

## A NEW METHOD FOR COMA-FREE ALIGNMENT OF TRANSMISSION ELECTRON MICROSCOPES AND DIGITAL DETERMINATION OF ABERRATION COEFFICIENTS

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### Abstract

A simple method of finding the coma-free axis in electron microscopes with high brightness guns and quantifying the relevant aberration coefficients has been developed which requires only a single image of a monocrystal. Coma-free alignment can be achieved by means of an appropriate tilt of the electron beam incident on the crystal. Using a small condenser aperture, the zone axis of the crystal is adjusted parallel to the beam and the beam is focused on the observation plane. In the slightly defocused image of the crystal, several spots representing the diffracted beams and the undiffracted one can be detected. Quantitative values for the beam tilt required for coma-free alignment, as well as for the aberration coefficients of interest, can directly be obtained from measured coordinates of spots that correspond to diffraction angles of equal sizes.

**Key Words:** Coma-free alignment, wave aberration, defocus, spherical aberration, coma, astigmatism, aberration curve, beam tilt, diffraction spots, spot position.

### Introduction

Electron micrographs are affected by coma when the direction of illumination does not coincide with the direction of the true optical axis of the electron microscope. Hence, elimination of coma is of great importance in high-resolution work. With increasing resolution, the effects of three-fold astigmatism and other axial aberrations grow also in importance. To reduce the effect of aberrations, methods for coma-free alignment of electron microscopes and accurate determination of aberration coefficients are, therefore, required.

A method for coma-free alignment was originally proposed in an early paper by Zemlin *et al.* (1978). In this paper, the three-fold astigmatism had also been taken into account. The proposed method is somewhat tedious, however, since it is based on the production and evaluation of several optical diffractograms. When modern electronic cameras and image-processing systems are employed, the process of coma-free alignment can be performed automatically. A simple method of finding the coma-free axis and quantifying the relevant aberration coefficients has now been developed which is an extension of our previous on-line alignment method (Lauer and Ade, 1994). In contrast to other methods (Heinemann, 1971; Koster and de Ruijter, 1992; Krivanek and Fan, 1992; Krivanek, 1994; Ishizuka, 1994), where the information is used which is contained in beam-tilt-induced image shifts or in astigmatism and defocus changes in a set of images or diffractograms, the new method requires only a single image of a monocrystal with known lattice plane parameters, such as a gold foil in [002] orientation. In the following, the mathematical background of the method is presented, and some preliminary results are given.

### Principle of the Method

We take advantage of the high brightness of field emission guns to form tiny probes with high intensities. Using a small condenser aperture (e.g., 30  $\mu\text{m}$  in diameter) the zone axis of the crystal is adjusted parallel to the beam, and the crystal is imaged with a magnification of at least 100 000 times onto the observation plane. If the beam is focused

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on this plane and a slight defocus (up to 1  $\mu\text{m}$  overfocus) is induced by the objective lens, several spots representing the diffracted beams and the undiffracted one can be observed in the image. In a well-aligned microscope, all spots corresponding to diffraction angles of equal size will be located on a circle with the undiffracted beam in its center (Fig. 1a). When two-fold astigmatism is present, this circle degenerates into an ellipse, but the undiffracted beam remains a center of symmetry (Fig. 1b). This symmetry is lost, however, when coma and three-fold astigmatism are present. The diffracted beams then arrange in an asymmetrical oval shape around the undiffracted beam (Figs. 1c, d).

Coma-free alignment can simply be achieved by an appropriate tilt of the electron beam incident on the specimen. As schematically depicted in Figure 2 and theoretically explained in the following section, the spot coordinates are used to determine the amount and orientation of the required beam tilt and to obtain quantitative values for the aberration coefficients of interest.

### Theoretical Background

To review the underlying theory, we start with the expression for the wave aberration  $W$  which describes the phase of the complex transfer function for the object wave. Using complex notations, and taking third-order spherical aberration and all axial aberrations up to the third order of the diffraction angle  $\alpha$  into account, the wave aberration can be written as

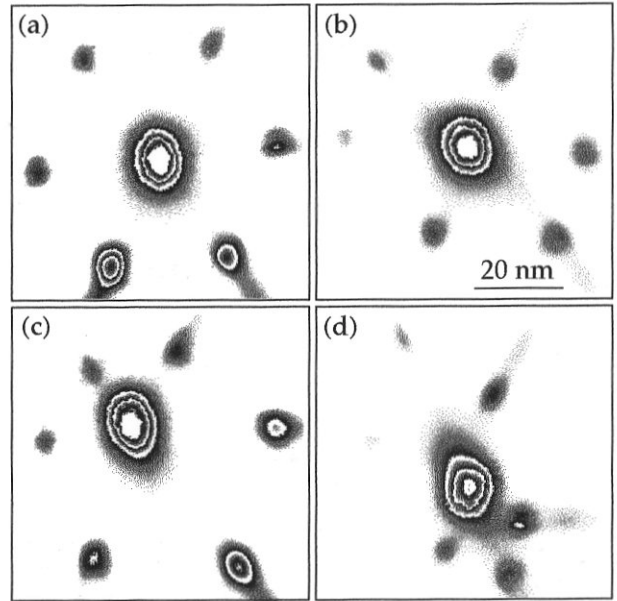
$$W(u) = (1/\lambda) \{ C_s u^2 \underline{u}^2 / 4 + \Delta z u \underline{u} / 2 + \text{Re}(A_2 \underline{u}^2 / 2 + B u \underline{u}^2 + A_3 \underline{u}^3 / 3 + \mathbf{x}_v \underline{u}) \} \quad (1)$$

in terms of the complex variable

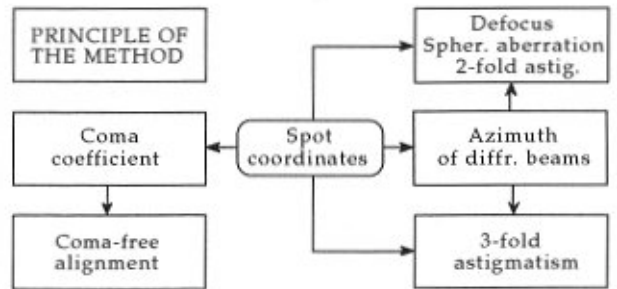
$$u = u_x + i u_y = \alpha \exp(i\phi) \quad (2)$$

where  $\phi$  represents the azimuth of the diffracted beam (see, e.g., Zemlin *et al.*, 1978). In Equation (1),  $\lambda$  is the electron wavelength,  $C_s$  the coefficient of spherical aberration, and  $\Delta z$  the defocus. The complex coefficients  $A_2$ ,  $A_3$ ,  $B$ , and  $\mathbf{x}_v$  represent the (normal) two-fold astigmatism, three-fold astigmatism, axial coma, and image shift, respectively. Underlining is used here to denote complex conjugation, i.e.,  $\underline{u}$  is the complex conjugate of  $u$ . The complex notation is particularly useful when dealing with tilted illumination. If a beam tilt (misalignment)  $u_k$  is introduced, for example, the modified wave aberration can directly be obtained from Equation (1) by replacing the variable  $u$  with  $u + u_k$ .

As already mentioned, the procedure of finding the coma-free axis and quantifying the aberration coefficients is based on the determination of the positions of the



**Figure 1.** Effect of different aberrations on the arrangement of diffraction spots in the image plane of a silicon crystal with zone axis in [111] orientation. To visualize the true positions of the spots, a pseudocontour contrast mode was selected. (a) Microscope aligned for coma and two-fold astigmatism. (b)-(d) Effect of two-fold astigmatism, coma, and coma and astigmatism, respectively.



**Figure 2.** Schematic diagram of the digital procedure for coma-free alignment and determination of aberration coefficients.

diffracted spots in the image plane of the crystal. These positions can easily be calculated from Equation (1) by differentiation with respect to the variables  $u_x$  and  $u_y$ . If the complex notation is retained and a misalignment  $u_k$  is taken into account, the following result is obtained for the spot position  $r(u)$  of the diffracted beam relative to that of the undiffracted one:

$$r(u) = C_s \underline{u} u^2 + \Delta z_c u + A_{2c} \underline{u} + 2B_c \underline{u} u + B_c u^2 + A_3 \underline{u}^2 \quad (3)$$

where

$$\Delta z_e = \Delta z + 2C_s u_k u_k + 2(B u_k + \underline{B} u_k) \quad (4a)$$

(defocus)

$$A_{2e} = A_2 + C_s u_k^2 + 2B u_k + 2A_3 u \quad (4b)$$

(2-fold astigmatism)

$$B_e = B + C_s u_k \text{ (coma)} \quad (4c)$$

Note that the effective (or apparent) aberration coefficients given in Equation (4) are functions of the misalignment  $u_k$ .

### Aberration figures

Equation (3) can be used to calculate the geometric curves determined by different aberrations. For this purpose, it is necessary to express the variable  $u$  by the diffraction angle  $\alpha$  and the azimuth  $\phi$  according to Equation (2). This leads to the result

$$r(\alpha, \phi) = (C_s \alpha^3 + \Delta z_e \alpha) \exp(+i\phi) + A_{2e} \alpha \exp(-i\phi) + \alpha^2 [2B_e + \underline{B}_e \exp(+i2\phi)] + A_3 \alpha^2 \exp(-i2\phi) \quad (5)$$

If we consider diffracted beams with a fixed value of  $\alpha$ , the azimuth  $\phi$  varies between zero and  $2\pi$ . The corresponding spots in the image plane are then expected to lie on curves whose shape strongly depends on the type of aberrations involved. As stated in the legend of Figure 3, spherical aberration and defocus lead to a circular aberration curve. The circle becomes an ellipse, however, if two-fold astigmatism is also present. Furthermore, a „three-pointed“ star (or oval) is obtained if three-fold astigmatism (or coma) is considered instead of two-fold astigmatism. Finally, the aberration curve assumes a somewhat complex form if all aberrations are taken into account.

### Coma-free alignment

According to Equation (4c), a misalignment  $u_k$  leads to an effect similar to that of the axial coma  $B$  of the objective lens. Hence, coma-free alignment can simply be achieved by introducing an additional beam tilt which results in a vanishing effective coma  $B_{er}$ . Thus, if we require that

$$B_{er} = B_e + C_s u_\tau = 0 \quad (6)$$

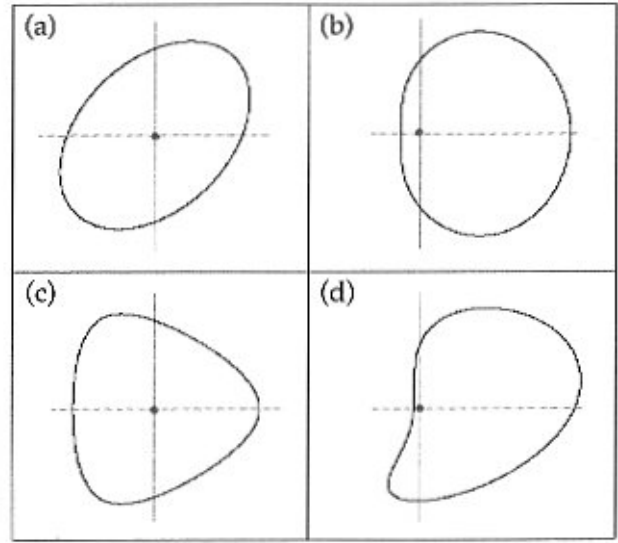
the coma-free direction of the incident beam is given by:

$$u_\tau = -B_e / C_s \quad (7)$$

To determine the amount and orientation of this direction, numerical values for the coefficients  $B_e$  of apparent coma and  $C_s$  of spherical aberration are required.

### Determination of aberration coefficients

Examination of Equation (3) reveals that the first three



**Figure 3.** Aberration curves in the presence of different aberrations. If only defocus and spherical aberration are present, the aberration curve is expected to be circular. The circle changes to an ellipse (a) if two-fold astigmatism is included. It assumes the shape of an oval (b) or three-pointed star (c) if either coma or three-fold astigmatism is taken into account. An aberration curve of somewhat complicated shape (d) is obtained if all aberrations mentioned are present.

terms are odd functions of the variable  $u$  while the remaining terms are even functions of  $u$ . By proper combination of the spot positions corresponding to  $+u$  and  $-u$ , the two groups can, therefore, be separated.

In the following, we assume that the crystal foil employed is oriented in such a way that at least two sets of crossed lattice planes can be viewed. For simplicity, we require that the lattice planes of each set are orthogonal and have the same spacing. Different sets may have different spacings. Since each set of parallel lattice planes leads to a pair of diffraction spots, corresponding to  $\pm u$ , two pairs of spots corresponding to each orthogonal set of lattice planes can be found in the image. In an ideal microscope, these spots will be arranged in a quadratic raster. Due to aberrations, however, a distorted spot pattern is usually observed (Fig. 4).

### Determination of coma and three-fold astigmatism

As depicted in Figures 4 and 5, we denote the positions of one pair of spots by  $r_1(+u)$  and  $r_3(-u)$ . The spot positions of the other pair are denoted by  $r_2(+u)$  and  $r_4(-u)$ . By means of Equation (3), we then have

$$(r_1 + r_3)/2 = 2B_e u u + \underline{B}_e u^2 + A_3 u^2 \quad (8)$$

and

$$(r_2 + r_4)/2 = 2B_e u - B_e u^2 - A_3 u^2 \quad (9)$$

From these equations, the aberration coefficients  $B_e$  and  $A_3$  can easily be calculated. If we express the variable  $u$  by means of the diffraction angle  $\alpha$  and the azimuth  $\phi$  according to Equation (2), we obtain the results

$$B_e = (r_1 + r_2 + r_3 + r_4)/8\alpha^2 \quad (10)$$

for the coma and

$$A_3 = \{2(r_1 - r_2 + r_3 - r_4) \exp(i2\phi) - (r_1 + r_2 + r_3 + r_4) \exp(i4\phi)\}/8\alpha^2 \quad (11)$$

for the three-fold astigmatism. In contrast to  $B_e$ , the coefficient  $A_3$  is seen to be dependent on the azimuth  $\phi$ . However, this is only an apparent dependency since  $\phi$  can be expressed as

$$\phi = \arctan\left\{\frac{\text{Im}[(r_1 - r_3) - i(r_2 - r_4)]}{\text{Re}[(r_1 - r_3) - i(r_2 - r_4)]}\right\} \quad (12)$$

by means of the spot positions under consideration.

#### Determination of two-fold astigmatism, defocus, and spherical aberration

As in the preceding subsection, we start with Equation (3). By proper combination of the spot positions corresponding to  $+u$  and  $-u$ , we obtain the following equations:

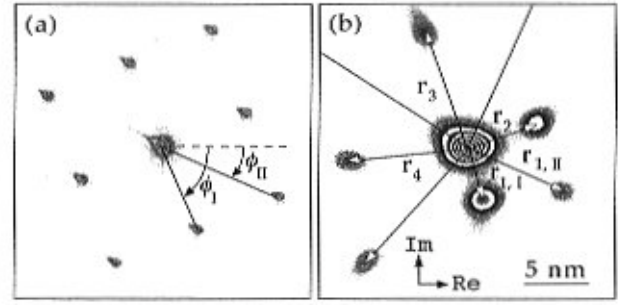
$$A_{2e} = (1/4\alpha)[(r_1 - r_3) + i(r_2 - r_4)] \exp(+i\phi) \quad (13)$$

$$C_s \alpha^3 + \Delta z_e \alpha = (1/4)[(r_1 - r_3) - i(r_2 - r_4)] \exp(-i\phi) \quad (14)$$

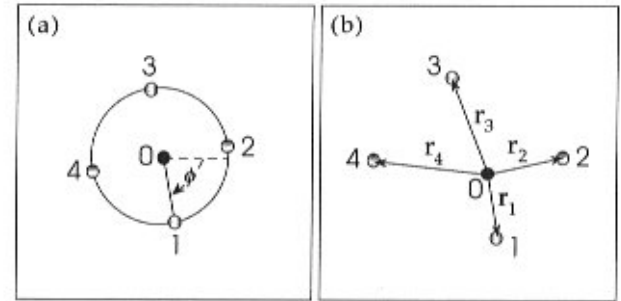
While the coefficient  $A_{2e}$  of two-fold astigmatism can directly be calculated by means of the diffraction angle  $\alpha$  and the measured spot positions  $r_1$  to  $r_4$ , the coefficients  $\Delta z_e$  of apparent defocus and  $C_s$  of spherical aberration cannot be determined independently. To remove this difficulty, a second set of spots which correspond to a different diffraction angle  $\alpha_{II}$  is needed. The diffraction angle  $\alpha_{II}$  and the azimuth  $\phi_{II}$  of the second set of spots can in general be expressed as

$$\alpha_{II} = m\alpha; \phi_{II} = \phi + \beta \quad (15)$$

in terms of the parameters  $\alpha$  and  $\phi$  from the first set used above. If a gold foil in [002] orientation is used as a specimen, for example, the numerical values  $m = \sqrt{2}$  and  $\beta = \pi/4$  are valid (Fig. 4). In analogy to Equation (14), we can therefore



**Figure 4.** Diffraction images of a gold foil in [001] orientation. To visualize the true positions of the spots, a pseudo-contour contrast mode was selected. (a) Usual diffraction image resulting in the back focal plane of the objective lens. (b) Diffraction image obtained in a slightly defocused image plane.



**Figure 5.** Spot positions in the image plane of a crystal foil with two orthogonal sets of lattice planes with the same spacing (schematically). (a) Without coma and astigmatism, (b) In the presence of coma and astigmatism.

write

$$m^3 C_s \alpha^3 + m \Delta z_e \alpha = (1/4)\{(r_1 - r_3) - i(r_2 - r_4)\}_{II} \exp[-i(\phi + \beta)] \quad (16)$$

From these equations we then obtain directly the results

$$\Delta z_e = -\frac{(m^2/[4(m^2 - 1)\alpha])\{\text{Re}[(r_1 - r_3) - i(r_2 - r_4)]_{II} / [m^3 \cos(\phi + \beta)] - \text{Re}[(r_1 - r_3) - i(r_2 - r_4)]_I / \cos(\phi)\}}{m^2} \quad (17)$$

for the apparent defocus and

$$C_s = \frac{(1/[4(m^2 - 1)\alpha^3])\{\text{Re}[(r_1 - r_3) - i(r_2 - r_4)]_{II} / [m \cos(\phi + \beta)] - \text{Re}[(r_1 - r_3) - i(r_2 - r_4)]_I / \cos(\phi)\}}{m^2} \quad (18)$$



for the spherical aberration. The indices I and II refer to the first and second set of spots, respectively. With these results, we have established a complete set of equations for the determination of the aberration coefficients of interest.

### Experimental Results

In the preceding sections, the theoretical background of the method and the relevant equations for the determination of aberration coefficients have been presented. In the following, some preliminary results will be given.

The proposed method was tested by evaluating images of a gold foil with zone axis in [002] orientation. The images were produced in a Philips electron microscope CM 200 SuperTwin with field emission gun (Philips Electron Optics, Eindhoven, The Netherlands) at an accelerating voltage of 200 kV and were digitally recorded by means of a Gatan slow-scan camera. Subsequently, the images were evaluated with the aid of the DigitalMicrograph image-processing system of Gatan.

The numerical values for the amount and orientation of the beam tilt required for coma-free alignment, which were obtained by means of measured spot coordinates in the foil image, were found to be in accordance with the corresponding values of intentional beam misalignments ( $\approx 5 \times 10^{-4}$ ) induced for test purposes. Furthermore, a numerical value of  $|A_3| = 0.53 \mu\text{m}$  was derived for the coefficient of three-fold astigmatism using the same coordinates. Finally, a spherical aberration coefficient of  $C_s = 1.21 \text{ mm}$  was obtained which agrees well with the value of 1.2 mm given by Philips.

### Summary

We have presented a simple method for coma-free alignment of transmission electron microscopes which requires only a single image of a crystal foil. Using a small condenser aperture, the electron beam is focused on the observation plane and a slight defocus is induced by the objective lens. The coordinates of the diffraction spots occurring in the defocused image of the crystal are the observation plane and a slight defocus is induced by the objective lens. The coordinates of the diffraction spots occurring in the defocused image of the crystal are used to (i) determine the beam tilt required for coma-free alignment and (ii) to obtain numerical values for the aberration coefficients of interest. The determination of the precision attainable with this method and the automation of the whole procedure remain subjects for future work.

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### Discussion with Reviewers

**E. Voelkl:** It would be helpful to have an answer on the following questions: (1) How accurately can the position of the diffracted beams be detected? Is it plus/minus one pixel on the CCD camera? (2) How large and homogeneous does a crystal have to be to allow the suggested procedure? (3) Is this a method that can be applied on an „everyday“ material? (4) Does a bent crystal cause problems when evaluating the aberrations?

**Authors:** (1) According to the possibilities of Digital-Micrograph, the position of the diffracted beams can be determined with an accuracy of plus/minus one pixel. (2) A plan-parallel crystal foil should be used as a specimen. The illuminated area is usually in the nanometer range. (3) Yes. A crystalline gold foil or a very sharp silicon wedge can be used for alignment purposes. (4) A bent crystal is not expected to cause problems since the illuminated area is extremely small. Such a crystal is, on the contrary, very helpful in finding the zone axis.

**K. Scheerschmidt:** Can you briefly characterize the sensitivity of your method with respect to a possible disturbance by the crystalline object given by, e.g.,

dynamical phase shifts and due to the beam tilt?

**Authors:** Dynamical effects and beam misalignments may have an influence on the diffraction angle of the beams if a thick crystal of prismatic shape is employed. In our method we suggest the use of thin plan-parallel crystal foils.

**D. Smith:** Could you provide some estimate of the errors or reproducibility of the parameter measurements?

**M. Kundmann:** Could you give some information regarding the precision or reproducibility of the measurements given in the section entitled „experimental results“?

**Authors:** It has been stated in the text that the determination of the precision attainable with the proposed method is a subject of future work and that the given values 1.21 mm for the spherical aberration coefficient  $C_s$  and 0.53 mm for the coefficient  $|A_3|$  of three-fold astigmatism are only preliminary. However, the measurements were repeated several times using different sample areas. The experimentally obtained results show that the above values for  $C_s$  and  $|A_3|$  can be reproduced with a precision of 0.04 mm and 0.11 mm, respectively.