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DOI: 10.1017/S1431927606067511
Microsc Microanal 12(Supp 2), 2006

Advanced tuning algorithm for crystalline materials in scanning transmission electron microscopy

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The successful operation of modern aberration-corrected microscopes requires the accurate determination of the illumination conditions. However, the aberration measurement tools used on most practical set-ups rely on the numerical analysis of diffractograms obtained from amorphous regions of the sample [1, 2]. Going back and forth between the area of interest and a possibly distant patch of amorphous material is cumbersome, and a method was recently developed and tested at the SuperSTEM laboratory that can allow a partial fine re-tuning *in situ* in the crystalline area.

Ronchigrams, or shadow images, obtained from a crystalline sample characteristically present many sets of fringes due to the coherent interference between the various Bragg-diffracted discs as they overlap in the diffraction plane [3]. It can be shown that the intensity recorded along a line situated at midpoint between two overlapping discs is independent of defocus [4]. Experimental evidence of such behaviour is most elegantly seen by recording a through-focus series of electron Ronchigrams and adding up the obtained images, thus smearing out all focus-dependent contrast: Fig. 1. The new numerical scheme uses the contrast variations along those “achromatic lines” to obtain experimental values for the non-round aberration coefficients up to the desired order. Very successful tests on silicon and gallium nitride on the Nion-corrected VG HB501 microscope of the SuperSTEM facility indicate comparable performances to AutoSTEM, the Nion Co. tuning algorithm: table 1. Probe drift during the acquisition of the achromatic patterns may however be introducing systematic errors in the measurement [5] and steps are being taken to study and incorporate these effects in the algorithm. For samples of known lattice constants, the scheme does not rely on any microscope-specific parameter or calibration, thus providing an “absolute” measurement of the aberration coefficients. This could also become extremely useful for calibration of other amorphous-based methods for instance.

Although it was initially feared that the high brightness and coherence of the VG HB501 cold field emission gun was necessary to observe the achromatic lines, preliminary results obtained on a non-corrected Tecnai F20 prove that achromatic patterns can indeed be observed on machines equipped with a Schottky emitter: Fig. 2. It is interesting to note that a complementary numerical scheme allowing the measurement of spherical aberration with crystalline Ronchigrams in conditions akin to those necessary for “achromatic imaging” was recently reported [6]. This, combined with the present method, should pave the way to a complete *in situ* fine-tuning package for aberration corrected STEMs. A full implementation of the achromatic tuning algorithm will now be tested on a CEOS-corrected instrument at Brookhaven National Laboratory.

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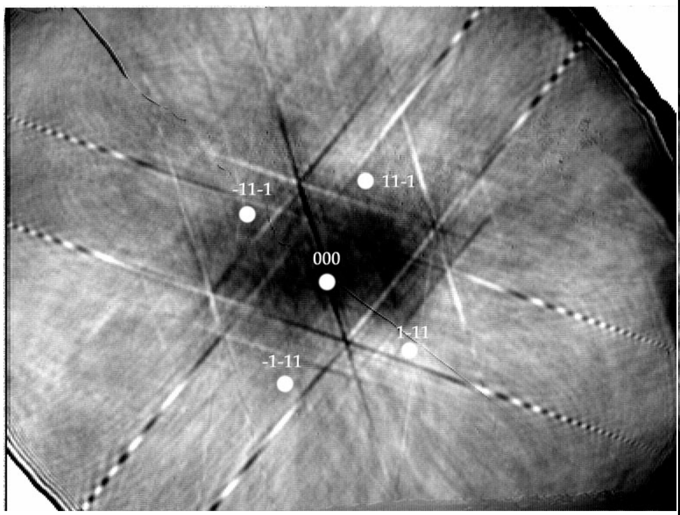

Fig. 1**:** Achromatic lines observed the aberration-corrected VG HB501 SuperSTEM instrument by integrating a through-focus series of Ronchigrams (silicon in 110 orientation).

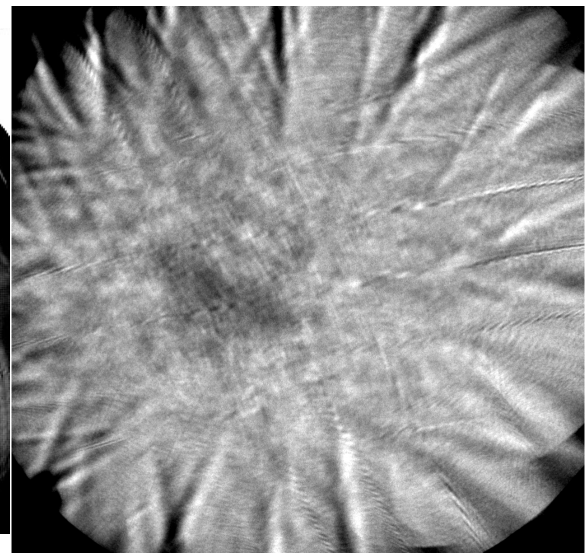

Fig. 2**:** Similar achromatic lines may be obtained with a Schottky emitter, here on a non-corrected Tecnai F20 at LBNL, on silicon (110).

Table 1**:** Results of the test of the "achromatic tuning" algorithm. The indicated target values are the non-round aberration coefficients measured with the Nion tuning algorithm. Cases A and B correspond to consecutive measurements, in an identical situation, on a silicon sample. Cases C and D correspond to measurements on a GaN sample for a well-tuned and badly-tuned system respectively.

	Target A and B	Measured A	Measured B	Target C	Measured C	Target D	Measured D
$C_{1,2a}\ (m)$	1.25×10^{-9}	3.42×10^{-9}	-7.37×10^{-10}	-2.94×10^{-10}	1.97×10^{-9}	6.97×10^{-8}	1.79×10^{-8}
$C_{1,2b}\ (m)$	6.49×10^{-9}	-4.75×10^{-9}	9.80×10^{-10}	-6.49×10^{-10}	-1.41×10^{-10}	-1.66×10^{-8}	9.40×10^{-9}
$C_{2,1a}\ (m)$	-1.18×10^{-6}	-4.02×10^{-7}	1.16×10^{-6}	-2.09×10^{-7}	-8.63×10^{-7}	-1.25×10^{-6}	-3.70×10^{-6}
$C_{2,1b}\ (m)$	-2.84×10^{-6}	6.21×10^{-7}	5.11×10^{-7}	-1.00×10^{-7}	-3.13×10^{-7}	-1.03×10^{-6}	9.18×10^{-7}
$C_{2,3a}\ (m)$	-4.57×10^{-7}	3.90×10^{-7}	1.54×10^{-7}	7.67×10^{-8}	-7.77×10^{-8}	-1.82×10^{-7}	-7.13×10^{-7}
$C_{2,3b}\ (m)$	-3.34×10^{-7}	-5.49×10^{-7}	-4.75×10^{-7}	-3.94×10^{-8}	9.64×10^{-8}	1.20×10^{-6}	1.51×10^{-6}
$C_{3,2a}\ (m)$	1.24×10^{-5}	-1.12×10^{-4}	-5.29×10^{-5}	3.96×10^{-6}	-2.46×10^{-6}	4.69×10^{-5}	7.23×10^{-7}
$C_{3,2b}\ (m)$	-1.36×10^{-5}	4.81×10^{-5}	1.41×10^{-4}	-1.19×10^{-6}	4.68×10^{-6}	8.54×10^{-6}	-4.08×10^{-5}
$C_{3,4a}\ (m)$	1.08×10^{-6}	7.43×10^{-5}	3.47×10^{-5}	4.83×10^{-6}	2.22×10^{-6}	8.59×10^{-6}	-8.36×10^{-6}
$C_{3,4b}\ (m)$	-2.99×10^{-6}	1.83×10^{-5}	1.06×10^{-5}	6.53×10^{-6}	5.14×10^{-6}	5.94×10^{-6}	3.85×10^{-6}

https://doi.org/10.1017/S1431927606067511 Published online by Cambridge University Press