Controlling Magnetic Anisotropy in Amplitude Expansion of Phase Field Crystal Model

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The amplitude expansion for a magnetic phase-field-crystal (magnetic APFC) model enables a convenient coarse-grained description of crystalline structures under the influence of magnetic fields. Considering higher-order magnetic coupling terms, the possibility of tuning the magnetic anisotropy in these models is demonstrated. This allows reproducing the easy and hard directions of magnetization. Such a result can be achieved without increasing the computational cost, enabling simulations of the manipulation of dislocation networks and microstructures in ferromagnetic materials. As a demonstration, the simulation of the shrinkage of a spherical grain with the magnetic anisotropy of Fe is reported.

1. Introduction

In order to explore the possibilities external magnetic fields offer to manipulate microstructure in ferromagnetic materials,[1] a detailed understanding of the interactions between magnetic fields and solid-state matter transport is required. Various modeling approaches have been introduced to describe magnetostructural interactions in a multiscale framework. A promising approach is the phase-field-crystal (PFC) model,[2,3] describing crystal lattices through a periodic density field, which was extended to capture the fundamental physics of magnetocrystalline interactions.[4,5] In ref. [4], the PFC density is coupled with magnetization to generate a ferromagnetic solid below a critical temperature, while in ref. [5] this PFC model is extended to multiferroic binary solid solutions and used to demonstrate the influence of magnetic fields on the growth of crystalline grains. Based on symmetry arguments, magnetostructural interactions are incorporated phenomenologically. This model, which consists of a system of evolution equations for the rescaled atomic density field $\phi$ and an averaged magnetization $m$, is used in refs. [6,7] in a simplified form to study the role played by external magnetic fields on the evolution of defect structures, grain boundaries, long-time scaling behaviors, and various geometrical and topological properties in grain growth. While the microscopic details are resolved with the considered magnetic PFC model and experimentally relevant time scales can be reached, the required spatial resolution restricts simulations to 2D settings. Furthermore, all previous investigations have considered generic material parameters for magnetic anisotropy.

The complex amplitude PFC (APFC) model initially introduced in refs. [8,9] provides a framework to overcome the restrictions resulting from the spatial resolution required by PFC models. The idea is to model the amplitude of density fluctuations instead of the density itself. This allows for reaching larger spatial scales while retaining essential microscopic effects,[10-12] thus enabling mesoscale investigations of crystalline systems. For a recent review of APFC models, we refer to another study.[13] In ref. [14], a magnetic APFC model is introduced, and its applicability in a simple 3D setting has been demonstrated. Together with advanced numerical approaches,[15] this enables the description of magnetostructural interactions in multiscale simulations, combining the dynamics of defects, dislocation networks, and grain boundaries with experimentally accessible microstructure evolution on a diffusive time scale.[13]

Here, we modify the magnetic coupling energy based on this approach. In the proposed modification, the magnetic anisotropy can be tuned, and the easy and hard directions of ferromagnetic materials can be reproduced. We demonstrate this for body-centered-cubic (BCC) and face-centered-cubic (FCC) crystals. This modification essentially overcomes the limitations of previous approaches and enables the modeling of material-specific magnetic anisotropies.

The article is structured as follows. In Section 2, we briefly review the magnetic PFC and APFC models. We describe the numerical approach to solving the magnetic APFC model, discuss a modification of the magnetic coupling energy that does not increase the computational cost, and introduce the few-mode approximation and the minimal energy surface in the reciprocal space as a tool to analyze the impact of magnetic coupling. In Section 3, we analyze the magnetic properties of BCC and FCC crystals and demonstrate the possibility of tuning the easy and hard directions with the modified magnetic coupling energy. We further apply this enhanced setting to study the magnetic impact on grain growth. In Section 4, we draw conclusions.
2. Magnetic PFC and APFC Models

2.1. Magnetic PFC Model

In refs. [4,5], a magnetic PFC model was proposed. This model describes the basic phenomenology of magnetostructural interactions in crystals, namely, magnetic anisotropy and magnetostriiction. In the limit of constant magnetization or strong external magnetic fields, the free energy, on which the magnetic PFC model builds, reads

\[
\mathcal{F}[\phi, m] = \int_\Omega \left[ \frac{B_0^2}{2} \phi (\delta_0^2 + \nabla^2)^2 \phi + \frac{\Delta B_0}{2} \phi^2 - \frac{t}{3} \phi^3 + \nu \phi^4 \right] \, \text{d}r + \mathcal{F}_m[\phi, m] \tag{1}
\]

\[
\mathcal{F}_m[\phi, m] = \int_\Omega \left[ \frac{\partial \Omega}{2} \mathbf{m} \cdot \mathbf{m} \cdot (\mathbf{m} \cdot \nabla \phi)^2 \right] \, \text{d}r \tag{2}
\]

where \( \phi \) denotes the scaled particle density and \( m \) the magnetization. \( \delta_0 \) defines the lattice spacing at equilibrium, \( \Omega \) is the domain of integration, and \( B_0, \Delta B_0, t, \) and \( \nu \) are parameters as introduced in ref. [16]. Together with the average density \( \bar{\phi} \), they define crystal structure and physical properties. \( \mathcal{F}_m[\phi, m] \) accounts for the magnetocrystalline interactions with magnetization \( \mathbf{m} \), which is assumed to be constant and scaled to unit length, \( |\mathbf{m}| = 1 \). Even powers are considered in the expansion due to the required mirror symmetry \( m \rightarrow -m \). The parameters \( \delta_0 \) can be tuned to control magnetic anisotropy. Nevertheless, by setting \( \delta_0 = 0 \) and considering the expansion only to the lowest order, resulting in \( \mathcal{F}_m[\phi, m] = \int_\Omega \frac{\partial \Omega}{2} (\mathbf{m} \cdot \nabla \phi)^2 \, \text{d}r \), leads to magnetic anisotropy and can be considered as a minimal model. In this setting, \( \delta_0 \) simply controls the strength of the magnetic interaction. Integrating by parts allows us to rewrite this as \( \mathcal{F}_m[\phi, m] = \int_\Omega -\frac{\partial \Omega}{2} (\mathbf{m} \cdot \nabla \phi)^2 \, \text{d}r \), which is numerically advantageous and has been considered in refs. [6,7]. The evolution equation for \( \phi \) reads

\[
\frac{\partial \phi}{\partial t} = \nabla^2 \frac{\delta \mathcal{F}}{\delta \phi} \tag{3}
\]

As the expansion in Equation (2) is phenomenological, we can also propose a different expansion fulfilling the same symmetry constraints

\[
\mathcal{F}_m[\phi, m] = \int_\Omega \left[ \frac{\partial \Omega}{2} \mathbf{m}^2 m^2 \phi^2 + \sum_{i=1}^{\infty} \frac{\partial \Omega}{2} (\mathbf{m} \cdot \nabla \phi)^2 \phi \right] \, \text{d}r \tag{4}
\]

We again set \( \delta_0 = 0 \) and, as already seen, in lowest order \( \mathcal{F}_m[\phi, m] = \mathcal{F}_m[\phi, m] \) if \( \delta_0 = -\delta_x \). However, in this formulation, higher-order terms can be also considered in a numerically efficient manner. We will consider expansions to the order \( i = 2 \) and demonstrate the possibility of tuning the magnetic anisotropy.

2.2. Magnetic APFC Model

In the crystal phase, the density field \( \phi \) described by the PFC model is periodic, with maxima at the atomic lattice sites, thus encoding the crystal structure directly. In the amplitude expansion of the PFC model, this density is expanded in terms of a small set of Fourier modes

\[
\phi(r) = \bar{\phi} + \sum_{j=1}^{N} \left[ A_j(r) e^{i k_j \cdot r} + A_j^*(r) e^{-i k_j \cdot r} \right] \tag{5}
\]

with \( \bar{\phi} \) the overall mean density and \( \{k_j\}_{j=1}^{N} \) defining the symmetry of a reference crystal usually corresponding to a bulk, relaxed lattice. The reference crystal is then described by real and constant amplitudes \( A_j \in \mathbb{R} \). Complex and space-dependent amplitudes account for deviations from the reference crystal. \( |A_j| \) entails information on local ordering while the phase of the complex amplitudes accounts for displacement with respect to the reference crystal.

Thus, some of the amplitudes vanish at defects where singular displacement occurs, namely, the ones having singular phases. Far from defects, both the phase and argument of \( A_j \) vary typically on a larger length scale than the distance between particles in the crystal. Only at defects the amplitudes vary on a similar scale.

In ref. [14], a corresponding magnetic APFC model using Equations (1), (3), and (4) has been derived. It results from substituting Equation (5) into Equation (1) and (4) and averaging fluctuations on small scales. The resulting free energy reads

\[
\mathcal{F}[\{A_j\}] = \int_\Omega \sum_{j=1}^{N} B_0^2 (A_j^* G_j^2 A_j + A_j^* C_j A_j) + g^s(\{A_j\}) \, \text{d}r \tag{6}
\]

with

\[
g^s(\{A_j\}) = \sum_{j=1}^{N} \left( -\frac{3V}{2} |A_j|^4 + \frac{\Delta B_0}{2} A_j^2 + \frac{3V}{4} A_j^4 + f^s(\{A_j\}) \right) \tag{7}
\]

and \( A_j^2 \equiv 2 \sum_{j=1}^{N} |A_j|^2 \), \( A_j^4 \equiv (A_j^2)^2 \), \( G_j \equiv g_0 - |k_j|^2 + \nabla^2 + 2 i k_j \cdot \nabla \), and \( C_j \equiv \frac{1}{2} \sum_{j=1}^{N} \partial \Omega \delta_0 (m \cdot \nabla + i m \cdot k_j) \). The magnetic coupling is considered in the terms \( A_j C_j A_j \). In ref. [14], only the lowest order \( (i = 1) \) is considered, leading to a minimal magnetic APFC model. The equilibrium crystal without magnetization is chosen as a reference, which leads to \( |k_j| = g_0 \). \( f^s \) is a polynomial function in \( \{A_j\} \) and \( \{A_j^*\} \). It depends on the reference crystal structure, see other studies [13,15,17].

The evolution equations for each amplitude read

\[
\frac{\partial A_j}{\partial t} = -|k_j|^2 \frac{\delta \mathcal{F}}{\delta A_j^*} \tag{8}
\]

with

\[
\frac{\delta \mathcal{F}}{\delta A_j^*} = B_0^2 \left[ G_j^2 + C_j \right] A_j^* + \frac{\partial g^s(\{A_j\})}{\partial A_j^*} \tag{9}
\]

Considering \( C_j = \mathcal{M}_j^2 + Q_j \) we can write

\[
\left[ G_j^2 + \mathcal{M}_j^2 \right] = (G_j + i \mathcal{M}_j)(G_j - i \mathcal{M}_j) = N_j^+ N_j^- \tag{10}
\]

and Equation (8) can be written as systems of second-order equations.
\[ \frac{\partial A_j}{\partial t} = -|k|^2 \left[ B_\alpha^i N_j^\alpha \mu_j + B_\alpha^i Q_j A_j + G_j((A_j)) \right] \]  \hspace{1cm} (11)

\[ \mu_j = -N_j^\alpha \dot{A}_j \]

with \( G_j((A_j)) = d_{\alpha}^i \left( A_j^i \right) / \partial A_j \) the nonlinear terms. In the following sections, we consider two specific forms of the magnetic coupling term

Model MC1: \( \mathcal{M}_j^2 = \alpha_2 \left( \mathbf{m} \cdot \nabla + i \mathbf{m} \cdot \mathbf{k}_j \right)^2 \) and \( Q_j = 0 \)  \hspace{1cm} (12)

Model MC2: \( \mathcal{M}_j^2 = \alpha_2 \left[ g_m^2 + (\mathbf{m} \cdot \nabla + i \mathbf{m} \cdot \mathbf{k}_j)^2 \right] \)

\[ Q_j = -\alpha_2 g_m^4 \] \hspace{1cm} (13)

with \( \alpha_2 = \alpha_2 / B_0^2 \). Model MC1 corresponds to the lowest-order expansion \( i = 1 \) and has been considered in ref. [14]. Model MC2 also considers the next higher-order term \( i = 2 \). It is only reformulated by completing the square. It introduces an additional parameter \( g_m \), which can be used to tune the magnetic anisotropy. The original parameters are obtained by \( \alpha_2 = 2 \alpha_2 g_m^2 B_0^2 \) and \( \alpha_4 = \alpha_2 B_0^2 \). Model MC1 and Model MC2 can be solved with almost the same computational cost. We follow the numerical approach in ref. [14], see other studies\([15,17]\) for further details. The finite element (FEM) discretization is implemented in the parallel and adaptive finite-element toolbox AMDiS\([18,19]\).

2.3. Minimum Energy Surface (MES)

In order to analyze the influence of \( \mathbf{m} \) in a bulk system, it is not necessary to solve Equation (11). The density of a single crystal without defects and under homogeneous deformations can be expanded as in Equation (5). The deformation is accounted for by deformed reciprocal space vectors \( \mathbf{k}_j = \mathbf{D}_j \mathbf{k}_0 \), with \( \mathbf{D}_j = (\mathbf{D}^{-1})^T \) and \( \mathbf{D} \) the deformation matrix. As a result of symmetry considerations, only deformations parallel and perpendicular to \( \mathbf{m} \) are permitted. The deformation matrix can be written as \( \mathbf{D} = R^T \mathbf{D}^i R \) with \( \mathbf{D} \) a diagonal matrix with \( D_{11} = d_0 \) and \( D_{22} = D_{33} = d_1 \). These values correspond to the deformations perpendicular \((d_1)\) and parallel \((d_0)\) to \( \mathbf{m} \). \( \mathbf{R} \) rotates \( \mathbf{m} \) in the direction of the first eigenvector of \( \mathbf{D} \) with eigenvalue \( d_0 \).

This description is exact for homogeneous deformed single crystals and provides a valid approximation in more general situations\([20]\). For Model MC1, the free energy, Equation (6), simplifies to

\[ \mathcal{F} \left[ \{A_j\}, \{k_j\} \right] = \Omega \left[ \sum_{j=1}^{N} A_j B_0^2 \left( q_j^0 - k_j^2 \right)^2 - \alpha_2 \left( m \cdot k_j \right)^2 \right] \]

(14)

where \( \Omega \) is the volume of the integration domain. The kernel \( w(q = k_0) \) estimates the energy contribution of a single mode defined by \( k_0 \) and \( A_j \). It governs the elastic and magnetic properties of the model. The first term in \( w(q) \) corresponds to the approximation of the excess free energy of classical density functional theory\([21-23]\) and resembles the approximation of a correlation function in the reciprocal space. It is invariant on the orientation of \( q \) and, thus, on the orientation of the crystal structure. The rotational symmetry of the crystal is broken by the magnetization, \( \mathbf{m} \). This is reflected in the second term of \( w(q) \), which depends on the relative orientation of \( \mathbf{m} \), \( \mathbf{q} \), and the coupling strength \( \alpha_2 \).

Model MC2 leads to a similar expression with a modified kernel

\[ w(q) = B_0^2 \left( q_j^0 - q_j^2 \right)^2 + \alpha_2 \left( g_m^2 \mathbf{m} \cdot \mathbf{q} \right)^2 \]  \hspace{1cm} (15)

To visualize the impact of magnetization on the crystal structure, we consider the energy contribution given by \( w(q) \). For a given orientation \( \mathbf{q} \) in the reciprocal space, \( w(q) \) is minimized by adjusting the length of \( \mathbf{q} \). The resulting lengths for all possible \( \mathbf{q} \)'s can be plotted as a surface in reciprocal space, referred to as the minimum energy surface (MES). Figure 1 shows such surfaces for different cases with a color map corresponding to the energy contribution from \( w(q) \). In Figure 1a, the contribution of the first term in \( w(q) \) is shown, corresponding to the case without

![Figure 1. Symmetry breaking due to magnetic interaction. Minimum energy surface (MES) without and with magnetic coupling. The red spheres correspond to the (shortest) \( \mathbf{k}_0 \)'s representing an undeformed BCC crystal. The color indicates the energy contribution in the reciprocal space due to the kernel \( w(q) \). Every mode \( \mathbf{k}_j \) has an energy contribution proportional to \( w(\mathbf{k}_j) \). a) \( \alpha_2 = 0 \), no magnetic coupling. The MES is a monochrome sphere. All \( \mathbf{k}_j \) are on the sphere, and the energy contribution due to \( w(q) \) is independent of the rotation of the crystal. b) Model MC1 and \( \alpha_2 < 0 \). The energy contribution is increased in the direction of \( \mathbf{m}||\langle 100 \rangle \), and the MES is compressed in this direction. c) Model MC2 and \( \alpha_2 > 0 \). The energy contribution also varies with orientation. However, \( q_m = 1/\sqrt{3} \) is chosen such that all the \( \mathbf{k}_j \)'s are still on the MES for \( \mathbf{m} \) in \[001\]. Here the deformation of MES is exaggerated by considering unrealistic large \( \alpha_2 \). 0.75 (Model MC1) and \( \alpha_2 = 0.75 \) (Model MC2) for illustration purposes.]
magnetization. Considering only this part, $\alpha_2 = 0$, a monocrome sphere with radius $q_0$ is obtained. The $k_j$ vectors lie on this sphere, as illustrated for a BCC crystal by the red dots in Figure 1a (see Figure 2 for details), and any rotation of the crystal does not change the energy. This reflects the rotational symmetry of the model inherited from the correlation function. Deformations shift the vectors $k_j$ away from the MES, thus leading to an increase in energy. When considering magnetic coupling, the scenario changes. The energy depends on the relative orientation with respect to $m$. For directions aligned with $m$, $w(q)$ is increased for $\alpha_2 < 0$ in Model MC1, Figure 1b. In addition, the MES is deformed. For $\alpha_2 < 0$, it shrinks in the direction of $m$. The $k_j$ vectors describing a relaxed crystal then do not lie anymore on the MES. This leads to additional effects, such as the tendency of the crystal to deform in order to minimize the energy or in other words, the deviation from the MES leads to magnetostriction. We note that this effect is relatively small. As the $k_j$ vectors cannot vary independently, they cannot always lie on the MES, as seen in Figure 1b, where the effect is exaggerated with large values of $\alpha_2$ for illustration purposes. In Model MC2, the MES is also deformed, see Figure 1c. But, $q_m = 1/\sqrt{2}$ is chosen such that all the $k_j$’s are still on the MES for $m$ in [001] direction. That is, no deformation is induced. However, the model features magnetic anisotropy, which can be noted from the shape of MES deviating from a sphere, and the color map illustrating the energy contributions, which vary on the MES. With all the information it conveys, the MES can then be used as a suitable tool to study the impact of magnetic coupling.

3. Analysis and Simulation

3.1. Magnetic Properties

We consider Equation (14) to calculate the free energy of a single crystal. We minimize the free energy with respect to $\{A_j\}$ and compute the deformation along and perpendicular to $m$, that is, we determine the eigenvalues of the deformation matrix, $d_0$ and $d_1$, see another study. This deformation defines magnetostriction. The energy dependence on the direction of $m$ defines the magnetic anisotropy. The direction with the lowest and highest energy are called easy and hard directions of magnetization, respectively. BCC and FCC crystals are considered.

3.1.1. BCC Crystals

For BCC crystals, the structure-dependent part in Equation (7) reads

$$f_{\text{BCC}}(\{A_j\}) = -2t(A_1^2A_2A_4 + A_2^2A_1A_3 + A_3^2A_4A_6 + A_4^2A_1^2A_6 + \text{c.c.})$$
$$+ 6\nu(A_1A_3^2A_4^2 + A_2A_1^2A_3^2 + A_3A_1^2A_2^2 + A_1A_2A_3^2 + \text{c.c.})$$

(16)

with $k_j$ vectors defined as, see, for example

$$\{k_j\} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 1 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \begin{bmatrix} 0 \\ 1 \end{bmatrix} \begin{bmatrix} 0 \\ -1 \end{bmatrix} \begin{bmatrix} -1 \\ 0 \end{bmatrix} \begin{bmatrix} 0 & 1 \end{bmatrix}$$

(17)

where we have used $q_0 = 1$. They are numbered from $j = 1, \ldots, 6$ and shown in Figure 2(left).

In Figure 3 we compare Model MC1 and Model MC2 for $m$ in [001] direction and different $\alpha_2$. The free energy is minimized in terms of amplitude $\{A_j\}$ and deformation $D$. Figure 3a,b shows Model MC1 and in panel a) are shown the amplitudes $\{A_j\}$ as a function of $\alpha_2$ while in panel b the deformations along and perpendicular to $m$, $d_0$ and $d_1$, are shown for different $\alpha_2$. Figure 3c,d shows the same quantities for Model MC2 with $q_m = 1/\sqrt{2}$. For $\alpha_2 = 0$, all amplitudes are equal, and there is no deformation. However, with magnetic coupling, differences occur. The amplitudes $A_j$ depend on $\alpha_2$ according to the relative orientation of the corresponding $k_j$ vectors to $m$. The influence is less pronounced for those amplitudes whose $k_j$ are perpendicular to $m$ as $m \cdot k_j = 0$. Therefore, the MES is not influenced by magnetic coupling in these cases. All other contributions equally behave. This behavior is qualitatively the same for Model MC1 and Model MC2, but with opposite signs. Differences between the models are found in the deformation. While Model MC1 leads to an expansion in the $m$ direction for negative $\alpha_2$ and the opposite behavior perpendicular to $m$, there is no deformation in Model MC2. This difference is due to the double-well structure of $w(q)$ in Model MC2 and the choice of $q_m = 1/\sqrt{2}$. The parameter $q_m$ is chosen so that $k_j$ is on the MES regardless of $\alpha_2$. Therefore, there is no induced deformation ($d_0 = d_1 = 1$) for $k_j$. As a result, Model MC2 allows for decoupling magnetic anisotropy and magnetostriction effects and allows these properties to be tuned through $q_m$ and $\alpha_2$. The influence of magnetization aligned along the [001] direction of the grain is shown in Figure 3. This analysis can be repeated for other magnetization directions. Then the magnetic anisotropy can be calculated by comparing the free energy for various magnetization directions. Figure 4 and 5 show the magnetic anisotropy of the system for different values of $\alpha_2$ using Model MC1 and Model MC2, respectively. For Model MC1 the hard directions are always the (100) directions. The easy directions are (111). Only for positive $\alpha_2$, (110) directions become energetically comparable to (111). However, we can conclude that for BCC crystals, the hard direction of magnetization cannot be controlled by Model MC1. This changes for Model MC2.
With the considered set of parameters, for $\alpha_2 > 0$, the easy directions of magnetization are \langle 100 \rangle, and the hard directions are \langle 111 \rangle.

### 3.1.2. FCC Crystals

For FCC crystals, the structure-dependent part in Equation (7) reads

$$ f_{\text{FCC}}(\{A_j\}) = -2I[A_1^2A_5^2 + A_1^2A_7 + A_2^2A_6^2] $$

$$ + A_2^2(A_1^2A_6^2 + A_2^2A_7^2) + A_1^2A_2^2 + c.c. $$

$$ + 6I[A_1^2A_1^2A_3^2 + A_2^2A_6^2A_7^2] $$

$$ + A_1A_2A_3A_5A_7 + A_1A_2A_5A_6 + A_2A_3A_5A_6 + A_2A_3A_4A_6 + A_4A_5A_6 + c.c. $$

(18)
The phase of the complex amplitudes in the grain is given by

$$\phi_g = \frac{1}{\sqrt{3}} \begin{pmatrix} -1 \\ 1 \\ 1 \\ 1 \\ -1 \\ -1 \\ 2 \\ 0 \\ 2 \\ 0 \\ 0 \\ 2 \end{pmatrix}$$

(19)

with \( q_0 = \{1, \frac{2}{3}\} \) for the different sets of \( k_j \) vectors in Model MC1 and analogously in Model MC2. Model MC2 now allows us to tune the parameter \( g_m \) independently for the different sets of \( k_j \) vectors. We consider \( g_m = \{1/\sqrt{3}, 1/\sqrt{2}\} \) for \( k_{1-4} \) and \( k_{5-7} \), respectively. With these modifications the same analysis as for BCC crystals can be done. Figure 6 and 7 show the results concerning magnetic anisotropy for Model MC1 and Model MC2, respectively. For FCC, Model MC1 describes the easy directions always aligned to the \( \langle 100 \rangle \) directions. The hard directions are \( \langle 111 \rangle \). Only for highly positive \( \alpha_2 \) \( \langle 110 \rangle \) becomes energetically comparable to \( \langle 111 \rangle \). However, we can conclude that for FCC crystals the easy direction of magnetization cannot be controlled by Model MC1. As for BCC, this changes for Model MC2. With the considered set of parameters for \( \alpha_2 > 0 \), the easy directions of magnetization are \( \langle 111 \rangle \) and the hard directions are \( \langle 100 \rangle \).

3.1.3. Magnetic Anisotropy of Ferromagnetic Materials

Tuning the easy and hard directions of magnetization becomes necessary as the magnetic anisotropy of various ferromagnetic materials features \( \langle 100 \rangle \) or \( \langle 111 \rangle \) the easy direction of magnetization for BCC and FCC crystals, respectively. Model MC1 does not allow for such versatility. Thus, to account for the proper easy and hard directions of magnetization, Model MC2 has to be considered. Table 1 shows such directions for Fe, Ni, and Co.

These properties also hold for various alloys, for example, Fe_{1-x}Ga_{x}. PFC and APFC models for alloys have been introduced in ref. [29], and magnetic coupling for these models has been introduced in ref. [5]. The same modifications suggested for Model MC2 can also be applied to these models.

3.2. Impact of Magnetization on Grain Growth

We study the effect of magnetization on the shrinkage of a relatively small, initially spherical grain with BCC crystal structure and Model MC2 parameters \( (\alpha = 0.1 \text{ and } g_m = 1/\sqrt{2}) \) that exhibits basic magnetic properties similar to those of iron. The grain has a radius of 60\( \pi \) and is rotated by 5° around the \( \langle 011 \rangle \) direction with respect to the surrounding matrix, as shown in Figure 8a. The phase of the complex amplitudes in the grain is given by taking

$$\alpha_2 = -0.05$$

\( \alpha_2 = 0.05 \)

and \( \alpha_2 = 0.1 \).
where \(\delta k(\theta) \cdot r\), where \(\delta k(\theta)\) is the difference between the rotated and unrotated wave vectors (see other studies\(^{12,17,30,31}\) for more information). In the matrix, the phase is zero.

Figure 8b illustrates the cubic unit cell. The rotational axis [011] coincides with the grain and the matrix, while magnetizations are defined with respect to the crystallographic axes of the matrix. For magnetization oriented along the [011], [001], and [011] directions, the free energy of the matrix and grain, computed as reported in the previous sections, varies as illustrated in Figure 8c. For the matrix, there are energy maxima

\[
\begin{align*}
\delta k(\theta) \cdot r &= \delta k(\theta) \cdot \hat{r} \\
\hat{r} &= \text{unrotated wave vector} \\
\delta k(\theta) &= \text{rotated wave vector}
\end{align*}
\]
for \( \mathbf{m} \) oriented along \((011)\) directions and energy minima for \( \mathbf{m} \) oriented along \((001)\) directions. For the rotated grain, magnetization in these directions results in slightly shifted easy and hard directions, owing to the (small) rotation of its crystallographic axes. Also, the energy difference between grain and matrix varies with the orientation of \( \mathbf{m} \). We select orientations of \( \mathbf{m} \), which maximize this difference. In particular, we consider \( \mathbf{m} \) between \([100] \) and \([011] \), \((\mathbf{m}_1)\), for which the crystal structure in the grain is energetically favored, and between \([\overline{0}11] \) and \([001] \), \((\mathbf{m}_2)\), for which the crystal structure in the matrix is favored, see Figure 8c.

Table 1. Material-specific magnetic anisotropies.

| Material | Crystal Structure | Easy Directions | Hard Directions | References |
|----------|-------------------|-----------------|-----------------|------------|
| Fe       | BCC               | \((100)\)       | \((111)\)       | [35]       |
| Ni       | FCC               | \((111)\)       | \((100)\)       | [35,36]    |
| Co       | FCC               | \((111)\)       | \((100)\)       | [35]       |

No preference for grain or matrix is achieved by choosing \( \mathbf{m} \) along the axis of rotation of the grain, \([011] \), \( \mathbf{m}_0 \), which will be considered for comparison.

Figure 9 illustrates the grain shrinkage for magnetization \( \mathbf{m}_0 \). In particular, the dislocation network formed between grain and matrix is shown. This is obtained by exploiting the decrease of amplitudes at defects. We consider here regions with \( A^2 < 0.083 \). The dislocation network shrinks anisotropically, as it has been observed in previous studies. For \( \mathbf{m}_0 \), \( \mathbf{m}_1 \), and \( \mathbf{m}_2 \), shrinking grains with very similar dislocation networks (as in Figure 9) are obtained, indicating negligible effects on the fine details of the dislocation network structure. This can be attributed to the unchanged incommensurability of the crystals in the grain and the matrix. Similar topological defects form, and small changes in the elastic interactions caused by the magnetostrictive force are obtained for the considered model parameters and magnetization direction. Qualitatively different results are indeed obtained if considering Model MC1, resulting however from an unphysically large magnetostriction and with easy and hard directions that cannot be tuned. Importantly,

![Figure 8](image-url)  
Figure 8. Setup for magnetic APFC simulation of grain shrinkage. a) Schematic of a spherical grain rotated about the \([011] \) axis (grain) and embedded in an unrotated crystal (matrix). b) Illustration of the crystal’s unit cell: the \([011] \) direction of both the grain and matrix is aligned with the axis of rotation. The direction of magnetization is defined according to the crystallographic orientation of the matrix. c) Magnetic anisotropy of matrix (blue squares) and grain (orange circles) along the lines defined by the \([0\overline{1}1] \), \([001] \), and \([011] \). \( \mathbf{m}_1 \) and \( \mathbf{m}_2 \) point at specific magnetizations considered in simulations, favoring the crystal structure in the grain and the matrix, respectively, while maximizing the corresponding energy difference.

![Figure 9](image-url)  
Figure 9. Defect networks of an initially spherical grain in a BCC crystal during shrinkage. A–F) The structures at representative times, also indicated in Figure 10. The magnetization is aligned with the axis of rotation \((\mathbf{m}_0)\). Measures of the grain extension, \( a_{0,2} \) are introduced to characterize its shape, corresponding to the axes of an ellipsoid approximating the grain–matrix interface extension. \( a_0 \) is parallel to the rotation axis, \([011] \), see Figure 8. \( a_{1,2} \) lies in the \((101) \) plane.
the shrinkage speed is largely influenced by the orientation of \( \mathbf{m} \). This is because magnetic anisotropy introduces an additional driving force related to the differences in bulk energy when moving across the grain boundary between grain and matrix.

A detailed analysis of this evidence is reported in Figure 10. Figure 10a shows the energy decay relative to the initial energy during grain shrinkage. Constant energy is obtained when the grain vanishes, at a time here referred to as vanishing time \( t_v \), which is found to depend on magnetization. The slowest shrinkage is achieved if the grain is energetically preferred, \( m_1 \). Here the additional driving force caused by magnetization tends to favor the crystal structure in the grain, opposing the shrinkage dynamics (well described by mean-curvature flow\(^{(32)}\)). The energy gain during shrinkage is the lowest. Consistently, the highest shrinkage speed and energy gain are observed when the grain has higher energy, \( m_2 \). Here the driving force introduced by magnetization enhances the underlying shrinkage dynamic favoring the crystal structure in the matrix. If matrix and grain are energetically equivalent, \( m_0 \), the vanishing time is between the two cases. Thus, magnetization enhances or hinders grain shrinkage according to its direction.

More details on the structure of the evolving dislocation network are reported in Figure 10b,c. The initially spherical small-angle (semicoherent) grain boundary is approximated as the surface of a 3D ellipsoid interpolating the dislocation network, whose axes are along \( x, y, \) and \( z \)-direction are denoted as \( a_{0,1,2} \). The corresponding surface area is computed by the so-called Knud Thomsen's formula\(^{(31)}\)

\[
S = \frac{\pi}{3^{1/3}} \left( (a_0 a_1)^p + (a_1 a_2)^p + (a_0 a_2)^p \right)^{1/p}
\]

with \( p = 1.6075 \). This quantity normalized by the surface area of the initial spherical grain as well as the axes \( a_{0,1,2} \) are reported in Figure 10b,c, respectively, against time normalized by \( t_v \). For all cases considered, the grain boundary between grain and matrix decreases nearly linearly (Figure 10b). Such a linear scaling, as well as a linear decrease in energy, is predicted by the classical theory of grain shrinkage driven by mean curvature flow\(^{(32)}\) and it is reproduced by PFC and APFC without magnetic interaction\(^{(30,31)}\). This indicates that evolution is still mainly governed by the minimization of interface energy, owing to the relatively small size of the grain and, thus, the relatively large mean curvature of the (small-angle) grain boundary between grain and matrix. Deviations are expected for larger systems\(^{(33)}\).

Nevertheless, the magnetic anisotropy may enhance or hinder evolution with a volumetric energy contribution that acts isotropically on the grain boundary and its dislocation network. This is further shown in Figure 10c where the evolution of the axes \( a_{0,1,2} \) are shown. \( a_0 \) shrinks nearly linearly with a constant speed, up to the end of the shrinking process, \( t/t_v > 0.9 \). A more complex evolution is observed for \( a_1 \) and \( a_2 \) due to defect annihilation. In particular, in the interval \( 0.6 < t/t_v < 0.8 \) indicated by C–E in Figure 9, dislocation lines vanish, and sudden changes in the dislocation network morphology occur. This stage can also be detected in the decay of energy and grain boundary surface, where the decay rate deviates the most from linear.

**Figure 10.** Influence of magnetization on grain shrinkage. a) Energy decay during shrinkage. Eventually, the energy becomes constant when the grain disappears. The grain vanishes at different times \( t_v \) depending on the magnetization direction. b) Decrease in the extension of the interface between grain and matrix, as approximated by Equation (21) and normalized by its initial value. The time scale is normalized by the vanishing time \( t_v \). c) Decrease over time of \( a_{0,1,2} \) (as defined in Figure 9).
scaling and could be ascribed to additional elasticity effects enhanced by short-range dislocation interaction. However, it has been found that the magnetization has a negligible effect on the anisotropy during the shrinkage of the grain as the values of \(a_{0,1,2}\) vary only slightly among the different chosen orientations of \(\mathbf{m}\).

4. Conclusion

We have reviewed and extended magnetic PFC and APFC models. Our focus has been on the control of magnetic anisotropy in these models. For various ferromagnetic materials, the easy direction of magnetization is [001] for BCC crystals and [111] for FCC crystals. Modeling this behavior requires an extension of magnetic couplings in existing models. By analyzing the MES, we explored the possibility of tuning the easy and hard directions of magnetization by including higher-order coupling terms. This can be achieved without increasing the complexity of the model significantly. The numerical realization only requires directional derivatives of order four and thus does not increase the order of the derivatives in the equations. The higher-order terms describe a double well in the direction of magnetization in reciprocal space. Therefore, the local extrema of the double scale \(q_m\). A different approach to decoupling both phenomena is addressed in ref. [34].

The model has been applied to the simulation of the shrinkage of a spherical grain in a matrix under the influence of a constant magnetic field and using the basic magnetic properties of Fe. Such a shrinkage is anisotropic and can be enhanced or hindered by magnetization. However, the details of the magnetic coupling influence the grain shrinkage only slightly in terms of the morphologies of dislocation networks and scaling laws. In this case, the curvature of the grain boundary between grain and matrix is dominant due to the small grain size.

Acknowledgements

A.V. and R.B. acknowledge support by the German Research Foundation (DFG) within SPP1959 under grant no. VO899/20-2. M.S. acknowledges support from the Emmy Noether Programme of the German Research Foundation (DFG) under grant no. SA4032/2-1. The authors further acknowledge computing resources provided at the Center for Information Services and HPC (ZIH) - TU Dresden and Jülich Supercomputing Center under grant PFAMDIS.

Open Access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

Fe, grain boundaries, magnetic anisotropy, modeling, phase field crystals

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