Effect of Spatial Distribution of Local Magnetization on Microstructure Formation in $L1_0$ -type Ferromagnetic Alloys under External Magnetic Field

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The effect of spatial distribution of local magnetization vectors on variant selection in $L1_0$ -type ferromagnetic alloys under a magnetic field has been investigated by micromagnetics in conjunction with phase-field modeling. The magnetic energy components, including Zeeman, exchange, anisotropy and dipole-dipole interaction energies resulting from spatial distribution of local magnetization vectors are separately evaluated to clarify which energy component is dominant during microstructure evolution of the alloys. The dependence of the local magnetization vector distribution on the amplitude of the external magnetic field is also carefully examined. It is found that, under an external magnetic field, the anisotropy and Zeeman energies are relatively large and dominant factors affecting the alignment of local magnetization vectors which completely align in the direction of the external field when its amplitude is as high as 6 T. This indicates that the distribution of magnetization is not important for the variant selection in $L1_0$ -type ferromagnetic alloys since it requires a high magnetic field. In fact, microstructure evolution examined using phase-field modeling remains unchanged by taking into account the spatial distribution of local magnetization vectors, suggesting that only the scalar magnitude of the external magnetic field and the uniaxial magnetic anisotropy are sufficient to form the microstructure.

Key words: Micromagnetics, $L1_0$ -type ferromagnetic alloys, Variant selection, Phase Field Modeling

1. INTRODUCTION

$L1_0$ -type ferromagnetic alloys are promising materials for magnetic storage devices due to their high magneto-crystalline anisotropy¹. The macroscopic magnetic anisotropy is determined by not only the microscopic anisotropy but also the microstructure of the material. This type of material exhibits three possible variants depending on the orientation of the c -axis of the $L1_0$ phase upon transformation from FCC to $L1_0$. When multiple variants exist in the resultant microstructure, the microscopic anisotropy is cancelled out on the macroscopic level. Thus single variant structure is desired to improve their macroscopic anisotropy. Extensive experiments have been carried out to realize a single variant structure. There are generally two approaches for doing this. One is to utilize an external stress field². The other is to utilize an external magnetic field^{3,4}. The former is based on the fact that the transformation strain which the FCC to $L1_0$ transformation introduces to the $L1_0$ phase is negative along the c -axis or positive along the a -axis. An external compressive stress field along one direction can induce preferential growth of the variant whose c -axis is along the direction of the external compressive stress. The latter is based on the fact that the c -axis of the $L1_0$ phase is the easy axis of magnetization. Under an external magnetic field, the variants whose easy axis of magnetization is aligned in the direction of the external magnetic field can grow preferentially. In either case,

successful results have been obtained. Nevertheless, our knowledge of the mechanism behind the variant selection is still limited.

Tanaka et al. have clarified that the magnetic or stress field is effective only at the initial stage of phase transformation³. Farjami et al. have found that the fraction of the preferred variant becomes slightly larger than the other variants after annealing under a magnetic field at the initial stage of phase transformation⁴. We have further clarified the mechanism in more detail with the aid of numerical calculations: A slight difference in volume fraction of a preferred variant becomes larger by the interplay between external magnetic field, transformation strain, and the energy components involved^{5,6}. We also clarified the duration for which a magnetic field is effective for variant selection. The details of the magnetic effect are, however, still unclear because a very simple model was used for magnetic energy, taking into account only its scalar magneto-crystalline anisotropy. Furthermore, although the magnetic shape anisotropy plays an important role in determining a material's magnetic properties, for example, modifying the shape of precipitates affects the magnetic properties⁷, the role of each magnetic energy component during microstructure formation is not sufficiently clear for studying variant selection under a magnetic field.

In this study, in order to obtain a more quantitative and detailed understanding of the effect of magnetic

field, the spatial distribution of local magnetization vectors has been examined according to the theory of micromagnetics. Its influence on microstructure formation has also been examined by phase-field modeling. By taking full advantage of the analytical models of micromagnetics, the contribution of each component of magnetic energy is examined both in terms of its modification by the spatial distribution of the local magnetization vectors and the magnitude of the overall magnetic energy.

2. METHODS

2.1 Micromagnetics calculation

Time evolution of the magnetization distribution is given by the Landau-Lifshitz-Gilbert (LLG) equation⁸⁻¹¹:

$$\frac{d\vec{m}}{dt} = -\gamma_0 (\vec{m} \times \vec{H}_{\text{eff}}) + \alpha \left(\vec{m} \times \frac{d\vec{m}}{dt} \right) \quad (1)$$

where α , γ_0 , and $\vec{H}_{\text{eff}}$ are the Gilbert damping parameter, the gyromagnetic ratio, and the effective field, respectively. The scaled magnetization vector is given by $\vec{m} = \vec{M}/M_s$, where $\vec{M}$ and M_s are the magnetization vector and saturation magnetization, respectively. Time evolution of the magnetization can be expressed as follows by solving (1) for the time evolution of the magnetization:

$$\left(1 + \alpha^2 \right) \frac{d\vec{m}}{dt} = -\gamma_0 \left[\vec{m} \times \vec{H}_{\text{eff}} + \alpha \vec{m} \times \left(\vec{m} \times \vec{H}_{\text{eff}} \right) \right]. \quad (2)$$

The effective field can be expressed by

$$\vec{H}_{\text{eff}} = -\frac{\delta E_{\text{total}}^{\text{mag}}}{\delta \vec{m}} \quad (3)$$

where $E_{\text{total}}^{\text{mag}}$ is the total magnetic energy. In our study, total magnetic energy is defined as the sum of Zeeman $E_{\text{external}}^{\text{mag}}$, exchange $E_{\text{exchange}}^{\text{mag}}$, anisotropy $E_{\text{aniso}}^{\text{mag}}$ and dipole-dipole interaction $E_{\text{dipole}}^{\text{mag}}$ energies. Each energy component can be written as follows:

$$E_{\text{external}}^{\text{mag}} = -K_d \int \vec{m} \cdot \vec{H}_{\text{external}} d\vec{r} \quad (4)$$

$$E_{\text{exchange}}^{\text{mag}} = -A \int \nabla \vec{m} \cdot \nabla \vec{m} d\vec{r} \quad (5)$$

$$E_{\text{aniso}}^{\text{mag}} = K_u \int \left\{ 1 - (\vec{m} \cdot \vec{e}_{\text{uni_aniso}})^2 \right\} d\vec{r} \quad (6)$$

$$E_{\text{dipole}}^{\text{mag}} = -\frac{1}{2} K_d \int \vec{m} \cdot \vec{H}_d d\vec{r} \quad (7)$$

where $K_d = M_s \mu_0$, with μ_0 being the magnetic permeability of space; $\vec{H}_{\text{external}}$ the amplitude of external magnetic field; A the exchange constant, which is related to the magnetic wall energy; K_u uniaxial anisotropy constant; $\vec{e}_{\text{uni_aniso}}$ the unit vector of the direction of the easy axis of magnetization; and $\vec{H}_d$ the demagnetization field given by

$$\vec{H}_d = -\int \left[\frac{m(\vec{r}')}{|\vec{r} - \vec{r}'|^3} - \frac{3(\vec{r} - \vec{r}') \{ m(\vec{r}') \cdot (\vec{r} - \vec{r}') \}}{|\vec{r} - \vec{r}'|^5} \right] d\vec{r}' \quad (8)$$

Normalized time can be obtained by dividing time by γ_0 . We used $M_s = 0.43$ T, $A = 10^{-11}$ J/m, $\alpha = 0.02$ and $K_u = 7.0 \times 10^6$ J/m³^{12,13}. A 32×32 mesh was used for computation with a mesh size of 2 nm².

2.2 Phase Field Modeling

The time evolution of microstructure during annealing of $L1_0$ -type ferromagnetic alloys under an external magnetic field was calculated using the phase-field modeling^{14,15}. The governing equation is

$$\frac{\partial \phi_i}{\partial t} = -M \frac{\delta F}{\delta \phi_i} \quad (9)$$

where M is mobility, F is free energy of the whole calculation region and ϕ_i are phase-field variables. We defined ϕ_1 and ϕ_2 as the existence probability of the variants whose easy axis of magnetization is along [100] and [010]. Free energy F is expressed as the sum of chemical energy F_{chem} , interface energy $F_{\text{interface}}$, strain energy F_{strain} and magnetic energy. In phase-field modeling, $F_{\text{interface}}$ is given by the sum of the gradient energy F_{grad} and the energy barrier of chemical energy. F_{chem} , F_{grad} and F_{strain} are given by

$$F_{\text{chem}} = \int_{\vec{r}} \Delta G_c \left\{ \frac{A}{2} \sum_i \phi_i^2 - \frac{B}{3} \sum_i \phi_i^3 + \frac{C}{4} \left(\sum_i \phi_i^2 \right)^2 \right\} d\vec{r} \quad (10)$$

$$F_{\text{grad}} = \frac{1}{2} \kappa_s \int_{\vec{r}} \left\{ \sum_i (\nabla \phi_i)^2 \right\} d\vec{r} \quad (11)$$

$$F_{\text{strain}} = \frac{1}{2} \int_{\vec{r}} C_{ijkl} \epsilon_{ij}^{\text{el}} \epsilon_{kl}^{\text{el}} d\vec{r} \quad (12)$$

Further details are given elsewhere⁶. As for the connection with micromagnetics calculations, $\vec{e}_{\text{uni_aniso}}$ is related to the phase-field variables by

$$\vec{e}_{\text{uni_aniso}} = (\phi_1, \phi_2) \quad (13)$$

Equation (1) is iterated to obtain the most stable distribution of magnetization vectors at every time step when solving equation (9). Free surface conditions are used for micromagnetics calculations and two dimensional periodic boundary conditions are used for calculations of microstructure evolution.

3. RESULTS AND DISCUSSION

At first, each magnetic energy component given by Eqs. (4)-(7) and its influence on microstructure formation were separately examined under no external magnetic field since local magnetization vectors spontaneously align or relax in ferromagnetic alloys.

Understanding the effect of anisotropy energy is straightforward: Anisotropy energy is low when the magnetization vector is parallel to the easy axis of magnetization. To examine the effect of this energy component, we compared two different simplified microstructures consisting of one variant or two variants, shown in Fig. 1(a) and (b), with the same distribution of magnetization as shown in Fig. 1(d) and (e). The x -variant in the middle of Fig. 1(b) is confined to one mesh wide to intentionally retain a single magnetic domain even in the presence of an x -variant in a y -variant matrix. The components of the overall magnetic energy are compared in Fig. 1(g). Anisotropy energy is lower in the single variant structure, indicating that the anisotropy energy leads the whole structure to the single variant whose easy axis of magnetization is parallel to the local magnetization vector as in Fig. 1(a).

To examine the effect of the exchange energy on the microstructure formation, the thickness of the x -variant was changed from that in Fig. 1(b), as shown in Fig. 1(c).

From the viewpoint of anisotropy energy, local magnetization vectors attempt to align along the easy axis of magnetization, i.e., x - or y -direction in dark or light gray region, respectively, in Fig. 1(c). However, the exchange energy which increases with the variation of directions of local magnetization vectors becomes large under that condition, since local magnetization vectors in the x -variant are allowed to deviate from the y -direction to decrease anisotropy energy. When the thickness of the x -variant is small, the magnetization vector becomes parallel to the y -direction to reduce the exchange energy with the small increase of anisotropy energy. When the thickness is large, the increase of anisotropy energy becomes large. Therefore, local magnetization vectors are not aligned along one direction as shown in Fig. 1(f). In other words, reduction of the exchange energy is less than the increase in anisotropy energy when local magnetization vectors are forced to align. These results indicate that when the variant becomes too thin to allow the deviation of local magnetization vectors from easy axis of magnetization, exchange energy promotes variant selection by aligning the magnetization vector.

Fig. 1. Microstructure dependence on spatial distribution of local magnetization vectors and resultant magnetic anisotropy and exchange energies. The structure shown in (a) is a single y -variant. The thin vertical stripe in (b) and (c) in the middle is the x -variant embedded in the y -variant matrix. Stable spatial distributions of local magnetization vectors corresponding to (a), (b), and (c) are shown in (d), (e) and (f), respectively. The magnetic energy components are compared in (g) for the three microstructures.

The influence of the magnitude of the external magnetic field on spatial magnetization vectors was examined by applying an external magnetic field along the x -direction on the simplified microstructure shown in Fig. 1(c). The most stable distributions of local

magnetization vectors for different amplitudes of the external magnetic field are compared in Figs. 2(a)-(d). The alignment of local magnetization vectors to the external magnetic field is promoted as the amplitude increases, as expected. The components of overall magnetic energy are compared in Fig. 2(e). Absolute values of the Zeeman energy and anisotropy energy significantly increase with the amplitude of the external field. In other words, the system's Zeeman energy reduces by sacrificing anisotropy energy when under the high external field. Figs. 2(a)-(d) indicate that the Zeeman energy dominates the determination of the distribution of local magnetization vectors when the strength is at least 6 T, although the remnant of the anisotropy energy after relaxation of the local magnetic vectors is larger than that of the Zeeman energy for a given microstructure. In that case, the magnetization vector aligns along the direction of the external magnetic field so that the variant whose easy axis of magnetization is parallel to the external magnetic field becomes magnetically preferred. In such a range of the external magnetic field amplitudes, interactions between the magnetization vectors, i.e., exchange and dipole-dipole interaction energies, become unimportant.

Fig. 2. (a)-(d) Dependence of spatial distribution of local magnetization vectors and (e) the magnetic energies on the amplitude of resulting from an external magnetic field along the x -direction. The microstructure used for this comparison is shown in Fig. 1(c).

The dipole-dipole interaction energy is related to magnetic shape anisotropy. To examine the effect of this energy, a 16×4 mesh was used. Fig. 3(a) shows an initial spatial distribution of local magnetization vectors. This distribution is less stable than that in Fig. 3(b) since local magnetization vectors are less aligned in the long direction than in Fig. 3(b). In these calculations, anisotropy energy was not taken into account to examine the effect of dipole-dipole interactions, which was found to be small. The energy change is shown in Fig. 3(c),

indicating that the variant elongated along easy axis is more stable. However, the effect of magnetic shape anisotropy is far smaller than the other magnetic energy components, being negligibly small for variant selection.

Fig. 3. Effect of spatial distribution on magnetic dipole-dipole interaction energy: (a) Initial spatial distribution of local magnetic vectors, and (b) their stable distribution. It should be noted that the a 16×4 mesh was used. The magnetic energy components are shown in (c), excluding the Zeeman energies.

Finally, evolution of variants is computed with the aid of phase-field modeling in conjunction with the detailed model for magnetic energies. The initial distribution of variants was randomly determined⁶. An external magnetic field of 6 T was applied along the x -direction, which means the x -variant is magnetically preferred. The temporal evolution of microstructure is shown in Fig. 4. The most stable distribution of magnetization vectors was calculated every time step in Fig. 4(a), allowing local magnetization vectors to change while keeping them fixed in the x -direction in Fig. 4(b). Therefore the interaction between magnetization vectors can be neglected in the calculation for variant selection in $L1_0$ -type ferromagnetic alloys under high magnetic field. This justifies our separate study for the variant selection, which does not take into account the effect of the interaction between magnetization vectors.

Fig. 4. Temporal evolution of microstructures (a) when the interaction between local magnetization vectors is taken into account or (b) not taken into account. Black, dark gray and light gray regions show FCC initial matrix, x - and y -variant, respectively. The difference between (a) and (b) is negligible, indicating that the interaction between magnetization vectors does not need to be taken into account to investigate the variant selection in $L1_0$ -type ferromagnetic alloys under high magnetic field.

4. CONCLUSIONS

The influence of the spatial distribution of local magnetization vectors on microstructure formation has been investigated using micromagnetics in conjunction with phase-field modeling. Micromagnetics calculations revealed that the effect on microstructure evolution is negligibly small and thus can be approximated as the scalar difference in magnetization for easy and hard axes of magnetization. Examination of the variant selection with or without taking into account the spatial distribution of magnetization vectors by phase-field modeling clarified that the effect of interactions between local magnetization vectors is negligibly small when the amplitude of the external magnetic field is as large as 6 T. This indicates that calculations without interactions between magnetization vectors are sufficient to investigate the variant selection in $L1_0$ -type ferromagnetic alloys under high magnetic field.

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