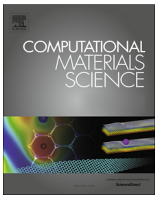
Computational Materials Science 105 (2015) 101–109

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci



Quantifying a two-mode phase-field crystal model for BCC metals at melting point

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ARTICLE INFO

Article history:
Received 1 December 2014
Received in revised form 7 March 2015
Accepted 31 March 2015
Available online 22 April 2015

Keywords:
Phase-field crystal
BCC
Fe
Melting
Solid–liquid

ABSTRACT

A recently developed two-mode phase-field crystal (PFC) model (Wu et al., 2010; Asadi and Asle Zaeem, 2015) is applied for quantitative modeling of body centered cubic (BCC) crystals at their melting points. This model incorporates the first two density wave vectors of BCC crystals in its formulation and consists of three model parameters (two independent and one dependent) in its dimensionless form. A systematic study is presented to show that the two independent parameters of the model control the material properties such as solid and liquid densities and the structure factor. An iterative procedure is presented to determine the PFC model parameters for specific BCC materials using their liquid structure factor and the fluctuation amplitude of atoms in their crystalline state. As a case study, the two-mode PFC model parameters are determined for Fe at its melting point. The calculated model parameters and results of the PFC model are validated by comparing the calculated expansion in melting, solid and liquid densities, elastic constants, and bulk modulus of Fe with the available experimental and computational data in the literature. In addition, to show the potential application of this PFC model, the solid–liquid interface free energy and surface anisotropy of Fe are determined and compared with their available counterparts in the literature.

1. Introduction

Phase-field crystal (PFC) model has drawn a great attention among materials scientists for the past couple of years [1–5]. The fascinating unique characteristic of PFC models is that it provides atomistic scale details acting on diffusive time scales by resolving the crystal density field; thus, elasticity, plasticity, vacancies, grain boundaries, and two-phase interfaces are naturally incorporated in PFC models. The free energy of the original PFC model is [1]

$$F = \int_V \left\{ \frac{1}{2}\phi(\mathbf{r})\left[\alpha + \lambda\left(q_0^2 + \nabla^2\right)^2\right]\phi(\mathbf{r}) + \frac{g}{4}\phi(\mathbf{r})^4 \right\} d\mathbf{r}, \qquad (1)$$

where $\alpha,\ \lambda,\ q_0$ and g are parameters and $\phi(\mathbf{r})$ is a function related to the density field which is constant in the liquid state and a periodic function in the crystalline state. The above-mentioned PFC model is also called one-mode PFC, because it damps the dynamics of the system except near the first density wave length. Examples of different applications of PFC models include simulating solidification [6], elastic deformation [7], spinodal decomposition [8], grain-boundary premelting [9], dislocation dynamics [10], Kirkendall effect [11], structural phase transformation [12,13], and stacking faults [14]. PFC models can be also used in quantitative modeling of materials if its model parameters are determined for the material of interest at a given temperature; e.g. the parameters $\alpha,\ \lambda,\ q_0$ and g for the one-mode PFC model. This quantitative modeling of materials using PFC models has been only explored for modeling of BCC (Fe) [15–18] and FCC (Ni) [19,20] metals at their melting points and their coexistence with the liquid state. For FCC crystals, Wu et al. [19] developed a two-mode PFC model and presented a method to determine its model parameters based on the liquid structure factor and solid density amplitude fluctuation. These quantitative studies used PFC models to simulate solid–liquid and grain growth microstructures, and to calculate properties such as solid–liquid interface free energy, and grain boundary free energy in reasonable agreement with the experimental data. These PFC models need additional improvements to better calculate some other properties such as expansion in melting and elastic constants, which will be explained in details in the next paragraphs. This article aims to improve quantitative modeling of BCC metals at their melting points; therefore, we will focus on quantitative PFC modeling of BCC metals hereafter.

The three main factors affecting the quantitative modeling of materials (including PFC models) are: I. the model formulations, II. the method used to determine model parameters, and III. the

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http://dx.doi.org/10.1016/j.commatsci.2015.03.051
0927-0256/

input material properties obtained from other methods, such as molecular dynamics (MD) or experiments for the case of PFC modeling for instance. The method to determine one-mode PFC model parameters ($\alpha,\ \lambda,\ q_0$ and g) was proposed for the first time by Wu and Karma [15]. They followed Ginzburg–Landau theory and a multi-scale analysis to propose relations to determine the PFC parameters suitable for modeling BCC metals near their melting points and their coexistence with the liquid state. Wu and coworkers provided relations between the parameters α, λ and q_0 and the liquid structure factor of the element at its melting point. The fluctuation amplitude of the atoms in the solid state were used to obtain the parameter g. These input material properties were determined for Fe form MD simulations performed by Sun et al. [21] using embedded atom method (EAM) [22,23]. The resulted quantitative PFC model calculated the solid–liquid interface free energy and surface anisotropy in reasonable agreement with MD results and experiments. The method to determine the one-mode PFC model parameters using the same input material properties was updated by Jaatinen et al. [16]. They used the common tangent line between solid and liquid densities and marginally improved the solid–liquid interface free energy calculations. Jaatinen and coworkers also calculated the grain boundary free energy of Fe near its melting point which was also in reasonable agreement with experiments. Recently, Asadi et al. [18] used a systematic procedure to model solid–liquid coexistence of Fe using one-mode PFC model. They modified the parameters of the modified-embedded atom method (MEAM) interatomic potentials of Fe in the concept of MD simulations [24] to make them suitable for modeling solid–liquid coexistence problems; they performed MD simulations at the melting point to provide input material properties for the PFC model using an iterative procedure. This one-mode PFC modeling resulted in improved calculations of solid–liquid interface free energy, expansion in melting, and grain boundary free energy for Fe.

Regardless of the utilized model parameter determination method, the one-mode PFC model is unable to calculate expansion in melting and elastic constants in agreement with experiments. The one-mode PFC model predicts the expansion in melting significantly higher than the experiments (three to four times higher) because the value of the liquid structure factor at zero cannot be included in the parameter determination procedure [16]. Jaatinen et al. [16] introduced an eight-order fitting PFC (EOF-PFC) model that predicts the expansion in melting of Fe in agreement with the experiment. EOF-PFC uses an additional PFC model parameter in order to include the value of liquid structure factor at zero in the fitting procedure. Nevertheless, the available PFC models for BCC metals under predict the elastic constants and result in zero tetragonal shear modulus [19] ($C' = (C_{11} - C_{12})/2 = 0$ because $C_{11} = C_{12} = C_{44}$ in available PFC models for BCC crystals).

In this article, we utilize a two-mode PFC model which is suitable for modeling BCC metals at their melting points. This model incorporates the first two density wave vectors in its formulation and damps out the dynamics of the system except near the first two density wave lengths, q_0 and q_1; the Helmholtz free energy of this two-mode PFC model is [19,25]

$$F = \int_V \left\{ \frac{\phi(\mathbf{r})}{2} \left[\alpha + \lambda \left[(q_0^2 + \nabla^2)^2 + r_0 \right] \left[(q_1^2 + \nabla^2)^2 + r_1 \right] \right] \phi(\mathbf{r}) + \frac{g}{4} \phi(\mathbf{r})^4 \right\} d\mathbf{r}, \tag{2}$$

where $\alpha,\ \lambda,\ q_0,\ r_0,\ q_1,\ r_1$ and g are parameters to be determined for the specific material.

In Section 2, the dimensionless form of the model is reviewed (Eq. (4a)) in order to study the properties of the model thoroughly. The dimensionless form of the model has two adjustable model parameters (ε and R_1) and a dependent model parameter (R_0). First, we systematically investigate the influence of varying ε and R_1 on dimensionless properties such as solid and liquid densities and density wave amplitudes which shows that the model has the capability to control these properties. Then, the relations to determine the elastic constants in the present model are derived which results in non-zero tetragonal shear modulus ($C' = (C_{11} - C_{12})/2 \neq 0$ because $C_{11} \neq C_{12} = C_{44}$) unlike the one-mode PFC model. In addition, an iterative method to determine the present PFC model parameters for BCC metals are presented.

In Section 3, the two-mode PFC model is used to quantitatively investigate the solid and liquid properties of Fe at its melting point. The model parameters of the present PFC model for Fe are determined. Expansion in melting, solid and liquid densities, elastic constants, and bulk modulus of Fe at its melting point are calculated and compared to their experimental counterparts to validate the results. The solid–liquid interface free energy and surface anisotropy of Fe are also determined and compared with the results of one-mode PFC models and their MD counterparts from the literature.

2. Two-mode phase-field crystal model

2.1. Basic equations

The standard time-evolution equation for conserved phase fields is valid for PFC models:

$$\frac{\partial \phi(\mathbf{r})}{\partial t} = M \nabla^2 \frac{\delta F}{\delta \phi(\mathbf{r})}, \tag{3}$$

where M is the mobility constant and F is the two-mode PFC free energy in Eq. (2). It is convenient to use the dimensionless form of the PFC model in analytical and computational procedures because it has less number of parameters to deal with. The dimensionless form of Eqs. (2) and (3) are

$$F^* = \int_V \left\{ \frac{\psi}{2} [-\varepsilon + \left[(1 + \nabla^2)^2 + R_0 \right] [(Q_1^2 + \nabla^2)^2 + R_1]] \psi + \frac{\psi^4}{4} \right\} d\boldsymbol{x}, \tag{4a}$$

$$\frac{\partial \psi}{\partial t^*} = \nabla^2 \frac{\delta F^*}{\delta \psi}, \tag{4b}$$

where $\varepsilon = -\alpha/\lambda q_0^8$, $R_0 = r_0/q_0^4$, $Q_1 = q_1/q_0$, $R_1 = r_1/q_0^4$, $\psi = \phi\sqrt{g/\lambda q_0^8}$, $\boldsymbol{x} = q_0 \mathbf{r}$, $F^* = (g/\lambda^2 q_0^{13})F$, and $t^* = tM\lambda q_0^7$. In Eq. (4a), ε is a small positive parameter, R_1 is the second parameter which can be positive or negative, and R_0 is a parameter which will be determined as a function of R_1 and ε. The first and second density wavelengths (q_0 and q_1) for BCC crystals are related to [1 1 0] and [2 0 0] density wave vectors, respectively; thus, $Q_1 = \sqrt{2}$. The corresponding dimensionless density field for BCC crystal is:

$$\begin{aligned} \psi = \psi_s &+ 4A_s(\cos qx\ \cos qy + \cos qx\ \cos qz + \cos qy\ \cos qz) \\ &+ 2B_s(\cos 2qx + \cos 2qy + \cos 2qz), \end{aligned} \tag{5}$$

where ψ_s is the average dimensionless density at solid, A_s and B_s are the density amplitudes for [1 1 0] and [2 0 0] density wave vectors, respectively, and $q = 1/\sqrt{2}$. It is worth mentioning that we have chosen the second density wave vector in our formulation to be [2 0 0], because it is the closest one to [1 1 0] wave vector. The choice of a farther second density wave vector is also possible, which results in a smaller periodic length for the density field (Eq. (5)). This means that the resulted model needs to use smaller mesh sizes for convergence of the results. The free energy density in the solid state is obtained by substituting Eq. (5) into Eq. (4a), integrating over a lattice cell, and dividing the resultant by the lattice volume as

$$\begin{aligned} f_s = -[\varepsilon - (1 + R_0)(4 + R_1)] \frac{\psi_s^2}{2} + \frac{\psi_s^4}{4} + 6(R_0 - \varepsilon + R_0 R_1 \\ + 3\psi_s^2)A_s^2 + 3(3\psi_s^2 + R_0 R_1 + R_1 - \varepsilon)B_s^2 \quad + 72A_s^2 B_s \psi_s \\ + 144A_s^2 B_s^2 + 144A_s^3 B_s + 48A_s^3 \psi_s + 135A_s^4 + \frac{45}{2} B_s^4. \end{aligned} \tag{6}$$