1. Introduction

Utilizing microscopic crystal properties on the macroscopic level is straightforward in concept while many issues needs to be overcome to do so in practice since materials is usually composed of multi-variants and multi-grains where the microscopic crystal properties are often canceled out and vanished. In order for the microscopic crystal properties to prevail on the macroscopic level, understanding of microstructure evolution is essentially needed.

There has been of great interest in L1 0 type ferromagnetic alloy since it is a prime candidate for the magnetic recording media due to its high magnetic crystalline anisotropy, which is one of the essential properties needed for high density magnetic storage.1) Therefore, many studies on FePd have been conducted so far.2–4) Among L1 0 type ferromagnetic alloys, FePt has the highest magnetic crystalline anisotropy,5) which would increase the information density in the magnetic storage. Meanwhile, the external field required for writing data on the FePt storage media is higher, which impedes the practical use of FePt for efficient recording. Then, since FePd requires smaller switching field it became the prime candidate for the material of high density magnetic storage.

FePd transforms its crystal structure from FCC to L1 0 at about 923 K when the temperature is lowered. The L1 0 FePd has a shorter c-axis than the a-axes of FCC FePd. In this transition, three variants having the shorter c-axis along one of three Cartesian axes are formed in L1 0 FePd from a single-variant single-crystal of FCC FePd. Without any external field, the formation probabilities of three variants are equal to each other, leading to a multi variant structure within a grain. This is a negative outcome for the application as the magnetic storage since the anisotropy would be canceled out when the multi variant structure forms. Many techniques to obtain the single variant structure have been reported thus far.6–9) Those studies demonstrated that the application of uniaxial external field, such as magnetic field or stress field, successfully produced the single variant FePd structure. However, the exact mechanism of the single variant formation remains unclear since it is very difficult to observe the process with high temporal and spatial resolutions. This is an impediment to optimize the process, and, in turn, the overall macroscopic property of FePd.

Phase field modeling10) is an attractive method to monitor and examine the time evolution of microstructure of materials under any circumstance specified. Calculations of numerous kinds of materials microstructures have been reported after the first calculation by Kobayashi.10) This method can easily incorporate the contributions of various energies from any origin and calculate their effects on microstructures.

This paper attempts to clarify the effect of external magnetic field on the formation of single variant of FePd as a...
first step. This is prerequisite for designing material processing to acquire desired properties of materials. Thus far, several phase field models for FePd have been reported.\textsuperscript{11,12) Elastic energy based on micromechanics and magnetic energy based on micromagnetics have been introduced to phase field model. In this study, we focused on the effect of magnetic crystalline anisotropy energy. Chemical free energy, interface energy, and magnetic crystalline anisotropy energy were taken into account in our model. This expression of the energy enables the calculations that intentionally exclude the effects of other energies, thereby focusing on the influence of those energy terms included, which is quite difficult to do by means of practical experiments. Conducting calculations under various conditions as computational experiments would reveal the detailed effect on the single variant formation.

2. Phase Field Modeling

We have adopted multi phase field model\textsuperscript{13)} to calculate the microstructures. This model enables us to calculate microstructure evolutions from the free energy of the materials. The time evolution equation of the phase field model is

$$\frac{\partial \phi_i}{\partial t} = M \left( \nabla \cdot \phi_i \frac{\partial f}{\partial \phi_i} - \frac{\partial f}{\partial \phi_i} \right)$$

where $\phi_i$ is phase field, $M$ is mobility, $f$ is free energy per unit volume. The subscript $i$ indicates the state of FePd: 0 corresponds to FCC FePd, 1, 2 or 3 corresponds to L1\textsubscript{0} FePd that has c-axis parallel to x-, y-, or z-axis, respectively. Hereafter the L1\textsubscript{0} FePd of which easy axis of magnetization is parallel to x-, y-, or z-axis is referred to x-, y-, or z-variant respectively. For example, when a variant FePd is specified by the phase field of 1 indicating it is x-variant, the value of $\phi_i$ is 0 outside of the variant and 0<$\phi_i$<1 at the variant boundary. In this study, chemical free energy, chemical inter-face energy $f_{\text{interface}}$ and magnetic crystalline anisotropy energy $f_{\text{magnetic}}$ were taken into account. These energies were calculated by following equations respectively:

$$f_{\text{chemical}} = \left[ 1 - \sum_{i=1}^{3} h(\phi_i) \right] f_0 + \sum_{i=1}^{3} h(\phi_i)(f_i + f_{\text{magnetic}})$$

$$f_{\text{interface}} = \sum_{i=1}^{3} \left( \phi_i \nabla \phi_i - \phi_i \nabla \phi_i \right)^2$$

$$+ \frac{\epsilon_i^2}{2} \sum_{j(i,\neq j)} \left( \phi_i \nabla \phi_j - \phi_j \nabla \phi_i \right)^2$$

$$+ w_i \phi_i^0 \sum_{j=1}^{3} \phi_j^1 + \frac{W_i}{2} \sum_{j=1}^{3} \phi_j^2$$

$$f_{\text{magnetic}} = - \frac{1}{2} \sum_{k=1}^{3} \mathcal{X}_d H_k^2$$

respectively, $\mathcal{X}_d$ is the magnetic susceptibility parallel to k direction of phase $i$, $H_k$ is the external magnetic field parallel to k direction. In our calculation condition, $e_0$ and $w_0$ are zeros, $e_i$ and $w_i$ are calculated as

$$e_i^2 = \frac{3}{2.2} \lambda \sigma_{i\gamma}$$

and

$$w_i = 6.6 \frac{\sigma_{i\gamma}}{\lambda}$$

respectively (see appendix for details). $\lambda$ is the width of interface, $\sigma_{i\gamma}$ is the interface energy between variants. The magnetic susceptibility parallel to easy axis of magnetization $\chi_{\gamma}$ was derived as

$$\chi_{\gamma} = \frac{C}{T - T_c} = \frac{N m^2}{3 k_b (T - T_c)}$$

where C is the curie constant, T is temperature, $T_c$ is the curie temperature, $N$ is the number of atoms per unit volume, $m$ is the magnetic moment of an atom, $k_b$ is Boltzmann constant.\textsuperscript{14) We assumed that the magnetic susceptibility parallel to hard axis of magnetization $\chi_{\lambda}$ is a tenth of $\chi_{\gamma}$. Parameters of material’s properties used in the calculations are summarized in Table 1. Lattice constant and molar volume of FePd are needed to calculate the number of atoms per unit volume. As for the mobilities for the phase field in Eq. (1), since their values are not known, they were set as 1 m/s. This assumption only brings about a change in time scale: Relative time is still meaningful. Phase, temperature and composition were initialized as a disordered state with random noise whose value is smaller than 2.5x10\textsuperscript{-3}, i.e., 50 K below the order-disorder transition temperature and equiatomic, respectively. Then, computation has been carried out by assuming that temperature was spatially uniform and chemical composition remains at initial state. Therefore only the distribution of phases was calculated at every time steps. In our computation, calculation area is 2.0 \textmu m x 2.0 \textmu m, mesh spacing is 10.0 nm x 10.0 nm, $\lambda$ is 50.0 nm and time step is 0.1 ns.

3. Results and Discussions

First of all, calculations were carried out under no magnetic field and evolution of phase profile and the volume fractions of variants were monitored. One of the results is shown in Fig. 1 and Fig. 2, where y-variant happened to emerge more than other variants at 10\textsuperscript{5} time steps. This is originated from not completely uniform initial states of the disordered phase. The average and standard deviation of the volume fractions at 999 900 step of ten calculations with statistically different initial states are shown in Table 2. It is confirmed that these variations are within statistical error.

Secondly, time dependence of volume fractions of variants and spatial phase profile were calculated under magnetic field of 5 T in x-direction. One of the results is shown in Fig. 3 and Fig. 4. At this magnetic field, magnetic crystalline anisotropy energy is as small as about 0.028% of the latent heat of order-disorder transition. The x-variant of which easy axis of magnetization is parallel to external magnetic field formed much more than other variants, sug-
suggesting that the magnetic crystalline anisotropy energy has large enough effect for variant selection as it is expected.

To reveal in more detail about the effectiveness of the magnetic field, extra calculations under magnetic field of 5 T for only 1 000 steps at various stages were carried out. Selected results are shown in Fig. 5. In the case of Fig. 5(a), magnetic field was applied from 0 to 1 000 time step and no magnetic field is applied from 1 000 to 106 time steps. Although the volume fraction of favorable variant was smaller than Fig. 3, it was much larger than statistical error. The average and standard deviation of the volume fractions at 999 900 step of ten calculations are shown in Table 3. It is confirmed from ten sets of calculations to achieve higher statistical accuracy that the volume fraction of favorable variant is larger than statistical error. Focusing on Figs. 5(b) and 5(c), it is found that the volume fraction of magnetically favorable variant is much smaller than in Fig. 5(a), smaller than the statistical error. Thus, for variant selection, it is much more effective to apply magnetic field at the initial stage of microstructure evolution than latter stages: Application of magnetic field only at latter stage exhibit little effect which is surpassed by variant coarsening driven by interface energy.

For better understanding these phenomena, we focused...
on the volume fraction at the time step when ordering is completed since Fig. 5(a) is done under exactly the same condition of calculation as Fig. 1 after 1 000 steps from the beginning. The magnified profile of volume fraction of variants is compared in Fig. 6. It is found that the volume fraction of the $x$-variant which dominates at the end of calculation has already been larger than the ones of other variants just at the end of the order–disorder transition. Since the free energy in this study includes the only three kinds of energy, i.e., chemical free energy, interface energy and magnetic crystalline anisotropy energy, interface energy is the only energy that affects the variant selection after completion of ordering when magnetic field has removed. It is expected that the variant that has smaller interface energy per unit volume evolves more than other variants at the later stages. As exemplified in Figs. 5(b) and 5(c), magnetic energy is not effective at later stages once variant coarsening governs overall microstructural evolution. Therefore, the small difference in the volume fraction of $x$-variant at the end of order–disorder transition shown in Fig. 6(b) would determine overall dominance of a variant at the end.

This indicates that, for variant selection, magnetic field is needed only at the early stage just to decrease interface energy per unit volume of the favorable variant relative to other variants by increasing its volume fraction slightly.

<table>
<thead>
<tr>
<th>Variant</th>
<th>Average Volume Fraction</th>
<th>Standard Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x$-variant</td>
<td>0.32</td>
<td>0.02</td>
</tr>
<tr>
<td>$y$-variant</td>
<td>0.36</td>
<td>0.03</td>
</tr>
<tr>
<td>$z$-variant</td>
<td>0.32</td>
<td>0.04</td>
</tr>
</tbody>
</table>

Fig. 3. Calculated volume fractions of variants as a function of time step under magnetic field of 5 T. Solid line, chain double-dashed line and broken line indicate $x$-, $y$- and $z$-variant, respectively. It is found that the $x$-variant, which is magnetically favorable, is formed more than other variants at 100 step though volume fractions of variant are almost equal to each other at 1000 step.

Fig. 4. Time evolution of phase profile under magnetic field of 5 T parallel to $x$-axis. The $x$-variant of which easy axis of magnetization is parallel to external magnetic field formed much more than other variants as it is expected.

Fig. 5. Volume fractions of three variants. (a) When magnetic field of 5 T parallel to $x$-axis was applied for 1000 steps from the beginning and no magnetic field was applied after 1000 step, the $x$-variant, which is magnetically favorable, was formed more than other variants. This difference is more larger than statistical error. (b) When magnetic field of 5 T parallel to $x$-axis was applied for 1000 steps from 1000 step and no magnetic field was applied other time steps, the $x$-variant was formed more than other variants. But this difference is smaller than statistical error. (c) When magnetic field of 5 T parallel to $x$-axis was applied for 10000 steps from 10000 step and no magnetic field was applied other time steps, Three variants are formed almost evenly.
Table 3. Averages and standard deviations of the volume fractions at 999,900 steps of ten calculations under no magnetic field of 5 T until ordering is completed.

<table>
<thead>
<tr>
<th></th>
<th>x-variant</th>
<th>y-variant</th>
<th>z-variant</th>
</tr>
</thead>
<tbody>
<tr>
<td>average</td>
<td>0.48</td>
<td>0.31</td>
<td>0.21</td>
</tr>
<tr>
<td>standard deviation</td>
<td>0.08</td>
<td>0.07</td>
<td>0.06</td>
</tr>
</tbody>
</table>

Fig. 6. Magnified figure around 1 000 time step of time evolution of volume fractions of variants. (a) Under no magnetic field, the y-variant is formed slightly more than other variants. (b) Under magnetic field of 5 T from 0 step to 1 000 step, the x-variant, which is magnetically favorable, is formed more than other variants. This difference is larger than the difference in the case of no magnetic field, which leads the dominance of x-variant in latter part of calculation.

Appendix

In this appendix, derivation of Eq. (5) and Eq. (6) are shown. Firstly, phase profile at equilibrium state is calculated. Secondly, interface thickness is represented by $\varepsilon_i$ and $w_i$. Thirdly, interface energy is represented by $\varepsilon_i$ and $w_i$. Finally, Eq. (5) and Eq. (6) are derived. [JSCES, 15 (2010) 2, 7]

1. Phase Profile

Let me consider a system consisting of two kinds of phases, represented by $\phi_1$ and $\phi_2$, under no magnetic field and steady state condition, that is $\lim_{t \to \infty} \phi(x) = 1$, $\lim_{t \to \infty} \phi(x) = 0$ and $\partial \phi(x)/\partial t = 0$. In this condition, free energy density is derived from Eq. (2) and Eq. (3) as

$$ f = \varepsilon_i^2 [(1 - \phi_1) \nabla \phi_1 - \phi_1 \nabla (1 - \phi_1)]^2 + w_i (1 - \phi_1)^2 \phi_1^2 + h(\phi_1) f_1 + h(1 - \phi_1) f_2 \quad \ldots \ldots \ldots \ldots \ldots (8) $$

where $\phi_1 + \phi_2 = 1$. In this study, this equation is expressed as

$$ f = \varepsilon_i^2 (\nabla \phi_1)^2 + w_i (1 - \phi_1)^2 \phi_1^2 + f_1 \quad \ldots \ldots \ldots \ldots \ldots (9) $$

by using the equations $h(\phi_1) + h(1 - \phi_1) = 1$ and $f_1 = f_2$. In the case of one-dimensional calculation under steady state condition, by substituting Eq. (9) into Eq. (1), we get

more than others, and then variant selection driven by interface energy spontaneously proceeds at the later stage, resulting in dominance of the favorable variant.

4. Conclusions

The calculation of variants evolution in FePd during and after order–disorder transition has been conducted by phase field model with the presence or absence of external magnetic field as a variable. Under no magnetic field, three variants formed almost evenly. Under magnetic field of 5 T, the variant of which easy axis of magnetization is parallel to external magnetic field prevailed in good agreements with experiments. Then, extra computational experiments under external magnetic field prevailed in good agreements with experiments.

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where \( g(\phi_i) = (1-\phi_i)^2 \phi_i \). By multiplying both sides of the equation by \( d\phi_i/dx \) and integrating both sides from \(-\infty\) to \( x \), we obtain

\[
2\varepsilon_1^2 \int_{-\infty}^{x} d^2\phi_i = w_1 \int_{-\infty}^{x} d\phi_i \quad \text{.................}(10)
\]

where \( g(\phi_i) = (1-\phi_i)^2 \phi_i \). By multiplying both sides of the equation by \( d\phi_i/dx \) and integrating both sides from \(-\infty\) to \( x \), we obtain

\[
2\varepsilon_1^2 \int_{-\infty}^{x} d^2\phi_i \phi_i \frac{d\phi_i}{dx} \cdot dx = w_1 \int_{-\infty}^{x} d\phi_i \quad \text{.................}(11)
\]

and integrating both sides from \(-\infty\) to \( x \), we obtain

\[
\varepsilon_1^2 \left( \frac{d\phi_i}{dx} \right)^2 = w_1 \phi_i^2 (1-\phi_i)^2 \quad \text{.................}(12)
\]

using partial integration. As \( \phi_i \) is a monotonically decreasing function, \( d\phi_i/dx < 0 \) is satisfied. Therefore Eq. (12) is rewritten as

\[
\varepsilon_1 \left( \frac{d\phi_i}{dx} \right) = -w_1 \phi_i (1-\phi_i) \quad \text{.................}(13)
\]

\[
\int \left( \frac{1}{\phi_i} - \frac{1}{1-\phi_i} \right) d\phi_i = \frac{-\sqrt{w_1}}{\varepsilon_1} \int_{-\infty}^{x} dx \quad \text{.................}(14)
\]

\[
\phi_i = C \frac{\exp \left( -\sqrt{w_1} \frac{x}{\varepsilon_1} \right)}{1 + \exp \left( -\sqrt{w_1} \frac{x}{\varepsilon_1} \right)} \quad \text{.................}(15)
\]

where \( C \) is an integral constant. By using boundary condition, \( \lim_{x \to -\infty} \phi_i(x) = 1 \), \( C = 1 \) is derived. Finally, \( \phi_i \) is expressed as

\[
\phi_i = 1 - \frac{1}{1 + \exp \left( -\sqrt{w_1} \frac{x}{\varepsilon_1} \right)} \quad \text{.................}(16)
\]

\[
\phi_i = \frac{1}{2} \left[ \exp \left( -\sqrt{w_1} \frac{x}{2\varepsilon_1} \right) + \exp \left( -\sqrt{w_1} \frac{x}{2\varepsilon_1} \right) \right] \quad \text{.................}(17)
\]

\[
\phi_i = \frac{1}{2} \left[ \exp \left( -\sqrt{w_1} \frac{x}{2\varepsilon_1} \right) - \exp \left( -\sqrt{w_1} \frac{x}{2\varepsilon_1} \right) \right] \quad \text{.................}(18)
\]

\[
\phi_i = \frac{1}{2} \left[ 1 - \tanh \frac{-\sqrt{w_1}}{2\varepsilon_1} \right] \quad \text{.................}(19)
\]

2. Interface Thickness

We define interface region as the region at \( 0.1 < \phi_i < 0.9 \). From Eq. (18), we obtain

\[
0.9 = \frac{1}{2} \left( 1 - \tanh \frac{-\sqrt{w_1}}{2\varepsilon_1} \right) \quad \text{.................}(19)
\]

\[
0.1 = \frac{1}{2} \left( 1 + \tanh \frac{-\sqrt{w_1}}{2\varepsilon_1} \right) \quad \text{.................}(20)
\]

By subtracting Eq. (20) from Eq. (19), we obtain

\[
0.8 = \tanh \frac{-\sqrt{w_1}}{2\varepsilon_1} \quad \text{.................}(21)
\]

\[
\lambda = \frac{2\varepsilon_1}{\sqrt{w_1}} \tanh \frac{-\sqrt{w_1}}{2\varepsilon_1} \quad \text{.................}(22)
\]

3. Interface Energy

Interface energy can be obtained by integrating interface energy defined in this study. Therefore \( \sigma_{\nu-v} \) is expressed as

\[
\sigma_{\nu-v} = \int_{-\infty}^{x} \left( \frac{1}{w_1} \beta(\phi_i) + \varepsilon_1 \left( \frac{d\phi_i}{dx} \right)^2 \right) dx \quad \text{.................}(23)
\]

By substituting Eq. (12) to Eq. (23), we obtain

\[
\sigma_{\nu-v} = 2 \int_{-\infty}^{x} \varepsilon_1 \left( \frac{d\phi_i}{dx} \right)^2 dx \quad \text{.................}(24)
\]

With the help of Eq. (18), Eq. (24) become

\[
\sigma_{\nu-v} = \frac{1}{8} w_1 \int_{-\infty}^{x} \frac{1}{\cosh^4 \frac{-\sqrt{w_1}}{2\varepsilon_1} x} dx = \frac{\varepsilon_1 \sqrt{w_1}}{3} \quad \text{.................}(25)
\]

4. The Relationship between \( \varepsilon_1, w_1 \) and \( \sigma_{\nu-v} \)

By multiplying Eq. (22) by Eq. (25), we obtain

\[
\lambda \sigma_{\nu-v} = 1.1 \frac{2}{3} \varepsilon_0 \quad \text{.................}(26)
\]

\[
\varepsilon_0 = \frac{3}{2.2} \lambda \sigma_{\nu-v} \quad \text{.................}(27)
\]

By dividing Eq. (25) by Eq. (22), we obtain

\[
\sigma_{\nu-v} = \frac{w_1}{\lambda} \quad \text{.................}(28)
\]

\[
w_1 = 6.6 \frac{\sigma_{\nu-v}}{\lambda} \quad \text{.................}(29)
\]