample.

338 **Acknowledgements**

339 We acknowledge Stefan Findeisen from the HZDR's mechanical engineering department and
340 Rachid Barrahma from LIST for the mechanical designs of several parts. We thank Olivier Bouton
341 from LIST for the technical support, and Ciarán Fowley from HZDR for sample preparation. We
342 also thank Pasqual Bernhard and Andreas Oelsner from Surface Concept GmbH for insightful dis-
343 cussions on MCPs and delay line detectors. Parts of this research were carried out at the Ion Beam
344 Center (IBC) at the Helmholtz-Zentrum Dresden — Rossendorf e. V., a member of the Helmholtz
345 Association.

346 **Funding**

347 This work has been supported by the H2020 Project npSCOPE under grant number 720964 and by
348 the Luxembourg National Research Fund (FNR) through project STHIM (C16/MS/11354626). ES
349 also received financial support from the PicoFIB network.

References

- 350 1. Hlawacek, G.; Veligura, V.; van Gastel, R.; Poelsema, B. *Journal of Vacuum Science & Technology B* **2014**, *32* (2), 20801. doi:10.1116/1.4863676.
- 351 2. *Helium Ion Microscopy*; Hlawacek, G., Götzhäuser, A., Eds.; NanoScience and Technology;
352 Springer International Publishing: Switzerland, 2016; doi:10.1007/978-3-319-41990-9.
- 353 3. Everhart, T. E.; Thornley, R. F. M. *Journal of Scientific Instruments* **1960**, *37* (7), 246–248.
354 doi:10.1088/0950-7671/37/7/307.
- 355 4. Petrov, Y.; Vyvenko, O. Secondary Electron Generation in the Helium Ion Microscope: Basics
356 and Imaging. In *Helium Ion Microscopy*; Hlawacek, G., Götzhäuser, A., Eds.; NanoScience
357 and Technology; Springer International Publishing: Switzerland, 2016; Chapter 5, pp
358 119–146.
- 359 5. van Gastel, R.; Hlawacek, G.; Dutta, S.; Poelsema, B. *Nuclear Instruments and Methods in
360 Physics Research Section B: Beam Interactions with Materials and Atoms* **2015**, *344*, 44–49.
361 doi:10.1016/j.nimb.2014.11.073.
- 362 6. Klingner, N.; Heller, R.; Hlawacek, G.; von Borany, J.; Notte, J.; Huang, J.; Facsko, S. *Ultra-
363 microscopy* **2016**, *162*, 91–97. doi:https://doi.org/10.1016/j.ultramic.2015.12.005.
- 364 7. van Gastel, R.; Hlawacek, G.; Zandvliet, H. J. W.; Poelsema, B. *Microelectronics Reliability*
365 **2012**, *52* (9-10), 2104–2109. doi:10.1016/j.microrel.2012.06.130.
- 366 8. Veligura, V.; Hlawacek, G.; van Gastel, R.; Zandvliet, H. J. W.; Poelsema, B. *Journal of
367 Physics: Condensed Matter* **2014**, *26* (16), 165401. doi:10.1088/0953-8984/26/16/165401.
- 368 9. Boden, S. A.; Franklin, T. M. W.; Scipioni, L.; Bagnall, D. M.; Rutt, H. N. *Microscopy and
369 Microanalysis* **2012**, *18* (6), 1253–1262. doi:10.1017/S1431927612013463.
- 370 10. Ogawa, S.; Iijima, T.; Awata, S.; Sugie, R.; Kawasaki, N.; Otsuka, Y. *Microscopy and Micro-
371 analysis* **2012**, *18* (S2), 814–815. doi:10.1017/S1431927612005922.
- 372
373

- 374 11. Wirtz, T.; Vanhove, N.; Pillatsch, L.; Dowsett, D.; Sijbrandij, S.; Notte, J. *Applied Physics*
375 *Letters* **2012**, *101* (4), 041601. doi:10.1063/1.4739240.
- 376 12. Dowsett, D.; Wirtz, T. *Analytical Chemistry* **2017**, *89* (17), 8957–8965. doi:{10.1021/acs.
377 analchem.7b01481}.
- 378 13. Wirtz, T.; De Castro, O.; Audinot, J.-N.; Philipp, P. *Annual Review of Analytical Chemistry*
379 **2019**, *12* (1), 523–543. doi:10.1146/annurev-anchem-061318-115457. PMID: 30699036
- 380 14. Klingner, N.; Heller, R.; Hlawacek, G.; Facsko, S.; von Borany, J. *Ultramicroscopy* **2019**, *198*,
381 10–17. doi:10.1016/j.ultramic.2018.12.014.
- 382 15. Lindhard, J. *Phys. Lett.* **1964**, *12* (2), 126–128. doi:10.1016/0031-9163(64)91133-3.
- 383 16. Gemmell, D. S. *Rev. Mod. Phys.* **1974**, *46*, 129–227. doi:10.1103/RevModPhys.46.129.
- 384 17. Hlawacek, G.; Veligura, V.; van Gastel, R.; Poelsema, B. Channeling and Backscatter Imag-
385 ing. In *Helium Ion Microscopy*; Hlawacek, G., Götzhäuser, A., Eds.; NanoScience and Tech-
386 nology; Springer International Publishing: Switzerland, 2016; Chapter 9, pp 205–224.
- 387 18. Veligura, V.; Hlawacek, G.; van Gastel, R.; Zandvliet, H. J. W.; Poelsema, B. *Beilstein Journal*
388 *of Nanotechnology* **2012**, *3*, 501–506. doi:10.3762/bjnano.3.57.
- 389 19. Hijazi, H.; Li, M.; Barbacci, D.; Schultz, A.; Thorpe, R.; Gustafsson, T.; Feldman, L. *Nuclear*
390 *Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials*
391 *and Atoms* **2019**, *456*, 92–96. doi:10.1016/j.nimb.2019.07.002.
- 392 20. Ohya, K. *AIP Advances* **2018**, *8* (1), 015120. doi:10.1063/1.5018126.
- 393 21. Ghaderzadeh, S.; Ghorbani-Asl, M.; Kretschmer, S.; Hlawacek, G.; Krashennnikov, A. V.
394 *Nanotechnology* **2019**, *31* (3), 035302. doi:10.1088/1361-6528/ab4847.
- 395 22. Holeňák, R.; Lohmann, S.; Primetzhofer, D. *Ultramicroscopy* **2020**, *217*, 113051. doi:https:
396 //doi.org/10.1016/j.ultramic.2020.113051.

- 397 23. Dissanaïke, G. *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of*
398 *Science* **1953**, *44* (357), 1051–1063. doi:10.1080/14786441008520363.
- 399 24. Lohmann, S.; Primetzhofer, D. *Phys. Rev. Lett.* **2020**, *124*, 096601. doi:10.1103/PhysRevLett.
400 124.096601.
- 401 25. Scipioni, L.; Ferranti, D. C.; Smentkowski, V. S.; Potyrailo, R. A. *Journal of Vacuum Science*
402 *& Technology B* **2010**, *28* (6), C6P18–C6P23. doi:10.1116/1.3517514.
- 403 26. Hall, A. R. *Microscopy and microanalysis : the official journal of Microscopy Society of*
404 *America, Microbeam Analysis Society, Microscopical Society of Canada* **2013**, *19* (3), 740–4.
405 doi:10.1017/S1431927613000500.
- 406 27. Scipioni, L.; Sanford, C. A.; Notte, J.; Thompson, B.; McVey, S. *Journal of Vacuum Science*
407 *& Technology B: Microelectronics and Nanometer Structures Processing, Measurement, and*
408 *Phenomena* **2009**, *27* (6), 3250–3255. doi:10.1116/1.3258634.
- 409 28. Emmrich, D. A. *Mikroskopie und Modifikation von Nanostrukturen mit Helium-Ionen*. Ph. D.
410 Thesis, Universität Bielefeld, 2019.
- 411 29. Notte, J. A.; Hill, R.; McVey, S. M.; Ramachandra, R.; Griffin, B. J.; Joy, D. C. *Microscopy*
412 *and Microanalysis* **2010**, *16* (05), 599–603. doi:10.1017/S1431927610093682.
- 413 30. Woehl, T. J.; White, R. M.; Keller, R. R. *Microscopy and Microanalysis* **2016**, *22* (Issue 3),
414 544–550.
- 415 31. Kavanagh, K. L.; Herrmann, C.; J. Notte, *Journal of Vacuum Science and Technology B* **2017**,
416 *35* (Issue 6), 1–5.
- 417 32. Wang, J.; Huang, S. H.; Herrmann, C.; Scott, S. A.; Schiettekatte, F.; Kavanagh, K. L. *Journal*
418 *of Vacuum Science and Technology B* **2018**, *36* (021203), 1–5.

- 419 33. Keller, H.; Klingelhöfer, G.; Kankeleit, E. *Nuclear Instruments and Methods in Physics Re-*
420 *search Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **1987**,
421 258 (2), 221 –224. doi:10.1016/0168-9002(87)90059-3.
- 422 34. Mende, S.; Heetderks, H.; Frey, H.; Stock, J.; Lampton, M.; Geller, S.; Abiad, R.; Sieg-
423 mund, O.; Habraken, S.; Renotte, E.; Jamar, C.; Rochus, P.; Gérard, J.-C.; Sigler, R.;
424 Lauche, H. *Space Science Reviews* **2000**, 91, 287–318. doi:10.1023/A:1005292301251.
- 425 35. Müller-Caspary, K.; Oelsner, A.; Potapov, P. *Applied Physics Letters* **2015**, 107 (7), 072110.
426 doi:10.1063/1.4927837.
- 427 36. Beni, A.; Ott, N.; Pawelkiewicz, M.; Ward, M.; Young, K.; Bauer, B.; Rajput, P.;
428 Detlefs, B.; Zegenhagen, J.; McGrath, R.; et al., *Electrochemistry Communications* **2014**, 46,
429 13–17. doi:10.1016/j.elecom.2014.05.024.
- 430 37. Uhm, H. S.; Choi, E. H.; Cho, G. S. *Applied Physics Letters* **2009**, 94 (3), 031501. doi:10.
431 1063/1.3073983.
- 432 38. De Castro et al., in preparation
- 433 39. Möller, W.; Eckstein, W. *Nuclear Instruments and Methods in Physics Research Section B:*
434 *Beam Interactions with Materials and Atoms* **1984**, 2 (1), 814 –818. doi:https://doi.org/10.
435 1016/0168-583X(84)90321-5.
- 436 40. Doyle, B. L. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interac-*
437 *tions with Materials and Atoms* **2016**, 371, 63 –68. doi:10.1016/j.nimb.2015.08.047. The 22nd
438 International Conference on Ion Beam Analysis (IBA 2015)