Multislice calculations for quantitative HAADF STEM analysis of germanium diffusion in strained silicon

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Abstract. For decades, microelectronic industry has reduced components size while increasing their performance and reliability. Actual systems rely on multilayer nanostructures that require a perfect control of composition at atomic scales. In this study, we focus on detection of germanium contaminants in a thin silicon layer, using High Angle Annular Dark Field (HAADF) with Scanning Transmission Electron Microscopy. We proceeded to multislice calculations in order to extract quantitative information from HAADF measurements. Models consist of Ge substitutional impurities introduced in 10 nm thick Si layers. We analysed atomic column HAADF intensities along \langle100\rangle zone axis, varying Ge impurities amount and position in an atomic column. Results show that for a reduced number of Ge atoms \(n<9\), column intensity varies linearly with \(n\). However depending on Ge atoms positions, a column containing \(n\) impurities can display a higher signal than a column containing \(n+1\) impurities. Therefore, a direct correlation between atomic column intensities and their Ge atoms content cannot be established without additional assumptions or complementary measurement.

1. Introduction
Strained silicon is intensively used in microelectronic industry to keep improving transistor characteristics [1]. For instance, Si channels of complementary metal-oxide-semiconductor transistors are grown on relaxed \(\text{Si}_{1-x}\text{Ge}_x\) layers. The stress induced by lattice mismatch increases carriers mobility and limits short channel effects [2–4]. It results in an enhancement of the drain current, a key parameter to gain switching speed. This technique is now applied to one or two dimensional nano-objects, to combine stress effects with quantum confinement. A new components generation emerges, like high-performance field effect transistors or solar cells, based on band-gap engineering [5–7]. Calculations show that an atomic layer-by-layer growth of alternating Si and Ge atomic planes with a specific sequence would lead to a direct band gap Si-Ge film [8–10]. Such a material represents a great interest for optoelectronic applications.

Considering devices dimensions, a perfect control of layer composition on short scales is required. Small fluctuations would drastically change their properties. Yet, many physical processes like diffusion during multilayers growth, can deteriorate interface sharpness. Scanning Transmission Electron Microscopy (STEM) with High Angle Annular Dark Field (HAADF) is a relevant technique for quantitative detection of impurities. Several studies proceeded to detection of dopant [11,12] and chemical mapping in semiconducting materials [13].

In this paper, we focus on Ge atoms detection in a thin silicon film. Quantitative calculations have been performed in order to analyse image intensity as a function of sample composition. We varied
substitutional Ge atom number and position in the Si lattice. We report simulation results and discuss quantitative information that can be extracted from HAADF measurements on this kind of materials.

2. Simulations setup
Quantitative HAADF STEM simulations have been carried out using the QSTEM software [14], employing the frozen lattice calculations. Results have been averaged over 30 frozen phonon configurations. Microscope characteristics were fixed, considering a 200 keV aberration-corrected probe: $C_3 = 4 \mu$m, $C_c = 1.4$ mm and energy spread $dE = 1.4$ eV. The probe-forming aperture’s half-angle has been set to 18 mrad, inner- and outer-angle of the annular detector to 60 and 160 mrad, respectively. Electron beam was focused in the middle of the layer i.e. $\Delta f = -5$ nm.

The layer model consists of a bulk 3x3 Si supercell, 10.5 nm thick. As the sample is observed along the $\langle 100 \rangle$ zone axis, each atomic column displayed in simulations (see Figure 1) contains 20 atoms. Ge atoms have been introduced as substitutional impurities into the Si lattice. For each simulation, all Ge atoms are located in the same atomic column. Considering the small amount of impurities introduced, we neglected potential lattice relaxation.

Using method of last squares, image intensity was then fitted with a sum of Gaussian functions corresponding to the 13 atomic columns contained in the simulated image:

$\sum_{i=1}^{13} \frac{a_i}{(2\pi s_i^2)^{1/2}} \exp\left[-\frac{(x-x_i)^2 + (y-y_i)^2}{2s_i^2}\right]$

$x_i$ and $y_i$ refer to the column coordinates, $a_i/(2\pi s_i^2)^{1/2}$ and $s_i$ to their amplitude and standard deviation, respectively (see Figure 2). $I_0$ was set to zero, as no noise was introduced during simulations. In the following, amplitude of the atomic column containing Ge impurities was reported as a function of simulation conditions, using pure silicon column amplitude as amplitude reference.

3. Results

3.1. Column intensity as a function of Ge atom number
In a first step, column intensity containing Ge impurities was investigated as a function of $n$, the number of germanium atoms. Theses atoms are located in the middle of the column, in successive planes. Calculations show that for $n \leq 8$, a linear increase of intensities by about 20 %, per additional atom impurity, is observed (see Figure 3, red circles). For higher Ge contents ($9 \leq n \leq 20$), additional Ge atoms are located far from the beam focused point, and progressively fill the column. As a consequence, the intensity variation is no longer linear and saturation arises for $n > 13$.
Using the same model but for Ge atoms located close to the specimen top, intensity evolution presents the same behaviour (cf Figure 3, blue triangles). A linear response is obtained for $3 \leq n \leq 11$, with a 20 % increase. This shift suggests that the intensity also depends on Ge atom positions.

3.2. Column intensity as a function of Ge atom position
For a single Ge substitutional atom, the intensity can vary from 8 %, if located at top or bottom edge of the sample, to 29 % if placed close to the centre. These variations are directly linked to the microscope focus conditions: signal is maximal when impurities are located at the focus point, i.e. the middle of the layer.

![Figure 3](image3.png)  
**Figure 3.** Column intensity as a function of $n$, number of Ge substitutional atoms. Ge atoms are inserted in successive planes, located in the centre of the column (circles) or at top column edge (triangles).

![Figure 4](image4.png)  
**Figure 4.** Column intensity increase as a function of Ge cluster position in the column. Ge cluster consists of 1 (circles), 2 (triangles), 3 (diamonds) or 4 (squares) Ge atoms placed in successive planes.

![Figure 5](image5.png)  
**Figure 5.** Column intensity increase as a function the gap between Ge substitutional atoms. Ge atoms are located in the same column, regularly spaced and symmetrically distributed around the column centre. Circles, triangle and diamonds correspond to a column containing 2, 3 or 4 Ge atoms, respectively.

Ge atoms clusters consisting of $n = 1$ to 4 adjacent Ge atoms aligned in a single column have been modelled, varying cluster position in the column. Results are displayed in Figure 4. Again, column intensity is maximal when the cluster is located at the microscope focus point, i.e. in the middle of the layer. It drastically decreases, by more than 50 %, when a cluster reaches layer surface. As a consequence, a column containing an $n$-Ge cluster located in the middle of the column can display a...
higher intensity than a column for which an \((n+1)\)-Ge cluster is located in the top or the bottom of the column. Calculations show that a column containing \(n\) atoms can even display a signal higher than a column with \(n+2\) Ge atoms for \(n \geq 4\).

This observation is confirmed by another set of atom position distributions. This time, Ge atoms are not located on adjacent positions, but are regularly spaced and symmetrically distributed around the column centre, varying the gap between them. As can be seen in Figure 5, column intensity decreases rapidly with gap and, again, signal arising from a column containing \(n+1\) Ge atoms can be weaker than from a column containing \(n\) Ge atoms.

4. Conclusion

In this study we investigate the possibility for HAADF to be used as a convenient tool for characterising simultaneously sample structure and composition. We focus on the mainstream materials used in microelectronics and optoelectronics industries: silicon and germanium.

The models consider Ge substitutional atoms in a Si lattice. Calculations show that a linear increase of column signal intensity is observed when Ge atoms are located close to the electron probe focus point. As a consequence, the linear regime can be observed only in very thin specimens. For thick ones, a direct correlation between columns’ intensity and their composition cannot be established. Complementary experiments would be necessary. Other STEM techniques like EDS would hardly combine high spatial resolution and chemical sensitivity. Silicon and germanium have relatively low and close atomic numbers which represents a major challenge for chemical quantitative investigation at atomic scale.

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