A Method to Determine the Critical Nucleus of Nonlinear Elastic Solids Described by the Landau-Ginzburg Theory

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1. ABSTRACT

We consider first-order phase transitions of finite, one-dimensional, nonlinear, elastic solids undergoing a change of volume and present a method for finding the critical nucleus of the system. In order to do that we represent the nonclassical strain critical nucleus profile by a function, which is a Gaussian probability distribution multiplied by a suitable parameter. So, the Landau-Ginzburg total potential energy of the system is written as a function of two variables, which are the maximum value of the function and the rms-deviation. We find that for a given undercooling, the total potential energy of the system has a saddle-point which separates one local and one global minimum. The saddle-point corresponds to the critical nucleus of the system, the local minimum to the metastable unstrained parent phase, and the global minimum to the stable fully developed product phase. Comparing the results of our method with the results from the nonlinear differential equation for the critical nucleus, we find that outside the region of equilibrium, where the nucleation behavior is nonclassical, the agreement between the two approaches is very good. In the vicinity of equilibrium, where the nucleation behavior tends to be classical and the strain critical nucleus profile has not a Gaussian form, a disagreement occurs.

Keywords: nonclassical nucleation, critical nucleus, undercooling, Landau-Ginzburg theory

2. INTRODUCTION

First-order phase transitions of nonlinear elastic solids have been studied within the framework of the time-dependent, nonlinear, nonlocal, Landau-Ginzburg theory /1-6/. It is known that, in contrast with the classical nucleation theory /7-10/, this theory describes well the nonclassical nucleation behavior, which appears outside the region of equilibrium /11/. The nonclassical nucleation behavior includes the divergence of the size and the vanishing of the nucleation energy barrier of the critical nucleus at lattice instability, where the free energy density barrier vanishes. Also, the strain within the critical nucleus is not constant and the interface between the critical nucleus and the parent phase is diffuse. However, the Landau-Ginzburg theory does not favor closed-form analytical solutions even in one-dimensional systems, and the use of complicated numerical techniques is therefore unavoidable /11,12/.

Motivated by this, we present a simple method for finding the critical nucleus of a system described by the time-dependent Landau-Ginzburg theory, using the fact that the critical nucleus is a saddle-point configuration of the system /13,14/. We consider the simplest kind of elastic phase transitions, namely first-order phase transitions of finite, one-dimensional, elastic solids undergoing a change of volume. The order parameter of the system is the dilatational strain and the driving force for nucleation is the difference in free-energy densities of the unstrained parent phase and the strained fully developed product phase. This kind of elastic phase transitions has been described by the time-dependent, nonlinear, nonlocal, Landau-Ginzburg theory, which states that the critical nucleus of the system is the lowest energy, saddle-point solution of the time-independent (static), nonlinear differential equation of motion /4/. Within the framework of our method it is not necessary to find and solve the time-independent, nonlinear, equation of motion in order to determine the critical nucleus of the system. We use the nonlinear Landau-Ginzburg potential and assume that the nonclassical strain critical nucleus profile can be represented by a function which is a Gaussian probability distribution multiplied by a suitable parameter. So, the total Landau-Ginzburg potential energy of the system is written as a function of two variables, which are the maximum value of the function and the rms-deviation. For a given undercooling, we find that the total potential energy of the system has a saddle-point, which separates two minima. One local which corresponds to the metastable unstrained parent phase and one global which corresponds to the stable fully developed product phase of constant strain. The saddle-point corresponds to the critical nucleus of the system for the given undercooling.

In section 3, we describe the method for finding the critical nucleus of the system and in section 4 we compare the results of our method with the results obtained by the time-independent, nonlinear differential equation, which describes the critical nucleus. Finally, in section 5 we present our conclusions.

3. THE METHOD

In this section we present the method for finding the critical nucleus of a finite, nonlinear, elastic solid of length L, undergoing a change of volume first-order phase transition. The method is based on the fact that the critical nucleus of the system is a saddle-point configuration separating the metastable unstrained parent phase and the stable fully developed product phase /13,14/. We assume that the nonclassical strain critical nucleus profile, which is a state localized at the edge x = L of the system /4/, can be represented by a function $e_{cr}(x)$ given by

$$e_{cr}(x) = P_0 P(x) \tag{1}$$

where

$$P(x) = \frac{\exp\left[-\frac{(x-L)^2}{2\sigma^2}\right]}{\sqrt{2\pi\sigma^2}} \tag{2}$$

is the Gaussian probability distribution, σ is the rms-deviation and P_0 a suitable parameter. Equation (1) is written as

$$e_{cr}(x) = M_0 \exp\left[-\frac{(x-L)^2}{2\sigma^2}\right] \tag{3}$$

where

$$M_0 = e_{cr}(L) = \frac{P_0}{\sqrt{2\pi\sigma^2}} \tag{4}$$

is the maximum value of the function $e_{cr}(x)$ (edge critical strain). The corresponding critical displacement field $u_{cr}(x) = \int e_{cr}(x)dx$ is given by

$$u_{cr}(x) = \sqrt{\tfrac{\pi}{2}} M_0 \sigma \left[1 + Erf\left(\tfrac{x-L}{\sqrt{2}\sigma}\right)\right] \tag{5}$$

where *Erf(x)* is the error function. We define the size x_{cr} of the strain critical nucleus as the value of x for which Equation (3) falls of to 1/e of its maximum value /4/. We find that it is given by the simple expression $x_{cr} = \sqrt{2}\sigma$. In terms of the function $e_{cr}(x)$, the total scaled Landau-Ginzburg potential energy F of the system /4/, is written as a function of the two variables M_0 and σ

$$F(M_0, \sigma) = \int_0^L \left(f_L\left[e_{cr}(x)\right] + f_{NL}\left[e_{cr}(x)\right]\right) dx \tag{6}$$

where

$$f_L\left[e_{cr}\right]=\frac{1}{2}\delta T e_{cr}^2-\frac{1}{3}e_{cr}^3+\frac{1}{4}e_{cr}^4 \tag{7}$$

is the scaled and dimensionless local elastic free energy density and $\delta T = T - T_C$ /4/. T_C is the lattice instability temperature where the parent phase becomes unstable and $T_C < T < T_1$ where T_1 is the first-order transition temperature. The nonlocal term

$$f_{NL}\left[e_{cr}\right]=\frac{1}{2}\left(\frac{de_{cr}}{dx}\right)^2 \tag{8}$$

is the scaled and dimensionless gradient contribution to the energy density /4/.

We find that for $0 < \delta T < 2/9$ (in order for the parent phase to be metastable), the total potential energy $F(M_0, \sigma)$ of the system has a saddle-point $(M_{0,sp}, \sigma_{sp})$ which separates two minima. One local at $M_0 = 0$ which corresponds to the zero energy unstrained parent phase, and one global at $(M_{0,min}, \infty)$ which corresponds to the fully developed product phase. The saddle-point corresponds to the critical nucleus of the system for the given undercooling and the nucleation energy barrier of the critical nucleus F_{cr} is given by $F_{cr} = F(M_{0,sp}, \sigma_{sp})$. The saddle-point and the global minimum are both depend on the specific value of δT. In Figure 1, a contour plot of $F(M_0, \sigma)$ is plotted for $\delta T = 0.18$ and $L = 100$, where the saddle-point and the two minima are shown.

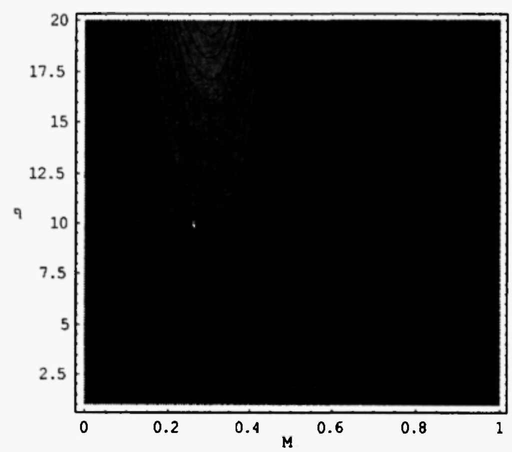

Fig. 1: A contour plot of the total potential energy $F(M_0,\sigma)$ of the system for $\delta T = 0.18$.

We find numerically a saddle-point at $(M_{0,sp}, \sigma_{sp}) = (0.373, 4.371)$, separating a local minimum at $M_0 = 0$ and a global minimum at $(M_{0,min}, \sigma_{min}) = (0.764, \infty)$. It is easy to show that within the framework of the time-dependent Landau-Ginzburg theory /4/, the strain critical nucleus profile $e_{cr}(x)$ and the corresponding displacement field $u_{cr}(x) = \int e_{cr}(x)dx$ are given by

$$e_{cr}(x) = \frac{3\delta T}{1+\sqrt{1-\frac{9}{2}\delta T}\cosh\left[(x-L)\sqrt{\delta T}\right]} \tag{9}$$

$$u_{cr}(x) = \sqrt{2}\ln\left[\frac{1-\frac{9}{2}\delta T+\left(1+\sqrt{\frac{9}{2}\delta T}\right)\sqrt{1-\frac{9}{2}\delta T}e^{(x-L)\sqrt{\delta T}}}{1-\frac{9}{2}\delta T+\left(1-\sqrt{\frac{9}{2}\delta T}\right)\sqrt{1-\frac{9}{2}\delta T}e^{(x-L)\sqrt{\delta T}}}\right] \tag{10}$$

Comparison of the displacement profiles $u_{cr}(x)$ obtained by our method and by Equation (10) is shown in Figure 2, whereas in Figure 3 the corresponding strain critical nucleus $e_{cr}(x)$ are shown. The agreement between the two approaches is very good.

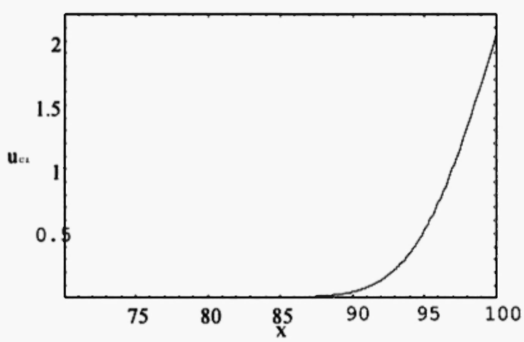

Fig. 2: The corresponding displacement profiles for $\delta T = 0.18$. The solid line for the method and the dashed line for Equation (10).

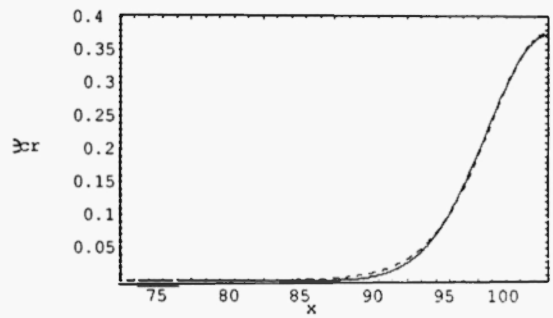

Fig. 3: The corresponding strain critical nucleus profiles for $\delta T = 0.18$. The solid line for the method and the dashed line for Equation (9).

4. COMPARISON OF THE TWO APPROACHES

We find that within the framework of the time-dependent Landau-Ginzburg theory /4/, the edge strain $e_{cr}(L)$, the size x_{cr} and the nucleation energy barrier F_{cr} of the critical nucleus, are given by the equations (in terms of the scaled and dimensionless constants and variables)

$$e_{cr}(L) = \frac{3\delta T}{1+\sqrt{1-\frac{9}{2}\delta T}} \tag{11}$$

$$x_{cr} = \frac{Arc\cosh\left[\dfrac{\left(1+\sqrt{1-\frac{9}{2}\delta\mathrm{T}}\right)e-1}{\sqrt{1-\frac{9}{2}\delta\mathrm{T}}}\right]}{\sqrt{\delta\mathrm{T}}} \tag{12}$$

$$F_{cr} = \int_0^L \left(f_L\left[e_{cr}(x)\right] + f_{NL}\left[e_{cr}(x)\right]\right) dx \tag{13}$$

where the strain critical nucleus profile $e_{cr}(x)$ is given by Equation (9). As in the previous section, we define the size x_{cr} of the critical nucleus as the value of x for which Equation (9) falls of to 1/e of its maximum value.

In order to illustrate the capability of our method, we determine numerically for a system of length L = 100, the dependence of the edge strain $e_{cr}(L)$, the size x_{cr} and the nucleation energy barrier F_{cr} of the critical nucleus on δT, and compare it with that obtained by Equations (11)-(13) respectively. This is shown in Figures 4 through 6 respectively. In Figure 7, the strain of the fully developed product phase, $M_{0,min}$ is plotted as a function of δT, and compared with the value

$$e_m(\delta\mathrm{T}) = \frac{1+\sqrt{1-4\delta\mathrm{T}}}{2} \tag{14}$$

obtained by the time-dependent Landau-Ginzburg theory [4].

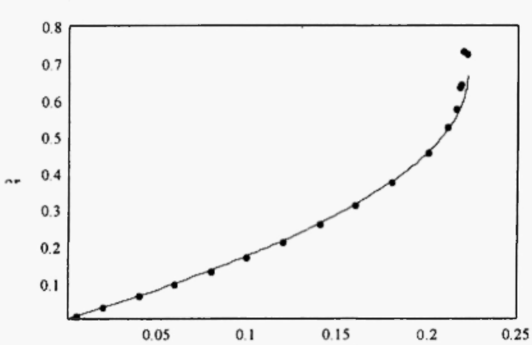

Fig. 4: The edge strain of the critical nucleus as a function of δT. The symbols for the new method and the solid line for Equation (11).

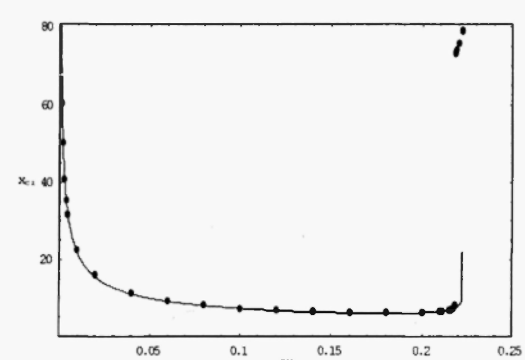

Fig. 5: The size of the critical nucleus as a function of δT. The symbols for the new method and the solid line for Equation (12).

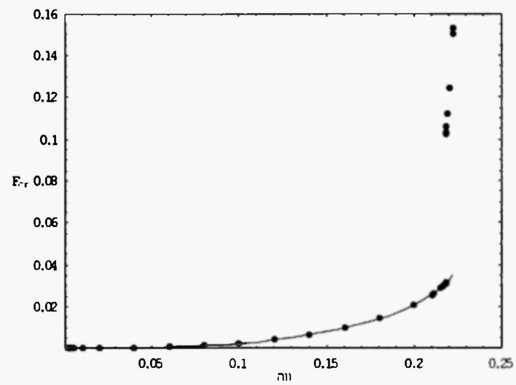

Fig. 6: The nucleation energy barrier of the critical nucleus as a function of δT. The symbols for the new method and the solid line for Equation (13).

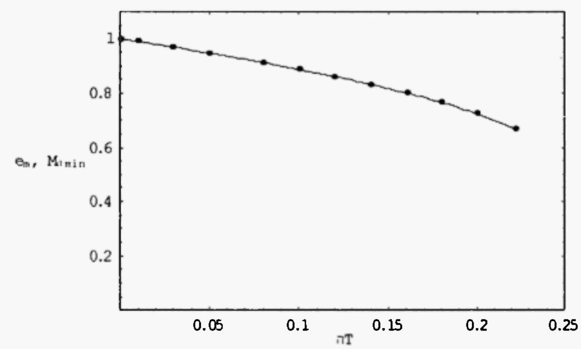

Fig. 7: The strain of the fully developed product phase as a function of δT. The symbols for the new method and the solid line for Equation (14).

As seen in Figure 4, within the framework of our method and outside the region of equilibrium (at equilibrium δT = 2/9), the edge strain of the critical nucleus decreases as δT decreases and tends to zero as the condition for lattice instability (where the parent phase becomes unstable) δT = 0 is approached. The agreement with Equation (11) is very good. In the vicinity of equilibrium where the nucleation behavior is classical, there is a disagreement between the two approaches.

In Figure 5 it is seen that according to our method, as the condition for lattice instability is approached the size of the critical nucleus diverges, in very good agreement with Equation (12). As δT increases, the critical size decreases initially to a minimum, and then increases to a finite value at equilibrium, in difference with Equation (12) according to which the size of the critical nucleus is infinite at equilibrium. The two approaches match very well outside the region of equilibrium.

As seen in Figure 6, according to our method, as δT decreases the nucleation energy barrier of the critical nucleus decreases and vanishes at lattice instability where δT = 0. The agreement between the two approaches outside the region of equilibrium is very good, whereas in the vicinity of equilibrium our method predicts a significant bigger nucleation energy barrier.

Finally, in Figure 7 it is seen that at lattice instability, $M_{0,min}$ = 1 and as the undercooling decreases the value of $M_{0,min}$ decreases to the value 2/3 at equilibrium. The agreement between the two approaches is extremely good in the whole range of δT. This justifies the capability of our method to determine the uniform fully developed product phase.

5. CONCLUSIONS

We have developed a method for finding the critical nucleus of a finite, one-dimensional, nonlinear, nonlocal, elastic solid undergoing a change of volume first-order phase transition. The method is based on the fact that the critical nucleus of the system is a saddle-point configuration, which separates the metastable unstrained parent phase and the stable fully developed product phase. In order to illustrate the capability of our method, we have compared it with the results obtained by the nonlinear differential equation for the critical nucleus, and we have found that outside the region of equilibrium, the agreement between the two approaches is very good. On the other hand, in the vicinity of equilibrium where the nucleation behavior tends to be classical, the Gaussian profile of the function $e_{cr}(x)$ given by Equation (3) is not appropriate to represent the strain critical nucleus profile and there is a difference between the two approaches. Although we have restricted to one-dimensional systems, the method we propose is quite general and simple, and it could be applied to more complex systems, where analytical progress is not possible and the existing numerical techniques are complicated and computationally intensive.

6. REFERENCES

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