The effects of elastic stress on microstructural development: the three-dimensional microstructure of a \(\gamma-\gamma'\) alloy

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Received 15 January 2002; received in revised form 25 February 2002; accepted 25 February 2002

Abstract

We investigate the three-dimensional structure of a model \(\gamma-\gamma'\) alloy using serial sectioning and digital reconstruction. The particles align in parallel two-dimensional sheets along the \(<100>\) crystallographic directions. The sheets in different regions of the microstructure are aligned along different \(<100>\) crystallographic directions. A variety of particle morphologies are observed. These morphologies are relatively independent of particle size, indicating a significant departure from single-particle equilibrium morphologies. The primary determinant of particle morphology appears to be the interparticle elastic interactions, both within sheets of particles and at the intersections of sheets. There is no evidence of particle coalescence despite the small interparticle separation. © 2002 Acta Materialia Inc. Published by Elsevier Science Ltd. All rights reserved.

Keywords: Phase transformations; Precipitation; Growth; Microstructure; Scanning electron microscopy (SEM); Image analysis

1. Introduction

Coarsening is the process by which a two-phase system reduces its total energy by decreasing its total interfacial area. In traditional coarsening theory the system is assumed to be stress-free, but this assumption is not valid in many two-phase mixtures, particularly in solid-solid systems where there is a coherent interface and misfit between the phases. This particle–matrix misfit induces elastic stress, which can have a significant effect on the coarsening behavior of a system, both on the coarsening kinetics and the microstructural evolution.

For example, stresses can give rise to changes in the shape of a particle with increasing particle size, inverse coarsening wherein a small particle can grow at the expense of less favorably located larger particles, and particle alignment [1–6].

To study the effects of elastic stress on microstructural evolution during coarsening we investigate the technically important \(\gamma-\gamma'\) system. This system consists of a FCC \(\gamma\)-matrix phase and an ordered \(L_1_2\), \(\gamma'\)-precipitate phase. The two phases are coherent but possess different lattice parameters, and the system is elastically anisotropic with elastically soft \(<100>\) crystallographic directions. As the average size of the \(\gamma'\)-particles increases during coarsening, their morphologies evolve from spheres, to cuboids, to non-equiixed rectangular parallelepipeds. At small sizes the iso-
tropic interfacial energy is the dominant energy and the particles are spherical. As they increase in size the elastic energy, that scales as the volume of the particle, increases and the particles assume more elastic energy minimizing shapes: cuboidal and parallelepiped shapes with faces parallel to the \( <100> \) crystallographic directions [1]. The particles also align with each other along the same \( <100> \) directions. This alignment is a result of the long-ranged elastic stress fields of the particles, which interact with one another and give rise to an interaction energy, or configurational force, that is attractive at long distances along the \( <100> \) directions and repulsive at short distances [7–9]. Unfortunately the particle shapes that are present in such systems and the nature of the particle alignment remain largely unexplored, as most experiments have examined the evolution of the microstructure using two-dimensional cross-sections of a three-dimensional microstructure [1,10–12]. Although some three-dimensional information can be extrapolated from such investigations [13], it is not possible to determine full three-dimensional information.

There have been some theoretical studies of the evolution of the three-dimensional microstructure of \( \gamma-\gamma' \) alloys that can serve as a guide for experiment. The equilibrium morphology of an isolated, misfitting precipitate has been examined by numerous researchers. Johnson et al. [14] assume the particle is an ellipsoid and determine the energy extrema of oblate, prolate, and spherical morphologies. They find that for the materials properties of Ni–Al \( \gamma-\gamma' \) alloys, spheres are favored at small particle sizes while either oblate or prolate shapes are energy minima at larger particle sizes. Mueller and Gross [15,16] apply a three-dimensional boundary element method to the problem, and thus do not constrain the particle morphologies to be ellipsoidal. They find that the equilibrium morphologies change from spherical to cuboidal as the size of the particle increases. Their calculations do not extend to non-cuboidal shapes, but they do report that at even larger particle sizes the cuboidal shape becomes unstable to a transition to oblate or prolate rectangular-parallelepiped shapes. Thompson and Voorhees [17] also observe a shape transition from spherical to cuboidal as the particle size increases. Their calculation does not extend into the range where the cuboidal shape becomes unstable.

Calculations have also been performed to examine the effects of elastic interactions between particles on particle morphology. Eckert et al. [18] expand the calculations discussed above [15,16] to simulate a periodic array of particles, and find that the particle volume fraction affects the curvature of the equilibrium particle shape. Several calculations have been performed on the evolution of three-dimensional arrays of interacting particles [19–23]. These calculations have been limited to either a relatively small number of particles, or to a larger number of particles with relatively few computational grid points per particle. A limited number of particles means large-scale alignment cannot be observed, while calculations using a small number of computational grid points per particle employ thick diffuse interfaces. However, given the increasing speed of computers it is likely that these limitations will be overcome. Thus, experimental results on the three-dimensional morphology of such alloys with misfitting particles will be particularly important as a comparison to these future three-dimensional calculations.

There have been a few experimental results on the microstructure of \( \gamma-\gamma' \) alloys in three dimensions. Small angle neutron or X-ray scattering have been performed [24,25], as well as deep etching of alloys [26]. However, small angle neutron or X-ray scattering require assumptions about the shape of the particles studied to extract specific information, while deep etching is limited to examining the morphology of partially exposed individual particles. Thus, these techniques are not able to extract full three-dimensional information on the microstructure.

A different method is needed to obtain such full three-dimensional information on the microstructure of \( \gamma-\gamma' \) alloys. It has been recognized for some time that the three-dimensional structure of materials can be determined through serial sectioning with subsequent reconstruction of the imaged sections into a three-dimensional volume [27,28]. However, until fairly recently it was not practical to perform a digital reconstruction of complex three-dimensional microstructures. As computing power has increased and algorithms for
reconstructing surfaces from volumetric data have improved, a number of such serial sectioning experiments have been performed. These have included three-dimensional reconstructions of the high volume fraction skeletal morphology in Pb–Sn [29], proeutectoid cementite precipitates in steel [30], particle reinforced metal matrix composites [31,32], and dendritic microstructures [33,34].

To our knowledge, no work has been published on the three-dimensional morphology or alignment of $\gamma$–$\gamma'$ alloys via serial sectioning. This technique allows full three-dimensional information on the $\gamma$–$\gamma'$ microstructure to be collected experimentally without a priori assumptions made with regard to the $\gamma'$-particle shapes or spatial distribution. In the following sections we present the procedures followed and results obtained from a serial sectioning study performed on a model $\gamma$–$\gamma'$ alloy. The results presented are a qualitative analysis of the particle morphology and spatial distribution; a future paper will present a quantitative analysis of these data.

2. Experimental procedure

The composition of the alloy investigated was Ni–24.0Co–5.0Cr–2.5Mo–4.0Al–4.0Ti by weight percent. The alloy was similar in composition to the alloys studied by Yoo et al. [26,35], with the Mo content adjusted to yield non-equiaxed rectangular-parallelepiped-shaped particles large enough to be studied by serial sectioning. The alloy was made by arc-melting a button from 99.99% Ni, 99.95% Co, 99.999% Cr, 99.98% Mo, 99.999% Al, and 99.995% Ti. The button was melted and flipped multiple times to ensure the best possible homogenization of the alloy. The alloy button was then solution treated at 1200°C for 135 h, and quenched into iced brine. Following the solution treatment, a portion of the alloy button was sectioned off and heat treated at 141.7 h at 1050°C to achieve the final microstructure investigated in this experiment. The heat treatment time and temperature were chosen so that distinctly non-spherical and non-cuboidal particle morphologies were present in the microstructure. At longer heat treatment times on the same alloy the particles were observed to be incoherent with the particle–matrix interfaces no longer parallel to the $<100>$ crystallographic directions. The $\gamma'$ volume fraction of the alloy following heat treatment was 0.272.

The heat-treated specimen was polished down to a 0.05 µm alumina suspension, and then marked with a series of Knoop hardness indents that were used both to realign one section with another and to measure the polishing depth. As serial sectioning proceeded these hardness indents needed to be periodically replaced as the cumulative polishing depth approached the depth of the indent. The serial sectioning consisted of repeatedly polishing, etching, and imaging the specimen surface. The polishing was performed using a 0.05 µm alumina suspension in an automatic polisher to ensure repeatability of the polish. The hardness indents used to measure the depth of the polish were protected from etching to ensure that etching effects did not confound the measurements. The polish was deeper than the depth of matrix material removed during the etching procedure, so measuring unetched hardness indents provided a reasonable measure of the polishing depth of the etched region. The polishing depth was 0.078 ± 0.018 µm per polish over 46 polishes. Very little deviation was observed in the normal to the sectioning plane; a total of approximately 0.005° over the course of the experiment.

After each polish, the area of the specimen to be imaged was electrolytically etched in a mixture of 17 parts deionized water, 1 part glacial acetic acid, and 2 parts nitric acid for 8 s at 2 V. This area was then imaged using a Hitachi S4500 scanning electron microscope (SEM). Since the sample had to be remounted in the SEM after each polish and etch, alignment of the sections with one another was necessary. This alignment was achieved using additional hardness indents placed within the field of view of the recorded area. The image was then cropped to the desired viewing area, and made binary.

The two-dimensional SEM images were combined into a volume measuring $11.33 \times 11.33 \times 3.6$ µm, with a resolution of 0.025 µm within the imaging plane, and a resolution of 0.078 µm along the polishing direction. It was not possible to reconstruct reasonably smooth $\gamma$–$\gamma'$ interfaces with such a large difference in resolution.
between the lateral and polishing directions, and thus the resolution in the lateral directions was reduced by a factor of two before rendering the interfaces. Due to the small \( \gamma' \)-interparticle separation distances, artificial bridging occasionally occurred between precipitates in the reconstructed images after this reduction in resolution. As this bridging is not present in the higher resolution lateral directions, and thus is a known artifact of the reconstruction process, such bridges are ignored in the analysis of the three-dimensional microstructure.

3. Results

A two-dimensional cross-section of the \( \gamma - \gamma' \) microstructure is shown in Fig. 1(a). If the serial sections are taken on \( \{100\} \) planar sections the particle cross-sections on consecutive sections are relatively constant and the end of the particle is abrupt, making it difficult to determine the particle dimensions in the sectioning direction. On serial non-\( \{100\} \) planar sections the particle cross-sections change gradually with each section, and the endpoint can be reasonably interpolated from this gradual evolution. Thus, the reconstruction plane was intentionally chosen not to be a \( \{100\} \) section of the microstructure.

For the digital reconstruction, each picture was converted into a binary image so that the matrix and particle phases could be easily differentiated. An example of this is shown in Fig. 1(b), which is the binary image obtained from Fig. 1(a). As mentioned in section 2, despite the small interparticle separation distances there are no connections between the particle cross-sections evident in Fig. 1. This is characteristic of all of the two-dimensional planar sections of the microstructure.

A portion of the three-dimensional volume obtained is shown in Fig. 2. The matrix has been made transparent, and the surfaces of the \( \gamma' \) particles are represented in metallic gray. As this is a two-dimensional projection of a three-dimensional volume, a traditional scale bar is not appropriate. Instead the volume has been framed in a box of known dimensions, which henceforth is referred to as the ‘box-frame’, and these dimensions are reported to give the approximate scale of the shown microstructure. In Fig. 2, the shortest dimension of the box-frame is parallel to the sectioning direction.

The degree of rounding of the particle edges seen in Fig. 2 is a result of the spatial resolution of the image. In this case, no radius of curvature less than or equal to 0.078 \( \mu \text{m} \) can be resolved consistently. The arrows in Fig. 2 point to examples of particles that have been truncated by the termination of the displayed experimental volume. Thus, the displayed morphologies of these particles are not representative of their full three-dimensional shape. Some of the characteristics of the three-dimensional \( \gamma - \gamma' \) structure can begin to be seen in Fig. 2. There are a variety of particle morphologies, and the morphologies are affected by the interparticle elastic interactions. The particles align with each other in three-dimensions along the three \(<100>\) crystallographic directions. The nature of this alignment would not be obvious from two-dimensional sections.

3.1. Spatial distribution

The truncated \( \gamma' \)-precipitates on the top and bottom of the microstructural volume shown in Fig. 2 make it difficult to discern the spatial alignment of the particles. Thus these particles have been removed in the portion of the microstructural volume shown in Fig. 3. The microstructural volume shown in Fig. 3(a) has been separated into three regions: A, B, and C. In all three regions particles...
align along the $<100>$ crystallographic directions. In region B in particular, most of this alignment occurs along two of the $<100>$ directions while there is very little alignment along the third $<100>$ direction.

In Fig. 3(b) the microstructure shown in Fig. 3(a) has been rotated, and the same regions A, B, and C are again labelled. From this viewing angle it becomes obvious that the particles in region B are aligned in two-dimensional sheets which are perpendicular to the $<100>$ direction along which no alignment appears to be occurring in Fig. 3(a). The separation distances between the sheets are far larger than the interparticle separation distances within the sheets, and appear to be relatively constant. In Fig. 3(c) the microstructure has again been rotated, this time so that the sheets in region B are approximately parallel to the viewing plane, and the regions A, B, and C have again been labelled. From this viewing angle it can be seen that the alignment of the particles into two-dimensional sheets observed in region B is also occurring in regions A and C, but that the sheets in regions A and C are perpendicular to a different $<100>$ direction than those in region B.

To more easily investigate the nature of the interparticle alignment within a two-dimensional sheet, a set of perpendicular two-dimensional sheets has been isolated in Fig. 4. Within this set there is a single large sheet labeled L and two smaller sheets, S1 and S2, which are parallel to each other and perpendicular to L. From the viewing angle shown in Fig. 4(a), it can be seen that within the two-dimensional sheets the particles tend to align themselves in one-dimensional strings. This results in open channels through the sheets with no particles; several of these channels in sheet L are noted by arrows.
In Fig. 4(b) the sheets of Fig. 4(a) have been rotated so that sheet L is approximately parallel to the viewing plane. From this viewing angle it can be seen that in addition to the particle alignment parallel to the channels highlighted in Fig. 4(a), there is additional alignment of the particles perpendicular to the channels already discussed. In both \( <100> \) directions within sheet L, it appears that the particles within L are aligning so that the corners of neighboring particles align with each other. The longer, thinner arrows in Fig. 4(b) point to examples of the artificial bridging between particles discussed in section 2 above; this is an artifact of the reconstruction procedure and does not represent true connectivity between the particles.

In Fig. 4(c) the sheets have been rotated again so that sheet L is now approximately perpendicular to the viewing plane and sheets S1 and S2 are parallel to it (with S2 obscured by S1). The characteristics of the intrasheet particle alignment in S1 are similar to what was observed in the sheet L, with the corners of the particles tending to line-up and particle-free channels running through the sheet. In addition, from this viewing angle it becomes apparent that the particles within sheet L all have approximately the same dimension along the direc-
Fig. 4. Two perpendicular two-dimensional sheets of \( \gamma' \) precipitates in a \( 7.5 \times 8.75 \times 3.6 \mu m \) box-frame (a) at an arbitrary orientation, (b) parallel to sheet L, and (c) parallel to sheets S1 and S2. The bridging between particles is an artifact of the reconstruction procedure.

Fig. 5. Two sections of the volume of Fig. 4 separated by 0.40 \( \mu m \).

There are a wide variety of particle morphologies present in the experimental volume. To facilitate discussion we use the following conventions: The aspect ratio of a particle is defined as \( a : b : c \), where \( a \) is the longest edge-length and \( c \) is the shortest edge-length. Particles with \( a \approx b \approx c \) are referred to as cuboidal, with \( a \approx b \gg c \) as plate-like, and with \( a \gg b \approx c \) as rod-like. All
other shapes are simply referred to as parallelepipeds.

As discussed above, the high curvature at the particle edges is below the resolution of the reconstruction technique. However, the overall particle morphologies are well within the resolution and can be meaningfully investigated. Shown in Fig. 6 are γ′ particles which have been isolated from the rest of the microstructure. The first column in Fig. 6 shows the γ′ particles at an arbitrary viewing angle, while the second and third columns show the same particles rotated so that the viewing planes are approximately parallel to the largest and smallest cross-sections of the particles respectively. From these views it can be seen that particle (d) is nearly cuboidal, particle (i) is plate-like, particles (a), (f), and (h) are rod-like, and the rest of the particles are simply rectangular parallelepipeds.

From the isolated particles shown in Fig. 6, it appears that size is neither the sole nor primary determinant of the particle morphology in this microstructure. To test this qualitative observation we compare the morphologies of particles (b), (d), and (j): the aspect ratio of particle (b) is approximately 2.4 : 1.5 : 1, of particle (d) approximately 1.2 : 1.2 : 1, and of particle (j) approximately 3.0 : 2.3 : 1. The volumes of particles (b) and (d) are within 15% of each other, and are approximately 6.5 times the volume of particle (j). The aspect ratios of (b) and (j) are similar, with one dimension far smaller than the other two dimensions, despite the wide disparity in volume. The aspect ratios of (b) and (d) are far different, with (b) being nearly cuboidal, despite being very similar in volume. It is clear from this comparison that the particle morphology is not solely a function of the particle size.

The particle morphologies must be considered in the context of their surroundings. In the previous section it was observed that alignment of the corners of neighboring particles appears to be energetically favorable, both within the sheets and at the sheet intersections. Also, the approximately uniform particle dimension along the thickness of a sheet seen in Fig. 4 must have a significant effect on the distribution of particle morphologies within a sheet. Thus, all three-dimensions of a particle within a sheet are constrained: its <001> dimensions within the sheet by its neighbors, and its <100> dimension perpendicular to the sheet by the thickness of the sheet.

An example of neighboring particles within a sheet is shown in Fig. 7, where a string of four particles has been isolated from within a sheet. The particles on the ends of the string are volumetrically far larger than the than the particles in between. However, the two larger particles are

Fig. 6. A selection of the γ′ particle morphologies present in the reconstructed three-dimensional volume. All the particles shown are scaled to the 2.45 × 2.45 × 1.58 μm box-frame around the first view of particle (a).
nearly cuboidal while the two smaller particles both have one dimension which is far smaller than the two other dimensions. The particle morphologies in this case clearly result from the volumetrically smaller and larger particles altering their morphologies so that their corners line up. This results in a procession of approximately matching parallel faces, and is driven by the interparticle elastic interaction energy rather than by the elastic self-energy of the particles.

The interactions between perpendicularly oriented sheets can also have a significant effect on the particle morphologies along the intersection of the sheets. This is demonstrated by the images shown in Fig. 8, which are higher magnification images of the intersections of the sheets shown in Fig. 4. Three of the particles which are located at this intersection have been highlighted with arrows and labelled 1, 2, and 3. The sheets have been labelled L, S1, and S2 as they were in Fig. 4 above.

Particle 1 is located at the intersection of sheets L and S2. In Fig. 8(a) the location of the top of particle 1 is determined by its neighbor in L, but the location of the bottom of particle 1 falls between the bottoms of its neighbors in L and S2. In Fig. 8(b) the view has been rotated so that the other dimensions of particle 1 are seen. In this case, both the top and bottom of particle 1 are located between the tops and bottoms, respectively, of its neighbors in L and S2. Thus, in both Fig. 8(a) and Fig. 8(b) the morphology of particle 1 is altered to provide a transition between sheets L and S2.

Particles 2 and 3 provide a similar transition, in this case between sheets L and S1. In Fig. 8(a), particle 2 is partially obscured by another particle, but both the top and bottom are fully visible. In this view, the bottom of particle 3 is parallel to the bottom of particle 2, while the top of particle 3 is located between the tops of its neighbor in L and particle 2. In addition, in this same view the top of particle 2 is sloping so that it comes close to lining up both with the top of the particle to its right in S1 and the top of particle 3. In this instance both particles 2 and 3 are providing a transition between L and S1, and the interparticle elastic interactions at the intersection of the sheets have caused particle 2 to no longer be a rectangular parallelepiped. Thus, the interparticle elastic energy is again acting as a stronger determinant of the particle morphology than the elastic self-energy.
4. Discussion

4.1. Particle alignment

We note above that all of the spatial alignment observed in three-dimensions involves parallel, matching particle faces aligned along the $<100>$ crystallographic directions wherein the corners of neighboring particles are aligned. This is a natural extension of the results of modeling in two-dimensions. Su and Voorhees [2,3] show that through translation or inverse coarsening, groups of two to three particles will evolve during coarsening to achieve a matching of the $\{10\}$ faces. Wang and Khachaturyan [36] have observed a similar phenomenon in their two-particle calculations. The tendency for corners to align follows from the configurational forces generated by misfitting particles in an elastically anisotropic system [2,3]. The configurational forces are attractive near the corners of a particle and repulsive near the center of the particle faces. Thus the corners of two neighboring particles attract one another, giving rise to the alignment observed in the experiments. Due to the repulsive force at the center of the particle faces, the attractive forces at the corners do not necessarily give rise to coalescence.

It is well known that misfitting particles align along $<100>$ directions because of a negative elastic interaction energy, while alignment along the $<110>$ or $<111>$ directions is not favored due to a positive elastic interaction energy [7,37]. Johnson and Voorhees [8] have examined the elastic energy of various arrangements of cuboidal particles in an elastically isotropic system where the particles have a smaller shear modulus than the matrix. They find that an alignment of five particles in a one-dimensional string along a single $[100]$ direction is more energetically favorable than an alignment in a cross, with one particle at the center of the cross and the remaining particles aligned one each along its $[001],[00\bar{1}],[100]$, and $[\bar{1}00]$ direc-

Fig. 8. Close-up views of the region where the sheets from Fig. 4 intercept. The sheets L, S1, and S2 are defined the same as in Fig. 4. The box-frame is $6.25 \times 3.75 \times 3.63 \ \mu m$. The bridging between particles is an artifact of the reconstruction procedure.
Consider cuboids arranged in evenly-spaced one, two, and three-dimensional arrays. For the sake of simplicity we only consider the nearest-neighbor interactions along each of the $\langle100\rangle$, $\langle110\rangle$, and $\langle111\rangle$ directions. If the nearest-neighbor spacing along the $\langle100\rangle$ is defined as $d_t$ then the nearest-neighbor spacings along the $\langle110\rangle$ and $\langle111\rangle$ directions will be $\sqrt{2}d_t$ and $\sqrt{3}d_t$ respectively. In an evenly spaced one-dimensional array, i.e. a string, each cuboid in the string has two $\langle100\rangle$ interactions with other cuboids and no $\langle110\rangle$ or $\langle111\rangle$ interactions. In an evenly spaced two-dimensional array, i.e. a sheet, each cuboid in the sheet has four $\langle100\rangle$ interactions with other cuboids, four $\langle110\rangle$ interactions, and no $\langle111\rangle$ interactions. In an evenly spaced three-dimensional array, i.e. a cluster, each cuboid in the sheet has six $\langle100\rangle$ interactions with other cuboids, twelve $\langle110\rangle$ interactions, and eight $\langle111\rangle$ interactions.

If the negative elastic interaction energy along the $\langle100\rangle$ directions and the positive elastic interaction energy along the $\langle110\rangle$ directions are of approximately the same magnitude, then the most favorable three-dimensional spatial alignment for misfitting cuboidal particles is an isolated one-dimensional string. This is the case for the isotropic system examined by Voorhees and Johnson [8], and explains why a string of five particles has a lower energy than a cross of five particles. In contrast, in more anisotropic materials such as Ni the magnitude of the negative elastic interaction energy along the $\langle100\rangle$ directions is larger than the magnitude of the positive elastic interaction energy along the $\langle110\rangle$ directions. When considering only the nearest neighbors as in the previous paragraph, the sheet alignment will become more energetically favorable than the string alignment when the magnitude of the negative elastic interaction energy along the $\langle100\rangle$ directions is more than twice that of the positive elastic interaction energy along the $\langle110\rangle$ directions. According to the calculations of Johnson and Lee [7] for spherical misfitting particles, this condition may be satisfied for cuboidal particles in the Ni-Al $\gamma-\gamma'$ system. Due to the large number of $\langle110\rangle$ and $\langle111\rangle$ interparticle interactions associated with the cluster alignment, it is unlikely that the elastic anisotropy would ever be sufficient to make it a lower energy configuration than the isolated string or sheet alignments for $\gamma-\gamma'$ alloys.

The spatial arrangements that are accessible to $\gamma'$ particles are also a function of the volume fraction of $\gamma'$ in the system. At low volume fractions isolated strings, isolated sheets, and clusters are all accessible arrangements. At intermediate volume fractions it would no longer be possible for strings to be sufficiently isolated to avoid interstring interactions, so only the isolated sheets or cluster arrangements are accessible. Similarly, at high volume fractions isolated sheets are no longer possible, and a cluster arrangement is the only accessible arrangement of the three possibilities. The definitions of low, intermediate, and high volume fractions will vary from alloy to alloy depending on the strength and range of the interparticle elastic interactions which, in turn, are primarily dependent on the degree of misfit, elastic anisotropy, and elastic inhomogeneity [7,8,25].

Taking into account both accessibility and energetic favorability, the spatial arrangements of $\gamma'$ particles can be predicted based on volume fraction. At low volume fractions the particles will align in isolated strings or sheets depending on the degree of elastic anisotropy. At intermediate volume fractions the particles will align in isolated sheets, and at high volume fractions the particles will align in a nearly continuous three-dimensional cluster. The spatial arrangement consisting of isolated sheets observed in this experiment at an intermediate volume fraction of 0.272 agrees well with this analysis.

The three-dimensional arrangements discussed above would lead to characteristic microstructures viewed on $\{100\}$ cross-sections. When sectioning a three-dimensional arrangement consisting of isolated strings of particles, one would expect to see isolated particle cross-sections that result from sectioning perpendicular to a string and isolated lines of particle cross-sections which result from sectioning parallel to a string. When sectioning a three-dimensional arrangement consisting of isolated sheets, one would expect to see isolated lines
of particle cross-sections that result from sectioning perpendicular to a sheet, and arrays of particle cross-sections of limited extent that result from sectioning parallel to a sheet. Finally, when sectioning a three-dimensional arrangement consisting of clusters, one would expect to see arrays of particle cross-sections, likely also of limited extent due to imperfections in the alignment. Thus, while the actual particle arrangement can only be determined through three-dimensional visualization of the microstructure, some comparisons can still be made based on the arrangement of particle cross-sections seen on two-dimensional sections.

In the work of Yoo et al. [26] on an alloy similar to the one studied in this experiment, the cross-sectional microstructure predicted for a particle arrangement consisting of isolated strings is observed on {100} sections of their low volume fraction alloys at long coarsening times. For their intermediate volume fraction alloys at long coarsening times, the {100} cross-sectional microstructures agree with the predictions for a particle arrangement consisting of isolated sheets. Yoo et al. did not produce any high volume fraction alloys in their experiment that can be compared to these predictions. MacKay and Nathal [38] did study such high volume fractions in Ni–Mo–Al–Ta alloys, and observe the {100} cross-sectional microstructures agree with the predictions for a particle arrangement consisting of isolated sheets. Yoo et al. did not produce any high volume fraction alloys in their experiment that can be compared to these predictions. MacKay and Nathal [38] did study such high volume fractions in Ni–Mo–Al–Ta alloys, and observe the {100} cross-sectional microstructures agree with the predictions for a particle arrangement consisting of isolated sheets. Yoo et al. did not produce any high volume fraction alloys in their experiment that can be compared to these predictions. MacKay and Nathal [38] did study such high volume fractions in Ni–Mo–Al–Ta alloys, and observe the {100} cross-sectional microstructures agree with the predictions for a particle arrangement consisting of isolated sheets.

As seen in Figs. 2–8, the γ’-particle morphology is not always cuboidal as assumed in the above analysis. Johnson and Voorhees [8] include a limited investigation of the energetics of rod or plate alignment in their work, and their results indicate that there are additional constraints on the favored alignments between particles of such morphologies. However, as the favored alignments for these morphologies are still along the <100> directions, the presence of non-cuboidal morphologies would most likely affect the alignment within a given string, sheet, or three-dimensional cluster of particles rather than affect the relatively favorability of the isolated string, sheet, or cluster arrangements themselves.

It was noted above that the sheets in region B of Fig. 3 appeared to have a relatively constant intersheet separation distance. This suggests that the sheets as a whole interact with each other to reach an equilibrium separation distance, while the elastic interactions between individual particles are much shorter range and thus do not set the intersheet spacing. As such, one may a view sheet of particles as single large plate-like particle, and the separation distance between the sheets can be expected to be similar to that predicted for large plate-like particles of the same size. Khachaturyan et al.[39] and Miyazaki et al. [40] calculate equilibrium separation distances between neighboring individual γ’ plates, and using similar calculations the lateral dimensions of sheets of particles could likely be estimated based on a measurement of the intersheet separation distances, or vice versa. These equally spaced parallel sheets are also similar to the “one-dimensional modulated structure” of parallel plates of infinite extent that Khachaturyan [37] predicts will be a stable structure for elastically anisotropic systems with misfitting precipitates.

4.2. Particle morphology

The equilibrium shapes of isolated, elastically stressed precipitates have been determined in both two and three dimensions [14–17,39,41–43]. Although the details of the methods used to determine particle morphology vary, all predict a dependence of the particle morphology on the particle size. Furthermore, the progression of shapes as particle size increases is predicted to be spherical, to cuboidal, to non-equiaxed rectangular parallelepiped, or the two-dimensional equivalents of those shapes. In Fig. 6 above we show a variety of particle morphologies observed in the reconstructed volume, and it is clear that the morphology does not have a particularly strong dependence on the particle size. This lack of a size dependence in the particle morphology is at odds with the predictions of the single particle equilibrium calculations, but the single particle equilibrium calculations do not account for the interparticle elastic interaction energies present in the system.

It is clear from the literature that interparticle elastic interactions can have a significant impact on the particle morphology [2,3,6,36,44–47]. In
particular, the recent large-scale calculations by Thornton et al. [48] show that interparticle elastic interactions in systems with large numbers of particles can alter significantly the particle morphologies from those that are predicted by single-particle equilibrium calculations. Thus, the particle morphologies observed within a $\gamma - \gamma'$ microstructure should not be expected to be the same as the equilibrium shapes predicted for isolated particles. This explains the lack of a clear dependence of the particle morphology on particle size seen in this experiment, and also agrees with the observed tendency for particle corners to align.

When coalescence of $\gamma'$ particles occurs in two-dimensional calculations [6,36] or is observed experimentally [49], the cross-sections of the coalesced particles have more than four sides. In three-dimensional calculations an analogous result is observed when particles coalesce: the resulting particles are parallelepipeds with more than six sides. In the three-dimensional calculations of Vaithyanathan and Chen [21] coalescence is widespread and many parallelepipeds with more than six sides can be observed, including unusual morphologies which appear to be the result of plates oriented along different $<100>$ directions coalescing. In three-dimensional calculations by Rubin and Khachaturyan [22] particles with more than six sides resulting from coalescence are also observed, although not to the extent seen in the calculations of Vaithyanathan and Chen. In both these calculations the anti-phase domain boundary energy is far higher than the $\gamma - \gamma'$ interfacial energy, and thus prevents the coalescence of neighboring particles with different order parameters.

We see exclusively six-sided parallelepipeds in our microstructure, and thus it can be reasonably concluded that no coalescence is occurring. Moreover, there is no coalescence despite the small interparticle separations within a sheet. The results of Vaithyanathan and Chen [21], Rubin and Khachaturyan [22], and Wang and Khachaturyan [36] emphasize the importance of anti-phase domain boundaries in preventing coalescence. Since it is unlikely that all neighboring particles in a sheet have different order parameters, the lack of coalescence that we observe is likely the result of the repulsive configurational forces acting on the centers of particle faces as discussed above.

The details of the spatial alignment and particle morphologies observed in the experimental microstructure above may not translate to other $\gamma - \gamma'$ alloys. It is well known that factors such as the degree of elastic inhomogeneity and elastic anisotropy, the magnitude of the particle-matrix misfit, and the volume fraction of coarsening phase can all, to some degree, effect particle alignment and morphology [18,19,25,26,50,51]. However, our observation that the elastic interaction energy can be as important as or more important than the elastic self-energy in determining the particle morphology certainly translates into all $\gamma - \gamma'$ alloys. Similarly, the observations on the qualitative character of the large-scale alignment of the microstructure, which is dependent only on the elastic interaction energy, can also be expected to apply to other $\gamma - \gamma'$ alloys.

5. Conclusions

We investigated experimentally the three-dimensional microstructure of a model $\gamma - \gamma'$ alloy using serial sectioning. This investigation led to the following conclusions:

- The corners of neighboring particles tend to align, resulting in neighboring parallel $\{100\}$ faces with aligned edges.
- The particles align with each other in parallel, two-dimensional sheets. In different regions of the microstructure the two-dimensional sheets of particles are aligned along different $<100>$ crystallographic directions, giving rise to unusual particle morphologies at their intersections.
- The particles within a sheet tend to share the same dimension perpendicular to the sheet, and the intersheet separation distance is approximately constant.
- Interparticle elastic interactions within sheets and at the intersections of differently oriented sheets appear to be the strongest determinants of particle morphology, and thus the particle
morphology does not depend strongly on the particle size.

- There is no microstructural evidence for coalescence, despite the small interparticle separation distances within a sheet of particles.

Acknowledgements

This work was supported through NSF grant number DMR-9707073. We thank J. Alkemper and K. Thornton for assisting with the three-dimensional rendering of the microstructures.

References