

Rotation-limited growth of three-dimensional body-centered-cubic crystals

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According to classical grain growth laws, grain growth is driven by the minimization of surface energy and will continue until a single grain prevails. These laws do not take into account the lattice anisotropy and the details of the microscopic rearrangement of mass between grains. Here we consider coarsening of body-centered-cubic polycrystalline materials in three dimensions using the phase field crystal model. We observe, as a function of the quenching depth, a crossover between a state where grain rotation halts and the growth stagnates and a state where grains coarsen rapidly by coalescence through rotation and alignment of the lattices of neighboring grains. We show that the grain rotation per volume change of a grain follows a power law with an exponent of -1.25 . The scaling exponent is consistent with theoretical considerations based on the conservation of dislocations.

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I. INTRODUCTION

In polycrystalline materials, the microstructure, given by the grain sizes, shapes, and coordination, is an important control parameter for macroscopic material properties such as the yield stress, conductivity, and brittleness. Polycrystalline materials typically form from the nucleation and growth of grains with different lattice orientations in a quenched or annealed melt. If the annealing temperature is sufficiently high, the grain boundaries will be mobile and rearrange in a way that leads to an overall coarsening of the polycrystalline matrix with time.

In classical models of grain growth in two dimensions, such as the von Neumann–Mullins model, grain boundary migration is driven by surface energy and the average grain area grows linearly with time [1] $\langle A \rangle \sim t$. In three dimensions, however, a correspondingly simple universal growth law cannot be established [2,3] because of the increased geometrical complexity. For models whose dynamics is driven by the minimization of a free energy constrained with a continuity equation, general arguments relying on energy dissipation [4] set the upper bound for the coarsening dynamics in D dimensions to $\langle L^D \rangle \sim t^{D/2}$, which coincides with the von Neumann–Mullins law in two dimensions. Experimentally it has been observed that the grain size growth can be described by a power law with an exponent that depends on a variety of experimental factors such as the annealing temperature [5]. It has further been found that the grain growth is sublinear, i.e., the growth in area is described by a power-law exponent less than unity [6]. Another possible mechanism, which results in grain coarsening, is the coalescence of neighboring grains. The rotation of crystals might lead first to the lattice alignment of neighboring grains and second to the elimination of the grain boundaries between them. Highly convoluted grains can be formed in this way. Grain rotation has been observed in experiments [7] and has recently also been observed in molecular-dynamics simulations [8].

In many materials, the coarsening dynamics further differs from normal grain growth, in the sense that the grain size distribution varies in time. These abnormal grain growth systems are characterized by a minority of abnormal grains growing faster than the mean size leading to an inhomogeneous size distribution [9]. In contrast to classical grain growth laws,

grain growth is also known to stagnate in time [10]. In two-dimensional systems, it has been suggested that the stagnation might be a combination of high kinetic barriers, preventing mass migration across grain boundaries, and a locking of individual grains preventing grain rotation and subsequent lattice alignment [11]. In this article we present a numerical study of the influence of grain rotation on the coarsening dynamics in three-dimensional polycrystalline systems with a body-centered-cubic (bcc) symmetry.

II. MODEL AND ANALYSIS

To study the coarsening dynamics, we use the phase field crystal (PFC) model. The PFC model describes the evolution of a continuous order parameter field, which is spatially periodic with atomistic resolution. The model is based on the minimization of a phenomenological Swift-Hohenberg free-energy functional given by

$$\mathcal{F} = \int d\mathbf{r} \left(\frac{1}{2} \psi (1 + \nabla^2)^2 \psi + \frac{a_2}{2} \psi^2 + \frac{\psi^4}{4} \right), \quad (1)$$

with the order parameter ψ representing the crystal density field [12]. The parameter a_2 and the mean density $\bar{\psi}$ are related to the melting temperature according to the phase diagram in Fig. 1. The points in Fig. 1 show the parameters used in the simulations presented in this article. While we change both the mean density and the parameter a_2 in our simulations presented here, we shall for convenience only refer to the a_2 value in the figures and in the text. The corresponding values for the mean density can then be found from the phase diagram. Alternatively, the functional form of Eq. (1) can be derived from classical density functional theory [13]. In three dimensions a free energy on this form results in a rich phase diagram with multiple equilibrium phases. Here we shall focus only on the bcc phase. The evolution equation of the density field ψ is assumed to obey an overdamped diffusion equation

$$\frac{d\psi}{dt} = \nabla^2 \frac{\delta \mathcal{F}[\psi]}{\delta \psi}, \quad (2)$$

thereby establishing a link between the microscopic length scales and diffusive time scales. The PFC model has been used to study a wide range of phenomena including phase transitions [13] and plastic deformation [14] and has been

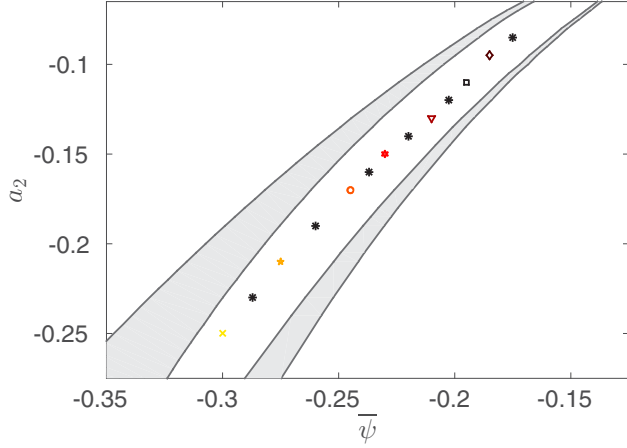


FIG. 1. (Color online) The PFC phase diagram with points indicating the parameters used in the simulations. The region in the middle has a bcc lattice as its equilibrium state, while the equilibrium states of the regions to the left and right are given by a uniform liquid state and a rod state, respectively. The gray areas indicate the coexistence regions.

shown to successfully predict grain boundary energy as a function of misorientation [12]. We solve the dynamical equation using an exponential time integration scheme [15] in a three-dimensional box of size $L \times L \times L$ with $L = 512dx$ or $1024dx$. Time and space are discretized with $dx = \frac{\pi}{3}$ and $dt = 1/2$. We initialize our system from an undercooled melt by introducing small crystal seeds of uniform size at random points and with random lattice orientations. For $L = 512dx$ the system was seeded with 120–150 seeds, while for the larger $L = 1024dx$ the system was initialized with 1200 seeds. After the initial crystallization phase, where the seeds grow to cover the whole melt, a polycrystalline structure is formed that coarsens over time. During the coarsening stage, we track the volume and lattice orientation of all grains.

For the segmentation of the grains, the peaks of the density field are located by a thresholding procedure from which the coordinates of the center of mass of each peak can be calculated. Grain boundary detection is performed using a Voronoi tessellation and a centrosymmetry parameter [16]. The angle and axis of rotation for each grain is found by taking the mean over the orientation of all the unit cells in the interior of the grain. The misorientation angle of two grains A and B is calculated by constructing the rotation matrix G for the grains and form the product $\Delta G_{AB} = G_A G_B^{-1}$. The misorientation angle θ is then given by $\theta = \arccos\left[\frac{\text{Tr}(\Delta G_{AB}) - 1}{2}\right]$.

To get a measure of the typical grain size in our systems, we calculate a coarsening parameter by fitting a Lorentzian squared to the structure factor of the system [17] $\langle |F[\psi]|^2 \rangle$, where $F[\psi]$ is the Fourier transform and the angular brackets denote averaging over all orientations in k space. In the averaged spectrum, the width $w(t)$ around the typical lattice scale provides a measure of the ordering wave number for the system, which is inversely proportional to the mean linear grain size. A small width implies fewer larger grains, while a large width implies a larger number of smaller grains. Defining $\xi(t) = w(t)^{-3}$, we have $\langle V(t) \rangle \propto \xi(t)$.

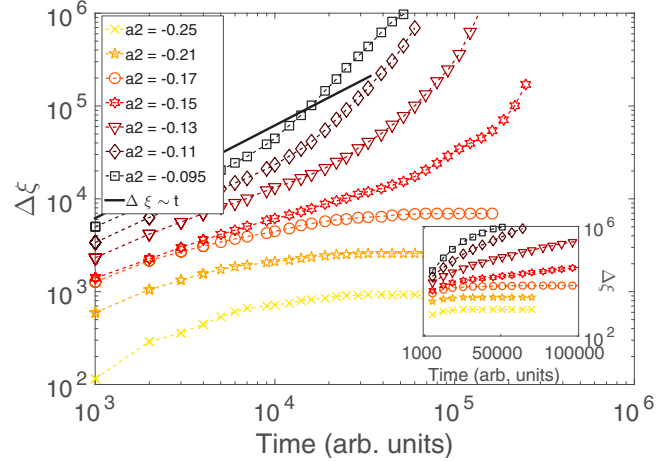


FIG. 2. (Color online) Mean size of grains on double logarithmic axes. The inset shows the same data on semilogarithmic axes.

III. RESULTS

In general, the polycrystalline structure coarsens at a rate that depends on the quenching depth, the a_2 parameter in Eq. (1). In Fig. 2 we show the change in the ordering length $\Delta\xi = \xi(t) - \xi_i$ as a function of time for different quenching depths, where ξ_i is the initial ordering scale measured right after the system has fully crystallized. For deep quenching parameters, the system dynamics is described by a short period of grain growth followed by stagnation. As the quenching depth is decreased, the time it takes to reach the stagnation stage is increased and eventually (for $a_2 \gtrsim -0.15$ and $\bar{\psi} \gtrsim -0.22$ in the phase diagram, Fig. 1) the stagnation stage is replaced by a stage of rapid grain growth where the ordering scale has an exponential growth. In this regime, the abnormal grain is large enough to coalesce with several smaller grains simultaneously. By tracking the orientation of individual grains, we observe that the stagnation is accompanied by a general decrease in the change of the lattice orientation. The change in orientation eventually drops to zero when the system stagnates (see Fig. 3). In contrast, the rapid coarsening regime shows little to no decay in the rotation of the individual grains.

Independent of the system size, the average grain size, here represented by $\Delta\xi$, can in the early state be approximated by a power law in time $\langle \Delta\xi \rangle \sim t^n$ with an exponent n that depends on the quenching parameters a_2 and $\bar{\psi}$. From simulations we find values of the exponent in the interval $n \sim 0.5-1$. In order to estimate the variation of the coarsening exponent and the transition point to exponential coarsening we have performed repeated simulations for the same quenching depths and different initial conditions. We find a standard deviation in the power-law exponents of the order of 0.1, while the location of the transition point has a standard deviation of 0.35 decade.

In general, the coarsening dynamics exhibits abnormal grain growth where a few grains grow significantly faster than the rest of the crystal matrix, as can be seen in Fig. 4. The figure shows a late stage snapshot of a $L = 1024dx$ system. The faster growth of a few grains compared to the rest of the matrix leads to a grain volume distribution that is not self-similar. For self-similar growth the standard deviation on

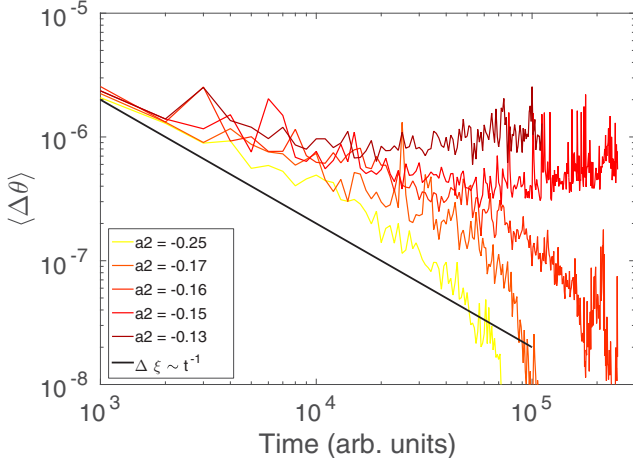


FIG. 3. (Color online) Average rotational velocity for individual grains calculated using a time window of 1000 time units. The straight line corresponds to a power law with an exponent $n = -1$. The stagnating regime shows a rapid decrease in grain rotation in contrast to the fast coarsening regime where grain rotation continues.

the grain volumes increases proportionally to the mean grain volume. In Fig. 5 the ratio of the standard deviation of the grain volumes and the mean grain volume is plotted, which shows that $\delta\langle V \rangle / \langle V \rangle$ is not a constant indicating abnormal growth.

The transition to fast grain growth is initiated by the mobilization of small grains, which align in lattice orientation with the larger so-called abnormal grains and finally coalesce into an even larger grain. This is supported by the observation that the amount of rotation per change in volume V increases as the small grains get even smaller. In order to quantify this effect we calculated the orientational velocity $\Delta\theta$ using the

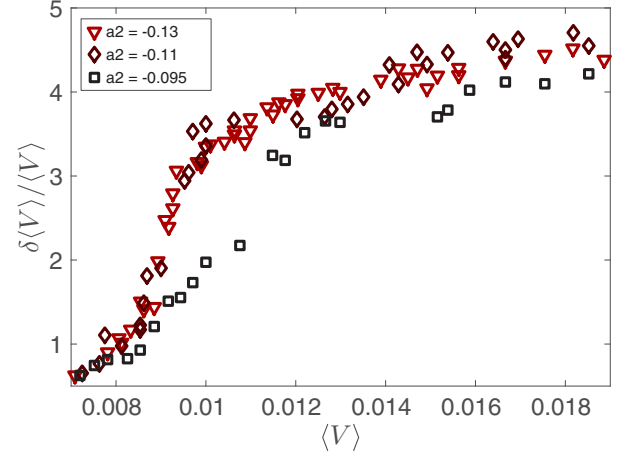


FIG. 5. (Color online) Ratio of the standard deviation in grain size and the mean size as a function of mean size.

misorientation angle and the volume change ΔV of individual grains for $\Delta t = 1000$ time units. Using the mean volume of the grain between two time points, we created scatter plots of $|\Delta\theta/\Delta V|$ versus V . The best fits across multiple quenching depths and initial conditions suggest a scaling behavior

$$\left| \frac{\Delta\theta}{\Delta V} \right| \sim V^{-\beta}, \quad (3)$$

where the scaling exponent is estimated to $\beta = 1.25 \pm 0.06$. The fitted exponent as a function of a_2 is shown in Fig. 6. The uncertainty is taken to be the standard deviation found from the weighted least-squares fit.

IV. DISCUSSION

In our simulations, we observe two distinct types of coarsening dynamics. For deep quenches the grain growth is typically accompanied by a decreasing rotation of the individual grains. When the grain rotation stops, the system

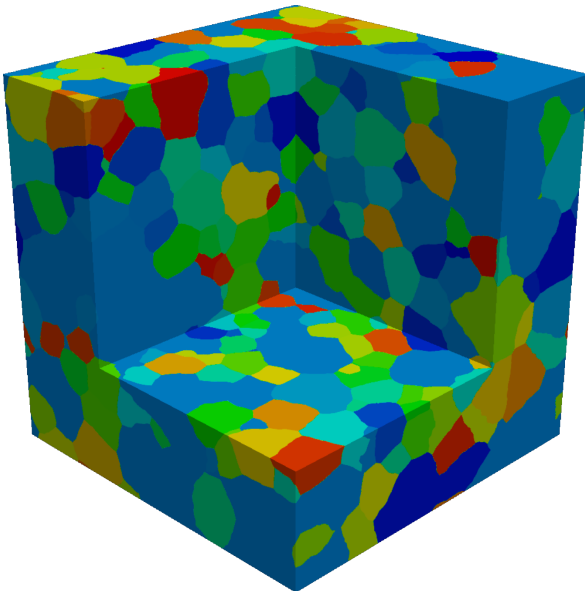


FIG. 4. (Color online) Snapshot of an $L = 1024dx$ simulation performed with $a_2 = -0.15$ and with periodic boundary conditions. Note the heterogeneous grain distribution that can be observed at the transition to abnormal grain growth.

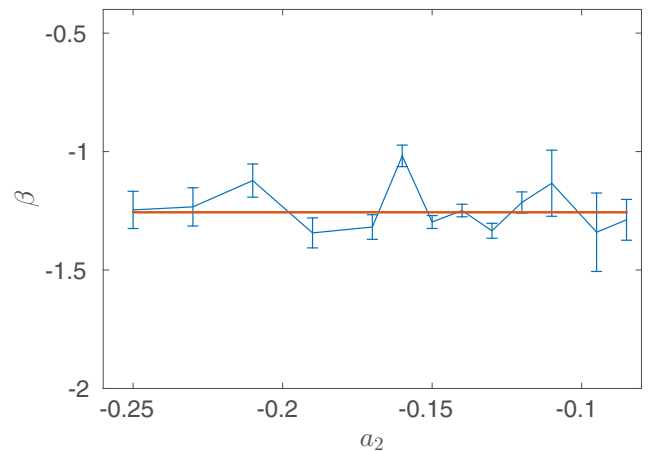


FIG. 6. (Color online) Power-law coefficients obtained from best fits to scatter plots of $|\Delta\theta/\Delta V|$ vs V for different values of a_2 . The uncertainty is the standard deviation calculated using a bootstrap method.

has reached a stagnation state. Grain growth stagnation has been reported in two-dimensional PFC studies [11,18] as well as in three-dimensional molecular-dynamics simulations [19]. For shallow quenches, we find that the early stage coarsening is described by a power-law growth of the form $V \propto \Delta\xi \sim t^n$ with $n \sim 0.5-1$, in reasonably good agreement with experimental studies [6], which finds $n \sim 0.4-1.2$, as well as with multiple two-dimensional PFC studies [11,18,20], which find exponents in the range $n \sim 0.2-0.8$. In contrast to classical grain growth models, the atomistic nature of the density field allows for bulk dissipation due to elastic relaxation of the lattice. Additionally it was found in [20] that a time-varying grain rotation introduces another mechanism for bulk dissipation suppressing large-scale rotation as well as slowing down grain coarsening. In the present study we find a large degree of rotation of grains on all scales as well as fairly rapid coarsening dynamics, implying that the anomalous dissipation mechanism is not too severe for the grain sizes investigated here.

Further, the grain growth is found to be abnormal, resulting in a crystal matrix consisting of a few large convoluted grains. Abnormal grain growth has been seen in other numerical experiments [21] as well as in experiments [22]. To analyze the late stage coarsening dynamics, we consider a simple model where the grain growth is mediated purely by grain coalescence. The grain coalescence follows from the continuous rotation of predominantly small grains, which tend to align their lattice with larger neighboring grains. Following [23], we introduce a characteristic time t_l it takes for two grains to coalesce. The dynamics of the number of grains can then be described by

$$\frac{1}{N} \frac{dN}{dt} = -\frac{1}{t_l}. \quad (4)$$

Since coalescence between two grains happen when their lattices are rotated to be aligned, we assume t_l to be proportional to the inverse of the mean grain rotation $\frac{1}{\langle \Delta\theta \rangle}$. Using the conservation equation between the total number of grains and mean volume $N(t)\langle V \rangle = V_{\text{sys}}$ and differentiating with respect to time and using the coalescence assumption, we establish the relation

$$\frac{1}{\langle V \rangle} \frac{d\langle V \rangle}{dt} = -\frac{1}{N(t)} \frac{dN(t)}{dt} = K_\theta \langle \Delta\theta \rangle, \quad (5)$$

where K_θ is a dimensionless constant describing the time scale over which rotation of a grain will result in coalescence between neighboring grains. Assuming $\langle \Delta\theta \rangle = Ct^\gamma$,

we find the two solutions

$$\langle V(t) \rangle = V_0 \left(\frac{t}{t_0} \right)^{CK_\theta}, \quad \gamma = -1 \quad (6)$$

$$\langle V(t) \rangle = V_0 \exp \left(\frac{CK_\theta}{\gamma+1} (t^{\gamma+1} - t_0^{\gamma+1}) \right), \quad \gamma \neq -1. \quad (7)$$

From these solutions we see that if the degree of rotation in the system does not fall off quickly enough grain coalescence will lead to exponential coarsening in time. In Fig. 3 we observe that for the late stages of the grain growth, the decay of $\langle \Delta\theta \rangle$ is in general much slower than t^{-1} and consequently we expect from Eq. (7) an exponential grain growth. To test the model we measure C using Fig. 3 in the late regime. In this regime we get that $\gamma \sim 0$ for the systems that fully coarsen. The product $K_\theta C$ is measured by fitting the region in which the systems coarsen exponentially using the semilogarithmic plot in the inset of Fig 2. In both cases we find that C and CK_θ scale linearly with a_2 . For $K_\theta \sim 40$ both curves collapse, which is consistent with the assumption that grain growth is mediated by grain rotation.

We can further estimate the coupling between grain growth and rotation given in Eq. (3) by assuming that the dynamics of grain rotation obeys conservation of dislocations, which then implies that [24] $r(t)\theta(t) = \text{const}$ or, equivalently, $\theta(t) \sim V^{-1/3}$, from which it follows that $\beta = 4/3$, in reasonably good agreement with our numerical results. The fact that simulations agree with this simple estimate might be an indication that dislocations are indeed conserved up to the point where grains coalesce.

V. CONCLUSION

From simulations of grain growth in polycrystalline materials, we have identified two distinct dynamical regimes. One regime is for deep quenching parameters where grain rotation is quickly suppressed and therefore leads to an overall stagnation of growth. The other regime is observed for more shallow quenches where grains continue to rotate and therefore will be able to align their lattices. The alignment eventually leads to the coalescence of neighboring grains and simultaneously allows for a few abnormally large grains to form. The grain rotation and coalescence cause an exponential increase in grain growth with time.

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