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A new systematic framework for crystallographic analysis of atom probe data

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ABSTRACT

In this article, after a brief introduction to the principles behind atom probe crystallography, we introduce methods for unambiguously determining the presence of crystal planes within atom probe datasets, as well as their characteristics: location; orientation and interplanar spacing. These methods, which we refer to as plane orientation extraction (POE) and local crystallography mapping (LCM) make use of real-space data and allow for systematic analyses. We present here application of POE and LCM to datasets of pure Al, industrial aluminium alloys and doped-silicon. Data was collected both in DC voltage mode and laser-assisted mode (in the latter of which extracting crystallographic information is known to be more difficult due to distortions). The nature of the atomic planes in both datasets was extracted and analysed.

1. Introduction

Atom probe tomography is a technique that is capable of providing the chemistry and spatial coordinates of atoms in small volumes, approx 100 nm × 100 nm × 300 nm, of solid material [1–3]. While being used extensively as a tool for 3D compositional mapping at the nano scale, it is not commonly used for crystallographic analysis. Atom probe data has been known to contain crystallographic information, the full potential of which has not been realised [4–6]. While progress has been made in this area in recent years, there remains a need for fast and highly accurate crystallographic analysis tools in order to obtain grain orientations for advanced analytical techniques such as lattice rectification [7] or for reconstruction approaches based on crystallography. Most atom probe crystallography methods rely on the observation of reconstructed crystallographic planes in a dataset, their identification, and measuring their orientation relative to the atom probe detector [5,8–11]. These planes are found in the vicinity of poles in the datasets. By determining the orientation and identification of two sets of crystallographic planes relative to the detector, the orientation of the grain relative to the detector can be determined and hence the orientation of objects within the detector can be determined relative to the crystal grain.

Manual methods of observing crystallographic planes and measuring their orientation were implemented by Liddicoat et al. [12] and the orientation of different crystal planes with the same Miller indices in adjacent grains were compared. This method is highly labour intensive and in some cases unfeasible as it is difficult to see the crystal planes in the cases of poorly resolved planes (common in silicon data) or high order planes which have low planar densities and small planar spacings.

3D Fourier methods were first proposed for atom probe data by Camus et al. [13] and were first used for analysis of atom probe data by Warren et al. [14] but these were at the time computationally expensive to study large datasets. This method was adjusted by Vurpillot et al. [15] to reduce the computational expense but this approach is still significantly time consuming.

Spatial distribution maps (SDM) have also been developed and applied to atom probe data [16–18]. This is a real space method which splits the three-dimensional radial distribution function into a one-dimensional distribution of atoms along an analytical axis as well as the 2D distribution of atoms perpendicular to that same axis. This technique is commonly used in determining reconstruction parameters for atom probe tomography data as it is capable of accurately measuring the crystal plane spacing [18]. It has also been used to measure the difference in orientation of crystallographic planes with the same identification in different grains in the same dataset [9], but this method requires knowledge of the location of the crystal planes, making it highly manual.

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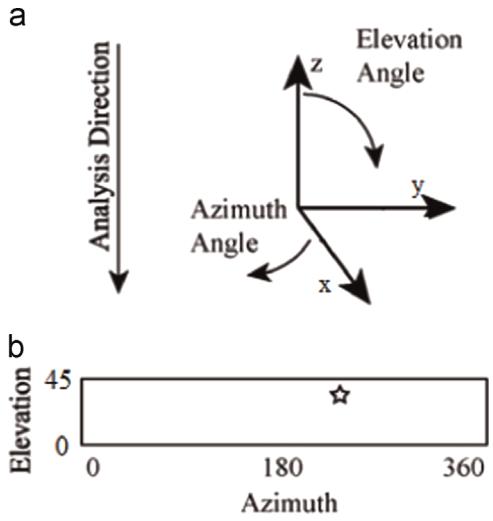

Fig. 1. (a) Schematic showing the convention of azimuth and elevation angle used here and (b) depiction of a signal vs plane orientation plot where the star marks a specific orientation.

3D Hough transforms are capable of determining the orientations of planes in 3D point cloud data and have been adjusted and used in conjunction with 1D Fourier transform techniques to analyse crystal planes in atom probe data by Yao et al. [8]. This approach is highly automated and efficient at obtaining the orientation of sets of planes in small volumes (20 nm × 10 nm × 10 nm) and their crystallographic spacing. It was recently used in several studies to determine grain orientation and grain boundary misorientations [19–21]. However, this technique also requires knowledge of the location of atomic planes in a dataset in order to extract the region of interest (ROI) to input into the transform.

In a recent work by Larson et al. [22] a method was presented which maps the location of planes across a slice of an APT reconstruction. This method used a plane of best fit approach to determine plane orientation coupled with Fourier transform methods to determine the location of atomic planes. This method is unfeasible for many datasets due to the the reliance on a fit that can be problematic with noisy data.

In the aforementioned studies, there are no true automated means of extracting crystallographic data from a previously unexamined dataset. This is due either to the inability of the above methods to deal with large datasets (greater than ~1 million ions) caused by computational constraints, the required input of a specific, manually extracted, ROI containing planes, or the manual search for plane orientation as in the case of SDMs.

This paper outlines a series of methods to automatically extract crystallographic information from atom probe data that enables us to answer the following three questions: (1) Is there crystallographic information in the form of planes present in a reconstructed atom probe dataset? (2) If planes are present, what are the Miller indices and the experimentally measured orientation and spacings of those planes? (3) Which atoms in the atom probe datasets are lying on which planes? Answering these questions would greatly reduce the current difficulties related to crystallographic analysis of atom probe data.

2. Methods

2.1. Conventions

Throughout this article we will use the conventions used in Yao et al. [8] to display the orientations of crystal planes in atom probe data. Fig. 1a depicts the convention of azimuthal and elevation angle and their relation to the common Cartesian coordinates relative to the atom probe detector which is commonly used. Fig. 1b is a depiction of a crystallographic direction in angular space. This convention is particularly useful in analysing plane normal orientations relative to each other. As most sets of reconstructed atomic planes in atom probe data have an elevation angle of less than 45°, these plots will generally have an elevation angle range from 0° to 45° and an azimuthal angle range from 0° to 360°. The elevation angle limits are chosen as the field of view of atom probe data is in general 45° or less and atomic planes are unlikely to be resolved beyond this range.

2.2. Plane orientation extraction

The input to the plane orientation extraction (POE) is a selected region of an atom probe dataset. It places a single analysis point, P, in the dataset, creates a region of interest (ROI) around P and determines the orientation of reconstructed planes (if they are present) in the dataset according to the following protocol:

(1) Place an analysis point, P, within the input dataset. The coordinates of P used here are the midpoint between the maximum and minimum X, Y and Z coordinates of the input dataset. For a spherical dataset, it would be the centre of the sphere.
(2) Create a spherical ROI containing all atoms within a radius, R_{POE}, of P.
(3) Create list of all possible plane orientations $N(\theta,\varphi)$ where θ is the azimuthal angle and φ is the elevation angle from a given range. A standard set of possible plane orientations for atom probe data would range the azimuthal angle from 0° to 360° and elevation from 0° to 45° at 1° increments as planes at inclines greater than 45° are unlikely to be resolved in atom probe data. Decreasing the degree increment provides greater accuracy but increased computation time.
(4) For each plane normal $N(\theta,\varphi)$:
    (a) Calculate the perpendicular distance of each atom in the ROI to the plane defined by the plane normal $N(\theta,\varphi)$ and the analysis point as depicted in Fig. 2a.
    (b) Create a histogram of the distances, as seen in Fig. 2b for a pure aluminium specimen.
    (c) Apply a 1D Fast Fourier Transform (FFT) to the histogram and plot the absolute value. Extract the location and height of peak with highest signal, as seen in Fig. 2c. The characteristic noise on the left hand side of the FFT is filtered out by selecting a peak to the right of where the FFT first reaches the median value from all values of the FFT.
(5) Plot peak strength vs $N(\theta,\varphi)$, as can be seen in Fig. 2d.

This protocol efficiently identifies the orientation of planes in a region known to contain them, such as a ROI extracted from a pole. Further accuracy in determining plane orientation can be obtained by refining the plane orientation search parameters. The planar spacing can be determined by examining the FFT plot of the plane normal orientation that provided the highest signal.

Fig. 3 shows the output of the above procedure applied to a model fcc aluminium data with R_{POE} of 2 nm in order to test the effectiveness of the protocol. The angular range was set at 0° to 360° azimuth angle and –90° to 90° elevation angle with an angular binning of 1°. This is similar to a Mercator projection and depicts the data's plane orientations. The POE is limited by the choice of binning for the histogram, $R_{POE,}$ and the orientation space inputted. The variations in peak strength shown are dependent on planar densities of the sets of planes. In real atom probe data