

## Nucleation in Systems with Elastic Forces

W. Klein,<sup>1,\*</sup> T. Lookman,<sup>2</sup> A. Saxena,<sup>2</sup> and D. M. Hatch<sup>2,†</sup>

<sup>1</sup>*CNLS, Los Alamos National Laboratory, Los Alamos, New Mexico 87545*

<sup>2</sup>*Theoretical Division, T-11, Los Alamos National Laboratory, Los Alamos, New Mexico 87545*

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Systems with long-range interactions when quenched into a metastable state near the pseudospinodal exhibit nucleation processes that differ from classical nucleation seen near the coexistence curve. In systems with long-range elastic forces the description of the nucleation process can be quite subtle due to the presence of bulk and surface elastic compatibility constraints. We analyze the nucleation process in a simple 2D model with elastic forces and show that the nucleation process generates critical droplets with a *different* structure than the stable phase. This has implications for nucleation in many crystal-crystal transitions, specifically martensites and shape memory alloys, and for the structure of the final state.

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Nucleation in systems with long-range forces can be very different [1–5] than the process predicted by classical theory [6,7]. The reason for the difference is the presence of a pseudospinodal [8,9] that affects the structure of the critical droplet [2–4] and alters the dependence of the nucleation rate on the thermodynamic parameters [1,10]. An interesting and important class of materials exhibiting pseudospinodal behavior is that involving elastic forces, a subclass of which undergoes martensitic structural transitions [11]. Examples of such a transition are shape memory alloys [12], e.g., FePd, NiTi, and CuAlNi that transform on cooling from an “austenite” phase at high temperatures to a mesoscale twin phase (i.e., “martensite”) below the martensite transition temperature  $T_o$  [13]. This transition is of first order and takes place via nucleation [14]. However, the nucleation process in these systems is not well understood [15–22].

The purpose of this Letter is to present an analysis of nucleation near the pseudospinodal of a model with elastic forces. Within a strain only description, a complete specification of the critical droplet profile requires, in addition to the usual bulk terms [3,10], a consideration of elastic compatibility constraints. This is, to our knowledge, the first treatment that describes nucleation near the pseudospinodal in this class of materials and is the first indication that the compatibility constraint plays an essential role in this nucleation process. In addition, this is the first suggestion that homogeneous nucleation in a crystal-crystal phase transition takes place with a critical droplet that *does not* have the structure of the stable phase (e.g., martensite).

We analyze a model that exhibits a two-dimensional square to rectangle transition [23] and captures the essential physics of crystal-crystal transitions in systems with elastic forces. The order parameter (OP) is a rectangular or deviatoric strain, which is a symmetry adapted combination of the 2D strain tensor  $\varepsilon_{\mu\nu}$  ( $\mu, \nu = x, y$ ). The non-OP, or secondary strain components, are related to the OP through a compatibility equation. We can write a Ginzburg-Landau (GL) free energy in the form [23]  $F(\epsilon) = F_o(\epsilon) + F_{\text{grad}}(\nabla\epsilon) + F_{\text{cs}}(e_1, e_2)$ , where

$$F_o(\epsilon) = \int d\vec{r} \{ (\tau - 1)\epsilon^2(\vec{r}) + \epsilon^2(\vec{r})[\epsilon^2(\vec{r}) - 1]^2 \}, \quad (1)$$

$$F_{\text{grad}}(\nabla\epsilon) = \int d\vec{r} \left[ \frac{a}{4} (\nabla\epsilon(\vec{r}))^2 + \frac{b}{8} (\nabla^2\epsilon(\vec{r}))^2 \right], \quad (2)$$

$$F_{\text{cs}}(e_1, e_2) = \frac{1}{2} \int d\vec{r} [A_1 e_1^2(\vec{r}) + A_2 e_2^2(\vec{r})]. \quad (3)$$

Here  $\epsilon(\vec{r}) = \frac{1}{\sqrt{2}}[\varepsilon_{xx}(\vec{r}) - \varepsilon_{yy}(\vec{r})]$  is the OP strain,  $e_1(\vec{r}) = \frac{1}{\sqrt{2}}[\varepsilon_{xx}(\vec{r}) + \varepsilon_{yy}(\vec{r})]$ ,  $e_2(\vec{r}) = \varepsilon_{xy}(\vec{r})$  are the compression and shear strain, respectively, and  $\tau = \frac{(T - T_c)}{(T_o - T_c)}$ , where  $T_c$  is the fictitious temperature at which the relevant elastic constant would completely vanish. The coefficients  $A_1$  and  $A_2$  are elastic constants for the compression and shear, and  $a$  and  $b$  are strain gradient coefficients independent of  $T$ .

The St. Venant compatibility equations for the symmetric strain tensor are  $\nabla \times (\nabla \times \varepsilon(\vec{r}))^T = 0$ . Using the Lagrangian multiplier formalism [24] in  $d = 2$  we find for Fourier expandable strains  $\epsilon(\vec{k})$  that  $e_1(\vec{k})$  and  $e_2(\vec{k})$  are proportional to  $\epsilon(\vec{k})$ , the Fourier transform of  $\epsilon(\vec{r})$  with  $\vec{k}$  dependent coefficients. This result allows us to replace  $F_{\text{cs}}(e_1, e_2)$  in Eq. (3) with an OP potential  $F_{\text{cs}}^{\text{bulk}}(\epsilon) = \int d\vec{k} U^{\text{bulk}}(\vec{k}) |\epsilon(\vec{k})|^2$ , where  $U^{\text{bulk}}(\vec{k}) = [A_1(k_x^2 - k_y^2)^2] / [k^4 + 8(A_1/A_2)k_x^2 k_y^2]$ . In addition, the presence of a boundary (i.e., the “habit plane”) between the high symmetry austenite phase and the low symmetry martensite phase generates a surface term,

$$F_{\text{cs}}^{\text{surface}}(\epsilon) = \frac{A_2}{2} \int d\vec{k} \frac{\frac{1}{2}[1 + i\frac{l(\vec{k})}{J}]\epsilon(\vec{k})^2}{|k_y|} \times \delta(\vec{k} - \vec{k}_{\text{surface}}),$$

so that  $F_{\text{cs}} = F_{\text{cs}}^{\text{bulk}} + F_{\text{cs}}^{\text{surface}}$  [23,25]. The integral is over the wave vector modes of the interface between

phases,  $J$  is independent of  $\vec{k}$  and depends only on the ratio  $A_2/2A_1$ , and the orientation factor  $I(\vec{k}) \propto (k_x^2 - k_y^2)f(k_x, k_y)$  is a known function. Also,  $k_{x(y)}$  is the  $x(y)$  component of  $\vec{k}$  with respect to the underlying square lattice [23,25] and  $k = |\vec{k}|$ .

The free energy [Eq. (1)] has the following form: For  $\tau > 4/3$  there is one minimum at  $\epsilon = 0$ . For  $1 < \tau < 4/3$  there are three minima, but  $\epsilon = 0$  is the global minimum. At  $\tau = 1$  there are three degenerate minima. For  $0 < \tau < 1$  there are again three minima. The one at  $\epsilon = 0$  (the austenite phase) is metastable while the two minima, symmetric about  $\epsilon = 0$  (the martensite phase), are stable. The stable martensite phases have twinning and a striped structure [23,25]. For  $\tau < 0$  there is no longer a minimum at  $\epsilon = 0$ . Note that this analysis does not consider instabilities to perturbations with nonzero wave vectors. We return to this point below. We first investigate the nucleation process from the metastable minimum at  $\epsilon = 0$  near the spinodal at  $\tau = 0$ . From the spatially homogeneous free energy it is simple to calculate the OP exponent  $\epsilon \sim \tau^\beta$  with  $\beta = 1/2$ . Reinstating the Laplacian term in the free energy it can be seen that the correlation length diverges as  $\xi \sim \tau^{-\nu}$  with  $\nu = 1/2$ . Adding a spatially homogeneous external field to the free energy leads to a susceptibility  $\chi \sim \tau^{-\gamma}$  with  $\gamma = 1$  [26].

Turning to nucleation near  $\tau = 0$ , we first note that nucleation does not occur in systems with infinite range interactions. That includes mean-field systems [10]. However, systems with elastic forces do not have infinite range interactions due to the screening from defects [27]. Hence we are dealing with systems that have the same bulk and surface interactions except they have an exponential cut-off of the form  $\exp(-r/R)$ , where  $R \gg 1$  is the interaction range and  $r = |\vec{r}|$ . Because of this modification, the contribution in  $\vec{k}$  space is the convolution of the infinite range terms with a highly peaked Lorentzian which becomes a delta function in the limit  $R \rightarrow \infty$ .

Since the interaction range is large but finite the system is no longer mean field but near mean field [28,29]. In order for the mean-field approach, including the idea of a spinodal, to be a reasonable approximation when  $R \neq \infty$ , the system must satisfy the Ginzburg criterion [28,29], namely,

$$\frac{\xi^d \chi}{\xi^{2d} \epsilon^2} = \frac{\tau^{-1}}{R^d \tau^{-d/2} \tau} \ll 1. \quad (4)$$

Note that the correlation length,  $\xi$ , as are all lengths, is in units of  $R$ . The Ginzburg criterion can be rewritten as  $R^d \tau^{2-d/2} = A \gg 1$ , where  $A$  is a fixed large number. When the Ginzburg criterion is satisfied, many aspects of the mean-field spinodal are still present. However, the singularity has been smeared out [8,9]. The larger the  $A$  the better the spinodal is approximated by the pseudospinodal. Since  $A \gg 1$  for these systems the pseudospinodal is very close to a true spinodal.

To calculate the nucleation or critical droplet structure we use saddle point techniques [1,4,6,10,]. Near the pseudospinodal there is an incompletely softened mode that can be identified by examining the  $\vec{k}$  coefficient of the Gaussian term in the action. We take the action to be the free energy in Eqs. (1)–(3) [6] and initially we ignore the surface term. The structure factor  $S(\vec{k})$  is then

$$S(\vec{k}) \sim \left[ \tau + \frac{\pm |a|}{4} k^2 + \frac{b}{8} k^4 + U^{\text{bulk}}(\vec{k}) \right]^{-1}, \quad (5)$$

where the + (–) is for  $a$  positive (negative). Consider first  $a, b > 0$ . Since all terms in  $S(\vec{k})$  are positive semidefinite, the only divergence is when  $k \rightarrow 0$  and  $k_x = k_y$ . The surface term appears to strongly suppress the fluctuations that cause the divergence. However, this term was derived for a sharp surface boundary [23,25]. We need to extend this result to a surface with width  $\xi$ . The reason for this particular scale will become clear. We can consider the smooth surface to be a sequence of sharp surface boundaries or steps, each one contributing a term to the free energy of the form given for  $F_{\text{cs}}^{\text{surface}}$ . Since  $k_x = k_y$ ,  $I(\vec{k}) = 0$ . We assume a form for the surface as  $\epsilon_{\text{surface}} \propto \exp(-r/\xi)$  and  $\xi \gg R$ . The difference in order parameter amplitude between steps is

$$\Delta \epsilon_{\text{surface}} \propto \frac{d}{dr} \exp(-r/\xi) dr = -\frac{\exp(-r/\xi)}{\xi} R,$$

where we have chosen our differential step  $dr = R$ . This is justified since all lengths are in units of  $R$  and  $R$  is the coarse graining scale in the GL theory [4,10]. Taking  $k \sim \xi^{-1}$  and  $dk \sim \xi^{-2}$ , the free energy cost for the surface is  $F^{\text{surface}} \sim \frac{\xi^2 \epsilon^2(\vec{r})}{\xi}$ , where  $\xi \epsilon(\vec{r})$  scales as  $\epsilon(\vec{k})$  and the number of steps in the surface is  $\xi/R$ . As we will see, the dominant contribution to the free energy  $F$  scales as  $\int d\vec{r} \tau \epsilon^2(\vec{r}) = \xi^2 \tau \epsilon^2$ , where by  $\epsilon$  we mean to include only the dependence of  $\epsilon(\vec{x})$  on  $\tau$ . Comparing these two scaling forms we have  $F^{\text{surface}} = F \frac{C}{A^{1/2}} \ll F$ , where  $C$  is a constant of order 1 and  $A \gg 1$  from the Ginzburg criterion. The Ginzburg criterion also implies that, for a fixed but large  $R$ ,  $\tau$  cannot reach zero [10] and the system remains near mean field. The ratio  $F^{\text{surface}}/F = 0$  only as  $R$  and hence  $A \rightarrow \infty$ .

For the long-range potential we are using, the surface term is, in general, small enough to neglect. Note that if  $A$  is not infinite, then the surface term is added to  $S(\vec{k})$  eliminating the divergence at  $k = 0$ . Since  $A \gg 1$  the structure factor can be extremely large and the true spinodal is well approximated. In calculating the surface contribution we have assumed that there is a domain with nonzero  $\epsilon(\vec{r})$  with a linear size of the correlation length  $\xi$  embedded in the metastable  $\epsilon(\vec{r}) = 0$  phase. We now proceed to demonstrate the existence of this domain. First we note that  $a$  and  $b$  in Eq. (2) must have units of length to the second and fourth powers, respectively, and hence are proportional to  $R^2$  and  $R^4$  as all lengths must be proportional

to  $R$  [10]. The Euler-Lagrange equation for the critical droplet is obtained by setting the functional derivative of  $F(\epsilon)$  in Eqs. (1)–(3) equal to zero to obtain

$$-\frac{a}{2}\nabla^2\epsilon(\vec{r}) + \frac{b}{4}\nabla^4\epsilon(\vec{r}) + 2\tau\epsilon(\vec{r}) - 8\epsilon^3(\vec{r}) + 6\epsilon^5(\vec{r}) + \int d\vec{r}' \tilde{U}^{\text{bulk}}(\vec{r}, \vec{r}')\epsilon(\vec{r}') = 0, \quad (6)$$

where  $\tilde{U}(\vec{r})^{\text{bulk}}$  is  $U^{\text{bulk}}(\vec{r})$  multiplied by the exponential cutoff  $\exp(-\frac{r}{R})$ . We now assume a solution of the form

$$\epsilon(\vec{r}) = \sum_n c_n(\tau) \exp(i\vec{k}_{o,n} \cdot \vec{r}) \psi\left(\frac{\vec{r}}{L}\right) = G(\tau, \vec{r}) \psi\left(\frac{\vec{r}}{L}\right), \quad (7)$$

where  $L \gg R$ ,  $\vec{k}_{o,0}$  is the value of  $\vec{k}_{o,n}$  at which the mean-field structure factor [Eq. (5)] diverges, and  $c_0(\tau) \gg c_n(\tau)$  for  $n \neq 0$ . For  $\tau \sim 0$  the  $c_n$  for  $n \neq 0$  can be neglected [29]. We are near a pronounced pseudospinodal so that we expect the critical droplet to have an interior structure similar to spinodal critical fluctuations [4,10] [see Eq. (5)]. Since  $a > 0$  implies  $\vec{k}_{o,0} = 0$ , with the assumed form for  $\epsilon(\vec{r})$  in Eq. (7), the Euler-Lagrange equation becomes

$$-\frac{a}{2}\nabla^2\psi\left(\frac{\vec{r}}{L}\right) + \frac{b}{4}\nabla^4\psi\left(\frac{\vec{r}}{L}\right) + 2\tau\psi\left(\frac{\vec{r}}{L}\right) - 8\psi^3\left(\frac{\vec{r}}{L}\right) + 6\psi^5\left(\frac{\vec{r}}{L}\right) = 0. \quad (8)$$

Since  $k_x = k_y$ , the term involving  $\tilde{U}^{\text{bulk}}$  gives no contribution and the  $c_0(\tau)$  are chosen so that  $G^3(\tau, \vec{r}) = G(\tau, \vec{r})$  [4,30].

Since  $\tau \sim 0$  the solution of Eq. (8) has the scaled form

$$\psi(\vec{r}) = D\tau^{1/2}\tilde{\psi}\left(\frac{B\vec{r}}{\xi}\right), \quad (9)$$

where the  $\nabla^4\psi(\frac{\vec{r}}{L})$  and  $\psi^5(\frac{\vec{r}}{L})$  terms have been neglected since they are higher order in  $\tau$ ,  $B$  and  $D$  are constants determined from Eq. (8), and  $L = \xi$ . This form of the solution is what we assumed when we calculated the contribution of the surface term for a smooth surface. Hence the omission of the surface term is justified self-consistently as is the scaling of the bulk free energy used to compare with the surface contribution.

The nucleation barrier,  $\Delta F$ , is calculated by inserting the critical droplet solution, Eq. (7), into the free energy [6], Eqs. (1)–(3). It is straightforward to see that  $\Delta F \propto R^d \tau^{2-d/2} = A$ . Therefore, if  $A = \infty$  the system is mean field, rather than near mean field, and there is no nucleation.

Note that the saddle point object which is the nucleation droplet shows no evidence of the twin stripes seen in the simulation of the stable phase of this model [23]. The critical droplets near the pseudospinodal are unstable [2,10] and differ from the metastable phase by a strain order of

magnitude given by  $\tau^{1/2} \sim 0$ . Their initial growth phase is a “filling in” or an increase in the order parameter difference [2,10]. The filled in droplet has a sharp interface and hence must have twinning [23]. Therefore, the symmetry breaking which results in the twin stripes must appear in the growth phase. As we will see, if  $a$ , the coefficient of the  $(\nabla\epsilon(\vec{r}))^2$  term, is negative the case is somewhat different. We treat this next.

For  $a < 0$  and  $b > 0$  we take the minus sign in the structure factor in Eq. (5). Since  $U^{\text{bulk}}(\vec{k})$  is independent of  $k$ , the value of  $k$  where the structure factor diverges is

$$k = \pm \frac{1}{\sqrt{b}} \left[ \frac{|a|}{4} \pm \left( \frac{|a|^2}{16} - \frac{(\tau + \tau_o)b}{2} \right)^{1/2} \right]^{1/2}, \quad (10)$$

where  $0 \leq \tau_o \leq 4\rho$  is a fixed value of  $U^{\text{bulk}}(\vec{k})$ . For  $\tau > \frac{|a|^2}{8b} - \tau_o$  there is no divergence for real  $k$  and hence no instability. Since the largest value of  $\tau$  for which there is an instability is the spinodal, then for  $a < 0$  the spinodal is at  $\tau_s = \frac{|a|^2}{8b} > 0$ . The structure factor will now diverge at a nonzero value of  $k = k_o$ , where  $k_o$  is given by Eq. (10) with  $\tau_o = 0$ . Note that the additional instability generated by  $a < 0$  is at a value of  $\tau$  greater than  $\tau = 0$  expected from a simple thermodynamics calculation. It is straightforward to calculate the exponents of the correlation length and the order parameter which have the same values as those at the  $\tau = 0$  spinodal for  $a > 0$ .

Turning to the nucleation problem for  $a < 0$  and initially ignoring the surface term, the Euler-Lagrange equation has the form

$$\int d\vec{r}' \tilde{S}^{-1}(\vec{r}')\epsilon(\vec{r} - \vec{r}') - 4\epsilon^3(\vec{r}) + 6\epsilon^5(\vec{r}) = 0,$$

where  $\tilde{S}^{-1}(\vec{r})$  is given by the Fourier transform of the inverse of  $S(\vec{k})$  in Eq. (5) with  $U^{\text{bulk}}(\vec{k})$  replaced by  $\tilde{U}^{\text{bulk}}(\vec{k})$ . We again assume a solution of the form given in Eq. (7) where  $k_{o,0}$  is the vector at which the structure factor diverges with  $a < 0$ . Since  $\tau_o = 0$  implies  $k_x = k_y$  and  $|\vec{k}_{o,0}|$  is given by Eq. (10),  $\vec{k}_{o,0}$  is specified. We now expand  $\psi(\frac{\vec{r}-\vec{r}'}{L})$  in a gradient expansion about  $\frac{\vec{r}}{L}$  to obtain

$$\frac{-|a|}{2}\gamma_2\nabla^2\psi\left(\frac{\vec{r}}{L}\right) + (\tau - \tau_s)\gamma_0\psi\left(\frac{\vec{r}}{L}\right) - 4\psi^3\left(\frac{\vec{r}}{L}\right) = 0, \quad (11)$$

where, anticipating the scaling, higher order derivatives and higher powers of  $\psi(\frac{\vec{r}}{L})$  have been neglected. The constants  $\gamma_0$  and  $\gamma_2$  are  $\int d\vec{r} [\exp(i\vec{k}_{o,0} \cdot \vec{r}) + \exp(-i\vec{k}_{o,0} \cdot \vec{r})] \tilde{S}^{-1}(\vec{r})$  and  $\int d\vec{r} r^2 [\exp(i\vec{k}_{o,0} \cdot \vec{r}) + \exp(-i\vec{k}_{o,0} \cdot \vec{r})] \tilde{S}^{-1}(\vec{r})$ , respectively. As above, the  $c_n$  have been chosen so that  $G^3(\tau, \vec{r}) = G(\tau, \vec{r})$ . The solution of Eq. (11) is of the form

$$\psi\left(\frac{\vec{r}}{L}\right) = A(\tau - \tau_s)^{1/2}\tilde{\psi}\left(\frac{B\vec{r}}{\xi}\right), \quad (12)$$

justifying the omission of higher order terms.

The strain field of the critical droplet

$$\epsilon(\vec{r}) \sim A(\tau - \tau_s)^{1/2} \times [\exp(i\vec{k}_{o,0} \cdot \vec{r}) + \exp(-i\vec{k}_{o,0} \cdot \vec{r})] \tilde{\psi}\left(\frac{B\vec{r}}{\xi}\right),$$

where we have neglected terms with  $n \neq 0$ , is *not* that of the stable phase but *does* exhibit a spatial modulation of regions where  $\epsilon(\vec{r}) \neq 0$ . Consequently, the stable phase structure, as in the case  $a > 0$  where there is no spatial variation in the strain, must evolve during the growth phase. Note that the solution, Eq. (12), justifies the omission of the surface term via an argument virtually identical to the one given above.

We have calculated the first critical droplet structures for nucleation from an austenitelike phase to a twinned martensitelike phase near the pseudospinodal in a system with elastic forces. The droplets do not have the stable phase structure as expected from classical nucleation [6,7] and in the  $a > 0$  case (e.g.,  $V_3Si$ ,  $Nb_3Sn$ ) exhibit no spatial modulation. Martensitic materials such as  $FePd$  and  $InTi$ , with  $a < 0$  (and  $b > 0$ ) determined from their phonon dispersion curves, are known [17]. Droplets that do not have the stable phase structure have been predicted in the nucleation of the crystal from the melt [4,5], but this is the first indication of such a droplet structure in a crystal-crystal transition. It is also the first result that demonstrates the importance of the compatibility constraints to the phase transition kinetics.

It is important to note in systems with  $R \gg 1$  that classical nucleation is strongly suppressed. In the classical case the nucleation rate is proportional to  $\exp(-\frac{R^d \sigma^d}{\Delta f^{d-1}})$  [6,7] where  $\sigma$  is the surface tension between the droplet and the surrounding metastable state and  $\Delta f$  is the free energy density difference between the stable and metastable states. For classical nucleation, near the coexistence curve,  $\sigma \sim 1$  so that for  $R \gg 1$  nucleation is severely suppressed. In order to have nucleation in a reasonable time frame the quench must bring the system close to the pseudospinodal where  $\sigma \ll 1$ . Therefore, nucleation near the pseudospinodal will dominate the phase transition process in realistic experiments [14]. Finally, the form of nucleation discussed in this Letter allows the possibility of evolution into metastable crystallites with structures different than the stable phase.

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\*Present address: Department of Physics and Center for Computational Science, Boston University, Boston, MA 02215.

†Present address: Department of Physics and Astronomy, Brigham Young University, Provo, UT 84602.

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