Stereological analysis of triple line orientations

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Abstract. Determining underlying interface geometries in polycrystalline materials is a daunting task and one that is an ongoing field of materials research. This work utilizes a method of determining structural anisotropy through stereological methods. The orientations of simulated triple-lines were characterized by analysing intersections of the triple-lines on randomly orientated planar sections in a given volume. Using randomly oriented sections, the triple-line orientation distribution, \( L_V(\phi, \theta) \), can be estimated. After approximately 10-100 sections, the true triple-line orientations can be determined with reasonable accuracy. The stereological method proposed is being used to show how it may be useful to the characterization of triple junction character in a real material system. The efficacy of the method was discussed along with considerations to reducing the overall variance in the distributions as well as applying the stereological procedures to real polycrystalline triple junction networks.

1. Introduction

In materials science, the polycrystalline triple junction has often been neglected as a studied microstructural feature. Since triple junctions are inherently three-dimensional objects, studying the geometry usually results in a disregard of the spatial orientation and underlying geometries. There have been many ways of stereologically determining the spatial arrangement and orientation of lineal features [1–3] in three-dimensions by quantifying line-length per unit volume (\( L_V \)), but there has been relatively little mention of using these methods to study triple junctions in polycrystalline systems. In the 1960’s Hilliard presented a stereological method for extracting the orientation of lineal features embedded in an arbitrary volume [4–6] which bares resemblance to Buffon’s needle experiment [7]. Hilliard’s work had stated that when randomly sectioning a given volume, the distribution of the number of intersections (\( N_s \)) of the lineal features on the oriented section can be used to estimate the total line-length per unit volume, \( L_V \). Utilizing the relationship between the probabilities of a section intersecting a lineal feature and the orientation of the lineal feature, a distribution of line orientations, \( L_V(\phi, \theta) \), can be measured. This distribution will give some indication of the anisotropy of interfaces in a volume.

Determining the 3D structure of materials is an on-going and continuously advancing science. Being able to characterize the total area of interfaces or understand grain boundary character in polycrystalline systems is extremely useful to the design of materials [8–11]. Knowing the total length
of interfaces such as fibers in composites or triple junctions in polycrystals would be of great importance to modeling and study of the structure-properties-performance of a material. It is generally understood that the orientation of triple junctions with respect to an observed free surface has an impact on properties such as recrystallization, grain growth, precipitation and various mechanical properties [12–16]. However, the actual orientation of the triple junction geometry is difficult to determine, making the study of structure-property-performance effects of triple junctions cumbersome.

While there are methods of gathering three-dimensional information of interfaces in a material, i.e. X-ray chromatography (CT), micro-CT, atomic probe tomography (APT) or high-energy diffraction microscopy (HEDM), these methods can be timely and/or costly to use and tend to be limited by small sample sizes. The use of stereology has been employed for relatively large samples and can be used readily to extract statistical and geometrical information about a given material. The grain boundary character distribution (GBCD) is widely studied in polycrystalline materials using stereological methods [10,17–20]. Since most forms of crystallographic information come in two-dimensional forms like electron backscatter diffraction (EBSD), the use of stereology to characterize interfaces become easier to manage experimentally. However, applications of stereological methods to characterization of triple junctions has been slow to develop. While some researchers have used stereological methods to study specific types of triple junctions [21], most common forms of triple junction characterization are done by observing the available information from a single two-dimensional section [8,10,16,22–24], neglecting the underlying three-dimensional information, which was acceptable for the given application. If the underlying geometrical information is desired, then additional methods that provide statistically viable datasets must be developed.

The purpose of this work is to shed light on the efficacy of determining \( L_V(\phi, \theta) \) and whether the stereological method can be applicable to material interfaces. Simulations of the method are done using multiple geometrical scenarios. The results of the stereological simulations indicate that it is possible to measure the orientation distributions of the triple junction interface, with some limitations. The type of feature geometries and the number of cuts through the volume that are needed to reliably extract the orientations of lineal features affect the efficacy of the stereological method.

2. Methods

Using a spherical reference frame, the set of coordinates \((\phi, \theta)\) will represent the polar and azimuthal angles of the triple-line orientation or “line sense”. The section, which is used to intersect the volume with the embedded triple junctions, will be defined by a plane with a set of spherical coordinates \((\omega, \psi)\) and a centroid \(P_0\). The specification of \(L_V(\phi, \theta)\) according to Hilliard is

\[
L_V = \int_0^\pi \int_0^\pi L_V(\phi, \theta) \sin \phi \, d\phi \, d\theta
\]

It is assumed that randomly oriented sets of triple-lines have uniform density of the surface of the reference hemisphere, thus the result from Eq. 1 shows that for uniform density \(L_V(\phi, \theta) = L_V/2\pi\) [5]. The full derivation of the method can be found in [5] but, for the sake of brevity, we will present only the most pertinent equation that provides the basis for the stereological procedure which was used in this analysis.

To test this hypothesis, synthetic microstructures and triple junction geometries were generated via the open-source Dream3D microstructure analysis software [25]. Information of grain interfaces were exported and custom routines to reconstruct the triple junction networks were created in MATLAB R2019a.
A point \( P_0 \) was defined in the bounds of the volume with randomly chosen normal angles \((\omega,\psi)\) used to define a section centered on \( P_0 \). The number of intersections the plane makes with the triple-line network were counted and subsequently divided by the area of the section. The result would be a section plane with polar angles \((\omega,\psi) = (\varphi,\theta)\), and intensity of \( N_A/A \), where \( A \) is the area of the section. After a user-defined number of section planes, a harmonic representation of \( N_A(\omega,\psi) \), the number of sections per unit area, can be made and normalized by \( 1/2\pi \) to estimate line-length per unit volume \( L_V(\varphi,\theta) \).

The open-source crystallographic analysis package MTEX [26] for MATLAB was used to estimate \( N_A(\omega,\psi) \) and \( L_V(\varphi,\theta) \) and to visualize the orientation distributions. All distributions were estimated using spherical harmonics up to order \( M = 8 \). All distributions are observed with respect to the Z-axis with the X-axis to the east and Y-axis to the north.

### 3. Results

To test the efficacy of the solution, simple geometries were first tested to see if the method was able to extract the orientation of a single triple-line. Figure 2 presents three simple geometries of triple-lines. A series of section planes were made through the volume and \( N_A \) was calculated.

For each case of line geometry, 10-100,000 sections were made. After \(~10-100\) sections the appearance of peaks formed in close proximity to the true line orientation. Smoothing of the orientation distribution can be seen after 1,000 sections with 100,000 sections resulting in a uniform Gaussian like distribution centered at the location of the true line orientation.
Studying an increasingly complex system of triple-lines, Figure 3, a grouping of straight lines were created at random locations and orientations within a unit volume. After repeating the same stereological procedure as before it appears that 100 sections through the volume was enough to extract the orientation of the random set of features. It should be noted that the resolution of the \( L_V(\phi,\theta) \) is dependent on the harmonic order used. In the MTEX analysis software, the distribution of \( N_A \) was first estimated using a harmonic approximation up to order \( M=8 \), followed by convoluting the harmonic estimation with spherical de la Valle Poussin kernel of halfwidth 10°.

Figure 3: A selection of randomly oriented lineal features in a unit volume. \( L_V \) distributions indicate that between 100-1000 sections, the orientations of the lineal features were able to be extracted. Hilliard’s stereological method appears to work for increasingly complex geometries of lineal features.

Figure 4: A geometry of lineal features that was not able to be resolved through stereological procedures. Peaks were shifted \(-45°\) about the \( y \)-axis. The location of the peaks after 1,000 slices indicate that there are certain geometries of lineal features that may not be accessible via Hilliard’s stereological procedure.

The stereological method has additional drawbacks, however. Presented in Figure 4 is a case where the geometry of the triple-lines was not able to be accurately determined. Two lines perpendicular to each other and intersecting at their midpoints produced a stereologically-derived distribution that did not accurately portray the orientation of the lines. The peaks in the resultant distribution were shifted by 45° in-plane of the triple junctions. Comparing the true distribution to the distributions after 1,000 section planes, it is apparent that the peaks show a \( \phi=45° \) shift. In real material systems, triple junction geometries such as that presented in Figure 4 are not possible. These could be possible with other lineal systems of interest, however, such as fiber orientations.

4. Discussion
Line-length per unit volume is a measure of the average orientation of a lineal feature, more specifically to the current work, triple-lines. The resultant distribution \( L_V(\phi,\theta) \) is a density of these orientations having no knowledge of their true orientations \textit{a-priori}. What \( L_V(\phi,\theta) \) does not tell the observer is the physical topology of the interfacial network. Much in the same way that a grain boundary character distribution (GBCD) tells the researcher the statistical distribution of grain boundary parameters, it does not indicate the connectivity of the grain boundary network in three dimensions. It is theoretically possible for two triple junction networks to have completely different topologies but result in the same orientation distribution, \( L_V(\phi,\theta) \). The spatial locations of triple junctions are indeterminate using this technique. It is anticipated that this method is material dependent since highly anisotropic grain shapes will affect the ability to accurately determine \( L_V(\phi,\theta) \) in real systems. For materials that exhibit grain size gradients, this method may be limited in its ability.
In [5], Hilliard points out that there are issues of which the observer must be aware when estimating $L_V(\phi, \theta)$. First, the choice of section plane can have an impact on minimizing the standard deviation of the distribution. The section used in this work was a section plane; a more useful section would be a hemispherical surface or sphere [3]. Using a hemispherical section instead of section planes could be used in a similar manner and would result in minimizing the overall variance of $L_V(\phi, 0)$. It is noted that a hemispherical section is near impossible to do practically. Second, the optimum number of section planes depends on two factors; first being identifying the degree at which the triple junctions deviate from that of a random distribution. For a random distribution, a single section plane would suffice to determine $L_V(\phi, \theta)$, but increasing anisotropy would elicit the need to have some prior knowledge of the grain morphology. The other factor is keeping a balance between the nonuniformity of the orientation distribution and spatial distribution of the section. Ideally, one would use section planes that sweep over all Cartesian and polar space as this would capture the triple junction orientations the best, but this is experimentally unreasonable. Thus, care must be taken when determining the type of sectioning done and having additional grain morphology information would help determine the spatial distributions of section planes and triple junction orientations. Real material systems are never truly random. Any deviation from “randomness” would negate the stereological assumption that the method is homogeneous. When determining $L_V(\phi, 0)$, the observer must understand that there are limitations since the material is more often than not inhomogeneous and may require other forms of creating an unbiased estimation of $L_V(\phi, 0)$.

The work presented here shows the efficacy of the stereological method of determining $L_V(\phi, \theta)$ for triple-line orientations. While the results indicate that more than ten to one hundred randomly oriented planar sections would be needed to determine $L_V(\phi, \theta)$, for practical purposes this is a daunting task, and likely should be substituted with a more systematic means of determining $L_V$.

Knowing the orientation distributions of triple junction networks would be of great benefit to interfacial science and real materials systems. Since the geometries of polycrystalline triple junctions are inherently 11-dimensional [21,27], additional geometries will need to be added to $L_V(\phi, \theta)$. By showing that it is possible to stereologically produce the orientation distribution of triple-lines, a way of characterizing real triple junction geometries can be developed.

5. Conclusion
The efficacy of stereologically determining line-length per unit volume was studied on simplistic and simulated triple junction networks from the use of randomized section planes. The number of intersecting points on the section planes were used to construct a distribution of line-length per unit volume $L_V(\phi, 0)$. It was determined that for simplistic triple-line geometries, the stereological procedure was able to capture the line directions of the interface network after ~10-100 randomized section planes. By showing $L_V(\phi, 0)$ can be determined from isotropic-uniform-random sections in simulated triple-line system, applications towards a triple junction characterization method for real polycrystalline systems can be developed.

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