Direct measurement of two-dimensional and three-dimensional interprecipitate distance distributions from atom-probe tomographic reconstructions

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Edge-to-edge interprecipitate distance distributions are critical for predicting precipitation strengthening of alloys and other physical phenomena. A method to calculate this three-dimensional distance and the two-dimensional interplanar distance from atom-probe tomographic data is presented. It is applied to nanometer-sized Cu-rich precipitates in an Fe-1.7 at. % Cu alloy. Experimental interprecipitate distance distributions are discussed. © 2007 American Institute of Physics. [DOI: 10.1063/1.2753097]

Many physical properties of materials depend on the edge-to-edge interprecipitate distance, $\lambda_{e-e}$. The applied stress required for a dislocation to glide past or climb over precipitates depends on $\lambda_{e-e}$, as does precipitate coarsening and electrical conductivity. Frequently, $\lambda_{e-e}$ is merely approximated by assuming that the precipitates form a cubic array or a square array in a plane. It is also assumed that precipitates are spherical with a known precipitate size distribution (PSD) usually either all precipitates are the same size or they obey the PSD derived by Lifshitz and Slyozov and Wagner (LSW). Real materials are almost always more complicated.

Much of the past work on calculating the distance between precipitates or other microstructural features of interest (whether interprecipitate distances, mean free paths or chord lengths, or nearest-neighbor distribution functions) has been theoretical. Experimental characterization of $\lambda_{e-e}$ requires a microscopic technique that has (i) a high enough spatial resolution to define clearly each and every precipitate and (ii) a large enough analysis volume to capture many precipitates and to exclude boundary effects. Furthermore, three-dimensional (3D) information (without suffering from precipitate overlap or truncation) makes determining the 3D distance distributions easier. For nanometer-sized precipitates, the local-electrode atom-probe (LEAP) tomograph (Imago Scientific Instruments) satisfies these requirements. Despite these capabilities, it has not been previously utilized to gather this information and the little available experimental data for $\lambda_{e-e}$ come from two-dimensional (2D) techniques. These cannot be compared directly to models of 3D microstructure, but only to 2D slices from theoretical 3D microstructures (although a stereological transformation might be possible).

In this article, an algorithm to calculate $\lambda_{e-e}$ from LEAP tomographic reconstructions is presented and applied to a binary Fe–Cu alloy. This alloy and many other steels are strengthened by a high number density of nanometer-sized copper-rich precipitates. Many of the proposed precipitate strengthening mechanisms depend on $\lambda_{e-e}$. Atom-probe tomography has been used to study the size, morphology, and chemical composition of Cu precipitates, but not to measure $\lambda_{e-e}$. An Fe-1.7 at. % Cu alloy was solutionized at 1000 °C for 1 h and 845 °C for 6 h. It was subsequently aged for 2 h at 500 °C. This treatment leads to a high number density $[(1.2\pm0.1)\times10^{24} \text{ m}^{-3}]$ of nanometer-sized precipitates (with a mean radius $\langle R \rangle$ equal to 1.3 nm). The specimens were cut, ground, and then electropolished into tips. The LEAP tomographic experiment was conducted with a 50 K specimen temperature, a 5–10 kV specimen voltage, pulse fraction of 15%, and a pulse repetition rate of 200 kHz to collect $\sim 1.3 \times 10^6$ ions in a 148 × 66 × 62 nm$^3$ volume [Fig. 1(a)]. The computer program IVAS (Imago Scientific Instruments) was used to analyze the data. Precipitates are isolated using a modified envelope algorithm. Because Cu partitions strongly to precipitates, an isoconcentration surface was not necessary to distinguish the 546 precipitates in this data set.

The interprecipitate distance algorithm begins by representing these precipitates with simpler geometric shapes. While $\lambda_{e-e}$ between spheres is simple (it being the difference of the center-to-center distance and the precipitate radii), spheres do not adequately represent many precipitate morphologies. Instead, best-fit ellipsoids to the precipitates are calculated [Fig. 1(b)] employing a recently presented algorithm. The $4 \times 4$ transformation matrix calculated with that algorithm translates, rotates, and scales a unit sphere centered at the origin to an ellipsoid that preserves the centroid, principal axes, and moments of inertia of a precipitate.

A Delaunay tetrahedral mesh is generated from the precipitate centroids [Fig. 1(c)]. The Delaunay mesh is the geometric dual of the Voronoi diagram; mesh segments connect neighboring precipitates whose Voronoi cells touch. It decreases the number of precipitate pairs for which $\lambda_{e-e}$ is calculated to a group of neighbors. The mesh also finds the 75 precipitates that make up the convex hull. These outermost precipitates are allowed to be nearest neighbors of the inner precipitates, but their own nearest neighbors are not calculated as they might fall outside the volume of the analysis.

The distance between two ellipsoids is found utilizing the constrained optimization by linear approximation (COBYLA) algorithm. This general optimization algorithm is

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chosen over more efficient algorithms that calculate explicitly the distance between ellipsoids, so that it can be used with other abstractions of precipitate morphology (such as the isoconcentration surface) and additional constraints (such as calculation of interplanar edge-to-edge distances) and because a gratis implementation exists. COBYLA minimizes the distance between two points, and in the analysis space, \( \sqrt{\sum_j (x_j - y_j)^2} \). The constraints are that and must fall on the ellipsoid. This is simplified by the fact that applying the inverse transformation of ellipsoids transforms them back into unit spheres, centered at the origin (so \( \Sigma j x_j^2 = 1 \) and \( \Sigma y_j^2 = 1 \), where the superscript \( T_i \) is the inverse transform of the best-fit ellipsoid for precipitate \( i \)). The initial guess is chosen as the two closest points that satisfy these constraints that lie on the line that connects the precipitate centers. Inter precipitate distance distributions (IDDs) may be generated using different combinations of Delaunay neighbors, as in Fig. 2. An IDD is the convolution of a PSD and the center-to-center distances. In Fig. 2(a), an IDD for all 6771 Delaunay neighbor distances yields a mean 3D interprecipitate distance, \( \langle \lambda_{c-e}^{3D} \rangle \), of 16 nm. Figure 2(b) displays two subsets of this IDD, each with 471 lengths because there is one distance associated with each of the 471 interior precipitates that are not on the convex hull. The distance between nearest precipitates is often used to calculate precipitate-dislocation interactions. The IDD for this is much sharper and \( \langle \lambda_{c-e}^{3D} \rangle = 2.6 \text{ nm} \). Precipitates that are very close to one another might be bypassed as a pair by a dislocation. The longest Delaunay distances provide an upper bound to the interactive distance. This is probably not physically important for plastic deformation, but may be relevant for other physical phenomena. The IDD for this case is broader, does not overlap the shortest distances, and has a mean value that is an order of magnitude larger \( \langle \lambda_{c-e}^{3D} \rangle = 25 \text{ nm} \). While 69% of the distances are shorter than this mean value, the distribution has a long tail that extends to 58 nm. Although the length of this tail is on the order of the diameter of the analysis volume, this is coincidental and it is the large variance in Delaunay distances that causes it. This tail is quite short and less than 5% of the lengths are 50 nm or longer.

In certain cases, it is not \( \langle \lambda_{c-e}^{3D} \rangle \) that is of interest, but rather the interplanar edge-to-edge distance, \( \langle \lambda_{c-e}^{2D} \rangle \). This might, for instance, be a glide plane of a dislocation. This 2D distance can be calculated by imposing an additional constraint for COBYLA—that and values must fall on a particular plane. For comparison, \( \langle \lambda_{c-e}^{3D} \rangle \) and \( \langle \lambda_{c-e}^{2D} \rangle \) can be calculated from one another by assuming precipitates are distributed on a cubic lattice:

\[
\langle \lambda_{c-e}^{3D} \rangle = \left( \frac{1}{\phi} \right)^{3/2} \langle \lambda_{c-e}^{2D} \rangle,
\]

where \( \phi \) is the volume fraction of precipitates. Assuming a square array of precipitates,
We are in the midst of applying this approach to calculate the strength of different alloys using analytical equations. We are also using it to evaluate the statistical accuracy of simulated microstructures, which are used in a continuum dislocation dynamics simulation that calculates a stress-strain curve.

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