A Method to Characterize and Correct Elliptical Distortion in Electron Diffraction Patterns

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Introduction

One of the main obstacles to performing electron crystallography analysis in a TEM is that the acquired electron diffraction data often exhibits some form of distortion introduced by the lens system. Recognizing this problem, Capitani *et al.*[1] has proposed a method to detect such distortion, which is primarily elliptical, by using a single crystal standard. Once such elliptical distortion is characterized, electron diffraction data acquired later can then be corrected by means of image processing. However, it may be desirable to correct such distortion at the instrument level. In this article, a different approach to measuring diffraction elliptical distortion is proposed by characterizing diffraction ring patterns and it is demonstrated that by varying the objective lens stigmation settings, it is possible to eliminate this elliptical distortion completely.

Obtaining "Good" Diffraction Rings – How to Adjust Diffraction Stigmation

The first step to obtain sharp and continuous diffraction rings is to have a good sample. In this study, a nickel oxide sample with a fine grain size is used (specifically the NiOX standard [2], Fig. 1). Other samples such as sputtered Au particles on a carbon film can also be used as long as the specimen can produce sharp and continuous rings. The microscope used in this study is a JEOL2010F TEM. "Parallel" illumination conditions can be achieved by using (1) a small condenser aperture (20um) and spreading the beam, or (2) a suitably sized selected area aperture. Standard TEM alignment procedures were performed prior to diffraction pattern acquisition with special attention on sample eucentric height, voltage/current centering, and condenser aperture centering. Diffraction patterns were recorded with a Gatan CCD camera.

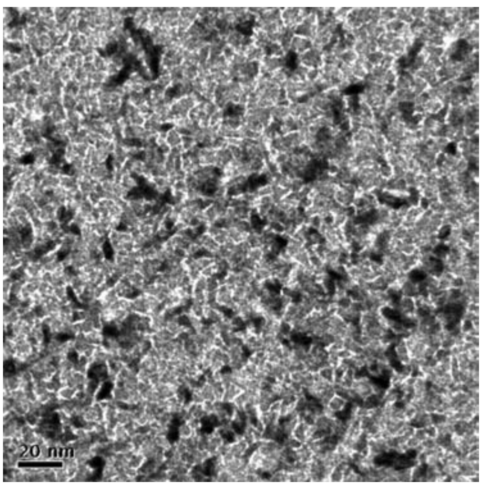

***Fig. 1**. TEM bright-field image of NiOx sample used as diffraction ring standard. It is noted that the dense and small grain size (≈10nm) of this Ni oxide film results in continuous diffraction ring pattern (Fig. 2a, 5a and 7a).*

Fig. 2a shows a "not-yet-focused" ring pattern. For the case where the diffraction lens stigmation (Diff-Stig) has not been adjusted correctly, the pattern in fig. 2b will result. If the Diff-Stig is well adjusted, the rings will be focused in all directions and Fig. 2c is obtained. Readers are encouraged to try adjusting Diff-Stig and focusing to get a feel for these adjustments. It is important to note that the primary effect of adjusting Diff-Stig is not to change the shape of diffraction rings but to change diffraction lens focus in various directions.

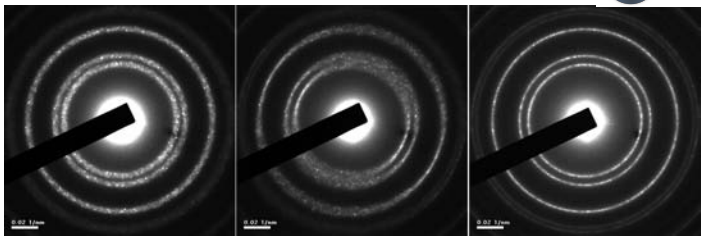

***Fig. 2**. Diffraction ring patterns with different diffraction stigmation and focus conditions: (a) not focused, (b) focused but stigmation not optimized, and (c) focused and stigmation optimized so the rings are in focus in all directions.*

A better way, to optimize Diff-Stig is to utilize the "caustic spot" resulting from spherical aberration of the objective lens. In this adjustment, no sample is needed, the largest condenser aperture should be used, and condenser 2 lens current maximized. This produces an illumination with a very large incident angle (alpha). Fig. 3 show a typical caustic spot pattern as observed in diffraction mode with an appropriate diffraction lens defocus value. Optimized Diff-Stig is achieved when the caustic spot pattern is adjusted from Fig. 3a to Fig. 3b. Diff-Stig settings obtained by this caustic spot method will also result in well-focused ring patterns. In addition, Diff-Stig adjustment is much more sensitive utilizing the caustic spot method compared to the "focusing the diffraction rings method." Therefore, the caustic spot method can be considered as the "fine" adjustment for diffraction lens stigmation.

Last, it is very important to note that in almost all TEM's, the objective lens stigmation (Obj-Stig) setting remains unchanged when the microscope is switched from imaging mode to diffraction mode. Usually the objective stigmators are set to yield a stigmation free image. The diffraction ring patterns acquired, after adjusting Diff-Stig, with such Obj-Stig setting are said to be in an "imaging-optimal" setting. As shown later, the Obj-Stig setting has a direct effect on the elliptical distortion of diffraction patterns so it is important to keep track of the Obj-Stig settings when diffraction patterns are acquired.

Distortion? What distortion?

The advantage of using diffraction ring patterns, as compared to spot patterns from a single crystal, is because it is easier to identify elliptical distortion. This is true only when the distortion is pronounced. Fig. 4a shows a software generated "circle" with large distortion. It is easy to identify its center and to estimate the lengths

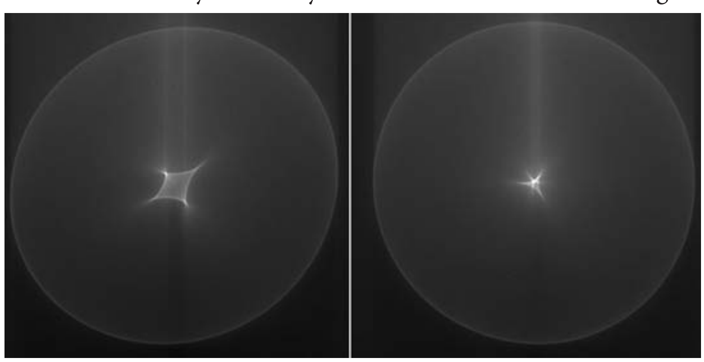

***Fig. 3**. Images of "caustic spot" in diffraction mode with different diffraction stigmation settings: (a) diffraction stigmation not optimized, and (b) diffraction stigmation optimized. Diffraction lens is intentionally defocused in order to show the patterns more clearly.*

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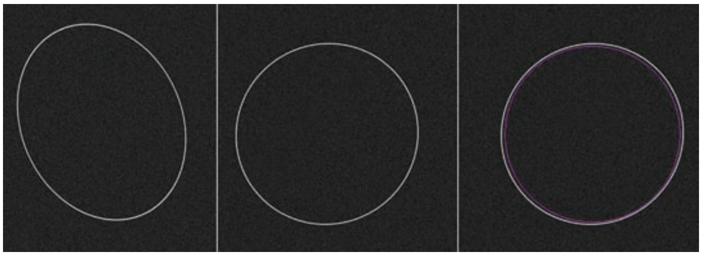


Fig. 4. Software generated ellipses. Both images are of size 512x512 pixels. (a) Ellipse with axes of 175±20 pixels and long axis at 65° CW from horizontal. (b) Ellipse with axes of 175±2 pixels and long axis at 30° CCW from horizontal. (c) Same as (b) but superimposed with a reference circle to help identify the elliptical distortion

of two axes. However, the same cannot be said to Fig. 4b. Without any visual reference, it is difficult to recognize whether or not elliptical distortion exists. By overlaying a reference circle to the ring, as shown in Fig. 4c, it is then possible to recognize and characterize such distortion. There are two major problems with this manual approach of distortion characterization. First, the results are quite subjective to the person doing the measurement. Secondly, the results will be less accurate as the distortion amount gets smaller. It is then necessary to use a software routine in order to get precise and consistent measurements of elliptical distortion.

Elliptical Distortion Characterization by a DigitalMicrograph™ Script

A software routine, written in DigitalMicrograph script language, is developed specifically to characterize elliptical distortion. The analysis of diffraction ring distortion is carried out in the *r*-θ polar coordinate system, as suggested by Braidy[3]. The process involves (1) digitizing points on a selected ring, (2) finding the center position of the ring, and (3) fitting an ellipse to the ring. This software, including source code and user instructions, can be downloaded from the DigitalMicrograph Script Database[4] . The algorithms used in the software are briefly described here:

(1) Digitize a diffraction ring of interest

A diffraction ring pattern obtained under the "imaging-optimal" settings is shown in Fig. 5a. First, this pattern is transformed from a *X*-*Y* Cartesian coordinate system into a *r*-θ polar coordinate system with respect to a specified point as the coordinate system origin, as shown in Fig. 5b. At present, an angular resolution of 0.5 degree per pixel is chosen so there are 720 pixels in the *r* coordinate. One of these diffraction rings, which is to be fitted to an ellipse later, can then be selected in this *r*-θ image by a rectanglular region of interest. In this pattern, the third ring from center is selected for analysis. This ring is digitized as an *r*-θ curve by finding the pixel with maximum intensity at each given θ. More details on this digitizing algorithm are given in the software's user instructions.

(2) Refine/find diffraction ring center

When an ellipse is transformed to *r*-θ space with respect to its center, the difference between *r*(θ) and *r*(θ+π) will be zero. Therefore, the center of the experimental ring can be obtained by minimizing this difference with iterations on different center locations. A fit parameter for finding center points is then defined as

$$\chi^2_{(center)} = \sum_i \frac{\left[r_i(\theta_i) - r_i(\theta_i + \pi)\right]^2}{r_i(\theta_i)} .$$

After the center location is found (*i.e.* where the minimum value of χ^2 is obtained), the final *r*-θ curve would represent the

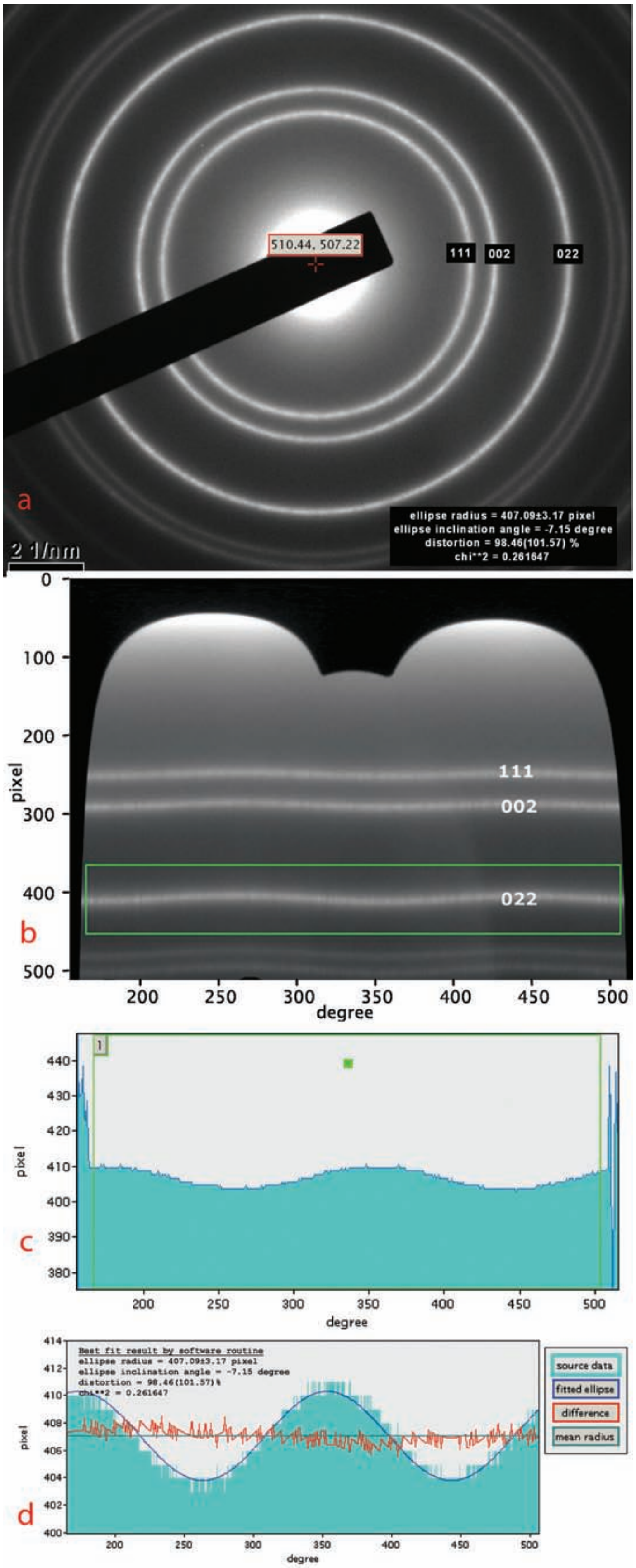
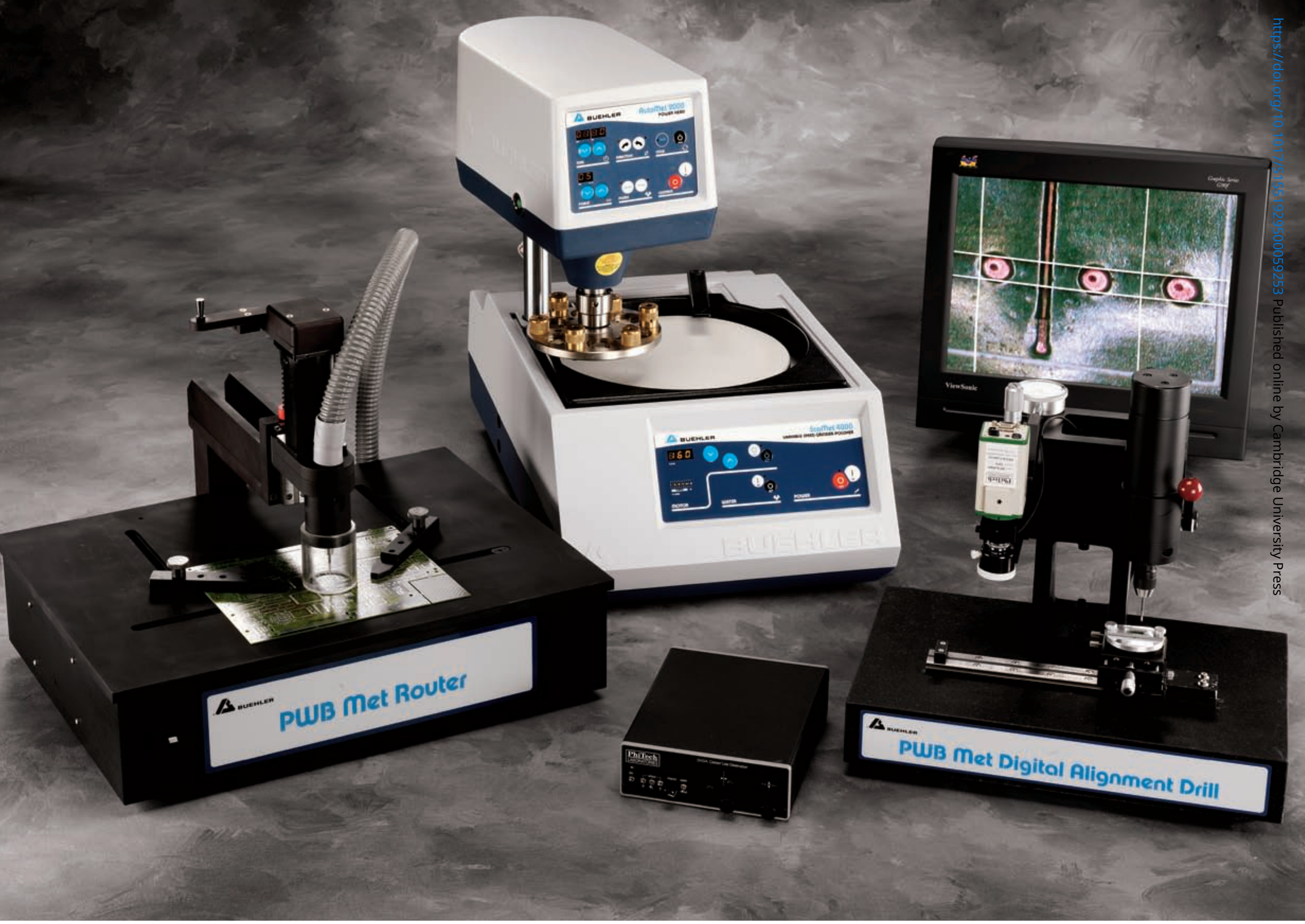


Fig. 5. Experimental and distortion analysis results obtained with objective stigmation set to "imaging optimal" condition: (a) Diffraction ring pattern, (b) the r-θ image transformed w.r.t. center position of (510.44, 507.22), (c) the r-θ curve of third ring, d_{022}, and (d) distortion analysis results by software routine. Note that an elliptical ring will result in a sinusoidal curve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experimental diffraction ring. In this case, it is the r-θ curve shown in Fig. 5c. Please note that a shift of θ has been applied to the r-θ image to move the shadow of beam stopper to the edge.

(3) Fitting the ellipse and characterizing distortion

Fitting an ellipse to the experimental r-θ curve is straightforward after the center is determined. An ellipse function in r-θ space can be expressed as:

$$\rho_i(\theta_i) = \left(R^2 - \Delta R^2\right) \Big/ \sqrt{R^2 + \Delta R^2 - 2 \cdot R \cdot \Delta R \cdot \cos\left[2\left(\theta_i + \alpha\right)\right]}$$

where α, R and ΔR are parameters pertaining to the ellipse: α as the ellipse inclination angle (rotation of major axis counter-clockwise from horizontal), $R+\Delta R$ and $R-\Delta R$ the half length of major and minor axis, respectively. The distortion is defined as the ratio of minor axis to major axis, i.e. $(R-\Delta R)/(R+\Delta R)$. The goodness-of-fit ($\chi^2$) is defined as

$$\chi^2_{(\alpha,R,\Delta R)} = \sum_i \frac{\left[r_i(\theta_i) - \rho_i(\theta_i)\right]^2}{\rho_i(\theta_i)}$$

Once a minimum χ^2 is achieved by iterations of α, R and ΔR, the elliptical distortion of the diffraction ring is then characterized. In addition to the numerical output, the fitting results are also displayed graphically (Fig. 5d) so users can examine the results visually. As shown in this figure, the experimental data agrees with the fitted ellipse very well. Under this "imaging-optimal" condition, the elliptical distortion is found to be 98.46% with an inclination angle of 7.15 degree (defined as the clockwise rotation of the long axis from the horizontal).

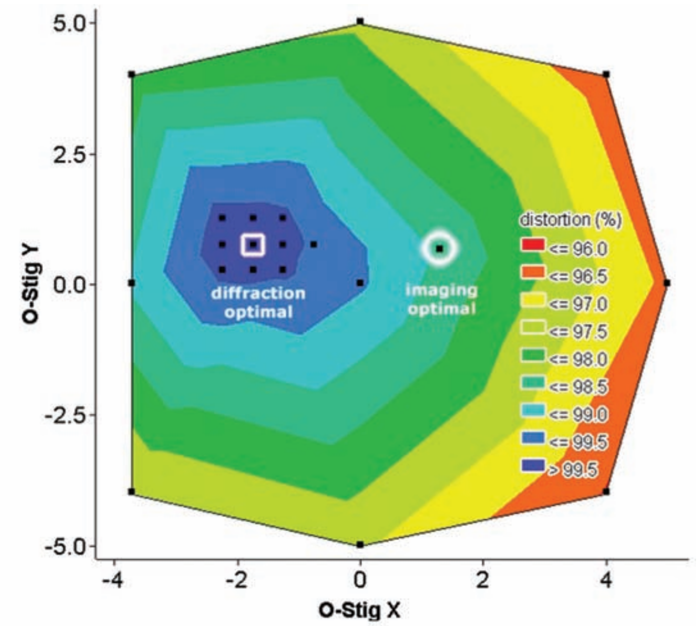

Fig. 6. *2D contour chart from data points in Table 1. The "center of gravity" at which least distortion located can be clearly visualized. It is also apparent that "imaging-optimal" setting is quite different than "diffraction-optimal" setting.*

This software also includes a routine to create test patterns so users can create rings with desirable elliptical distortion. In fact, the patterns in Fig. 4 were created with this software. Users are strongly encouraged to try the analysis routine on simulated patterns first before moving on to actual experimental data.

Correcting Elliptical Distortion in Diffraction

Once the distortion is characterized accurately, it can be cor-

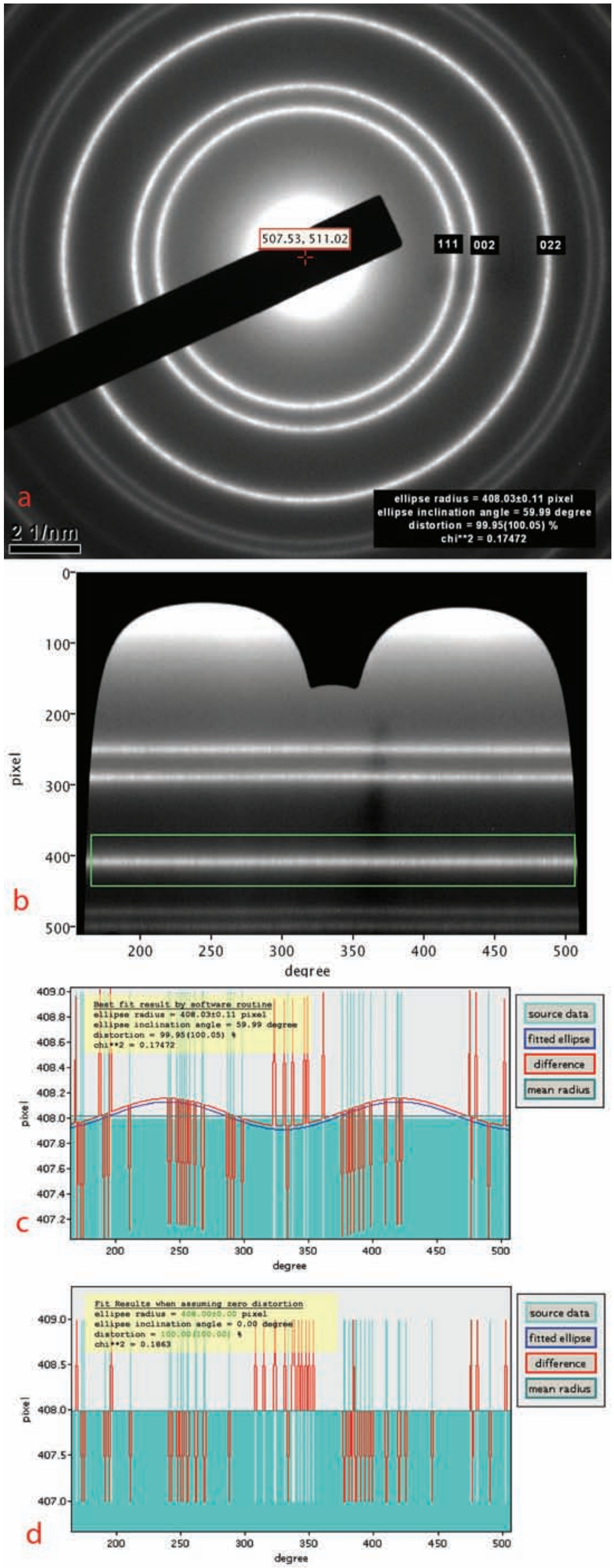

Fig. 7. *Experimental and distortion analysis results obtained with objective stigmation set to "diffraction optimal" condition: (a) Diffraction ring pattern, (b) the r-θ image transformed w.r.t. center position of (507.53, 511.02), (c) distortion analysis results by software routine, and (d) fitting result when assuming zero distortion. Note that true circular rings will result in a straight lines as shown in (b).*

rected by simple image processing methods such as rotate and resize. Alternatively, a "hardware" solution is shown here that can yield a distortion free diffraction pattern directly without the need of image processing. This solution is based on the finding that the elliptical distortion in a diffraction pattern is related to the objective lens stigmation (Obj-Stig). Therefore, a trial-and-error experiment can be performed to find an "optimized" Obj-Stig setting that can result in a distortion free diffraction ring pattern. Each iteration consists of three main steps, (1) set the Obj-Stig arbitrarily (starting from the "imaging-optimal" setting), (2) adjust the Dif-Stig accordingly, and then (3) measure the elliptical distortion. Results of such a trial-and-error optimization experiment (utilizing a JEOL 2010F TEM) are shown in Table 1. These data can also be displayed as a 2D contour map (Fig. 6). There is clearly an Obj-Stig setting where distortion-free diffraction patterns can be obtained. This setting is then named "diffraction-optimal," to differentiate it from the "imaging-optimal" Obj-Stig setting. It is expected that each TEM will have its own "diffraction-optimal" setting, which needs to be determined individually.

O-Stig X (kV)	O-Stig Y (kV)	distortion (%)
1.28	0.64	98.44
-3.71	4.00	97.76
-3.71	0.00	98.58
-3.71	-4.00	96.99
0.00	5.00	97.38
0.00	0.00	99.07
0.00	-5.00	97.09
4.00	4.00	96.23
5.00	0.00	96.38
4.00	-4.00	95.96
-2.25	1.25	99.71
-2.25	0.75	99.80
-2.25	0.25	99.61
-1.75	1.25	99.71
-1.75	0.75	99.95
-1.75	0.25	99.66
-1.25	1.25	99.63
-1.25	0.75	99.68
-1.25	0.25	99.58
-0.75	0.75	99.41

***Table 1**. Distortion as function of objective lens stigmation. "Imaging-optimal" setting is highlighted in green color and "diffraction-optimal" setting is highlighted in blue color (from a JEOL2010F TEM).*

A diffraction ring pattern obtained in this setting, and its r-θ transformed pattern and ellipse fitting results are shown in Fig. 7. It can be clearly seen that in the r-θ pattern (Fig. 7b) all the "rings" are transformed into straight lines, indicating they are indeed perfect circles. The final fitting results are shown in Fig. 7c and the numerical data show that there is still some residual distortion (ΔR= 0.11 pixel). However, the graphical presentation of the fitting results indicates that the residual distortion may stem from inherent noise in the experimental data. When forcing the distortion to be zero (*i.e.* ΔR= 0.0 pixel), the final goodness-of-fit, χ^2= 0.186, is still comparable with the software-derived χ^2 (0.175) with ΔR= 0.11. In addition, there is no significant discrepancy when comparing the graphical representations of the fitting result (*i.e.* comparing Fig 7d to Fig 7c). Therefore, for practical purposes, this diffraction ring pattern can be considered to be distortion free.

It is important to reiterate that the Obj-Stig setting in the "diffraction-optimal" condition is quite different than that in the "imaging-optimal" condition, so TEM images taken at this "diffraction-optimal" condition may exhibit strong stigmation. Therefore, the Obj-Stig should be set according to the microscope operation mode; "imaging-optimal" in imaging mode or "diffraction-optimal" in distortion-free diffraction mode.

Final Notes

In general, in a well-aligned TEM image, stigmation does not deviate much from the optimum condition over time. A touch-up adjustment of Obj-Stig is only needed when there is a need to get images with best quality. Hence, it is expected that the "diffraction-optimal" setting for Obj-Stig should also be stable. There should be no need to run the complete optimization routine before each diffraction experiment. Indeed, this is confirmed by the authors during a one-month period, where the distortion remained better than 99.5% with the same "diffraction-optimal" setting. However, since the distortion of the diffraction pattern is not easily recognized, it is necessary to check this distortion before the experimental session from time to time. It should also be noted that normal "image-optimal" Obj-Stig setting is sensitive to the beam tilt so cares must be taken to ensure the beam tilt (current/voltage centering) is consistent between optimization and experimental sessions.

Practically, this instrumental level distortion correction may not be necessary and the post-experiment correction may be sufficient. All it needs is to acquire a diffraction ring pattern at camera lengths of interest, characterize the distortion, and apply the correction via image processing. However, the direct correction method is preferred when the diffraction pattern is further "imaged" by another device such as a post column image filter (*e.g.* a Gatan Image Filter). In this case, the distortion in the final diffraction pattern is a convolution of distortion from TEM and the distortion from the GIF. Minimizing distortion in the diffraction pattern before it enters the GIF will have a great effect on the quality of energy-filtered CBED patterns[5].

It is shown here that by adjusting Obj-Stig it is possible to eliminate elliptical distortion in diffraction patterns. It is then natural to ask the obvious question: will adjusting the Diff-Stig also eliminate elliptical distortion in images? TEM images can be "distorted" by adjusting Diff-Stig. Therefore, it can be expected there should be an optimal Diff-Stig setting that would eliminate elliptical distortion in images. Work is under way to verify this conjugate method of correcting distortion in images. ■

References:

1. G.C. Capitani, P. Oleynikov, S. Hovmöller, M. Mellin, *Ultramicroscopy*, 106 (2006) 66.
2. R.F. Egerton, S.C. Cheng, *Ultramicroscopy*, 55 (1995) 43.
3. N. Braidy, private communication in the forum of DigitalMicrograph Script User Group (http://lists.asu.edu/archives/dmsug.html), 2006
4. DigitalMicrograph Script Database, "http://www.felmi-zfe.tugraz.at/dm_scripts"; URL to the software used in this article: "http://www.felmi-zfe.tugraz.at/dm_scripts/dm_scripts/freeware/programs/Diffraction-Rings-Distortion-Analysis.htm".
5. Vincent. D.-H Hou, "Distortion Minimization of CBED Patterns in TEM", paper submitted to MM2008 meeting.