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Advanced semiconductor characterization with aberration corrected electron microscopes

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Abstract. Spherical aberration (Cs) correctors were demonstrated in the last years of the twentieth century and became commercially available a few years later. In Grenoble, we received our first probe corrector on a TEM/STEM machine in 2006. Cs-correctors have allowed us to improve the spatial resolution and the contrast of high resolution images both in TEM and STEM. The aim of the article is not to give a detailed description of Cs-correctors or a thorough analysis of their pros and cons but to illustrate what the benefits of the Cs-correctors have been in four areas: (i) atomic structure determination, (ii) polarity measurement, (iii) strain determination and (iv) interface analysis. Emphasis is put on the probe corrector although some comments on image correctors are given as well.

1. Introduction

In 1936, Scherzer published a theorem that in electron microscopy, classical lenses with rotation symmetry are condemned to have positive spherical aberration (Cs>0) [1]. After the second world war, more than 10 years later he proposed several ways to build lenses without spherical aberration: non rotationally symmetric lenses, non static lenses, lenses containing space charge or a combination of lenses and mirrors. Soon after, some of his students started to work on the realisation of such Cs-corrected lenses. Later other groups in the world had also some trials (see [2] for a short history), but the real success came more than 50 years later. The first working Cs-corrector was demonstrated on a Scanning Electron Microscopy (SEM) in 1994 [3]. In Scanning Transmission Electron Microscopy (STEM), the first probe Cs-corrector was mounted on a VG microscope by Krivanek et al [4]. Soon thereafter the first image Cs-correctors was working on a Transmission Electron Microscope (TEM) [5] and commercialisation was quickly started. 15 years after the demonstration of the performance of correctors about 400 corrected microscopes are now working in the world. Of course there are great disparities between the different countries. France have 14 microscopes with Cs-correctors, half

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI. Published under licence by IOP Publishing Ltd 1 having probe correctors, half image correctors. England has about the same number, but most of them have a probe corrector, Germany has more than 60 and Italy 2 electron microscopes with Cs-correctors. My laboratory got the chance to have one of the first probe Cs-correctors of the German company CEOS. This Cs-corrector was installed by FEI on our existing TITAN in our laboratory.

The aim of this paper is not to describe in details how correctors work but, after 7 years of Cscorrector operation, to illustrate through four topics what the benefits have been for us of having Cscorrectors. Emphasis will be put on probe Cs-correctors although a few evocations to image Cscorrectors will be made. After a short presentation of Cs-correctors in practice, illustrations and comments will concern

- (i) atomic structure determination,
- (ii) polarity measurement,
- (iii) strain determination and
- (iv) interface analysis.

Most of the presented experimental data have been taken at CEA-Grenoble on three different microscopes: a JEOL 4000EX electron microscope equipped with a LaB₆ filament, a low base FEI TITAN microscope, first without Cs-corrector and then upgraded by FEI in our laboratory with a probe Cs-corrector, and an FEI TITAN Ultimate microscope having a monochromator and both image and probe Cs-correctors. A few tests were also performed using different microscopes in Canada and in the USA.

2. Cs-correctors in practice

Cs-correctors introduce some new complexities in the microscope as several non-symmetric lenses have to be aligned and tuned in order to reduce the aberrations of the objective lens. In fact, a Cscorrector does not only tune Cs to a desired value but also minimizes several low-order aberrations (appendix 1). The tuning is principally done by software. As aberrations are best visible when the beam is tilted out of the optical axis, the strategy of all software is to acquire series of images acquired with different tilts of the beam [5]. Typically, a table of 15 images with tilt angles up to 20 or 30 mrad in at least 8 different directions are taken and a special sample - gold particles on thin amorphous carbon films – is used for that. Hopefully, most of the aberrations are relative stable and only defocus (C1), astigmatism (A1) and coma (B2) (see appendix 1 for a list of aberrations) need a regular tuning if the microscope is kept in a given specific state, i.e. at the same high tension and same imaging technique. Tuning an image corrector is rather fast - about 20 minutes - while a probe corrector requires more time - at least 1 hour for the CEOS corrector. It is not cumbersome to have image and probe correctors on the same microscope as the tunings of both correctors are very similar and tuning the image corrector is done quickly. Up to now, only confocal experiments [6] have used the two correctors simultaneously. In normal use, only one corrector is optically active. Using a dedicated sample is not compulsory if the microscope is kept in a specific state and as there are ways to manually tune A1, C1 and B2 [7]. In our laboratory, where many users are working with different settings (for holography, TEM, STEM, diffraction), we generally prefer to start our session by introducing the 'gold on amorphous carbon' sample, which is constantly kept on a dedicated single tilt holder, and tuning accurately the aberrations.

For high resolution, either in TEM or in STEM, the benefits of the corrector are straightforward as far as image quality is concerned and the time spent in the tuning of the corrector is worth the improvements ([8], for STEM see Fig. 1). In STEM, the improvement in the transfer function is directly seen in the ronchigram which is just a diffraction pattern of an amorphous film obtained with a very high convergence angle ([9], see fig. 1e). The ronchigram gives a direct image of the aberration function $\chi(\theta, \phi)$ (see appendix for definition). One can see in fig. 1e that in the aberration corrected system, there is a large uniform region at the centre of the ronchigram (up to 20 mrad), whereas in the uncorrected system (i.e. a ronchigram obtained without the use of a Cs-corrector), the uniform region in the middle is much smaller. In STEM, the condenser aperture is introduced in order to include only

the uniform region of this ronchigram. In non-Cs-corrected microscopes, typical convergence angles will be about 9 mrad, whereas they reached 15 mrad in the first generation of Cs-correctors and up to 30 mrad in the last generations of Cs-correctors that tune aberrations up to the fifth orders (i.e higher order aberration than the ones given in appendix 1). By allowing higher convergence angles, the beam intensity is also increased which is important for chemical analysis and acquisition speed [10].

In TEM, the main effects of correctors are to increase the resolution (typically from 0.14 nm to below 0.1 nm), to reduce the noise and increase the contrast and reduce the delocalisation effect at interfaces [5]. In fact, with the introduction of Cs-correctors in TEM and STEM, manufacturers had to improve the electronics and mechanical stabilities of microscopes. These stability improvements are particularly important for high resolution STEM images as STEM is particularly influenced by instabilities. From a theoretical point of view there are many analogies between TEM and STEM images. Indeed, the principle of reciprocity [11] states that similar images are obtained in TEM and STEM if the convergence angles (α) and detection angles (β) in TEM and STEM satisfy the relation given in equations (1):

$$\alpha_{\text{STEM}} = \beta_{\text{TEM}}, \quad \beta_{\text{STEM}} = \alpha_{\text{TEM}}.$$
 (1)

In practice, however, equations (1) are rarely satisfied and in standard use, TEM and STEM images have different types of contrast.

As the first Cs-corrected microscope we got was probe-corrected, we have more practice with probe-correctors and most of the examples we will consider will be in STEM. Without starting a controversy or complete comparison of the two techniques, we have the feeling that more and more people, like us, are interested in STEM because STEM offers more possibilities in terms of chemical analysis and the image interpretation is often simpler. TEM is more and more focused on wave function reconstruction [12], low voltage electron microscopy – 80kV and lower – and small precipitates and thin membranes like graphene are more easily observed in TEM [13].



Figure 1. Comparison of STEM images of <110> silicon obtained on different microscopes: (a) FEI-TITAN 300kV without Cs-corrector, (b) the same microscope as in (a) but after the installation of a probe Cs-corrector, (c) FEI-TITAN ultimate 300kV equipped with a D-core probe Cs-corrector. d-e) Comparison of two ronchigrams, i.e. large angle diffraction patterns of amorphous carbon films, obtained on the same FEI-TITAN microscope before (d) and after (e) installation of the probe Cscorrector. The inner circle corresponds to a suitable position for a condenser aperture for STEM.

3. Atomic structure determination: the case of an incommensurate 90° Si grain boundary

The improvement in spatial resolution enabled by Cs-correctors has allowed the solution of unknown atomic structures of interfaces or defects and several examples can be found in the literature. For instance, in [14] it is shown how the atomic structure of material around SiN masks used in GaN growth was determined thanks to the use of an image Cs-corrector that allowed clear atomic images of the interface without blurring or possible delocalisation contrast as would have been observed in non-Cs-corrected HR-TEM. In this paper, we recall the personal experience of the first author of this paper to solve the atomic structure of an incommensurate 90° grain boundary (GB) obtained by wafer bounding. More details about this specific grain boundary can be found in [15]. This structure was first observed in 2001 on a JEOL 4000EX microscope (fig. 2a). With this microscope the Si dumbbells could not be resolved and as the interface is not periodic it was impossible to build a model of the interface. As one of us was in Chicago in 2001, the sample was observed in a JEOL 200kV STEM that was able to resolve the Si dumbbells [16]. However, the stability and the resolution of this microscope was not high enough to resolve simultaneously the silicon dumbbells in the two crystals rotated by 90°. This sample was also observed in an FEI microscope equipped with an image corrector installed at McMaster University in 2007. Here, the disappointment was that nice images could be obtained only for very thin areas. In most of the areas, the sample was too thick to provide interpretable images. We had to wait for the arrival of our TITAN microscope in 2005 and the installation of the probe Cscorrector in 2006 to solve the complex atomic structure of this grain boundary. Fig. 2 shows a part of this interface, with periods of 7/2[110] in grain 1 and 5[001] in grain 2 frequently observed in this non-periodic structure [15]. As high-angle annular dark field (HAADF) contrast does not vary much with thickness and defocus, we could observe large areas of the sample and determine at once without simulation the atomic projection of the structure (fig. 2b). The Si dumbbells are resolved in the two grains and single atomic columns can be clearly distinguished. Assuming that each Si atom has four neighbours situated approximately at the corners of a tetrahedron it was possible to build a 3D structure of the GB and minimize the energy of the configuration by using empirical potentials [15]. The projected optimized structure perfectly fits the experimental HAADF STEM image (fig. 2b). Once the structure was determined we inverted the HRTEM image of the JEOL 4000EX microscope obtained at Scherzer defocus and checked that this image was consistent with the determined structure.



Figure 2. Comparison of two images of an incommensurate 90° grain boundary in Si. Small red spots corresponding to atomic columns have been added to the images: (a) HRTEM image obtained on a JEOL 4000EX microscope at Scherzer defocus with atomic columns in black; the contrast has been inverted in order to compare it directly to (b) the STEM image obtained in an FEI TITAN microscope equipped with a probe Cs-corrector. Some bonds between atoms have been overlaid in green in (b).

4. Polarity measurement

In non-centrosymmetric crystals, there are directions along which the positioning of atoms is not symmetric leading to some special properties of these directions: electrostatic fields can build up along these directions, planes parallel to these directions can have different atomic terminations and so different properties. Determining the ordering of atoms along these directions is determining the polarity of the structure. In [17] we proposed a mnemonic way to define the polarity in a binary compound semiconductor AB, A and B being two chemical elements, for instance A=Ga and B=N. A given surface *n* of the AB compound is said to have A-polarity if the *AB* vector defined by the A-B bonds responsible of the asymmetry of the surface has an acute angle with n (Fig. 3). A couple of years ago, the traditional way to measure the polarity in TEM was to use convergent beam electron diffraction (CBED) and look at the asymmetry of some diffraction spots [17]. The difficulty was first to calibrate the microscope as the lenses can introduce a 180° rotation between the image and the diffraction pattern, and to obtain local diffraction maps. Alternative solutions have been proposed: HRTEM image analysis [18, 19, 20], HAADF-STEM image [21, 22], or more recently Annular Bright Field (ABF) STEM images [23, 24, 25, 26]. Before the introduction of Cs-correctors, the analysis of HRTEM was not very robust unless a high voltage microscope was used [18]. Indeed, the determining of polarity relied on the shape of a profile made along a narrow inclined line [19], but the difference between the two polarities was faint and the HRTEM contrast is very sensitive to tilt, thickness and defocus. More recently Bertoni et al [20] used focal series of Cs-corrected HRTEM images. From the series, they reconstructed the exit phase function and using a channelling approximation, built a phase image where the atomic columns could be directly positioned. A more simple solution is to use Cscorrected HAADF-STEM images and directly image the different atom columns. In AlN, this works very well because the difference between the atomic numbers of Al and N is small [21]. In [22], we showed that when the difference in atomic number is larger (for instance in GaN) it is easier not to locate directly the N-atomic columns but to locate the tunnels of the structure, which indirectly gives the N-column position and thus the polarity. Rather recently a new type of STEM image, called Annular Bright Field (ABF) [27], has been re-introduced by Findlay et al. [23]. It consists of adapting the camera length and convergence angle in order that the rim of the transmitted beam reaches an annular detector. In this way light atoms can be detected. The drawback of this method is that it requires a rather high convergence angle to resolve the GaN columns and that the ABF image suffers from similar drawbacks as TEM images, i.e. its pattern can change with defocus and thickness. Several publications have applied this ABF technique to the determination of polarity in nanowires [24, 25, 26].



Figure 3. a-b) The GaN crystal structure projection along the [2<u>11</u>0] direction. Bonds between atoms are in green. a) N-polarity orientation: the <u>Ga-N</u> vector is antiparallel to the surface normal <u>n</u> b) G-polarity orientation : the <u>Ga-N</u> vector is antiparallel to <u>n</u>. c-d) HAADF-STEM images of GaN observed along [2<u>11</u>0] with some atoms superimposed. c) Image obtained on TITAN 1, d) image obtained on TITAN Ultimate; the image has a better resolution and is less noisy. e) ABF-STEM image of GaN observed along [2<u>11</u>0]. f) Average profile taken from the horizontal rectangle shown in image d). This profile shows clearly where the N columns and the tunnels are located and thus allows determination of the polarity of the layers.

5. Strain determination

Our laboratory has been involved in strain measurement at interfaces and nanostructures for a long time. It started in 1984 with the analysis of HRTEM images acquired on a JEOL 4000EX microscope. First methods used interfacial profiles or direct space methods [28]. For instance, it was possible to detect half-filled semiconductor monolayers of MnTe in CdTe [29]. Then we preferred to use the Geometrical Phase Analysis (GPA) [30, 31] because of its simplicity to provide 2D analysis and its extreme sensibility. We wrote scripts in Digital Micrographs; scripts are available through collaborations. With the arrival of our first non-corrected TITAN microscope, we presented encouraging analysis of HR-STEM images at the 16th International Microscopy Congress in Sapporo [32], but the first physical results really came after the installation of the probe Cs-corrector on our first TITAN microscope. We could partly correct for scanning errors [33], which are intrinsic to HR-STEM images, and accurately determine the strain field around edge dislocations in AlInN layers [34]. Such an analysis would not have been possible in HR-TEM due to the quality of the plan-view samples that were very thick and had a thick amorphous layer on top of them. The analysis of HR-STEM became even better with the arrival of the TITAN ultimate. With the new FEI software, 4kx4k images could be acquired instead of the previous 2kx2k scanned images. With the Gatan Digiscan option, even 8kx8k images are possible. In addition, the stability of the TITAN Ultimate has greatly reduced the scanning errors and the newly designed probe corrector gives images with higher contrast (Fig. 1). This technique is now very useful to measure strain in microelectronic devices (fig. 4, [35]). Compared to HR-TEM images, HR-STEM images have several advantages: (i) patterns do not change as much with thickness and composition, (ii) so it is possible to analyse thicker samples or samples where the sample thickness is changing smoothly, which is very important in microelectronics where most of the samples are prepared by focused ion beam (FIB), (iii) large fields of view (800nm) with 8kx8k pixels are possible and (iv) in addition to strain, some chemical information can be obtained through the intensity of the Z-contrast image [36].



Figure 4. (a) 4kx4k HR-STEM image of a transistor with $Si_{0.64}Ge_{0.36}$ source and drain. At this scale the atomic column are not visible. (b) (220) strain map of (a). Some dislocations can be seen. A black arrow points to one of them. (c) (002) strain map of (a). (d) zoom on a dislocation present in the transistor of fig. 4a. It is situated at the SiGe/Si interface. The 60° dislocation is dissociated into two partial dislocations separated by a stacking fault. The atomic structure of the dislocation is directly determined and the step at the SiGe/Si interface introduced by the dislocation is clearly visible. (e) (002) strain map obtained from image (b). The reference of strain is here taken in the $Si_{0.64}Ge_{0.36}$ layer.

6. Interfacial profiles

As already mentioned in the previous part, the image pattern in Z-contrast images does not change much with thickness. These images can be effectively used for determining the chemical composition and the thickness at different semiconductor interfaces, by normalising these images by the incident beam intensity and by comparison with advanced simulations [36]. The method does not need a Cscorrector and can work with low or high resolution STEM images. The advantage of the Cs-corrector will be to acquire more easily high resolution images, which have better resolution and in addition higher contrast. However, when working with high convergence angles (i.e. angles greater than 15 mrad) one should take care of the reduced depth of field of HR-STEM images [37]. On the other hand, this limited depth of field can be used for depth sectioning, that is to say, 3D reconstructions [37]. We noticed that in FIB prepared samples or in samples having some amorphous layers at the surface of the TEM lamella, the best images are not obtainable from the thinnest areas of the samples. On the contrary, for enhanced quantitative analysis, it is better to use a very thin sample without amorphous layer at its surface. In our cases, the only method that achieved this was tripod polishing [38]. In [38], we succeeded in detecting single Tm dopant atoms in an AlN matrix. More recently, single Ge atoms could be detected at SiGe interfaces (Fig. 6). Quite recently, we applied a Template Matching Algorithm (TeMA) to analyse Z-contrast interfaces [39]. Such a method was already introduced some years ago for the analysis of some special HR-TEM images [40]. Its application in HR-STEM should be more general. The method starts by defining the image unit cell size of the local patterns I_i that will be used in the analysis; here *i* is an integer number used to identify the local pattern. For this a small region around an atomic column is selected with the mouse and forms an initial template, T. A crosscorrelation image, or C-map, is then calculated by making all the cross-correlations between T' and the local $I_{i,i}$ patterns; here *i,j* run over all the pixels of the initial image. The maxima of the C-map determine the locations where the local patterns P_i in the initial image will be taken and thus determine the number n of I_i patterns. By thresholding the C-map the regions where the local patterns I_i that will be taken for the calculation of the template T are selected; T is obtained by averaging these selected patterns. The above steps (i.e. calculation of a new C-map by using the new template as starting point and new threshold of this new cross-correlation) can be repeated to refine the template T. Generally, two templates T_1 and T_2 are defined, one for either side of the interface. Then each local image I_i is decomposed into these two templates (see equation 2):

$$I_i = a_{i1}T_1 + a_{i2}T_2 + \mathcal{E}_i$$
⁽²⁾

where normally ε_i should have small values and describe noise. Amplitude images a_1 and a_2 can then be constructed and provide semi-quantitative chemical information (fig. 5). For instance, the amplitude image a_{GaSb} of fig. 5 shows the asymmetry of the interfaces GaSb/InAs: following the growth direction, the second interface in GaSb is more extended than the first one. We check that this semi-quantitative information (the amplitudes are not real chemical values of the interface) has a physical meaning by comparing this profile to chemical profiles obtained with an atom probe [39]. For instance in fig. 5, the Sb atom probe profile has been added and shows a similar asymmetry, although the asymmetry appears greater in the atom probe data.

In the case of a SiGe/Si interface, two templates, T_1 and T_2 , were taken in the Si layer in order to have only one atomic column of the dumbbells (fig. 6). Each local image I_i was then expressed as function of either T_1 or T_2 , depending on whether I_i was extracted at the top (T_1) or bottom (T_2) atom of the dumbbell: $I_i = a_{i1}T_1 + \varepsilon_i$ or $I_i = a_{i2}T_2 + \varepsilon_i$. In this case the template matching is just an efficient way to extract for each atomic column an intensity value, either a_{i1} or a_{i2} , that can be calibrated in terms of Ge concentration (fig. 6). In our case, this calibration was done by using EELS maps that were simultaneously acquired and measuring precisely the thickness of the sample by PACBED (Position Averaged CBED) [41]. A variation of 6 nm in thickness between the top and the bottom regions of the HR-STEM image in fig.6 was found. Details of this analysis should be published soon.



Figure 5. a) HAADF-STEM image of GaSb/InAs multilayers. b) Amplitude map of GaSb as obtained with TeMA using the templates T1 and T2. c-d) Respective templates obtained in InAs and GaSb. In T_1 , the As atom is on the right, In is on the left. e) Superimposition of two profiles obtained on the multilayers: in dark, profile obtained from the amplitude map of fig. 5b (left scale), in grey profile obtained from the atom probe (right scale, percentage of Sb).



Figure 6. a) Zoom of a HAADF-STEM image showing the Si layer of SiGe interface. b-c) Templates taken at the atomic position of the silicon dumbbell. b) Template T_1 . c) Template T_2 . d) Amplitude image obtained at the Si/SiGe interface using the templates defined in b and c. By using EELS maps and the thickness of the sample the intensities of the amplitude map have been calibrated in multiples of integer numbers of Ge atoms.

7. Conclusion

By providing a few examples of semiconductor studies, we have tried to show what have been the benefits of Cs-correctors for our laboratory. Starting from a laboratory that was essentially doing HR-TEM, the probe Cs-corrector pushed us toward HR-STEM. We saw an important difference between HR-STEM done without a Cs-corrector (acquiring HR-STEM images demanded skills and patience) and HR-STEM done with a Cs-corrector (after its semi-automatic tuning, the Cs-corrector guarantees a good image). The introduction of Cs-correctors pushed the manufacturers to provide more stable microscopes.

Thanks to the Cs-correctors we were able to determine the structure of a complex incommensurate interface, to detect individual atoms in a column, to measure polarity and perform large field-of-view strain maps. Although not illustrated in this paper, the Cs-correctors also allow us to work at very low voltage (60kV and certainly soon below) while keeping very good spatial resolution and they permit the use of large pole piece gaps (for in-situ experiments). However, one must be aware that Cs-correctors introduce some addition aberrations (chromatic and high order aberrations) and distortions that can be annoying in some specific applications. Although Cs-correctors have boosted the electron microscopy technique and are very useful, it does not necessarily mean that Cs-correctors are and will be compulsory in a TEM. Nice experiments, for instance low resolution images, EDX chemical detection and diffraction studies can be done without Cs-correctors.

Appendix

Aberrations are introduced in the aberration function $\chi(\theta, \phi)$ which is the error in optical path length between a perfect planar wave and the actual wavefront as function of the diffraction angles θ (radial) and ϕ (azimuthal). Different notations for aberrations co-exist. For instance, in equation (3) the aberration of order *n* and angular multiplicity *m* is characterized by an amplitude $|C_{n,m}|$ and a phase $\phi_{Cn,m}$. As most of the TEM/STEM Cs-probe corrected microscopes use the correctors from CEOS company, Haider et al's notation is often used (3) although it is not very convenient as aberrations of the same order, like A2 and B2, do not always have the same pre-coefficients. In equation (3), A1, C1, A2, B2, C3, A3, S3 are respectively second-order astigmatism, defocus, third-order astigmatism, coma, spherical aberration (i.e. Cs), fourth-order astigmatism and star aberration. On a daily basis, all these aberrations are tuned. Some higher order aberrations are tuned during the build-up of the corrector in the factory.

$$\chi(\theta,\phi) = \sum_{\substack{m < n+1 \\ m+n \text{ odd}}} \frac{1}{(n+1)} \Big| C_{n,m} \Big| \theta^{n+1} \cos(m\phi - \phi_{C_{n,m}})$$
(3)

$$\chi(\theta,\phi) = \frac{1}{2}A_1\theta^2 \cos(2\phi - \phi_{A1}) + \frac{1}{2}C_1\theta^2 + \frac{1}{3}A_2\theta^3 \cos(3\phi - \phi_{A2}) + B_2\theta^3 \cos(\phi - \phi_{B2}) +$$
(4)
$$\frac{1}{4}C_3\theta^4 + \frac{1}{4}A_3\theta^4 \cos(4\phi - \phi_{A3}) + S_3\theta^4 \cos(2\phi - \phi_{S3}) + \dots$$

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