



# Semi-automatic structural labeling of HAADF-STEM images of hybrid precipitates

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Software overview

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**AutomAI 6000** is a stand-alone software tool for semi-automatic labelling of HAADF-STEM images of precipitates in the 6xxx aluminium alloys, developed as a part of my master project in physics at NTNU. Main features include:

## 1. Introduction

## 2. Background

## 3. Objective

## 4. Method

## 5. Summary

- Basic column detection.
- Column characterization algorithm
- Flexible statistical modeling from user data
- GUI
- Imports dm3, TIFF or AtoMap formats
- Exports png, csv or svg formats
- Open source

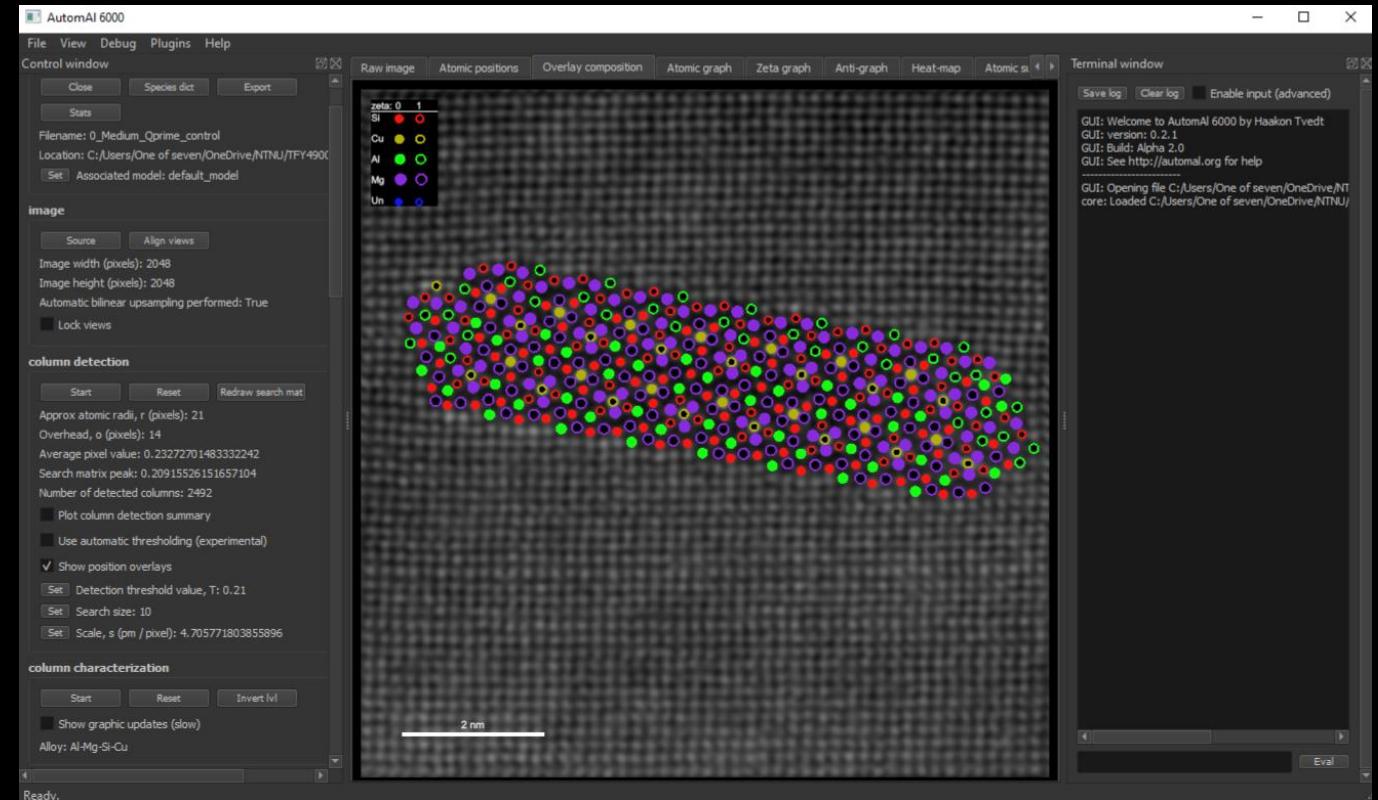


Fig. 1.1: The AutomAI 6000 GUI with an overlaid Q' precipitate.

## 1. Introduction

## 2. Background

## 3. Objective

## 4. Method

## 5. Summary

## 2.1 HAADF-STEM

## 2.2 Structural principles in Al-Mg-Si-(Cu) precipitates

**High angle annular dark field scanning transmission electron microscopy (HAADF-STEM)**, is a modern microscopy technique which scans a convergent electron beam across the surface of a sample while detecting electrons scattered to high angles (Rutherford and thermal diffuse scattering). The amount of incident electrons that are scattered to high angles, is approximately proportional to the square of the atomic number  $Z$  of the main column species and the distance between the column and the beam focus point. In this **Z-contrast**, columns with heavier atoms will appear brighter, and vice versa. If the probe forming lens is aberration corrected, atomic resolution can be achieved.

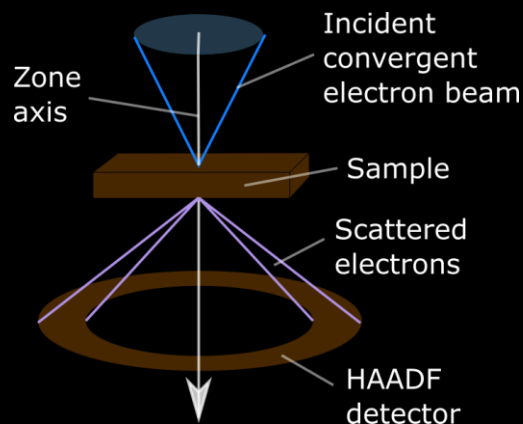


Fig. 2.1: A simple schematic of the HAADF-STEM imaging mode.

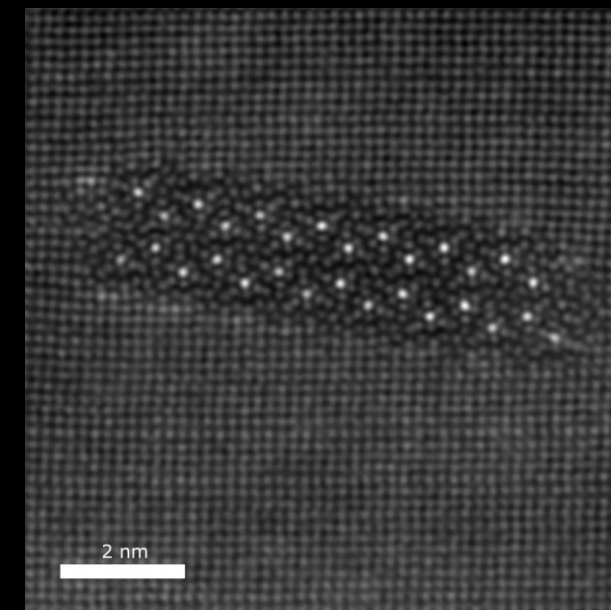


Fig. 2.2: An example of an atomically resolved HAADF-STEM image, showing a Q' precipitate in an Al-Mg-Si-(Cu) alloy. The bright columns are the Cu positions, which stand out because of the high  $Z$ -number of Cu. This image was acquired with the JEOL ARM 200, with a probe size  $<1$  Å at 200 kV high voltage.

## 2.1 HAADF-STEM

## 2.2 Structural principles in Al-Mg-Si-(Cu) precipitates

It has been found that the columns in the 6xxx system precipitates are commonly characterized by a set of simple structural principles or rules, that allow identification of the species and the relative displacement (longitudinal jumps) of the atomic columns [2].

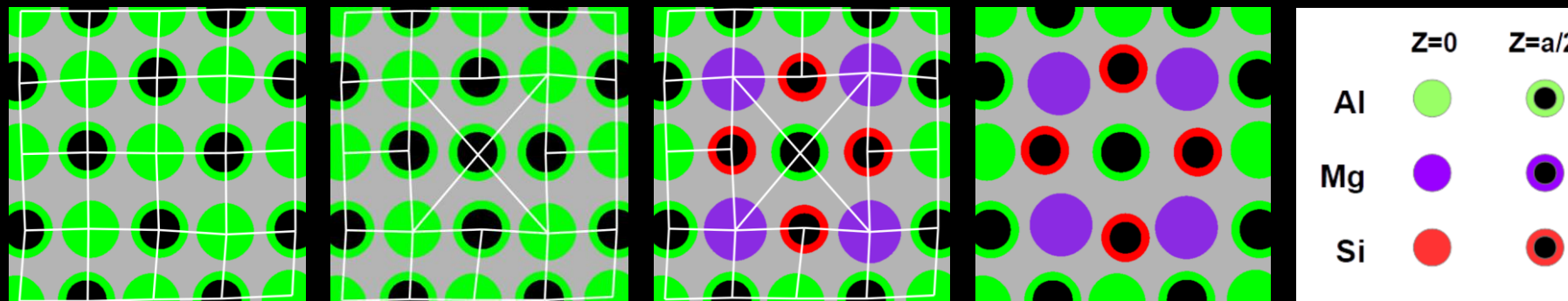


Fig 2.3: If a column is shifted into an interstitial position in the Face centered cubic (FCC) lattice (a line defect), the number of nearest neighbours (NN) of the surrounding columns change. In this figure, white lines show opposite plane NN's. Species with small atomic radii will favour positions with fewer NN's (Si and Cu with 3 opposite plane NN's), while larger species favors positions with more NN's (Al with 4 opposite plane NN's and Mg with 5 opposite plane NN's).

There are exceptions to these principles, for instance the interstitial Si-position in beta prime, but together with the Z-contrast information in HAADF-STEM images, and knowledge about the phases of the system, researches can create **atomic overlays**, which label the species and longitudinal position of each columns. However, if done manually, this is a highly tedious and time-consuming puzzle.

[2] Sigmund J. Andersen *et al.* "Directionality and column arrangement principles of precipitates in Al-Mg-Si-(Cu) linked to line defect in Al". *Materials Science Forum* 877 (Nov. 2016).



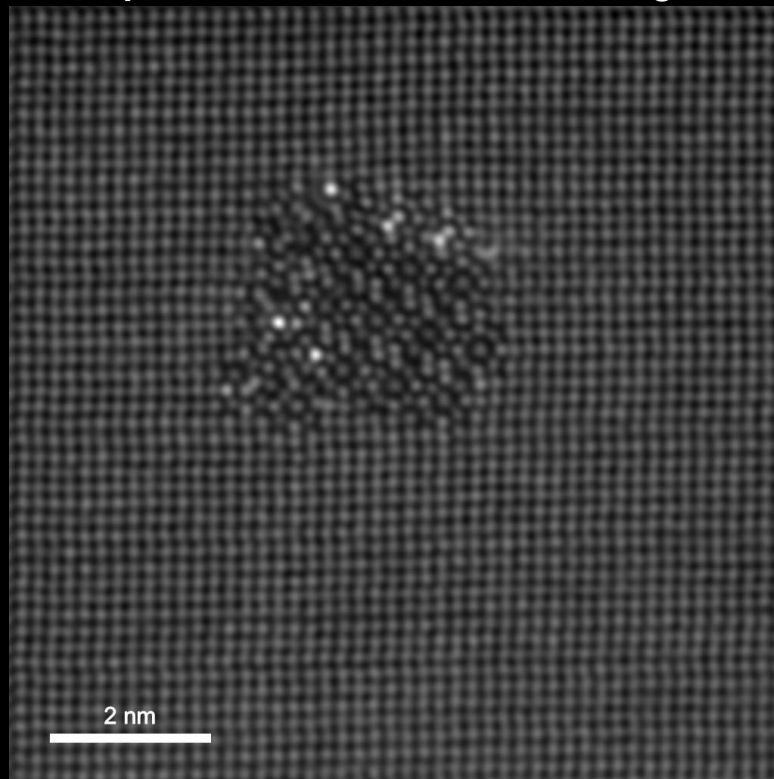
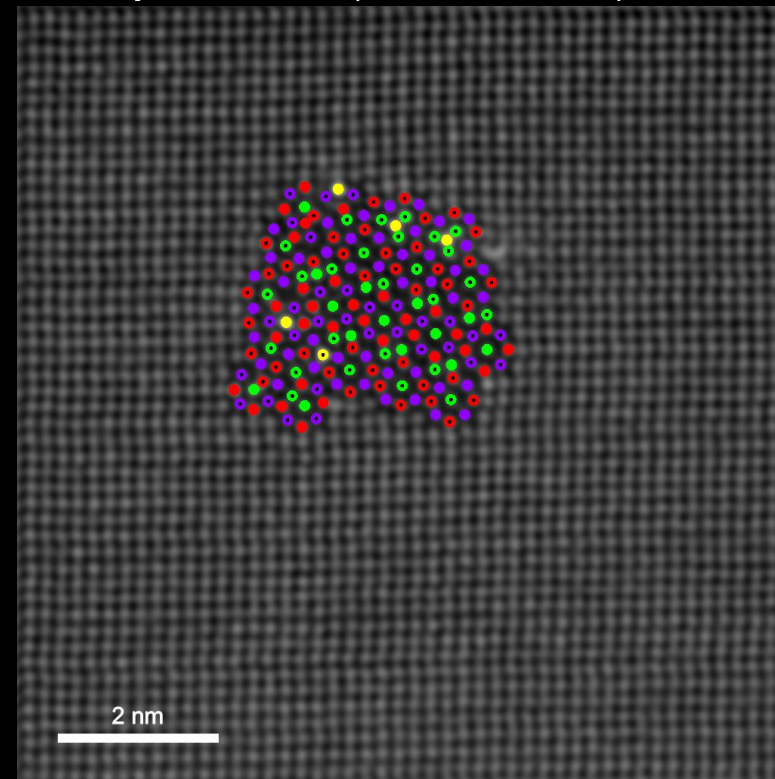
1. Introduction

2. Background

**3. Objective**

4. Method

5. Summary

**Input:** Filtered HAADF-STEM imageSemi-  
autonomous  
process**Output:** Atomic positions and species

*Fig 3.1:* The objective of AutomAI 6000. From an input HAADF-STEM image (appropriately noise-filtered), produce an atomic overlay showing the species and z-position of each column

## 4.1 Column detection

## 4.2 Atomic graphs

## 4.3 Statistical model

## 4.4 Column characterization

AutomAI 6000 features a basic **column detection** method based on calculating the **Center of Mass (COM)** of pixel intensities. The dm3 metadata for image scale is used to determine approximate atomic radii in pixels. A threshold value must be guessed by the user, but can be altered after column detection, which will incite the algorithm to either roll back on detection, or continue, depending on whether the threshold value has decreased or increased, respectively. The user can also refine the column detection by re-locating, adding or deleting columns with the GUI functionality.

No further efforts will be done to improve on column detection, because there already exists software which can do this well, particularly the widely used AtoMap by Magnus Nord [1]. For this reason, AutomAI 6000 can import column position data from files saved with AtoMap.

Feature	AutomAI 6000	AtoMap
Column detection	Yes	Yes
Automatic	Semi	Semi
Precision	Poor	Good

Table 4.1: Column detection software comparison

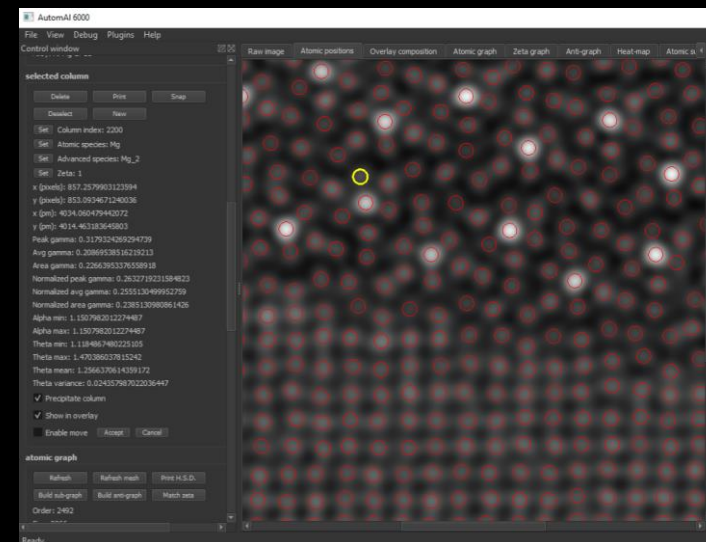


Fig 4.1: The column position tab of the AutomAI 6000 GUI. Information about the currently selected column (marked in yellow), is provided in the left control widget.

[1] Magnus Nord *et al.* "Atomap: a new software tool for the automated analysis of atomic resolution images using two-dimensional Gaussian fitting". *Advanced Structural and Chemical Imaging* 3.1 (Feb. 2017).

## 4.1 Column detection

## 4.2 Atomic graphs

## 4.3 Statistical model

## 4.4 Column characterization

### 4.2.1 Structure

### 4.2.2 Untangling

AutomAI 6000 uses **atomic graphs** to communicate its data-structure to the user, as well as a framework for its algorithms.

Atomic graphs are based on **simple digraphs** from graph theory, modified to fit the AutomAI 6000 application on structure data. In atomic graphs, vertices represents columns, and arcs represents the opposite plane NN structure.

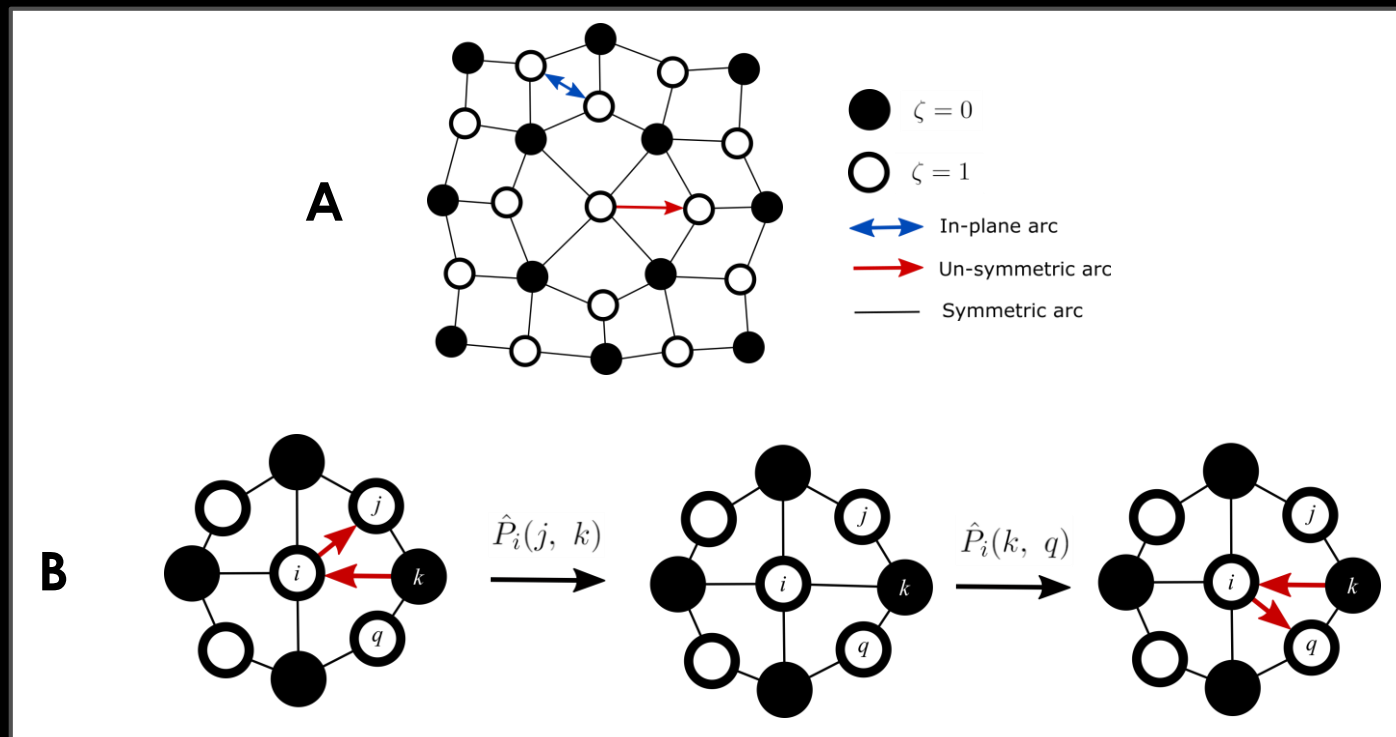


Fig 4.2: Atomic graphs. **A)** Filled vertices represents columns in the  $z=0$  plane, while hollow vertices represents columns in the  $z=a/2$  plane. Black lines represents double (symmetric) arcs, which means that both adjacent vertices agree that they are opposite plane NN's. Red arrows represents single (un-symmetric) arcs, meaning that the vertices do not agree that they are NN's. **B)** AutomAI 6000 will permute arcs during untangling. Here the central vertex  $i$  in a beta double prime "eye", erroneously believes the vertex  $j$  to be a opposite plane NN. A correct permutation operation will produce the correct NN structure for the beta double prime eye. Such permutations can also be performed by the user in the AutomAI 6000 GUI.

1. Introduction

2. Background

3. Objective

4. Method

5. Summary

## 4.1 Column detection

## 4.2 Atomic graphs

## 4.3 Statistical model

## 4.4 Column characterization

## 4.2.1 Structure

## 4.2.2 Untangling

A method termed **untangling**, uses a map of subgraph configurations to solve the structure such that the correct opposite plane NN's are connected by double (symmetric) arcs.

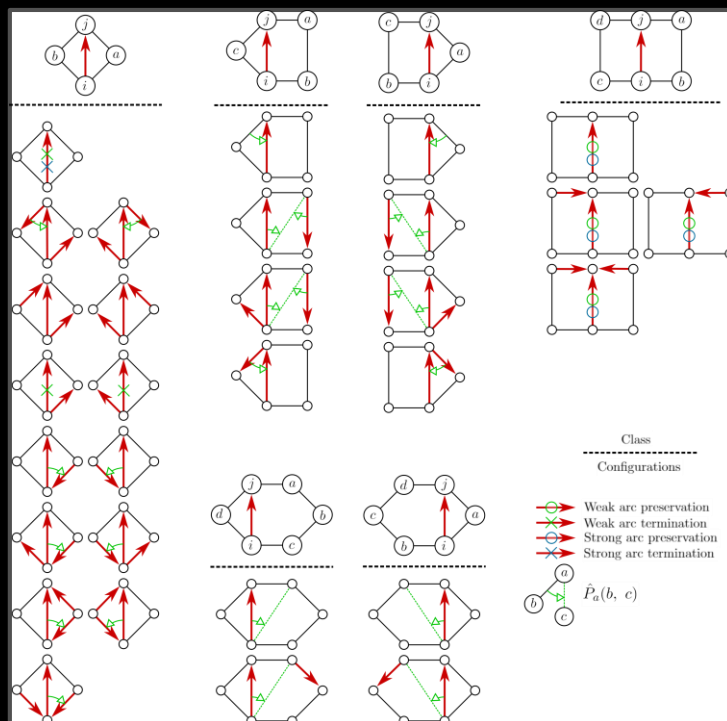
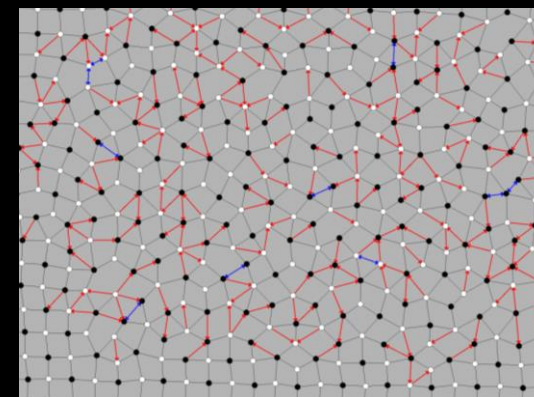


Fig 4.3: Configuration map used by AutomAI 6000 to untangle atomic graphs and attempt to find the correct NN structure.



Untangling

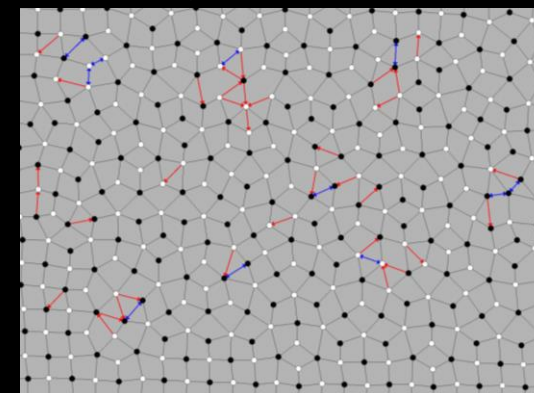


Fig 4.4: An atomic graph before and after untangling. The visual difference in the graphs provides some merit to the term "untangling".



1. Introduction

2. Background

3. Objective

4. Method

5. Summary

4.1 Column detection

4.2 Atomic graphs

4.3 Statistical model

4.4 Column characterization

4.3.1 Default model

4.3.2 User models

The atomic graph untangling method is good at determining the symmetries and NN structure of the graph, however this method cannot separate species with similar NN symmetries (3-fold Si and Cu look the same in an atomic graph). For a full column characterization method, all parameters (both symmetry indicators as well as Z-contrast information) must be considered statistically.

The default model preset is a **multinomial multivariate normal distribution**, which uses a nominal attribute termed **advanced species** for its categorization. The advanced species attribute captures more detail than pure atomic species categorization. This categorization scheme is termed the **species dictionary**. Each image might have different species dictionaries. The default model is calculated from a small set of images that include Q-prime, beta double prime, L, disordered and hybrid precipitates, and this model will only try to label species which are in the species dictionary of the current image.

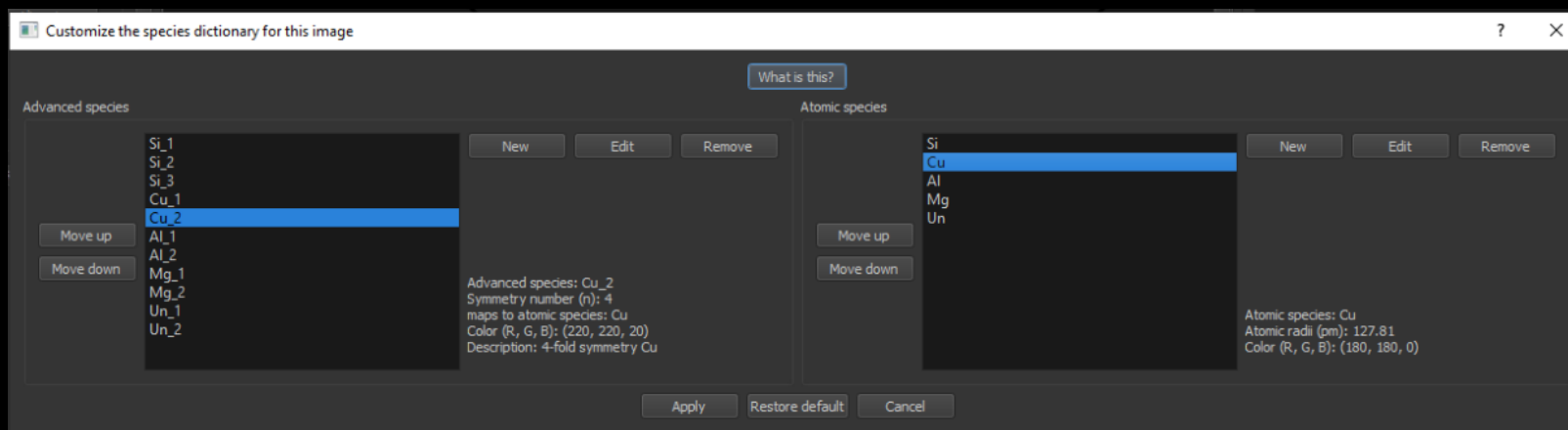


Fig 4.5: AutomAI 6000 species dictionary customization dialog window. Each advanced species must map to an atomic species, which is ultimately the assumed atomic overlay interest of the user.

1. Introduction

2. Background

3. Objective

**4. Method**

5. Summary

4.1 Column detection

4.2 Atomic graphs

**4.3 Statistical model**

4.4 Column characterization

4.3.1 Default model

**4.3.2 User models**

After observing some internal use cases here at NTNU, the conclusion was that researchers will typically be studying certain types of images or precipitates which are relevant to their current research projects. This altered the development philosophy of AutomAI 6000's column characterization algorithm, from being "a tool for everybody's problems", to being "a tool which can be configured for everybody's problems".

With AutomAI 6000's model wizard, a researcher can produce some overlays with the default model, which he or she must correct, and then calculate a new model from these overlays. In theory, this should produce a model which is ideal for the individual researchers' particular images.

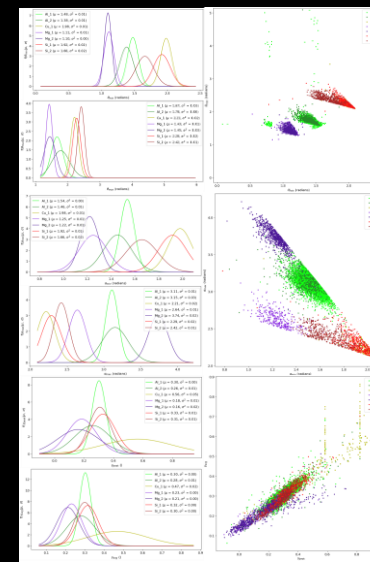
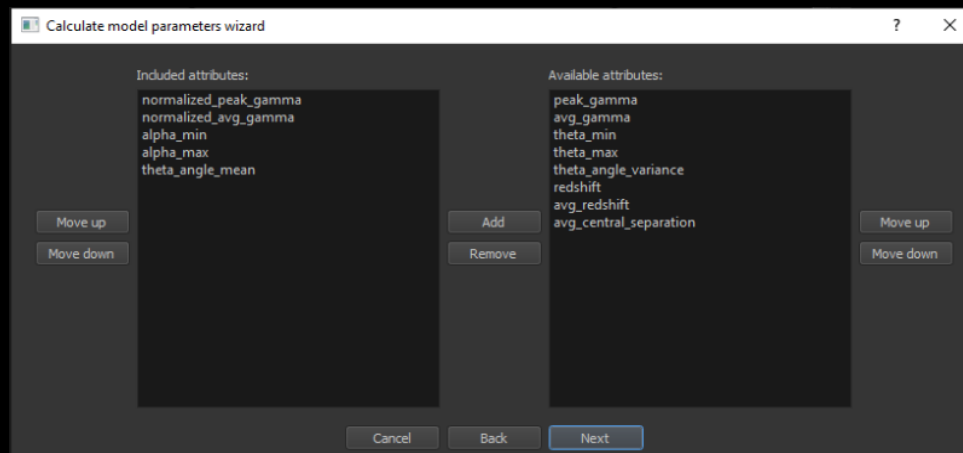
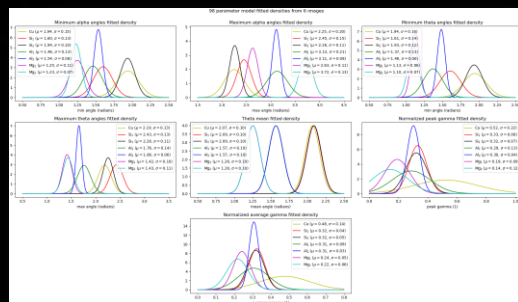
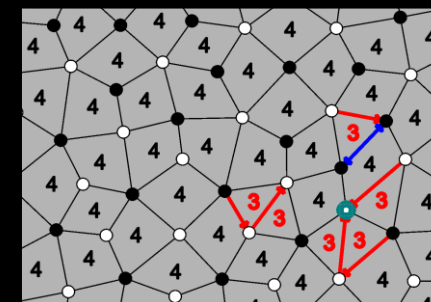


Fig 4.6: By using the model wizard, statistical models from user data can be calculated. These models can then in turn be used to overlay more images.

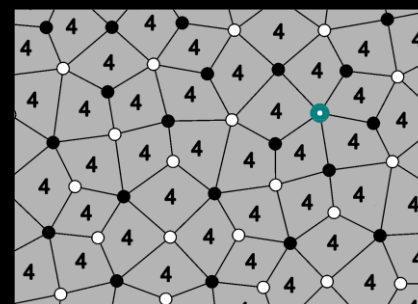
AutomAI 6000's **column characterization** algorithm is quite complex in the interplay between its many sub-methods. Qualitatively though, one might describe it as a symbiosis between the atomic graph untangling method, and the statistical predictions of the chosen model. On one hand, the statistical model needs correct NN structure information to give accurate species predictions, while on the other hand, the untangling needs correct species information to arrive at an accurate NN structure. The algorithm will move back and forth between the methods a few times, until the method converges at its best result.



Statistical species predictions will influence the number of arcs in the graph



A more correct NN structure will improve the accuracy of the statistical model.



Untangling will try to figure out the opposite plane NN structure based on species information and structural principles



Fig 4.7: A semi-accurate and qualitative description of AutomAI 6000's column characterization algorithm.

## 1. Introduction

## 2. Background

## 3. Objective

## 4. Method

## 5. Summary

## 4.1 Column detection

## 4.2 Atomic graphs

## 4.3 Statistical model

## 4.4 Column characterization

## 4.4.1 Algorithm

## 4.4.2 Interpreting results

Once the column characterization has run its course, result may vary, depending on many factors. If the literal atomic overlay had to be manually inspected atom by atom, then that would negate the purpose of the software. Luckily, the atomic graph is here to help. Problematic areas will stand out due to the red and blue arcs. A major effort has gone into making sure that the algorithm won't create symmetric NN structures that are not real, meaning that for the most part, the NN structure can be trusted wherever the graph is fully symmetric (no red arcs). However, keep in mind that correct NN structure does not guarantee a correct overlay, mostly due to the structural similarity between Si and Cu in the system. These columns should indeed be inspected, depending on how well the current model can separate the two species. Once the user becomes accustomed with the atomic graphs, and AutomAI 6000 in general, images with ~6000 columns can be processed from import to correct overlay in anything between 10 to 20 minutes, depending on the specific image details.

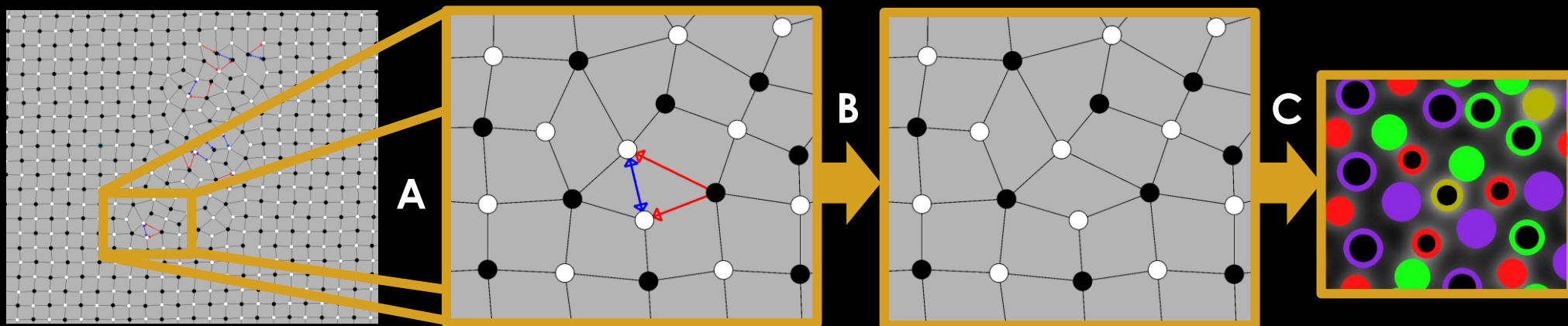


Fig 4.8: Manual inspection of the atomic graph. **A)** Locate any problematic areas. These will stand out with red and/or blue arcs. **B)** Apply your expertise and understanding of the atomic graphs to solve the area by permuting arcs and setting correct column species and z-height. **C)** Check Si and Cu columns against the raw image.



## 1. Introduction

## 2. Background

## 3. Objective

## 4. Method

## 5. Summary

## 5.1 Conclusion

## 5.2 Further work

## 5.3 Links

- Based on the structural principles in Al-Mg-Si-(Cu) alloys, we have created a tool which simplifies the overlaying task for researchers. However, since the method is only semi-automatic, the results must still be evaluated by individuals familiar with the 6xxx precipitation sequence. This manual revision is aided by visual clues in the atomic graphs.
- Easily extract the number density composition of your precipitates, as well as other precipitate attributes, like precipitate/FCC displacement or packing fractions. A whole slew of statistical insights that was previously time-consuming to extract, are now more easily available.
- Create atomic overlays that can be exported as layered **Scalable Vector Graphics** (.svg). Publishing quality overlays can then be created with software like Inkscape.
- Export the structural precipitate data as **Comma Separated Values** (.csv) that can be parsed for import into 3-d modelling software, simulation software or other analytical tools.
- Create a labelled database of your images, which can then be used to create statistical models, or (if large enough) serve as training data for machine learning. This data can then also be queried in many novel ways, for instance to create heat-maps, which is currently being considered as a potential path towards detecting clustering in 6xxx.

1. Introduction

2. Background

3. Objective

4. Method

5. Summary

5.1 Conclusion

5.2 Further work

5.3 Links

- Stable version
- Documentation and guides/tutorials
- Executable
- API design for non-GUI applications
- Bring the web-page up to date
- Improvements to the underlying methodology is a continuous process

- Web-page (Documentation, downloads, guides, etc): <http://automal.org>

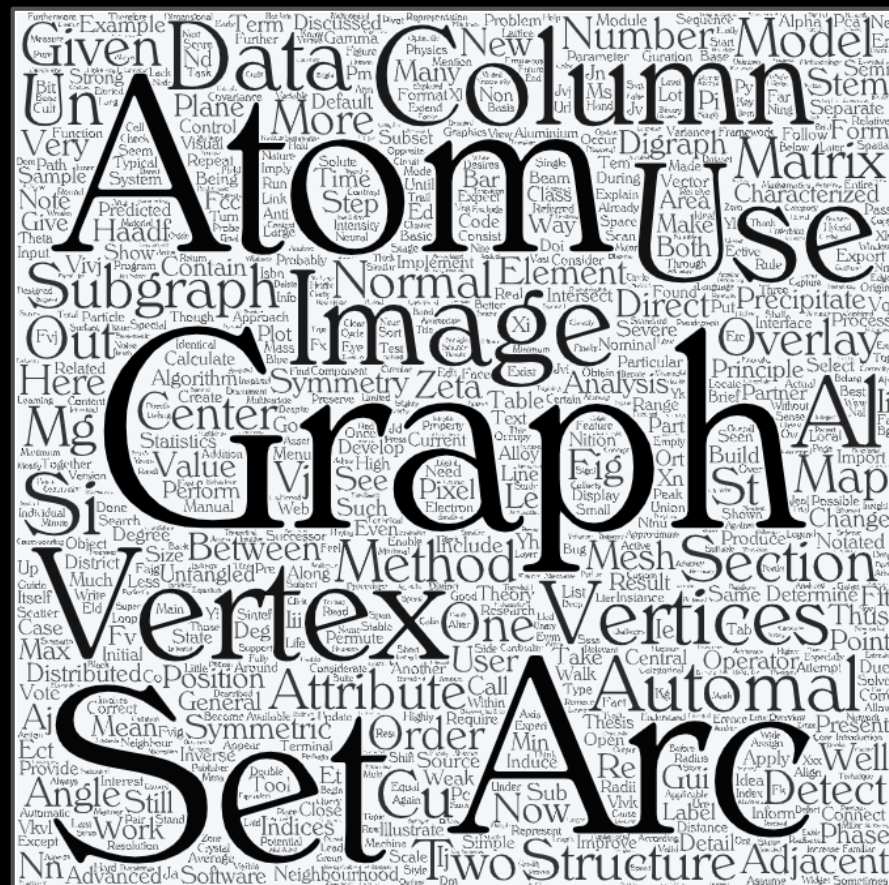


Fig 5.1: Word-cloud from my thesis