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Monte Carlo simulation of two-dimensional domain structures in magnetite

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Abstract. A Monte Carlo method was applied for micromagnetic studies of two-dimensional domain structures of a 1-μm magnetite cube in zero field. By using this method we could incorporate the effect of thermal agitation into micromagnetic modeling. Starting from an initial single-domain structure (a saturated state), we obtained a closure domain structure with three body and four closure domains. This structure is quite different from the checkerboard-like structure obtained by a conjugate gradient method, and it gives a much lower energy and saturation remanence ratio. An initial lamellar two-domain structure evolved into a vortex structure, which was also reached from an initial quasi-vortex structure. Although such a vortex structure has a lower energy than the closure domain structure, it was not attainable from an initial single-domain structure at room temperature. The Monte Carlo method is effective in finding a path to escape from unstable local energy minima and reach a stable local energy minimum, although not necessarily a global minimum, at a given temperature. The structure corresponding to such a stable local minimum should represent a realistic domain structure, comparable to what would be attained in nature with the aid of thermal fluctuations of spins.

Introduction

In recent studies of magnetic domain structures the concept of a local energy minimum (LEM) state has played a key role. *Moon and Merrill* [1985] showed that several LEM states exist for a given grain size and that these are separated by sufficiently high energy barriers (specifically >>25-50 kT) in their one-dimensional micromagnetic modeling. The existence of several LEM states was expected to give an explanation for the observed different domain structures for the same grain depending on its magnetic or thermal history [*Halgedahl and Fuller*, 1980, 1983]. Between theory and observation, however, there was a large discrepancy in the number of domains for a given grain size. The number of domains predicted by theory was significantly greater than the number observed [e.g., *Moon*, 1991]. The gap has become narrower by more sophisticated micromagnetic modelings. *Ye and Merrill* [1991] used a quasi-two-dimensional model, which allows near-surface magnetization directions to become parallel to the surface, to predict a smaller number of domains for the same grain size than *Moon and Merrill* [1985] had found. By using unconstrained two-dimensional micromagnetic modeling, *Xu et al.* [1994] predicted that closure domains should develop at surfaces, and the equilibrium number of body domains was predicted to be only two for 1-μm or four for 5-μm magnetite grains. The predicted closure domain structures were recently observed for a large magnetite crystal with precisely oriented viewing surfaces [*Özdemir and Dunlop*, 1993; *Özdemir et al.*, 1995].

The important effect of thermal fluctuations on magnetization processes has long been recognized [e.g., *Brown*, 1979]. Thermal fluctuations of atomic spins cannot be ignored except for extremely low temperatures near absolute zero. Most measurements of magnetic properties and domain observations have been made near room temperature, but micromagnetic calculations have not incorporated thermal fluctuation effects explicitly. The stability of LEM states was separately considered in the light of Boltzmann statistics in one-dimensional [*Dunlop et al.*, 1994; *Enkin and Dunlop*, 1987; *Moon and Merrill*, 1985] or constrained three-dimensional micromagnetic modelings [*Enkin and Williams*, 1994]. Meanwhile, most unconstrained two- or three-dimensional modelings have employed gradient methods for energy minimization to find LEM states [e.g., *Fabian et al.*, 1996; *Newell et al.*, 1993a; *Williams and Dunlop*, 1989; *Xu et al.*, 1994]. Such gradient methods allow only energy decreases. They yield the LEM state that is nearest to an arbitrarily chosen initial structure. In view of the many-valley configuration space, however, it is quite possible that such nearest LEM states correspond to shallow, that is, thermodynamically unstable, valleys.

Thermal effects were first introduced into micromagnetic modeling by using the simulated annealing method [*Thomson et al.*, 1994]. This method was developed by *Kirkpatrick et al.* [1983] to search for the global maximum or minimum of a function of many independent variables, although locating such an equilibrium state is not necessarily guaranteed. *Thomson et al.* [1994] found lower-energy structures by the simulated annealing method for a cubic magnetite grain with 5 x 5 x 5 resolution than by a conjugate gradient method. When considering magnetization structures, however, we need structures corresponding to LEM states that are relevant to the observed domain structure or measured magnetic properties at a certain temperature. Monte Carlo methods, especially when based on the Metropolis algorithm [*Metropolis et al.*, 1953], are useful techniques to incorporate thermal effects into structure calculations. This approach allows us to obtain a thermal equilibrium structure and to follow nonequilibrium relaxation phenomena, which are interesting in view of the

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expected many-valley configuration space for micromagnetism.

In this paper we apply a Monte Carlo method to two-dimensional micromagnetic modeling of a 1-μm cube of magnetite. The two-dimensional model, which divides the cube into a two-dimensional array of cells with finite lengths and square cross sections but allows the magnetization in each cell to rotate in three dimensions in space, was originally developed by *Newell et al.* [1993a]. We followed the coordinate system of *Xu et al.* [1994] in order to be able to compare our results directly with theirs. Two-dimensional modeling is far superior to one-dimensional modeling because it allows magnetizations to rotate so as to satisfy the pole avoidance principle [*Brown,* 1963] and enables closure domains to form at surfaces. In comparison with three-dimensional modeling [e.g., *Williams and Dunlop*, 1989], two-dimensional modeling requires less computation time and less memory. As the sizes of grains approach 1 μm, the three-dimensional LEM structures tend to become two-dimensional [*Williams and Dunlop* , 1990].

Model

In our model a cube of magnetite with linear dimension a = 1 μm was subdivided into a two-dimensional array of 50 x 50 cells. The coordinate system was chosen following *Xu et al.* [1994] (Figure 1). The *y-z* plane is the $(\bar{1}10)$ plane, which is energetically most favorable for a 180° Bloch wall in magnetite [*Lilley*, 1950]. The viewing plane was set as the *x-y* plane $(11\bar{2})$, perpendicular to the $(\bar{1}10)$ plane, which contains one of the <111> easy axes of magnetization. The magnetization direction $\mathbf{m}_i$ of each cell is given by the polar angle θ_i and the azimuthal angle φ_i,

$$\mathbf{m}_i = \sin\theta_i \cos\varphi_i \mathbf{x} + \sin\theta_i \sin\varphi_i \mathbf{y} + \cos\theta_i \mathbf{z} \qquad (1)$$

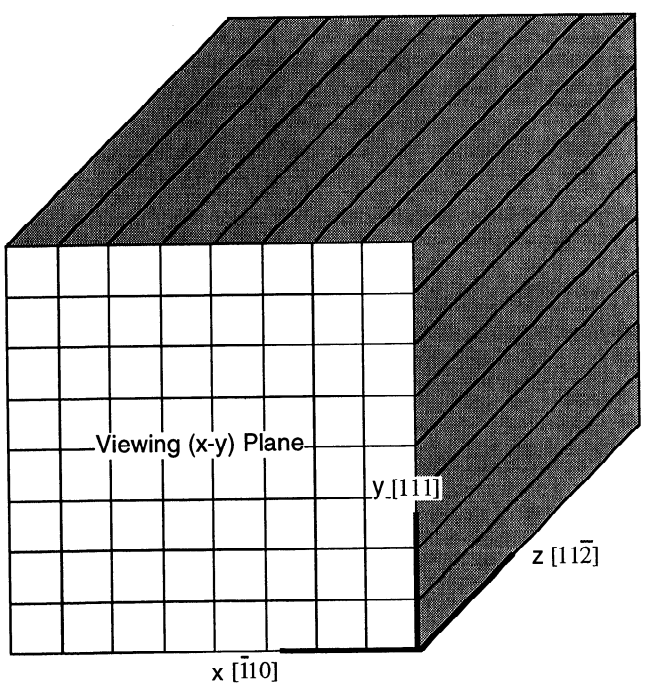

Figure 1. A model cubic grain and its crystallographic orientation. The viewing surface is the *x-y* plane.

where **x**, **y**, and **z** are unit vectors along the x, y, and z axes, respectively.

The total magnetic energy E_t for zero applied field is given by the sum of the exchange energy E_e, the magnetocrystalline anisotropy energy E_a, and the demagnetizing energy E_d. Following *Newell et al.* [1993a], we can express E_e, E_a, and E_d as

$$E_e = Aa\sum_{i=1}^{n}\sum_{j}\left(1-\mathbf{m}_i\cdot\mathbf{m}_j\right) \qquad (2a)$$

$$E_a = K_1 a^3\left[-\frac{1}{3}+\frac{1}{n}\sum_{i=1}^{n}\left(\alpha_i^2\beta_i^2+\beta_i^2\gamma_i^2+\gamma_i^2\alpha_i^2\right)\right] \qquad (2b)$$

$$E_d = \frac{\mu_0 a^3 M_s^2}{2n}\sum_{i,j=1}^{n}\mathbf{m}_i\cdot N_{ij}\cdot\mathbf{m}_j \qquad (2c)$$

where A, K_1, M_s, and μ_0 are the exchange constant, the magnetocrystalline anisotropy constant, the saturation magnetization, and the free-space permeability, respectively. E_e is calculated by assuming a linear change of spin directions between neighboring cells i and j, as shown in (2a). At the surface cells, E_e is calculated so as to satisfy the boundary condition, $d\mathbf{m}_i/dn = 0$ (see the appendix of *Newell et al.* [1993a]). In (2b), α_i, β_i, and γ_i are the direction cosines of $\mathbf{m}_i$ with respect to the three <100> crystal axes. When <111> are the easy axes of magnetization as for magnetite, K_1 takes a negative value. E_a is set to have a minimum value of zero by adding the constant term $-K_1a^3/3$ in (2b), when $\mathbf{m}_i$ of all cells are along a <111> easy axis of magnetization. In (2c), N_{ij} is the demagnetizing tensor between cells i and j [*Newell et al.*, 1993b], which is calculated as the interaction of surface charges of cells based on the method of *Rhodes and Rowlands* [1954]. The reduced energy is given by the energy value normalized by $E_{d0} = \mu_0 M_s^2 a^3/6$ for the single-domain state along <111>. The magnetoelastic energy is not considered in this model.

Monte Carlo simulation was performed as follows. First, an initial magnetization $\mathbf{m}_i$ was assigned for each of the 50 x 50 cells. The magnetization direction of one chosen cell was perturbed into a randomly chosen direction by a randomly chosen amount less than 5°. We calculated the difference in total energy ΔE_t caused by the perturbation and accepted the perturbation with the transition probability W,

$$W = \begin{cases} 1 & \Delta E_t < 0 \\ \exp\left(-\dfrac{\Delta E_t}{kT}\right) & \Delta E_t \geq 0 \end{cases} \qquad (3)$$

where k is Boltzmann's constant and T is absolute temperature. A perturbation is accepted or rejected by comparing W with a randomly chosen number between 0 and 1. This is called the Metropolis criterion, and it ensures that the system settles down in a thermal equilibrium state after numerous trials. Repeating such trial perturbations for each of the other cells constitutes one Monte Carlo step (MCS).

We used the effective field [e.g., *Berkov et al.*, 1993; *Schabes*, 1991] to calculate the difference in the total energy ΔE_t in the course of the Monte Carlo simulation. For the Metropolis algorithm we need only ΔE_t, not E_t itself, during the optimization. Even in calculating only ΔE_t the demagnetizing energy is still a computationally intensive

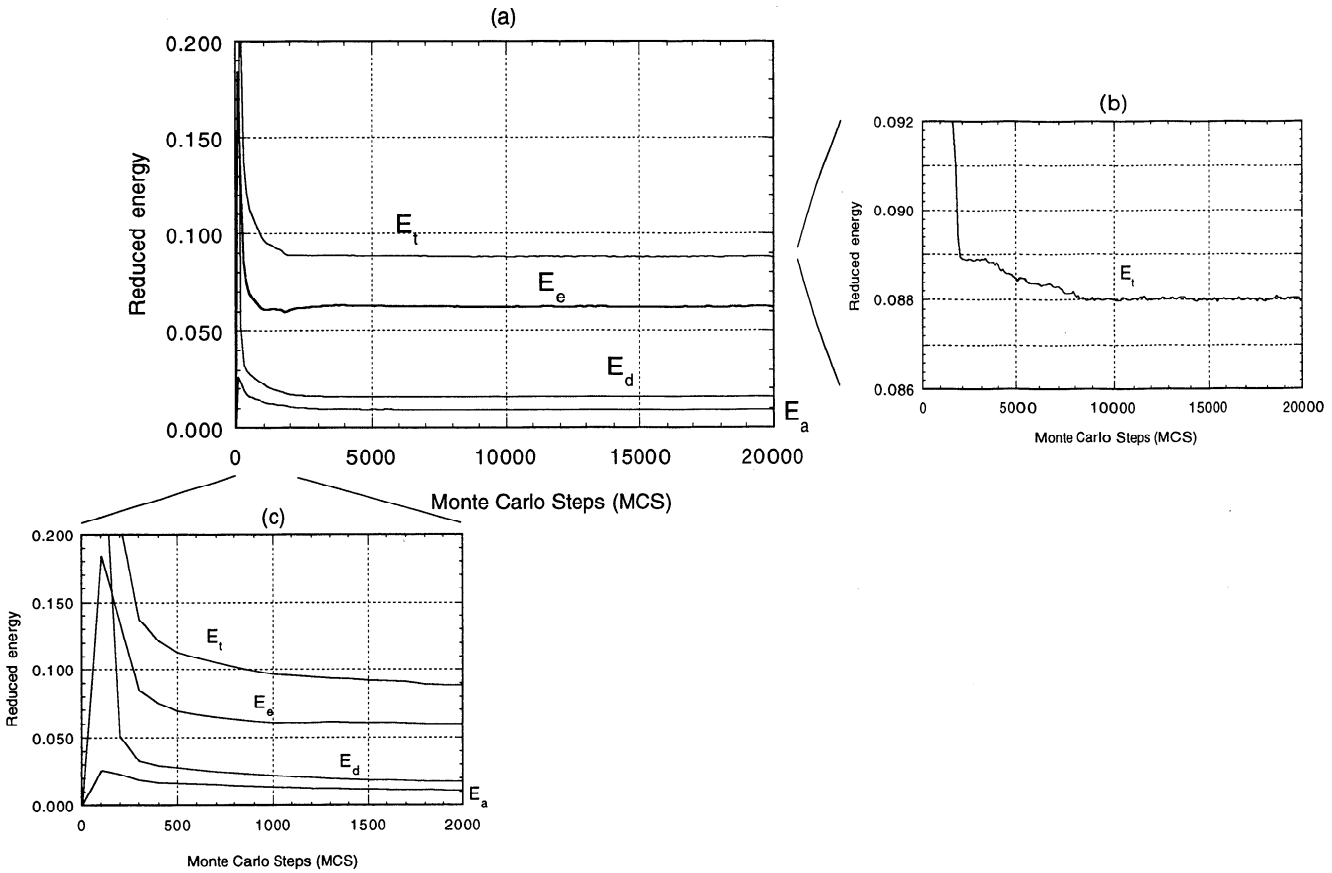

Figure 2. Energy change with Monte Carlo steps (MCS) for a 1-μm magnetite cube with an initial single-domain structure. The energy is presented as reduced energy normalized by the initial total energy. (a) Energy change between 0 and 20,000 MCS. (b) Total energy change expanded for the range of 0.086-0.092 in reduced energy value. (c) Energy changes enlarged for the interval 0-2,000 MCS. E_e, E_a, E_d, and E_t denote exchange, magnetocrystalline anisotropy, demagnetizing, and total energy, respectively.

term if we calculate the interactions between the perturbed cell and all other cells [*Thomson et al.*, 1994]. Instead of calculating such interactions we calculated ΔE_t caused by a perturbation of the magnetization direction $\mathbf{m}_i$ by using the effective field $\mathbf{H}_i$:

$$\Delta E_t = (\mathbf{m}_{i_{new}} - \mathbf{m}_{i_{old}}) \cdot \mathbf{H}_i \tag{4}$$

where

$$\mathbf{H}_i = -\frac{1}{\mu_0 V M_s} \frac{\partial E_t}{\partial \mathbf{m}_i}.$$

It was advantageous to use the effective field in reducing the computation time. Typically, we could reduce the computation time to only about 5% of that necessary to calculate all the interactions at each step.

Following *Xu et al.* [1994], we used four types of initial structures in this study: quasi-vortex, single-domain, and lamellar two- and three-domain structures. The initial quasi-vortex structure was given by $\varphi_i = \tan^{-1}(x/y)$ and $\theta_i = \pi/2$ for each cell, where (x, y) are measured with respect to an origin at the center of the model cube. The lamellar structures with two and three domains had walls of zero width placed at the positions determined from the one-dimensional nonmicromagnetic model of *Xu and Merrill* [1990]. In the case of single-, two-, and three-domain structures the initial magnetization directions were set along $+y$ or $-y$ directions (i.e., [111] easy axes of magnetization). The material constants, chosen for room temperature T = 298 K, were A=1.32 x 10^{-11} J/m, M_s = 4.80 x 10^5 A/m, and K_1 = -1.25 x 10^4 J/m^3. The computation was carried out with a KSR parallel computer using 10 or 20 processors. Each structure took about 1 hour to calculate.

Results

Variations With Monte Carlo Steps

To determine whether the system had reached a thermal equilibrium state, we examined the variation of E_e, E_a, E_d, and E_t with the number of MCS. In Figure 2, reduced values of E_e, E_a, E_d, and E_t have been plotted after every 100 steps, starting from a completely uniform magnetization structure along the [111] direction (i.e., a single-domain or saturated state). The reduced total energy E_t decreased rapidly with MCS within the first few hundred MCS down to less than 0.2, after which further changes became slow (Figure 2a). No further significant change of E_t was observed after about 8000 MCS, and the range of fluctuation of E_t was less than 0.1% of the initial value (Figure 2b). We therefore suppose that the system reached a thermal equilibrium state after about 8000 MCS.

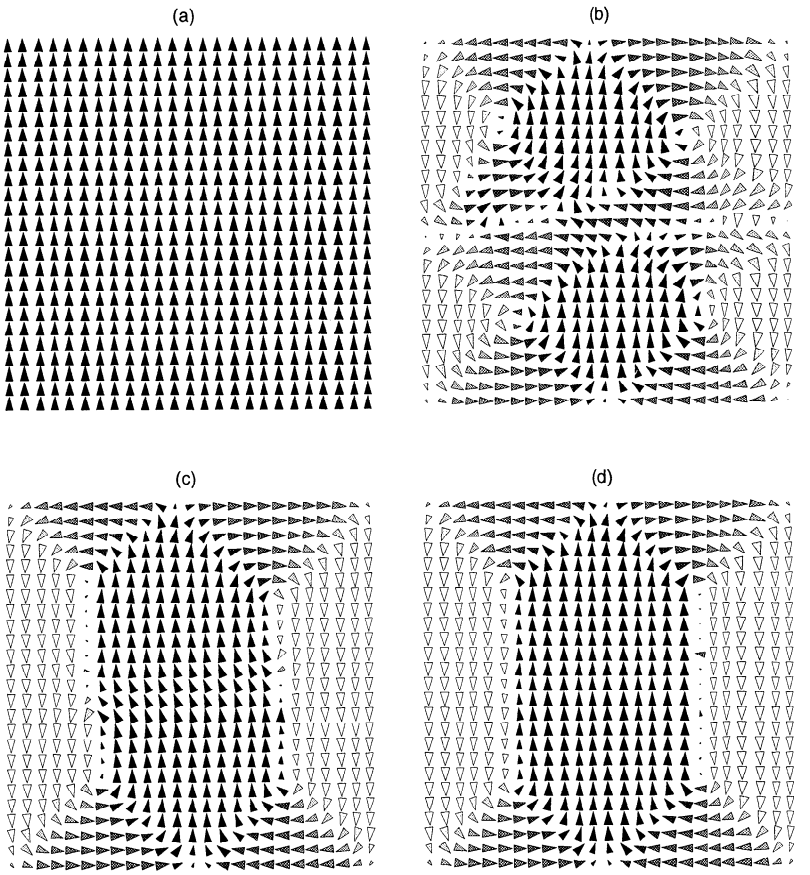

Figure 3. Variation of domain structures with the number of Monte Carlo steps (MCS) for a 1-μm magnetite cube, when starting from an initial single-domain structure. (a) Initial, (b) at 500 MCS, (c) at 2000 MCS, and (d) at 10,000 MCS.

On the other hand, the three energy terms constituting the total energy changed in different ways. E_e and E_a, starting from initial values of zero, increased rapidly in the first 100 steps and then decreased, whereas E_d steadily decreased from the initial value of unity (Figure 2c). Whereas E_d and E_a reached constant values at about 4000 MCS, E_e increased between 2000 and 4000 MCS, and then after 4000 MCS it slowly decreased (Figure 2a). Between 2000 and 3000 MCS the decrease of E_d and E_a was compensated by the increase of E_e, so that E_t remained almost constant. After 4000 MCS, E_e governed the slow change of E_t.

The variations in domain structures with MCS from an initial single-domain structure are shown in Figure 3. The calculated 50 x 50 cells have been reduced to 25 x 25 cells in order to have a simple presentation. Each arrow represents the projection on the x-y plane of the magnetization direction averaged over the four adjacent cells. The variation of structures with MCS represents how an initial domain structure relaxes into a final domain structure.

The initial uniform magnetization structure along [111] (Figure 3a) became a complicated checkerboard-like pattern at 500 MCS (Figure 3b). This structure can be viewed as consisting of four vortices and is similar to the final structure obtained for a 5-μm magnetite cube using an initial single-domain structure [*Xu et al.*, 1994]. At the surface cells the magnetizations were rotated to be parallel to the surfaces, and at the four corners the magnetization directions lie along the edge. Such features are favored, because they reduce magnetic poles appearing at the surfaces, as would be expected from the pole avoidance principle [*Brown*, 1963]. With this structure, E_d fell to 0.028 from 1.000 for the initial single-domain structure. However, the exchange energy E_e increased to a value of 0.069 from the initial value of zero as a result of the noncoherent structure. Also, E_a had an increased value of 0.016, because magnetizations in many cells deviated from the initial [111] easy axis of magnetization. Overall, the total energy E_t decreased greatly to 0.113, compared to the initial value of 1.000, because of the large decrease in E_d.

As MCS proceeded, the four vortices observed at 500 MCS coalesced in pairs to form two vortices elongated along the [111] direction, while keeping the magnetizations at surface cells parallel to the surface planes. By 2000 MCS the four vortices had almost disappeared, and a closure domain structure, consisting of three body domains capped with four closure domains, emerged (Figure 3c). At 2000 MCS, E_t further decreased to 0.089 from 0.113 at 500 MCS. The decrease came from the almost equal amounts of decreases in E_e, E_a, and E_d between 500 and 2000 MCS. The dissolution of vortices into a closure domain structure made magnetization vectors more parallel to each other in short and long ranges, reducing E_e and E_d, and also made $\mathbf{m}_i$ more nearly parallel to the [111] easy axis, resulting in a decrease of E_a.

After 2000 MCS, E_t was still decreasing until about 8000 MCS but very slowly (Figure 2a). In this interval the overall

structure remained almost unchanged from the closure domain structure seen at 2000 MCS (Figures 3c and 3d). In terms of energy, E_a and E_d remained almost constant. Only E_e changed and influenced the value of E_t. Such a change is interpreted as a process of fine tuning: The magnetizations of adjacent cells in the same domain changed their directions by small amounts to become more nearly parallel to each other.

To obtain thermal equilibrium structures, we averaged the structure over 1000 MCS in the interval in which the total energy reached a constant value. In the case of the initial single-domain structure we took the interval between 16,000 and 17,000 MCS as judged from Figure 2. When starting from the quasi-vortex and lamellar two- and three-domain initial structures, the relaxation was relatively fast, so that the equilibrium structures were obtained by averaging between 5000 and 6000 MCS.

Final Domain Structures

The final domain structures obtained by the Monte Carlo method were compared to those by a conjugate gradient method for 1-μm magnetite grains by using the same 50 x 50 cells and the same four initial states (Figure 4). We obtained the results by a conjugate gradient method using the parallel computer, and there are some differences from the structures found by *Xu et al.* [1994].

With an initial quasi-vortex structure, the final vortex structure by the Monte Carlo method (Figure 4a) is quite similar to that by the conjugate gradient method (Figure 4b). One minor difference is that the magnetization directions were parallel to the edges at cells near corners, and at the center of the cube the magnetizations deviated away from the viewing plane to form a vortex line. Such features were found in vortex structures by three-dimensional modelings for submicron magnetite cubes [*Williams and Dunlop*, 1995]. Although the energy difference between these two structures was small (Figure 5), the Monte Carlo method gave a lower reduced total energy (0.053) than the conjugate gradient method (0.062). Both structures gave very low M_{rs}/M_s of 0.001.

The ultimate structures developed from an initial single-domain structure were quite different between the two methods. The Monte Carlo method gave a domain structure with three body domains capped by four closure domains (Figure 4c) with a low reduced total energy of 0.088 (Figure 5). This structure is almost same as the structure obtained by using an initial lamellar three-domain structure by *Xu et al.* [1994]. Meanwhile, the checkerboard-like pattern, which was found for a 5-μm magnetite cube by *Xu et al.* [1994], emerged from the calculation by the conjugate gradient method (Figure 4d). Although this structure had a lower energy of 0.328 (Figure 5) compared to 0.680 for the structure with a single body domain and four corner domains, which was obtained from an initial single-domain state for a 1-μm magnetite cube [*Xu et al.*, 1994], the energy is still much higher than that of 0.088 by the Monte Carlo method. Also the checkerboard-like structure resembles the intermediate structure at 500 MCS (Figure 3b) in the course of relaxation. The final M_{rs}/M_s values were 0.002 for the Monte Carlo method and 0.201 for the conjugate gradient method. In the latter case the overall direction of magnetization still remained almost along the initially chosen [111] direction.

Starting from an initial lamellar two-domain structure, we found a vortex structure (Figure 4e) with a reduced total energy of 0.052 by the Monte Carlo method. The value of final total energy is almost the same as the value for the final vortex structure (Figure 5). Also there seems to be no significant difference between the two final structures (Figures 4a and 4e) obtained from initial quasi-vortex and lamellar two-domain structures. However, the M_{rs}/M_s value of 0.012 for the vortex structure from the initial two-domain structure is significantly higher than the value of 0.001 for the vortex structure from the initial quasi-vortex structure, and the net direction is almost perpendicular to the viewing plane. Such a difference is due to the following characteristics. The magnetizations parallel to the viewing plane cancel out each other by forming a single curl, whereas the magnetizations perpendicular to the viewing plane in the cells near the four corners and the center cancel out with opposite senses (Figure 4a) or sum up with same senses (Figure 4e).

By using the conjugate gradient method for an initial lamellar two-domain structure we obtained a two-body-domain structure with two associated large closure domains and a small quasi-vortex at each corner (Figure 4f), as found by *Xu et al.* [1994]. The reduced total energy was 0.139, which was higher than the value 0.052 by the Monte Carlo method (Figure 5). *Xu et al.* [1994] tested that their structure did not transform into a vortex structure with lower energy by perturbing the magnetization directions in the four corner cells within the viewing plane. To check the transformation process, we tried a simulation that accepted only decreases of energy (only $\Delta E_t<0$) resulting from perturbations. We again obtained a vortex structure as seen in Figure 4e from the initial lamellar two-domain structure, implying that the structure shown in Figure 4f does not represent even a LEM state. In the vortex structure, magnetization directions are almost perpendicular to the viewing plane at the corner cells. Probably the transformation of the initial two-domain structure into the final vortex structure was induced by rotating the magnetization away from the viewing plane, not within the plane.

By the Monte Carlo method, starting from an initial lamellar three-domain structure, we obtained a similar overall domain structure with three body domains capped with four closure domains (Figure 4g) as found by the conjugate gradient method (Figure 4h). When the structures are compared in detail, however, the magnetization directions at corners deviate from the viewing plane, and the Bloch walls are broader, extending over about 2 cells (=0.04 μm) in the case of the Monte Carlo method. Such small-scale features are associated with a significant difference in the reduced total energy: 0.086 by the Monte Carlo method compared to 0.156 by the conjugate gradient method (Figure 5). By taking account of the initially given zero-width walls and the eventual approximate wall width of 0.04 μm, close to the estimate of 0.03 μm using *Landau and Lifshitz*'s [1935] formula, the structure obtained by the Monte Carlo method (Figure 4g) can be regarded as more realistic than that obtained by the conjugate gradient method. The Monte Carlo method also gave a lower M_{rs}/M_s of 0.007, compared to 0.114 by the conjugate gradient method (Figure 5).

Discussion

By using a Monte Carlo method we have found lower energy states for a 1-μm magnetite cube than those obtained by a

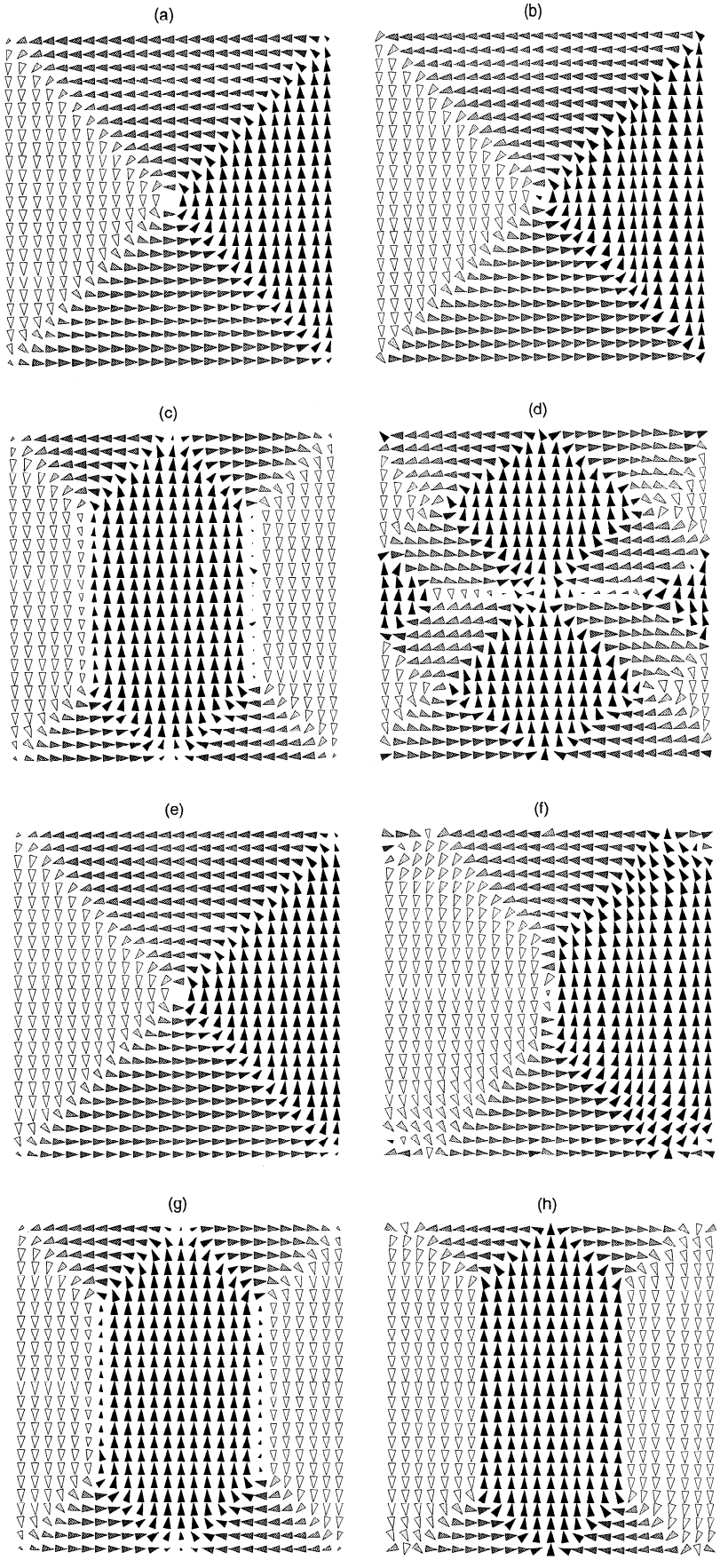

Figure 4. Comparison of final domain structures by (a, c, e, g) the Monte Carlo method and (b, d, f, h) the conjugate gradient method. The initial structures used are (a, b) quasi-vortex, (c, d) single-domain, (e, f) lamellar two-domain, and (g, h) lamellar three-domain structures.

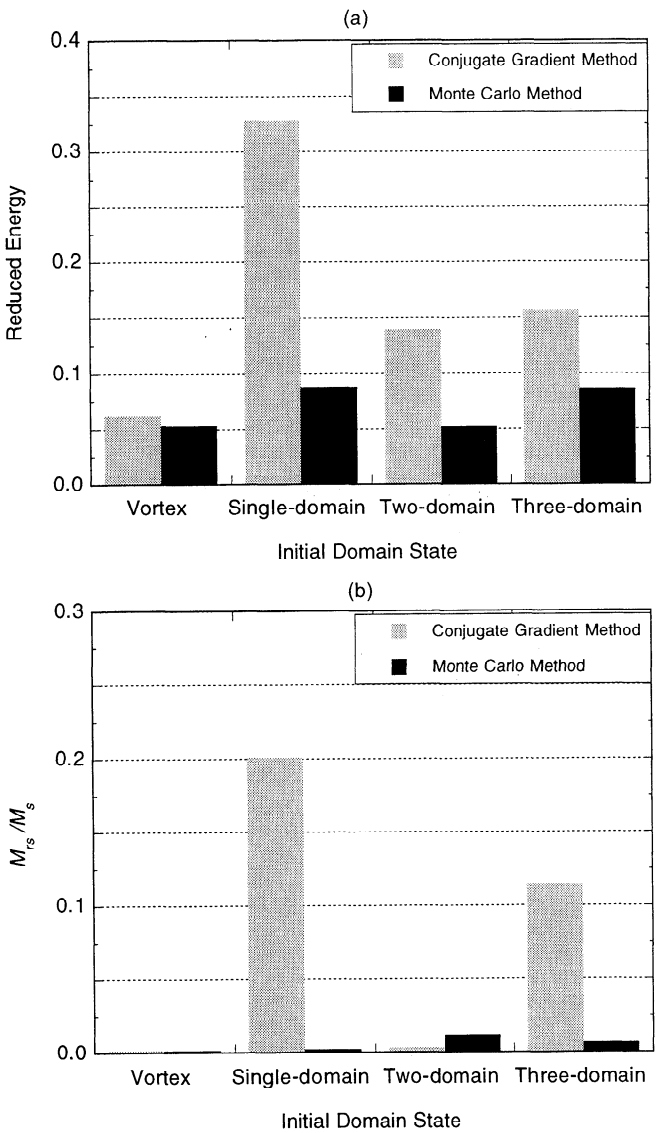

Figure 5. Histograms of (a) resultant reduced total energy and (b) M_{rs}/M_s for a 1-μm magnetite cube.

conjugate gradient method from all four initial states: quasi-vortex, single-domain, and lamellar two- and three-domains (Figure 5). The resultant two-dimensional structures showed reasonable fine features: Magnetizations were along the edges near corners and deviated from the viewing plane at the center of a vortex, and the domain walls have the widths expected from theory (Figure 4).

To obtain the final structures, we employed a Monte Carlo method based on the Metropolis algorithm. Conjugate gradient methods have been used in most previous micromagnetic studies [e.g., *Fabian et al.*, 1996; *Newell et al.*, 1993a; *Williams and Dunlop*, 1989; *Xu et al.*, 1994]. In general, it is very difficult to escape from the nearest local minimum by using the usual optimization techniques such as the conjugate gradient method [*Press et al.*, 1992]. Although simulated annealing methods have demonstrated important successes on a variety of global extremization problems and have been applied also in micromagnetics [*Thomson et al.*, 1994], it is of particular interest to us to obtain domain structures corresponding to LEM states starting from a specific initial state. The Monte Carlo method can locate a thermal equilibrium LEM state near the initial state by incorporating thermal agitation, avoiding capture by a possibly shallow nearest local minimum. Instead of setting an arbitrary criterion, which is usually determined on the basis of computation time, we examined the energy variation with MCS for each of the energy terms. After confirming that the system does not show any further reduction of energy after a certain number of MCS (Figure 2), we took the average structure for an interval of 1000 MCS with nearly constant energy in order to obtain the final structures. The Monte Carlo method does not necessarily guarantee a thermal equilibrium state under all conditions. When LEM states are separated by high energy barriers, such a LEM state would show no further energy change by small perturbations within a reasonable computation time. However, such a state should correspond to a stable (i.e., equilibrium) one for an appropriate choice of temperature and timescale. Thus we can regard the final domain structures obtained by the Monte Carlo method as being realistic and comparable to domain structures that one would expect to observe.

The four different initial domain structures converged to two final types of structures. Initial quasi-vortex and lamellar two-domain structures resulted in a final vortex structure (Figures 4a and 4e), and initial single-domain and lamellar three-domain structures resulted in a closure domain structure with three body domains capped by four closure domains (Figures 4c and 4g). Simulations employing the conjugate gradient method gave four different final structures for the same four initial structures (Figures 4b, 4d, 4f, and 4h). Some of these structures were quite unlike those obtained by the Monte Carlo method and had considerably higher energies (Figure 5).

Starting from the initial quasi-vortex and lamellar three-domain structure, the conjugate gradient method gave final structures almost identical to those obtained by the Monte Carlo method (Figures 4a, 4b, 4g, and 4h). It is probable that these final structures are the nearest or very near the corresponding initial structures in configuration space, judging from the similarity between the initial and final structures and the rapid relaxation. On the other hand, the final structures by the Monte Carlo method using initial single-domain and lamellar two-domain structures differ from those by the conjugate gradient method (Figures 4c, 4d, 4e, and 4f). The relaxation is relatively slow in these two cases, and the final structures are rather different from the corresponding initial structures. Also an intermediate structure during the Monte Carlo simulation resembles the final structure by the conjugate gradient method (Figures 3b and 4d).

Such comparisons would favor a Monte Carlo method when a final structure is obtained through a relatively long path in configuration space. The two final structures obtained by the conjugate gradient method from initial single-domain and lamellar two-domain structures (Figures 4d and 4f) might represent the states corresponding to the nearest shallow energy minima. However, a simulation that accepted only perturbations resulting in energy decreases also gave a final vortex structure from an initial two-domain structure. This result implies that the problem is not in the conjugate gradient method itself but in the particular scheme of implementation (e.g., the criterion for ending the iterations). This speculation is supported by the fact that *Williams and Dunlop* [1990] found a closure domain structure from an initial single-domain state in their three-dimensional simulation with the conjugate gradient method.

The closure domain structure resulting from an initial single-domain structure (Figure 4c) is particularly interesting, because such a domain structure would represent the structure for a saturation remanence state. Closure domains in magnetite have been observed on an appropriate viewing plane by *Özdemir et al.* [1995], who pointed out their importance in reducing the demagnetizing energy. *Xu et al.* [1994] obtained a closure domain structure from an initial lamellar three-domain structure in unconstrained two-dimensional micromagnetic modeling. We obtained a closure domain structure unlike the one-body-domain structure found by *Xu et al.* [1994] from the same initial single-domain structure. We could not obtain a metastable single-domain structure as observed for titanomagnetite grains of several tens of microns [*Halgedahl and Fuller*, 1980, 1983] or the U-shaped domain structure with large remanent magnetization observed for a 1-μm magnetite grain [*Geiß et al.*, 1996], both of which were observed in saturation remanence states. A metastable single-domain state, which was advocated to explain the high saturation remanence in the pseudo-single-domain region [*Halgedahl and Fuller*, 1980], has not yet emerged from modelings of a defect-free 1-μm magnetite cube. We obtained a much lower value of M_{rs}/M_s of 0.002 for a 1-μm magnetite by using the Monte Carlo method, compared to 0.201 (this study) or 0.640 [*Xu et al.*, 1994] by using the conjugate gradient method. However, our predicted value seems rather too low in comparison with experimental values, which range from 0.02 to 0.05 [*Amin et al.*, 1987].

The vortex structure is also interesting in two respects. First, the vortex structure is not attainable from an initial single-domain structure. Second, an initial lamellar two-domain state is unstable and transforms into the vortex structure. The final vortex structure gave the lowest energy for the model 1-μm magnetite cube we studied. *Worm et al.* [1991] concluded that a vortex-like four-domain structure should be of lowest energy for all multidomain grain sizes. However, in principle, such a global energy minimum state is not necessarily attainable from any point in a complicated configuration space with many variables. The closure domain structure from an initial single-domain structure never converted into a vortex structure with lower energy even with a very large number of MCS (Figure 2). This fact suggests that high energy barriers, which cannot be overcome by thermal energy available at room temperature, exist between a closure domain structure and a vortex structure. The LEM state with the closure domain structure should be a stable state of saturation remanence at room temperature in 1-μm magnetite cubes. Meanwhile, the initial two-domain structure eventually resulted in a vortex structure. A two-domain structure has often been assumed in one-dimensional nonmicromagnetic modelings [e.g., *Butler and Banerjee*, 1975] or assigned as an initial structure in one-dimensional micromagnetic modelings [e.g., *Moon and Merrill*, 1984]. *Geiß et al.* [1996] reported that two-domain states with relatively straight walls were observed with a Bitter pattern method in magnetite grains from 0.5 to 2.5 μm in size. However, such small grain sizes are near the resolution of the optical microscope, and they noted that a large number of grains in this size range displayed irregular Bitter patterns. As *Fabian et al.* [1996] pointed out on the basis of three-dimensional modeling of magnetite grains up to 0.23 x 0.63 μm, it is possible that a vortex structure attracts a line of colloid and gives the appearance of a two-domain state. The two-domain structure is unstable and easily converted to the vortex structure, so that it is unlikely to exist for a 1-μm magnetite grain.

Conclusions

1. The Monte Carlo method gave lower energy states for a 1-μm magnetite cube than the conjugate gradient method in all cases using the same initial structures (quasi-vortex, single-domain, and lamellar two- and three-domain structures).

2. Different initial states converged to an identical local energy minimum state for a 1-μm magnetite cube. Initial quasi-vortex and two-domain structures evolved to a final vortex structure, and initial single- and three-domain structures to a final closure domain structure.

3. Starting from an initial single-domain structure (i.e., a saturated state), we obtained a stable closure domain structure with three body and four closure domains. There should be no metastable single-domain state with large saturation remanence in 1-μm magnetite cubes.

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