A methodology for the extraction of quantitative information from electron microscopy images at the atomic level

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Abstract. In this paper we describe a methodology developed at the University of Cadiz (Spain) in the past few years for the extraction of quantitative information from electron microscopy images at the atomic level. This work is based on a coordinated and synergic activity of several research groups that have been working together over the last decade in two different and complementary fields: Materials Science and Computer Science. The aim of our joint research has been to develop innovative high-performance computing techniques and simulation methods in order to address computationally challenging problems in the analysis, modelling and simulation of materials at the atomic scale, providing significant advances with respect to existing techniques. The methodology involves several fundamental areas of research including the analysis of high resolution electron microscopy images, materials modelling, image simulation and 3D reconstruction using quantitative information from experimental images. These techniques for the analysis, modelling and simulation allow optimizing the control and functionality of devices developed using materials under study, and have been tested using data obtained from experimental samples.

1. Introduction: methodology overview

The extraction of quantitative information (such as strain, composition and morphology) from electron microscopy images is a challenging task. It has recently been shown an excellent agreement between experiments and theory in HAADF-STEM [1-3], demonstrating that current models of image formation are adequate in the modeling of both low and high-angle scattering, including thermal diffuse scattering, which dominates Z-contrast images.

Over the last decade, a set of tools for electron microscopy HAADF image analysis, nanostructure modeling and HAADF image simulation has been developed, giving rise to a methodology that enables us to obtain quantitative results for the understanding of materials at nanoscale.

The methodology first defines a 3D strained model using information coming from two sources: growing conditions and electron microscopy images. While growing conditions may give us detailed information about substrate composition, and expected nanostructures density, composition, size, shape, etc., more precise information may be obtained from the analysis of high-angle annular dark field (HAADF-STEM) electron microscopy images using both classical (EELS, strain mapping, etc.) and advanced techniques(compositional analysis with atomic resolution). The second step is the...
determination of a relaxed 3D atomistic model. This can be accomplished by calculating the
displacement fields in the model, and applying these displacements to atomic coordinates of crystal
positions in the bulk, or can be directly calculated using molecular mechanics. The final step is the
simulation of electron microscopy images that can be compared with experimental ones in order to
assess the accuracy of our model that might be refined in an iterative process. In the following
paragraphs we will discuss in detail these steps.

Finally, let us note that at the development stage, computationally intensive studies are usually
carried out using high performance computing. The development of compatible, portable, scalable,
and user-friendly computational tools running into Graphical Processing Units (GPUs) will make
possible its distribution to other universities, research centres and specialized companies.

Figure 1. Schematic diagram of the methodology developed at the University of Cadiz for the
quantitative extraction of information from electron microscopy images

2. Material characterization: strain and composition analysis techniques for 3D solid modelling
Two main techniques have been developed for the analysis of electron microscopy images. The first
one, under the name Peak Pairs, is an algorithm for the determination of strain from high-resolution
images. The second one is a technique to perform column-to-column compositional analysis of
materials from the analysis of intensities in atomic-column resolved HAADF-STEM images. Both
techniques have been implemented as plug-ins for the Digital Micrograph under the names PPA and
qHAADF respectively, and are distributed by HREM Research, Inc.
2.1. Peak Pairs
Strain mapping techniques constitutes a well-established numerical image processing technique that allows, from the analysis of high resolution electron microscopy images, the determination of the displacement of lattice fringes in a strained material with respect to a perfectly regular lattice measured in a reference (relaxed) material. By measuring the position of peaks in the image, it is possible to precisely determine distortions of the lattice with respect to a given reference.

Peak Finding techniques work in real space and for strain analysis were first introduced in 1993[4] and then developed by different researchers [5-7]. In 2007 the Peak Pairs [8] algorithm was proposed, where from a noise filtered image, pairs of peaks were detected along a predefined direction and distance in the affine transformed space. Since this transformation reduces potential errors in detecting adjacent maxima in heavily distorted areas and the procedure works without user’s intervention, Peak Pairs has become a useful tool for strain mapping [9,10].

PPA is the acronym of Peak Pairs Analysis, the plug-in for Digital Micrograph that implements the Peak Pairs algorithm, available from HREM Research Inc. Its key features are (a) Peak location at sub-pixel resolution using 2D filtering (Bragg, Wiener,...) and cubic interpolation techniques, (b) Robust Peak detection in noisy images using a small reference region, (c) Local strain tensor determination at atomic-column level, (d) Local representation of strain at the atomic-column level using color scales superimposed on HREM image, (e) Automatic peak intensity measurement and (f) Image distortion correction due to a projector lens system (CTEM) or a scanning system (STEM).

Figure 2. High-resolution transmission electron microscope (HRTEM) image of an InAs/GaAs (001) interface taken with a JEOL 2000 EX TEM at 200 kV. Point resolution = 0.21 nm.[9]

Figure 3. Components of the strain tensors exx , eyy , exy and eyx showing the presence of two well-distinguished materials corresponding to GaAs and InAs and a misfit dislocation network just at the InAs/GaAs interface.[9]
2.2. Column-to-column compositional mapping (qHAADF)

In [11] a phenomenological method to determine quantitatively the composition of a material with Atomic column spatial resolution was described. The method is based on the analysis of local integrated intensities of aberration-corrected Z-contrast images. A set of reference samples of known composition is used to quantify the relationship between the experimental Z-contrast intensities and the thickness and composition of the analysed material. The method has been applied to different alloys such as InAsP[11] and InGaAs[12] and has also allowed to determine maps of Bi content (expressed as number of atoms) in each atomic column of a GaAs$_{1-x}$Bi$_x$ epitaxial layer[13].

qHAADF is the acronym of Quantitative HAADF, the plug-in for Digital Micrograph that implements this technique, available from HREM Research Inc. It performs column-to-column compositional analysis of materials from the integrated intensities of an atomic-column resolved HAADF-STEM image, and its key features are (a) Gives a map of integrated intensities and/or composition in materials with atomic-column spatial resolution, (b) Experimental HAADF images are used to obtain the fitting parameters to determine composition, (c) Automatic finding intensity peaks and noise filtering tools, (d) Compositional segregation profiles can be obtained from epitaxial layers, (e) Counts atoms in each atomic column and (f) Locates interstitial atoms in materials.

3. Finite Element Analysis and Molecular Dynamics for 3D atomistic modelling

Several methods have been used to simulate the strain distribution in nanostructures, being the finite element method (FEM) the most extended. Among its advantages we can mention lower computing and time requirements, allowing the use of arbitrary shape structures with anisotropic properties and non-uniform composition.

3.1. Finite Element simulations to determine atomic positions

Finite element method (FEM) modelling may be used to calculate the stress and strain in a three-dimensional defining a model taking into account shape, composition, boundary conditions and elastic parameters of the materials. The approximate shape may be determined from electron microscopy images taken at different conditions, and is usually defined visually using a CAD tool using combinations of several volumes having different geometric shapes. Composition modeling should take into account segregation and diffusion across different materials, and different models may be considered, such as the Muraki model for segregation. When available, composition maps are preferred, such as those coming from EELS maps used frequently to estimate the composition in nanowires. Boundary conditions are defined using symmetries, periodicities, or fixing them against rigid body shift in a desired direction, as it is usually done on the substrate. The basic mesh distribution for finite element calculation is usually calculated using the Delaunay algorithm, as it can be applied to all geometry objects. Anisotropic behaviour and values at 300 K are assumed for elastic constants of known materials, and Vegard’s law is usually applied to estimate values at different compositions in the case of alloyed materials, such as ternaries or quaternaries, for which elastic constants are unknown. Finally, the displacement, strain and stress fields may be calculated by solving anisotropic elastic theory equations over the domain of continuum mechanics. The displacement field may then be applied to atomic coordinates of crystal positions in the bulk in order to determine atomic positions of the unstrained crystal [10].

3.2. Molecular Dynamics atomic position determination

Molecular Dynamics (MD) is a computer simulation technique allowing atoms and molecules to interact for a period of time under known laws of physics. The major advantage of this techniques as compared to the previous one(FEM) is that can MD may be used in the presence of defects, including point (vacancies, interstitials), linear (dislocations) and planar (grain boundaries, stacking faults) defects. This possibility is crucial for a better knowledge of their mechanical properties and therefore of a great technological interest.
Once all rules have been set up about how atoms interact with one another, initial positions and velocities of all system particles are defined. We can use two different techniques: methods minimizing the potential energy (often called energy minimization techniques) and methods modeling the system behavior with time propagation. The most usual model in semiconductor simulation breaks energy up into two parts: covalent and non-covalent terms. Covalent terms describe the deviation of bond lengths (stretching energy), bond angles (bending energy) and torsion angles (torsion energy) away from equilibrium values, while non-covalent terms include forces between all non-bonded atoms. It has been shown that FEM and molecular dynamics give similar results.[14]

![Figure 4. σ_xx component of the stress tensor (in bar): (left) Finite element model and (right) Molecular Dynamics (right)](image)

4. SICSTEM: a simulation software for HAADF-STEM images

High angle annular dark field scanning transmission electron microscopy (HAADF-STEM) is a powerful tool to quantify size, shape, position, and composition of nano-objects with the assessment of image simulation. A parallel software called SICSTEM[15] has been developed that can afford HAADF-STEM image simulations of nanostructures composed of several hundred thousand atoms in manageable time.

The input to SICSTEM software is a supercell described as a set of (x, y, z) coordinates for atom positioning, its composition, site occupancy, and Debye–Waller factor and the characteristics of the microscope beam energy, third and fifth order objective aberrations, objective aperture, detector angles, etc. At each desired position of the specimen, an incident focused probe wave function is generated, and then transmitted and propagated across the specimen taking into account phase grating and TDS until it reaches the exit surface. Finally, the electron intensity is integrated on the detector. In HAADF-STEM simulation, the calculation of each pixel requires a complete multislice simulation thus requiring more than 1x10^6 multislice calculations for a 1024x1024 image. Nevertheless, each pixel in the scanning process can be calculated independently from each other, making easy the division of tasks in parallel. The software has been designed to be able to generate not only one-dimensional line scans or two-dimensional images, but also focal series, very useful in three-dimensional HAADF-STEM research.

In order to test the accuracy across variations in thickness, the contrast ratio of Ga/As peaks has been determined for thickness values in the range of 0–20 nm. Figure 5 illustrates a comparison with Ishizuka’s Fast Fourier Transform (FFT) multislice approach, where the contrast ratio of As/Ga peaks as a function of specimen thickness is shown.

We run the SICSTEM software in order to get a HAADF-STEM simulated image of an InP capped InAsP /InP nanowire. The composition for the test nanowire assumes that the maximum composition in the nanowire is 100% pure InAs. The image was simulated considering a 100 kV dedicated VG
Microscope HB501UX STEM having a probe size of 0.85 Å. The number of atoms in the supercell was 422184. The simulated image of the nanowire is shown in Figure 6. This image has been obtained at the supercomputer of the University of Cadiz, a cluster composed of 320 cores and a peak performance of 3.75x10^{12} floating point operations per second. The processing time was 78 h for a 560x1024 pixels resolution image.

Figure 5. Contrast ratio of As/Ga peaks as a function of specimen thickness. Dots and squares represent the contrast ratios calculated using Izhizuka’s FFT multislice code and the theoretical estimates respectively while the line represents the As/Ga peak ratio obtained by SICSTEM software.[15]

Figure 6. HAADF-STEM simulated image of an In_{0.5}As_{0.5}P_{1-x} nanowire (560 x 1024 pixels) using SICSTEM software.[15]

5. Electron Tomography techniques for 3D atomistic reconstruction

Electron tomography (ET) can be used to determine the shape, and therefore the properties, of an object. It deals with retrieving 3D information from 2D projections of the object under study. While ET has been used widely in biology, its application to the nanoscale world is relatively recent because of the limitations in resolution and precision aberration of electron microscopes. Tomography 3D reconstruction can be used to estimate spatial parameters of the object of interest as its dimensions and spatial distribution. Despite a variety of reconstruction algorithms exists and good quality reconstructions can be achieved in theory, in practice the quality of the reconstruction depends strongly on the quality of the input data. In fact, the real imaging process supplies noisy images and consequently no suitable input data for the direct application of such algorithms. In the field of electron tomography, some of the problems arise from geometrical distortions suffered from the image collected by the CCD. In addition, there are other factors that affect the final reconstruction, as the unknown real tilt axis inclination, the actual value of the tilt angles and the actual specimen position with respect to the holder.

5.1. Needle-shaped samples reconstruction using quantum dots position as fiducial markers

The alignment of tomographic series is a mandatory step in order to obtain reliable reconstructions. The alignment can be carried out in several ways, depending on the amount of user interaction needed. At atomic scale, classical tomography reconstruction can be replaced by Tomography Through Points (TTP) reconstruction, that is, the 3D information can be obtained by applying image processing techniques to the projections and by using geometrical models instead of the classical integral approach.
5.2. Reconstruction of 3D atomic positions in simulated graphene
The marker-based model for alignment allows a direct application in all those cases in which markers can be easily detected. Therefore, a tomographic approach for the quantification of the graphene corrugation is a good choice. Indeed, a single graphene layer has a simple geometric configuration (honeycomb regular structure) and the atoms can be located with great accuracy [16]. It means that a kind of data simplification can be performed in order to optimize the whole reconstruction process. The main points of the proposed approach are (a) treating each atom as a marker, it is possible to obtain a tomographic reconstruction of the structure, (b) using a small angles range to obtain the tomography, focusing problems are avoided and more homogeneous images can be obtained, (c) exploiting an algebraic approach, the reconstruction speed can be improved without loss of precision and (d) estimating the positions of the atoms in 3D space instead of using the whole image, as it is done in classical tomography, problematic issues such as missing wedge or projections noise can be avoided or at least minimized.

![Figure 7. Original model(a) and reconstructed(b) graphene corrugation using Tomography through Points](image)

5.3. Discrete tomography – the ADART algorithm
DART is a heuristic Discrete Tomography technique, capable of computing reconstructions of high quality from a small number of projections. The algorithm is very effective for binary images, but it can also be used to reconstruct images that contain more than two gray levels [16]. In DART, the update operation is restricted to pixels in the boundaries across gray levels in the reconstructed object.

ADART [17] is an algorithm based on DART that is capable of computing high quality reconstructions from substantially fewer projections than required for conventional continuous tomography. ADART reconstruction errors are significantly reduced with respect to DART. This reduction is remarkable for binary images, especially in noisy environments, and is nearly constant and independent of noise level for multiple grey-level images.

![Figure 8. DART and ADART errors on the discrete reconstruction of the phantom image using 18 and 12 projections respectively.](image)
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