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Electron beam-induced current imaging with two-angstrom resolution

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Abstract

An electron microscope's primary beam simultaneously ejects secondary electrons (SEs) from the sample and generates electron beam-induced currents (EBICs) in the sample. Both signals can be captured and digitized to produce images. The off-sample Everhart-Thornley detector that is common in scanning electron microscopes (SEMs) can detect SEs with low noise and high bandwidth. However, the transimpedance amplifiers appropriate for detecting EBICs do not have such good performance, which makes accessing the benefits of EBIC imaging at high-resolution relatively more challenging. Here we report lattice-resolution imaging via detection of the EBIC produced by SE emission (SEEBIC). We use an aberration-corrected scanning transmission electron microscope (STEM), and image both microfabricated devices and standard calibration grids.

Keywords: aberration-correction, transmission electron microscopy, secondary electrons, STEM, EBIC

1. Introduction

Lattice resolution (< 1 nm) imaging with scanning transmission electron microscopy (STEM) in its standard mode was first demonstrated by Crewe and Wall in 1970 [1]. Since this milestone was achieved, efforts have been ongoing to extend such resolution to the auxiliary imaging and spectroscopic modes available to STEM instruments. Lattice resolution secondary electron imaging [2], electron energy loss spectroscopy [3], and energy dispersive X-ray spectroscopy [4] were first demonstrated in 1990, 2007, and 2010 respectively. Atomic-resolution (< 0.1 nm) versions of the same milestones were reached in 2009 [5], 2008 [6], and 2010 [7] respectively.

The subject of this paper, secondary electron beam induced current (SEEBIC) imaging, is closely related to the secondary electron imaging just mentioned, but also to electron-beam induced current (EBIC) imaging. In standard EBIC imag-

ing [8], the rastering STEM beam creates electron-hole pairs in the sample that are then separated in a local electric field, such as might be found in a p-n junction. The region where the pairs are generated is electrically connected to a transimpedance amplifier (TIA), which collects either the electrons or the holes, depending on the side of the circuit to which the TIA is connected. Associating the measured current with the beam position creates the EBIC image.

In SEEBIC imaging, on the other hand, there is no intrinsic electric field; the image contrast is generated by the production of secondary electrons and their associated holes [9] (see Fig. 1 and accompanying text). If a direct current path exists from the charge generation region to the TIA, the TIA will collect more holes and the contrast will be positive. If the TIA is instead connected to an electrode that is neighboring, but not directly connected to the charge generation region, the ejected SEs (or associated tertiary electrons) can travel through the

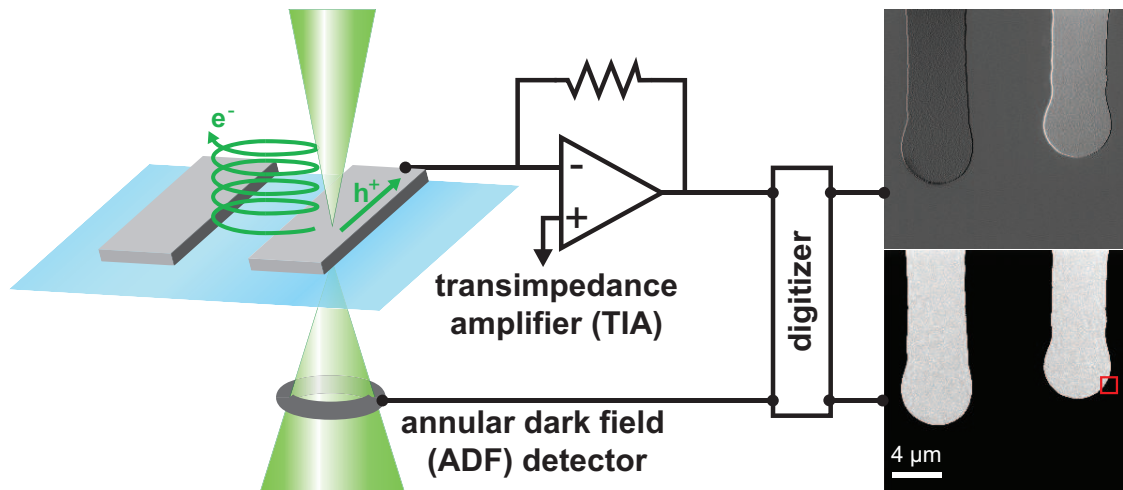


Figure 1: **Schematic of an experimental setup and corresponding low-magnification images.** A device consisting of two metal electrodes (each consisting of a 5 nm Ti adhesion layer covered with 25 nm of Pt) on a insulating, electron-transparent membrane is being imaged with scanning transmission electron microscopy (STEM). The lower signal chain generates the standard STEM annular dark field (ADF) image, which shows both contacts with the same contrast. The upper signal chain generates the SEEBIC image with its differential contrast: the electrode attached to the transimpedance amplifier (TIA) is bright while the other electrode is dark. A red box in the ADF image indicates the scale of the electrode-edge region shown in the leftmost frame of Fig. 2.

42 microscope vacuum to reach this neighboring electro-
 43 de. In this instance the TIA measures a negative
 44 current and generates negative contrast. Relative
 45 to standard EBIC, SEEBIC signals are typically
 46 smaller [9], but they are found throughout a de-
 47 vice, and not just in special regions that happen to
 48 support a non-zero electric field.

49 Standard SE imaging is, of course, the main
 50 imaging mode of the scanning electron microscope
 51 (SEM), and is sometimes employed in the TEM. In
 52 both cases SE liberated by the scanning electron
 53 beam are captured off-sample in a detector, and as-
 54 sociating the measured SE signal with the beam po-
 55 sition again produces the image. The off-sample de-
 56 tector most commonly used for SE was invented by
 57 Everhart and Thornley [10], and is a marvel of low-
 58 noise amplification. Using a kilovolt-scale positive
 59 potential, it accelerates the low energy ($\lesssim 10$ eV)
 60 SE into a scintillator, producing light that is sub-
 61 sequently detected with a photomultiplier tube. As
 62 described already in the 1960 publication announc-
 63 ing this invention [10], detectors based on this archi-
 64 tecture can have femtoampere (10^{-15} A) sensitivity
 65 with 10 MHz bandwidth. Compare these specifica-
 66 tions with those of a modern TIA used for the de-
 67 tection of EBIC: the DLPCA-200 made by FEMTO
 68 Messtechnik GmbH and used in this study, for in-

69 stance, has an integrated input noise current (rms)
 70 of 800 fA with 1.1 kHz bandwidth. These specifica-
 71 tions indicate that, as a device for measuring elec-
 72 trical currents, the Everhart-Thornley detector is
 73 superior to a modern TIA by more than two orders-
 74 of-magnitude in both noise current spectral density
 75 and bandwidth. From a technological standpoint,
 76 detecting free charges in vacuum is clearly easier
 77 than detecting them in a metal wire.

78 Given that off-sample SE detectors outperform
 79 TIAs by orders-of-magnitude, and that atomic reso-
 80 lution imaging using SE was achieved only recently,
 81 the question then arises: is it possible to achieve lat-
 82 tice resolution with an EBIC-based technique? The
 83 purpose of this communication is to provide an an-
 84 swer in the affirmative, and to describe how the
 85 STEM imaging and sample parameters can be op-
 86 timized to compensate for the fundamentally poor
 87 signal-to-noise performance of the TIA.

88 2. Experiment

89 Except for the data of Fig. 1, we used the
 90 TEAM 1 microscope at the National Center for
 91 Electron Microscopy (NCEM) at the Molecular
 92 Foundry in Lawrence Berkeley National Labora-
 93 tory (LBNL). This microscope is a modified FEI

94 Titan 80-300 equipped with a CEOS hexapole-type 146
95 probe corrector that provides full correction of 3rd 147
96 order ($C_3 < 0.5 \mu\text{m}$) and partial correction of 5th 148
97 order ($C_5 < 0.5 \text{ mm}$) spherical aberrations. Annu- 149
98 lar dark field (ADF) signals were collected with a 150
99 Fischione Model 3000 ADF detector, and digitized 151
100 by a Gatan Digiscan II to 12-bit precision simulta- 152
101 neously with the EBIC signal from a DLPCA-200. 153
102 Electrical connection to the sample was made with 154
103 a biasing sample holder (Hummingbird Scientific). 155
104 The images of Fig. 1 were acquired using the FEI 156
105 Titan 80-300 in the California NanoSystems insti- 157
106 tute at UCLA, which also has a Fischione Model 158
107 3000 ADF detector but does not have a corrector. 159

108 Both microscopes were operated at an accelerat- 160
109 ing voltage of 300 kV with a probe current of 200– 161
110 300 pA. In probe-corrected microscopes a smaller 162
111 accelerating voltage would likely give better SEE- 163
112 BIC performance, as the SE yield, and thus the sig- 164
113 nal, varies inversely with the beam energy [11, 9]. 165
114 The large probe current was chosen as a compro- 166
115 mise between having a small probe ($\lesssim 50 \text{ pA}$ is typi- 167
116 cal for high-resolution imaging) and a good signal- 168
117 to-noise ratio in the EBIC channel (the EBIC sig- 169
118 nal is proportional to the beam current [9]). The 170
119 data of Fig. 1 were acquired with a convergence an- 171
120 gle $\alpha \simeq 9 \text{ mrad}$, as is typical for high-resolution 172
121 imaging with an uncorrected microscope. All other 173
122 data were acquired with $\alpha \simeq 17 \text{ mrad}$. For high- 174
123 resolution imaging with a first generation spheri- 175
124 cal aberration corrector, $\alpha \simeq 25 \text{ mrad}$ would be 176
125 standard, but with the less-demagnified source the 177
126 smaller convergence angle more coherently fills the 178
127 probe-forming second condenser aperture. Typi- 179
128 cal dwell times were 1.5–2.5 ms/pixel, which corre- 180
129 sponds to 2–3 minutes for a 256×256 pixel image. 181

130 As SEEBIC imaging is most revealing in samples 182
131 that contain multiple electrically-disconnected re- 183
132 gions [9], we demonstrate lattice resolution imaging 184
133 in actual devices featuring lithographically-defined 185
134 metal electrodes. Figure 1 shows a basic experi- 186
135 mental arrangement, where the device consists of 187
136 two metal electrodes that have been defined via op- 188
137 tical lithography and are facing each other across 189
138 a 25 nm-thick silicon nitride membrane. The elec- 190
139 trodes have identical thicknesses and thus give the 191
140 same contrast in the ADF images, since they scatter 192
141 the beam electrons into the ADF detector with 193
142 equal efficiency. However, while they also gener- 194
143 ate secondary electrons (SEs) with equal efficiency, 195
144 these same electrodes give opposite contrast in the 196
145 EBIC images. Because the TIA is attached to one

146 electrode and not the other, the SE signal actu- 147
148 ally changes sign between the electrodes. When 149
150 the beam hits the electrode attached to the TIA, 151
152 more SEs are generated than return to the elec- 153
154 trode, and so the net (hole) current into the TIA is 155
156 positive and gives bright contrast. When the beam 157
158 hits the other electrode, some secondary and ter- 159
160 tiary electrons reach the TIA’s electrode (no holes 161
162 do) and the net (electron) current into the TIA is 163
164 negative, giving dark contrast. This SEEBIC image 165
166 of Fig. 1 demonstrates one of the major strengths 167
168 [9] of SEEBIC imaging relative to both standard 169
170 STEM imaging and off-sample SE imaging: it re- 171
172 veals electrical connectivity. 173

174 The contrast reversal between the electrodes is 175
176 not exact; the hole current in the electrode con- 177
178 nected to the TIA has greater magnitude than the 179
180 electron current generated from another electrode 181
182 [9]. Thus when imaging at high spatial resolution 183
184 or otherwise attempting to maximize the signal-to- 185
186 noise ratio, it is generally best to image using the 187
188 stronger hole signal. Under most circumstances this 189
190 optimization presents no difficulties. If the feature 191
192 of interest happens to be on or near an electrode 193
194 not attached to the TIA, one can either switch the 195
196 TIA to the electrode of interest, short the electrode 197
198 of interest to the TIA’s electrode, or add yet an- 199
200 other TIA and thereby add another SEEBIC imag- 201
202 ing channel. 203

204 3. Results

205 To demonstrate lattice resolution we first drop- 206
207 cast 5 nm diameter gold nanoparticles from a col- 208
209 loidal suspension (Ted Pella, part # 15702) onto a 210
211 device with 5/25 nm Ti/Pt electrodes. The gold 211
212 lattice provides a clean distance calibration stan- 212
213 dard, where a measured lattice parameter can be 213
214 identified with a known distance with certainty. 214
215 Such a standard is not available from the device it- 215
216 self, for in this case the three materials available are 216
217 unsuitable. Although crystalline, the metal elec- 217
218 trode materials might be alloyed, oxidized, or oth- 218
219 erwise chemically altered from their pure, elemen- 219
220 tal forms during the fabrication processing. The 220
221 Si_3N_4 support membrane is amorphous and thus 221
222 has no well-defined lattice parameter. And the sil- 222
223 icon wafer that frames the Si_3N_4 membrane, while 223
224 a single crystal, is in no place sufficiently thin to 224
225 allow lattice resolution imaging. 225

Two dropcast particles that have had their chem- 226
227 ical identities confirmed as Au via energy dispersive 227

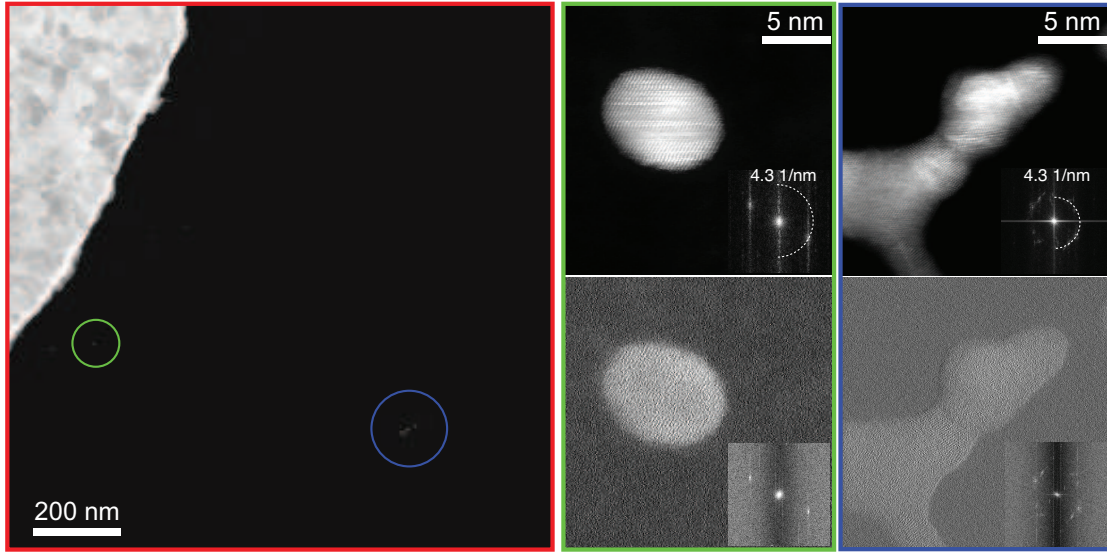


Figure 2: **ADF and SEEBIC images of regions adjacent to an electrode.** A Ti/Pt SEEBIC sense electrode has several gold nanoparticles nearby (left image, ADF). Two circled nanoparticles are shown at high-magnification (right images, indicated by the correspondingly colored frames). Both the ADF (upper row) and the SEEBIC images (lower row) show lattice resolution, as demonstrated by the peaks at the Au {111} spacing of 0.235 nm in the inset fast Fourier transforms (FFTs). As is observed generally and is the case here, the SEEBIC signal is stronger for nanoparticles closer to an electrode.

196 X-ray spectroscopy are highlighted in the leftmost 223
 197 frame of Fig. 2, one within 140 nm of the TIA's 224
 198 sense electrode, and the other 840 nm away. In 225
 199 both cases, high-resolution images of these parti- 226
 200 cles show the gold lattice in both the ADF and the 227
 201 EBIC channels. Fast Fourier transforms (FFTs) 228
 202 of the images identify the gold {111} Bragg peak 229
 203 at 4.25 nm^{-1} , which corresponds to an interpla- 230
 204 nar spacing of 0.235 nm [12]. For ADF and EBIC 231
 205 images acquired simultaneously, the images have 232
 206 strictly identical scaling. 233

207 Here the EBIC signals are positive, indicating the 234
 208 existence of a through-the-sample electrical path 235
 209 between the electrode and the nominally isolated 236
 210 nanoparticles. Although not well-characterized, the 237
 211 resistance of this connection is likely in the $T\Omega$ 238
 212 range. While such a connection is not robust 239
 213 enough to give a strong EBIC signal, it is, perhaps 240
 214 surprisingly, robust enough to give a net hole cur- 241
 215 rent. The contrast in the EBIC channel is smaller 242
 216 than that in the ADF channel, and it is decreasing 243
 217 with increasing distance from the sense electrode. 244

218 This decrease can be quantified. In the EBIC 245
 219 channel, the farther, dog-shaped particle generates 246
 220 $3\times$ less contrast than the closer, round particle, 247
 221 which itself generates $3\times$ less contrast than the 248
 222 sense electrode itself. In each case here 'contrast' is 249

defined as the difference between the signal from 223
 the metal and that from the neighboring Si_3N_4 . 224
 For comparison, in the ADF channel the farther, 225
 dog-shaped particle generates $1.5\times$ more contrast 226
 than the closer, round particle, which generates $6\times$ 227
 less contrast than the sense electrode itself. Thus, 228
 relative to ADF, the EBIC contrast is more sensi- 229
 tive to connectivity (and correspondingly to loca- 230
 tion), and less sensitive to the total thickness. To 231
 achieve the best possible EBIC signal-to-noise ratio 232
 and contrast, the region of interest should therefore 233
 be either part of the sense electrode, or electrically 234
 connected to it. 235

Imaging a 5/25 nm Ti/Pt sense electrode at 236
 higher magnification (Fig. 3) reveals that lattice 237
 resolution can be achieved not only in nanoparti- 238
 cles scattered over the device, but also in the actual 239
 components of the device itself. While the electrode 240
 is thin enough to be electron-transparent, the grains 241
 in the Pt layer are unlikely to be aligned with the 242
 grains of the Ti adhesion layer [9], which makes it 243
 very unlikely that both layers are aligned so as to 244
 allow the detection of lattice in the polycrystalline 245
 bulk. Consequently we image on the edge of the 246
 sense electrode, where the material is thinner than 247
 the nominal 30 nm. 248

To show both the electrode edge and the Si_3N_4 249

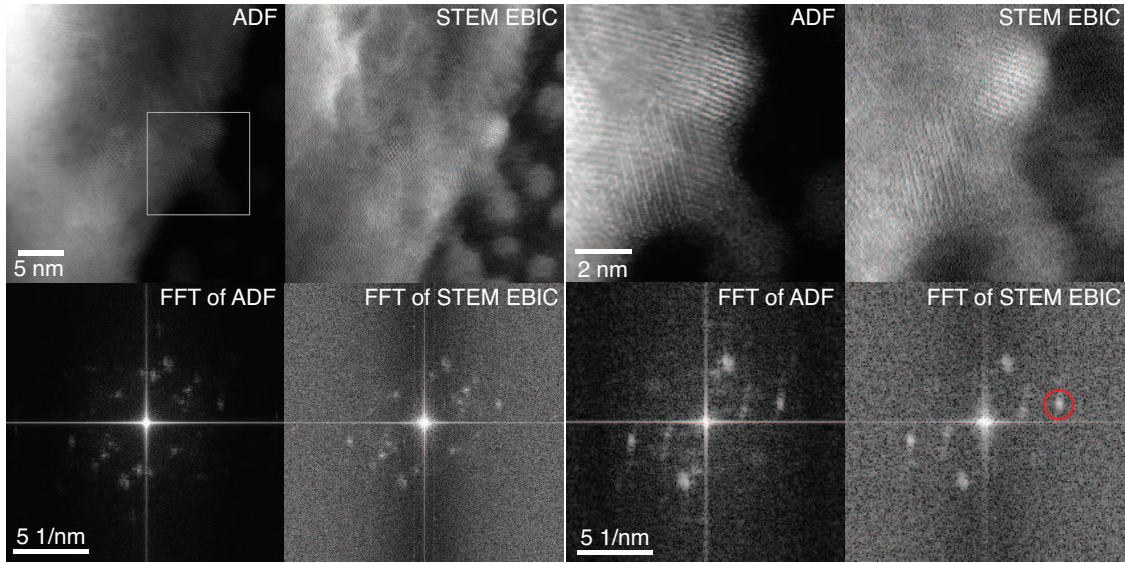


Figure 3: Two pair of lattice-resolution ADF and SEEBIC images of Ti/Pt contacts on silicon nitride (top row), and the FFTs of these images (bottom row). The real space images are acquired at two different magnifications (the grey box in the left ADF image indicates the full field of view of the right pair), but the FFTs are all shown with the same scale. A resolution of 200 pm is achieved, as indicated by the position of the circled peak in the FFT.

250 membrane, the first set of images (Fig. 3 left) have
 251 a slightly larger field of view. The ADF signal, being
 252 more sensitive to the sample's total thickness,
 253 shows the larger actual contrast variation between
 254 the electrode and the membrane. (These Fig. 3 im-
 255 ages have had their display contrast levels set with
 256 the default 'sparse' auto-contrast function set in ver-
 257 sion 2.3 of Gatan's Digital Micrograph software.)
 258 The EBIC signal, on the other hand, shows less ac-
 259 tual contrast change as the electrode gets thicker.
 260 Thus the EBIC image can better exploit the 8-bit
 261 gray-scale display range available: it reveals fine
 262 details that are nearly invisible in the ADF image,
 263 such as nanoparticles adjacent to the electrode on
 264 the Si_3N_4 membrane. An excellent insulator, the
 265 membrane itself gives little SE signal, and produces
 266 only a small EBIC background in comparison to a
 267 conducting support (see e.g. Fig 4 and discussion).

268 Higher-magnification images of the same region
 269 (Fig. 3 right) make the lattice obvious, even in
 270 the real space images. Again the ADF contrast
 271 is stronger, but the similarity between the ADF
 272 and the EBIC images, despite the completely differ-
 273 ent contrast mechanisms, indicates a common root
 274 cause. In a classical (e.g. Rutherford) model, the
 275 ADF contrast is generated by the nuclear cores,
 276 which scatter beam electrons more strongly at

277 smaller impact parameters. In the corresponding
 278 model of the EBIC contrast, the probability of SE
 279 emission varies with the sample's electron density,
 280 which is also greater nearer the nuclei. In more
 281 precise language one can say that the lattice sig-
 282 nal in the SEEBIC image is evidence that SE are
 283 produced by inner-shell excitations, which corre-
 284 spond to larger energy scales [13] than the peak
 285 (< 10 eV) of the SE distribution [11, 9], and which
 286 are not de-localized [14, 15]. Thus both ADF and
 287 EBIC techniques can image the crystal lattice as de-
 288 fined by the positions of the nuclear cores. Further
 289 complexities of generating SEs at atomic resolution
 290 are discussed in [16], particularly how screening can
 291 dampen states near the Fermi energy and decrease
 292 high resolution contrast from lower- Z elements such
 293 as oxygen.

294 Finally we show that, while desirable, the device
 295 structure is not necessary for lattice resolution SEE-
 296 BIC imaging: a simple TEM grid can be used in
 297 place of the device. For a test sample we use a stan-
 298 dard carbon diffraction grating replica on a copper
 299 Gilder grid, with Au/Pd shadowing (Ted Pella part
 300 # 607), such as is commonly used for magnification
 301 calibration. Images of the gold/palladium (Fig. 4
 302 top) show lattice in both the ADF and the EBIC
 303 channels. Because of its low atomic number Z , the

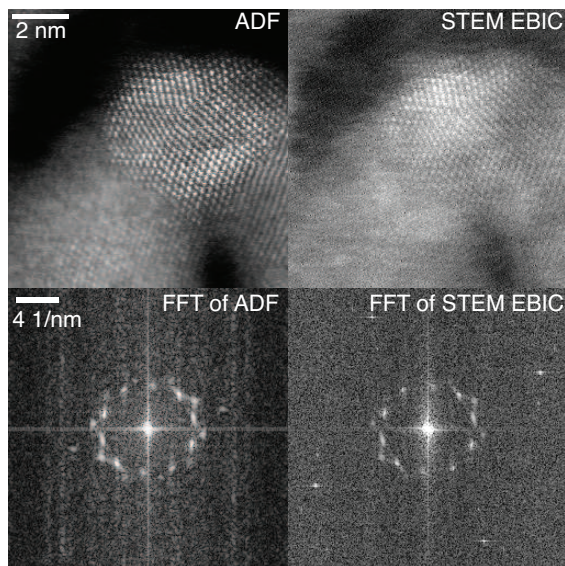


Figure 4: ADF and SEEBIC images of the Pd/Au of a standard magnification calibration (TEM grid) sample, along with the FFTs of each image. The Bragg reflections in the FFTs indicate a resolution of 200 pm. The apparent reflections at 11 nm^{-1} (which would correspond to an interplanar spacing of 90 pm) are due to 60 Hz pickup by the EBIC detection circuit.

carbon ($Z = 6$) film is less effective at scattering beam electrons than the bulk gold ($Z = 79$) and palladium ($Z = 46$), and thus it is not evident in the ADF image. The (conducting) carbon film is, however, visible in the EBIC image, because SEEBICs are generated more effectively from surfaces than from the bulk [9].

The corresponding FFTs (Fig. 4 bottom) show that both channels are detecting the same lattice, with a characteristic inter-plane spacing of 200 pm. The EBIC image shows some additional peaks that look as if they correspond to scattering angles about twice those of the main lattice peaks. These peaks are spurious and due to AC line noise. The noise is small and, if desired, could be easily removed from the image by masking the spurious peaks in reciprocal space and performing the inverse FFT. None of the data presented in this paper have had any such filtering applied.

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