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Effect of noise-induced nucleation on grain size distribution studied via the phase-field crystal method

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Abstract
We contribute to the more detailed understanding of the phase-field crystal model recently developed by Elder *et al* (2002 *Phys. Rev. Lett.* **88** 245701), by focusing on its noise term and examining its impact on the nucleation rate in a homogeneously solidifying system as well as on successively developing grain size distributions. In this context we show that principally the grain size decreases with increasing noise amplitude, resulting in both a smaller average grain size and a decreased maximum grain size. Despite this general tendency, which we interpret based on Panfilis and Filiponi (2000 *J. Appl. Phys.* **88** 562), we can identify two different regimes in which nucleation and successive initial growth are governed by quite different mechanisms.

1. Introduction and motivation

The phase-field crystal method (PFC) has recently been introduced [1] as a novel variant of the established phase-field method for describing nucleation processes on the atomistic scale by using a free energy functional which is minimized by a periodic hexagonal state. While this method, like molecular dynamics (MD) simulations, is used for investigating processes at the atomistic scale, it has the advantage of resolving the atomic-scale structure of polycrystalline materials, at the same time averaging out the fluctuations which are inherent in MD simulations [2]. This model has proven highly useful for describing elastic effects, multiple orientations, and the formation and movement of dislocations [1, 3, 4], and has even been extended to binary alloys as well [5].

Due to the novelty of the phase-field crystal method, not all of its aspects and elements are completely understood. One element which is frequently included in such models is a stochastic noise term representing thermal fluctuations. However, the precise effects of these fluctuations has generally been neglected and not yet examined in-depth in simulation studies [2, 5, 6]—in some studies even removing the noise term entirely to ensure that it does not randomly distort the results of the parameter studies [7]. Normally, in such studies initial crystal seeds are set at the start of the simulation, and in such studies it may be acceptable to ignore the effect of noise on the results. However, for one type of study the inclusion of thermal noise is essential: the study of homogeneous nucleation, where thermal fluctuations trigger spontaneous nucleation from a material (such as a liquid). The influence of thermal noise on nucleation has been of considerable interest to material scientists for decades [8–10], and it has been studied via Monte Carlo approaches [11–14], and via the phase-field method [15, 16] before. Moreover, nucleation processes have also been studied with density functional theory (DFT) [17, 18], however to the best of our knowledge not yet with the PFC method. This appears to not yet fully exploit the potential of the PFC method, which operates on diffusive timescales and atomic length scales, and thus allows for computations which are a factor of 10^6–10^8 faster than other atomic approaches [19]. This implies that it is an excellent method to study the initial stages of solidification, from nucleation to microstructure formation, coherently with one single method. Here we will demonstrate this by presenting a study which investigates, via PFC simulations, the influence of thermal noise on nucleation rates and average grain sizes in a homogeneously nucleating system.

To do so the paper is organized as follows: in the following section, we explain the model approach which we employ in our simulation studies. Afterwards, we discuss the numerical simulations themselves and their results, and finally we conclude with a summary and an outlook.

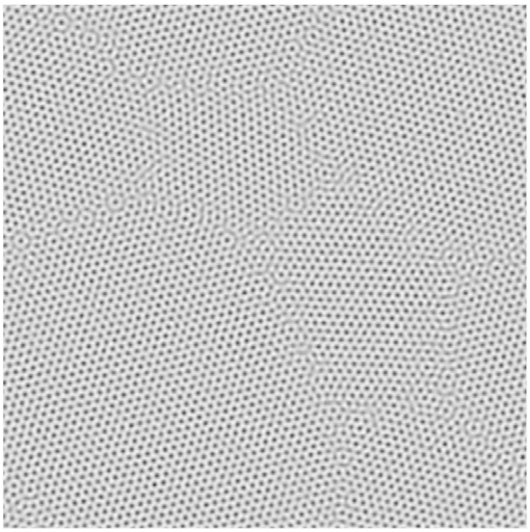

Figure 1. Sample simulation snapshot of the entire simulation domain for $G = 0.35$.

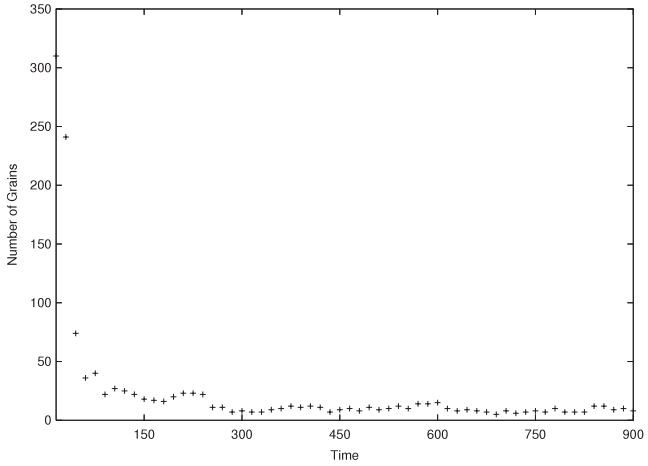

Figure 2. Plot of the number of grains over time for $G = 0.02$.

2. Underlying simulation approach

Our simulations are based on the PFC model formulation originally introduced by [1]. It employs the following free energy functional:

$$\mathcal{F} = \int \mathrm{d}\vec{r}\{\Psi[(q_0^2 + \nabla^2)^2 - \epsilon]\Psi/2 + \Psi^4\}, \qquad (1)$$

where Ψ represents the mass density field. The lattice constant of the structures described by the system is equal to $2\pi/q_0$ unit lengths [21]. ϵ is the dimensionless undercooling of the system.

A dimensionless equation of motion can be derived from the functional according to

$$\partial\Psi/\partial t = \nabla^2(\delta\mathcal{F}/\delta\Psi) + \eta, \qquad (2)$$

where η is a noise term of the form $\langle\eta(\vec{r}, t)\eta(\vec{r'}, t')\rangle = -G\nabla^2\delta(\vec{r} - \vec{r}')\delta(t - t')$. The square of the noise amplitude is linearly proportional to the temperature [1].

For a two-dimensional study we now present, the parameter q_0, which determines the length of the periodic state of the system, is set to 1 (thus, the lattice constant of the system is equal to 2π unit lengths, which in this case was set to be equal to the grid spacing of the simulation). The undercooling ϵ was set to 1 as well, the maximum value for this q_0 which still yields a stable periodic solution (see equation (24) in [19]), and which represents undercooling so strong that even the smallest disturbance from the equilibrium of the liquid results in nucleation. A calculation grid with a size of 400×400 individual cells and a cell spacing of $\Delta x = 1$ is chosen. The mass density Ψ was initialized with a uniform value of $\Psi = 0.07$. This choice of ϵ and the resulting average mass density $\overline{\Psi}$ ensured that the simulation results stayed within the hexagonal, equiaxis nucleation regime.

In order to examine grain size distributions, an algorithm was developed which identified individual grains. This was done by first identifying all calculation cells which were recognizable as part of a crystal structure (which was the case when $\Psi_{x,y} > \Psi_{\max}/2$), and then identifying groups of such cells by going from neighbouring cell to neighbouring cell checking the above criterion category until no further connecting cells could be found. In this way, it was possible to track the evolution of grain sizes over time.

3. Numerical simulations

3.1. Simulation set-up and numerical results

In our study, we varied the noise amplitude G over the following values: $G = 0.02, 0.04, 0.08, 0.1, 0.15, 0.2, 0.25, 0.3, 0.325, 0.35, 0.375, 0.4$. Using smaller noise amplitudes than $G = 0.02$ would not have yielded useful results in this study, as the average grain size would have been larger than the size of the calculation grid could have encompassed. Noise amplitudes larger than 0.4, on the other hand, generally resulted in a large number of grain 'fragments' instead of standard grains with recognizable hexagonal patterns.

The resulting grain sizes were evaluated at $t = 900$, as the number of grains as well as the grain size had stabilized after this time period for all grain sizes (see figure 1). After the calculation of the individual grain sizes, the average grain size was derived as well, although 'grains' below the size of 30 cells were discarded for this, as these do not form a complete hexagonal shape and merely represent minor dislocations instead of true grains.

As depicted in figure 2 (representing the simulation at $G = 0.02$), the number of grains is initially high after the nucleation process, but gradually decreases over time until it reaches a steady state. Minor fluctuations in the form of grain fragments persist at the boundaries due to the noise term, but the overall number of grains remains fairly stable.

To examine the impact of the noise amplitude on grain formation, the number of grains at which the simulation runs stabilized at $t = 900$ was examined. This number was plotted over G in figure 3. The grain number clearly increases with higher noise amplitudes, with the exception of the value for

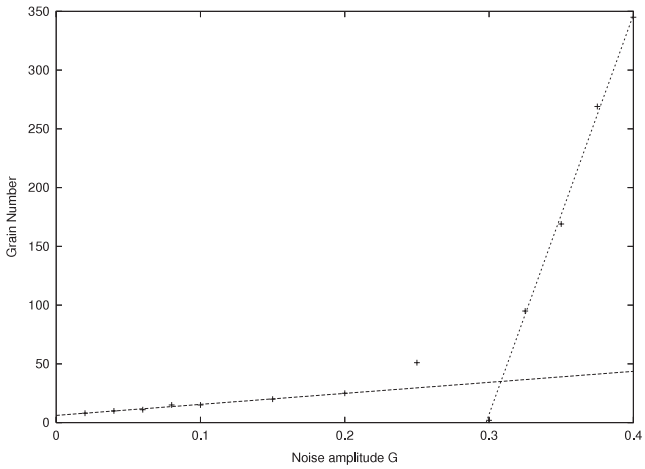

Figure 3. Plot of the number of grains dependance on noise amplitude.

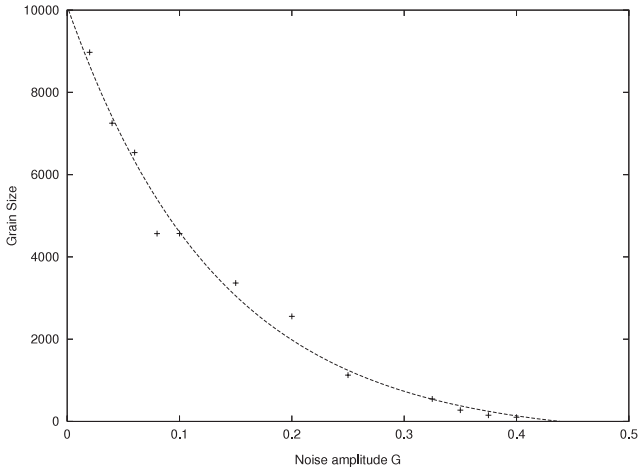

Figure 4. Plot of the average grain size dependance on noise amplitude.

$G = 0.3$. Upon closer examination of the development of the simulation run, we could reveal that three of the grains forming during the initial nucleation process had nearly identical grain orientations. This resulted 'accidentally' in a scenario where they connected with each other and turned into a single large crystal. Thus, the respective data point can be considered as an aberration when evaluating our results for general trends towards understanding underlying mechanisms.

3.2. Analysis and interpretation of our results

Analysing our results we can, at first sight, identify three regimes of different slope, one at low noise amplitudes with $G \leqslant 0.2$, where $\frac{dN}{dG} = 93.72$, another at high noise amplitudes with $G \geqslant 0.325$, where $\frac{dN}{dG} = 3400$, and an intermediary regime at $0.2 < G < 0.325$. However, as we have discussed above, the value for $G = 0.3$ can be considered anomalous and should be ignored for fitting purposes. Thus two regimes remain ('low' and 'high'), which can be fitted linearly as indicated in figure 3. The fits are

$$N_{\text{low}} = 6.154 + 93.73 \times G \tag{3}$$

$$N_{\text{high}} = -1013 + 3400 \times G. \tag{4}$$

This is equivalent to the nucleation rate.

Another value of interest is the average grain size, which is similar to the nucleation rate as it also depends on the number of grains growing, as each grain reduces the available space for others, but not identically as higher noise values result in a larger number of dislocations and incomplete grains at the grain boundaries. The relationship between the noise term and the average grain size can be found in figure 4. The data points can be approximated by the following exponential equation:

$$\bar{A}_{\text{Grains}} = -408.8 + 10\,505.2 \times e^{-7.4G}. \tag{5}$$

If we now return to figure 3 and try to derive a mechanism based understanding of the regimes we can identify, it appears that the average grain size is not the only measurement of interest. Another rather important indicator for the underlying mechanisms is given by the distribution of grain sizes. As figure 5 reveals, at lower noise amplitudes (a) the grains consist of a single large grain and a number of very small grains, inside the grain boundary region. At higher noise amplitudes, larger and larger grains become evident (b), until no grains exist in the formerly largest grain category. This occurs at noise amplitudes of $G > 0.3$ ((c) and (d)), starting with $G = 0.325$. This noise amplitude represents a 'break point' in the development of the grains. These results reveal that it makes sense to assume two regimes for the development of specific grain sizes from stochastically induced nucleation governed by the two different scenarios above (a) and (b).

As in classical nucleation theory and experiments (see equation (1) in [22]), the nucleation rate is higher at high temperatures (assuming the same amount of relative undercooling), though the classical equation for the nucleation rate converges towards a relatively constant value for high temperatures, while equation (4) increases rapidly with high G values. At first glance, the result might seem surprising—after all, the square of G is linearly proportional to the temperature. However, while the noise amplitude is temperature-dependent, the main driving force of the nucleation processes is the undercooling of the liquid, which was left constant in this study. One can therefore conclude that while a low undercooling (that is, a comparably high temperature) will result in a low nucleation rate, the thermal fluctuations caused by the same high temperatures will counterbalance this effect to some degree. Thus, neglecting the noise amplitude in nucleation studies may skew the final results, depending on the observed system. The different regimes observed also suggest that the magnitude of this effect will be different for studies at different temperatures—it may be possible to neglect it at low temperatures, but at high temperatures (less undercooling) it will be more likely to be large enough to be noticeable.

Finally, we carried out a series of simulation runs with successively smaller noise amplitudes, down to $G = 2.0 \times 10^{-16}$ (which was the smallest possible value permitted by the numerical accuracy of the simulation code) to investigate

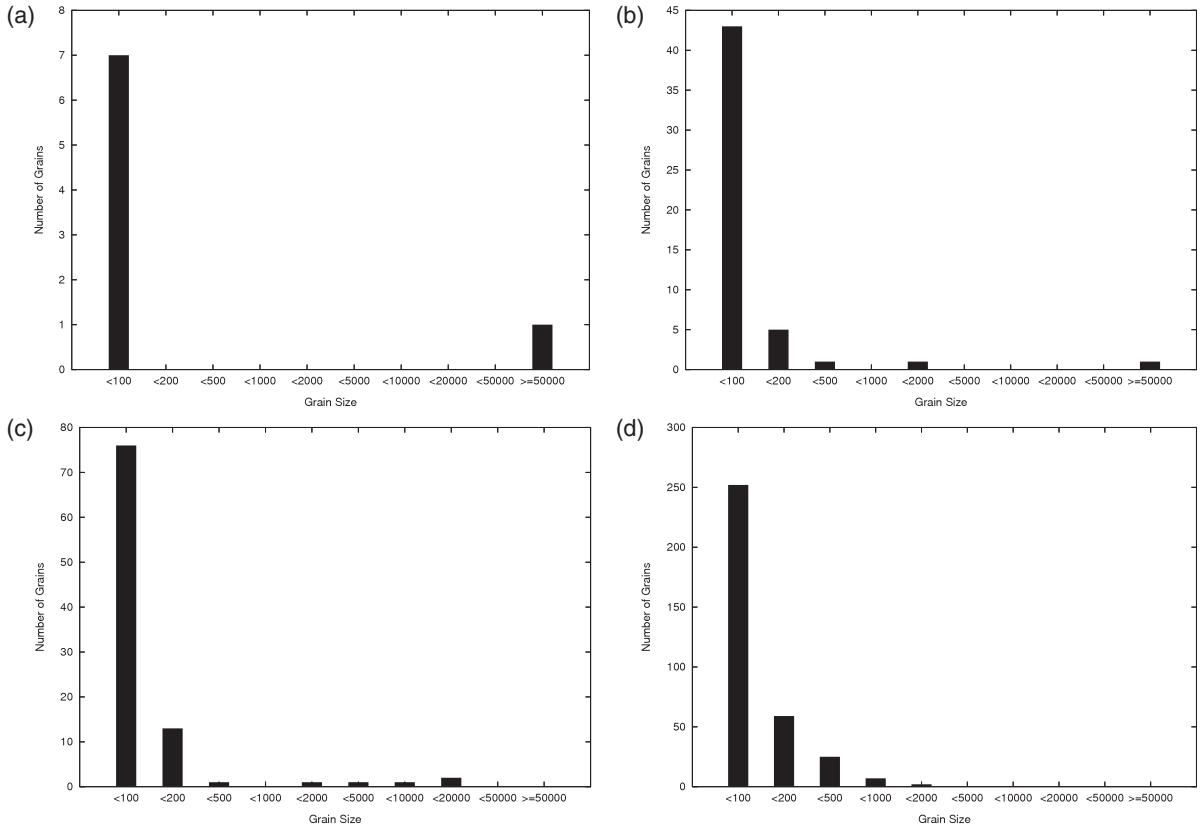

Figure 5. Grain size distribution for $G = 0.02$ (a), 0.25 (b), 0.325 (c), and 0.4 (d).

whether we can find a threshold for nucleation or not. As mentioned above, these noise amplitude values were too small to deliver useful measurements of grain size and numbers at $t = 9000$. Therefore we first assumed that there might indeed be a threshold for the noise amplitude below which nucleation would not be triggered. To address this question we carried out longer simulations with the following results: while nucleation took longer to start, the smaller the noise amplitude was chosen, eventually all simulation runs showed evidence of nucleation. Thus, unlike with conventional phase-field models [23], identifying a finite threshold for nucleation was not possible with our approach. We will demonstrate how our approach can be extended for that purpose in a subsequent upcoming paper.

4. Summary and outlook

In this paper, we presented a study examining the influence of noise on the nucleation process with simulations using the phase-field crystal method. As could be demonstrated, the average grain size decreased as the noise amplitude increased, with the correlation between the two values falling into two distinct regimes: the first is a regime dominated by a single large grain, which is surrounded by a number of smaller ones inside the grain boundary region. The second regime is characterized by the vanishing of individual outstanding large grains at large noise amplitudes. Furthermore, a relation between the noise amplitude and the nucleation rate was derived as given by equations (3) and (4). Unlike with conventional phase-field models [23], identifying a finite threshold for nucleation was not possible with our approach. We will demonstrate how our approach can be extended for that purpose in a subsequent upcoming paper.

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