An advanced study of the response of ADF detector

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Abstract: The detailed characteristics of the Annular Dark Field detector and acquisition system may play a strong role in quantitative analysis. This article discusses the role in high angle experiments of a non-uniform detection efficiency and of the saturation. The former can be neglected in most comparative experiment while the latter can produce hidden nonlinearities that reduce the reliability of quantitative analysis.

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1. Introduction
The development of quantitative High Angle Annular Dark Field (HAADF) promises to be the most interesting tool in semiconductor compositional analysis on the nanoscale. At present two families of approaches exist for quantitative analysis based on relative (intensity comparison with a reference material) and absolute intensity measurements [1],[2]. The absolute methods are based on the characterization of the detector response and do not require a reference material. Conversely, the relative methods are more robust and more suitable for the comparison between neighbouring materials and in particular semiconductor heterostructures.

The aim of this contribution is to highlight a few technical and physical considerations that can justify using relative techniques for quantitative analysis. The role of the detector saturation and of non-uniform efficiency will be also critically analyzed. The calibration of angular detection efficiency may prove to be quite complicated in some JEOL machines and it is relevant to evaluate to which extent the inhomogeneous detector response can be ignored.

2. Theory
2.1 Saturation effects of the acquisition system
For this study images were acquired by use of a DIGISCAN2 [3] system connected to the analog output of the STEM, but in principle many acquisition systems present similar dynamic limitations. In the detection system attached to the microscope the light from the scintillator is converted to an electrical signal and subsequently amplified by an internal amplifier and directly transformed into a TV signal. With old systems operating with a direct analog visualization of the image the synchronous movement of the scanning coils in the STEM optical system and in the monitor creates the image on the monitor. In digital systems the signal is directly matched to the Analog Digital Converter (ADC) range that converts it to an integer to be stored in the memory of the controlling PC system. The ADC sampling is completed within a short time $t_s$ (every 0.8 \( \mu \)s in a DIGISCAN2) and is repeated several times for each scanning position until the dwell time is reached. The details of the acquisition are actually system dependent. Hereafter, the whole analog part will be modeled as a linear system:

\[ I_{\text{analog}} = g n_s + b \] (1)
where the analog current $I_{\text{analog}}$ is proportional to the number of detected electrons in the sampling time by means of an overall gain $g$ and a DC offset $b$. Whereas many factors concur to the overall gain the proportionality factor can be always regulated by adjusting the contrast knob, unfortunately this has also a minor effect on $b$. Assuming that the ADC is 8 bit (a different number of bits does not change the results hereafter but only the discretization), $I_{\text{analog}}$ is an integer between 0 and 255; signals below 0 or larger than 255 are clipped by the system. However, the typical response of a detector presents a smooth saturation curve (see ref. [2],[4],[5]). Whereas this dependence has been largely observed, a detailed modeling of the saturation process has not been proposed.

![Figure 1. Schematic representation of acquisition system. The analog signal from the internal amplifier is driven to the ADC slot having its own amplifying system whose gain and DC offset can be also regulated by trim.](image)

Figure 1. Schematic representation of acquisition system. The analog signal from the internal amplifier is driven to the ADC slot having its own amplifying system whose gain and DC offset can be also regulated by trim.

The reason for the smooth saturation is that many ADC acquisitions are summed together. Due to statistical fluctuations in the number of counts per acquisition in proximity of the maximum allowed signal, only part of the sum is saturated. In a given point the expected number of counts is $<n_s>$ and the counts are distributed according to Poisson statistics $\mathcal{P}(i)$ with deviation $\sqrt{<n>}$). A convenient formula for the average intensity on the ADC (normalized to 255 levels) as function of $<n_s>$ is

$$I = \frac{1}{255} \sum_{i<n_s} (gi + b) \cdot P_{\mathcal{P}}(i) + \sum_{i\geq n_s} P_{\mathcal{P}}(i) + \sum_{i>n_s} b \cdot P_{\mathcal{P}}(i)$$

where $n_{\text{max}} = \frac{1}{g}$ and $n_0 = \frac{-b}{g}$. Experimentally with a positive value of $b$ its value can be gauged from regions without sample, provided that the relation between the read intensity and the ADC value is known. The value of $g$ however may require information on the total beam current.

### 2.2 The relevance of angular detector efficiency in comparative experiments

Whereas the evaluation of the detector efficiency has been a strong progress in quantitative analysis, this is not strictly necessary for a quantitative analysis when relative methods of quantification are used based on the comparison between nearby regions. The justification is given by the assumption that $I = M(t,x)\sigma$ [6]. Here $M$ contains the thickness dependence and $\sigma$ is the cross section. The ratio of two intensities is therefore

$$R = \frac{I_A}{I_B} = \frac{M_A\sigma_A}{M_B\sigma_B}$$

As the dependence on the detection angle is only contained in the cross-section, this can be written as

$$R \propto \frac{\int g(s) f_A^2(s) (1 - e^{-M_As^2}) ds}{\int g(s) f_B^2(s) (1 - e^{-M_As^2}) ds} = \frac{Z_A^2}{Z_B^2} \frac{\int g(s) f_A^2(s) ds}{\int g(s) f_B^2(s) ds}$$

where $g$ is the angular efficiency of the detector, $s$ is the in plane momentum and $f_{a,b}$ are the atomic scattering factors of atoms of type A,B. The simplification written here is valid for sufficiently high angles so that the exponential can be neglected. Moreover it has been assumed that the angular dependence of $f(s)$ on angle is unscreened Mott-Rutherford and therefore independent of $Z$. For sake of comparison the chemical contrast between InGaP and GaAs has been calculated for an annular detector starting in the range 75-210 mrad (corresponding to a camera length of 8 cm in our JEOL...
2200 FS) with a flat and a slowly varying efficiency (fig. 2a). Simulations have been performed with an updated version of STEM_CELL [7] parallel computer software for Frozen Lattice multislice. It permits to store data for different angular sectors and add detection efficiency to the simulation post processing. The plot of the two almost superimposed curves is shown in fig. 2b. The additional curve is their ratio. The error in $R$ is at most 3\% at low thickness but otherwise about 0.3\%. This explains the success of quantitative ADF methods not accounting for varying detection efficiency [1, 8].

2.3 Estimation of the thickness from comparative experiments

While it is not necessary for relative compositional HAADF analysis, the calibration of the detector efficiency may be useful to evaluate specimen thickness [4]. An alternative can be adopted in samples with strong thickness gradient and using a small outer detector angle (in this case 46-128 mrad). In such conditions it’s easy to find a thickness for which the ADF intensity of relatively heavy materials (i.e. GaAs) reaches a maximum and the starts to decrease. According to simulations in fig. 2c this occurs at a thickness of about 450nm but in the case of gold this would be much less.

This critical thickness can be determined by simulations and used to gauge the ratio in regions with lower intensity. This is not an absolute intensity measurement of the beam, yet it can be useful to measure the thickness at any position in GaAs. In fact, the two curves in fig. 2b obtained for different angular efficiencies of the detector produce similar shapes and only differ by a constant factor. Therefore the ratio between the maximum intensity and the actual value at low thickness is largely independent of the detector response. This ratio can be used to gauge the specimen thickness.

3. Experimental

STEM experiments have been performed on a JEOL 2200 FS equipped with a Schottky FEG and working at 200keV. The spherical aberration of the objective lens was 0.5 mm. A nominal 0.7 nm spot was used to achieve high beam currents. Image acquisition has been performed by DIGISCAN2 attached to the lower ADF detector. The sample was a GaAs substrate with epitaxial InGaP grown by MOCVD which will not be discussed in this work. For STEM observation the sample has been thinned by mechanical grinding and final polishing in a GATAN DUO MILL.

4. Results and discussion

Fig. 3a shows an ADF image of the GaAs sample where two gain settings are used. The solid line corresponds to the unsaturated image while the dashed line corresponds to the saturated image. The saturated profile has been normalized in order to have 1 on the y-axis in the saturated region; the solid profile has been scaled in order to be coincident with the dashed profile in the low thickness region. A point to point comparison of saturated and unsaturated profiles gives a saturation curve as in fig. 3b. The most striking result is that saturation occurs before the actual flat region. The reason is the relatively low count in the time sampled of electrons scattered to the high angle regime (in this case 115-322 mrad). In the present case a good fit is obtained with $g=10/255$ while $b$ has been set close to 0 and therefore neglected.
This means that the value of $<n_s>$ corresponding to the saturation signal is 25 electrons for each 0.8 µs sampled. The thickness at the saturation point is estimated to be roughly 200 nm and the detection corresponds to 15% of the impinging beam. This would correspond to a beam current of about 0.04 nA, compatible with a Schottky source. It is clear that a signal of 25 electrons per sample has a large statistical variability (20%) so that, even at 20% distance from the saturation, the count distribution has tails in the saturated region; an intensity evaluated at 10% from the saturation limit gives an error of 13%, but even an apparently safe intensity of 40% [5] from the limit signal produces quite a low but still appreciable 0.8% error. This is therefore potentially a much larger source of error in HAADF quantitative analysis rather than the effect of a non-constant angular efficiency evaluation.

**Figure 3.** a) Split ADF image of the GaAs sample where two gain settings were used. The intensity profiles from the same area are also shown as solid and dash lines. The two curves are normalized in order to be coincident in the low thickness region. b) The saturation curve deduced from profiles in (a) and compared with simulated curves according to eq. 2 for two values of the gain, $g$.

Finally, an experiment of comparative thickness measurement is sketched in fig. 3c where an ADF image (46-128 mrad) of a GaAs sample is shown. An intensity profile normalized to its maximum is also shown superimposed. Both show that, after reaching a maximum, the intensity decreases further from the edge. HAADF measurements (115-322 mrad) demonstrate that the thickness is continuously increasing so this maximum should corresponds to a thickness of about 450 nm as discussed in fig. 2c. This appears consistent with preliminary thickness measurements by EELS.

**5. Conclusions**

In conclusion, I have demonstrated, with simulations and experiments, that relative methods based on the comparison of intensities in different regions can be used for quantitative chemical analysis and thickness determination even if the detailed angular efficiency of the detector is unknown. The error involved in the non-flat response of the detector is potentially lower than the non-linearity errors caused by high gain settings. This is particularly the case for very high angle ADF without a Cs corrector on the probe where the number of electrons collected is lower and fluctuations are large. Care should be taken therefore to avoid subtle non-linearity even when saturation is not evident.

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