Acta Materialia 149 (2018) 193–205

Contents lists available at ScienceDirect

Acta Materialia

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

Full length article

Micromagnetic analysis of crystallographic texturing and substrate-induced strain effects in $NiFe_2O_4$ and $CoFe_2O_4$ thin films

Youness Alvandi-Tabrizi [a, *], Justin Schwartz [b, c]

[a] Department of Mechanical and Aerospace Engineering, North Carolina State University, Raleigh, NC, United States
[b] Department of Materials Science and Engineering, North Carolina State University, Raleigh, NC, United States
[c] Department of Engineering Science & Mechanics, The Pennsylvania State University, University Park, PA, United States

ARTICLE INFO

Article history:
Received 11 December 2017
Received in revised form
15 February 2018
Accepted 15 February 2018
Available online 2 March 2018

Keywords:
Phase-field micromagnetics
Magnetic hysteresis
Magnetic domains
Polycrystalline films
Epitaxial strain
Ferrites

ABSTRACT

A three-dimensional continuum based micromagnetic model is developed to simulate the magnetization process in polycrystalline thin films and address the influence of crystallographic texturing, grain size and the substrate-induced strain on the spontaneous domain structure and hysteresis curves of $NiFe_2O_4$ and $CoFe_2O_4$ thin films. The model employs the Landau–Lifshitz–Gilbert equation along with mechanical equilibrium and Gauss' Law for magnetism to calculate the temporal and spatial distributions of the magnetic moments. Thus, this approach falls within the category of phase-field methods used for non-conserved systems. The finite element method is used to solve the partial differential equations in fully coupled fashion while using a different discretization method for each equation. The results demonstrate how the magnetization process is altered by adopting different microstructural orientations revealing stronger sensitivity in $CoFe_2O_4$ thin films than in $NiFe_2O_4$ thin films. Moreover, it is shown that the substrate-induced compressive strain favors in-plane magnetization, whereas the tensile strain switches the easy axis from the in-plane to the out-of-plane direction. The validity of the model is verified by comparing the results with recently published experimental data for sol-gel deposited $NiFe_2O_4$ thin films.

1. Introduction

Since its introduction in the 1960s [1], the continuum theory of micromagnetics has been widely used to capture the complex magnetization processes in various material systems at the meso-scale. The computational micromagnetics is based on discretizing the continuum magnetic media into sub-micrometer cells and using an equation of motion for magnetic medium within those cells. Gauss' Law for magnetism is then used to explain the magnetostatic interactions of the magnetic moments with each other and with external magnetic field. Modern micromagnetics uses the Landau–Lifshitz–Gilbert (LLG) equation first developed in 1935 [2], and later modified by Gilbert in 1956 [3], as the equation of motion. It also uses the balance law of linear momentum to account for the inhomogeneous local stress distribution caused by the elastic incompatibility of the magnetostrictive strain [4].

Micromagnetic modeling, even without consideration of magnetoelastic coupling, falls into the class of phase-field modeling used over the past two decades to solve similar problems involving mobile sharp interfaces [5,6]. The LLG equation takes the same role as the evolution equation in phase-field methods and predicts the evolution of the interfaces, i.e. the motion of the magnetic domain walls. In fact, the LLG equation can be classified as an Allen-Cahn type equation used for predicting the kinetics of non-conserved fields [6].

The magnetization field vector with its constant magnitude is the order parameter of the evolution equation in micromagnetics. It orients itself uniformly within the magnetic domains and continuously changes its direction across the domain walls. Similar to other phase-field models, a free energy functional couples the order parameter to other field variables such as strain and magnetic field. This is a polynomial functional describing the total energy of the system; an integral representation of the magnetization vector, its derivatives, and other field variables that are all functions of geometry, material properties, temperature, etc.

The LLG equation in phase-field micromagnetics is accompanied by two other fundamental balance law equations for stress and magnetic field. Incorporating the free energy functional into these

* Corresponding author.
E-mail address: yalvand@ncsu.edu (Y. Alvandi-Tabrizi).

https://doi.org/10.1016/j.actamat.2018.02.039
1359-6454/

equations leads to set of equations that describe the temporal and spatial distribution of the magnetization vector for a given magneto-mechanical boundary condition.

Phase-field micromagnetics has been employed in various studies of ferro/ferri-magnetic materials including thin films [4,7–9]. Due to its meso-scale and multiphysics nature, micromagnetic simulations are computationally expensive and hence most simulations are restricted to nanostructures or two-dimensional cases. With the improvement in the computational resources, more sophisticated problems have been simulated recently which include strain-mediated switching [10–12], electrically induced magnetization [13,14], ferromagnetic shape-memory alloys [15,16], exchange coupling [17], and more [18]. At the same time, new numerical techniques are being adopted to lower the computational cost and expand the horizon to larger scales [19].

Although the crystallography of the magnetic films are known to have significant effect on the magnetic behavior of the films, it has received little attention from the micromagnetics community and most studies are limited to monocrystalline films. In the few cases considering polycrystallinity, the simulation was simplified [20–22] or the focus was only on the final equilibrium domain structure rather than the hysteretic behavior [23]. Similarly, the literature suffers from the lack of a comprehensive study on the effect of substrate-induced strain. In the few available studies concerned with the substrate-induced strain, the hysteresis behavior was not included [24,25].

The purpose of this study is to develop a three-dimensional phase-field micromagnetic model to analyze the magnetic behavior of polycrystalline thin films. The model utilizes the formulation proposed by Landis [26] and incorporates it into finite element code COMSOL Multiphysics. The goal is to develop a comprehensive design tool for predicting the effect of crystallographic texturing, grain size, and substrate-induced strain on the spontaneous and hysteretic behavior in thin films. Nickel ferrite ($NiFe_2O_4$) and cobalt ferrite ($CoFe_2O_4$) are selected for this study due to their range of interesting physical properties such as room temperature ferrimagnetism and small gap insulation. To evaluate the validity of the model, the obtained results are compared with recently published experimental data for sol-gel deposited $NiFe_2O_4$ films [27,28].

2. Model development

2.1. Free energy functional

The free energy functional for phase-field micromagnetic modeling is a state function for magnetization derived from continuum thermodynamics and crystal symmetry considerations. Assuming the temperature is constant and well below the Curie temperature it can be written as:

$$h = h(\varepsilon_{ij}, H_i, m_i, m_{i,j}) \tag{1}$$

where the primary field variables are the strain field tensor ε_{ij}, magnetic field vector H_i, magnetization unit vector (also known as direction cosines) m_i, and its gradient $m_{i,j}$. Standard index notation with summation convention over repeated indices is used throughout this paper. The indices are running over the range of 1–3. The comma in the subscript denotes partial differentiation with respect to spatial coordinate x_i.

Under the assumption of linear kinematics, the strain tensor in a material body can be calculated from mechanical displacement u_i.

$$\varepsilon_{ij} = \frac{1}{2}\left(u_{i,j} + u_{j,i}\right) \tag{2}$$

The magnetic field and magnetization unit vector are related to the magnetic field B_i, via

$$B_i = \mu_0(H_i + M_s m_i) \tag{3}$$

μ_0 is the permeability of the free space and M_s is the saturation magnetization. The magnetic field can be expressed as the gradient of the magnetic scalar potential ϕ.

$$H_i = -\phi_{,i} \tag{4}$$

Assuming constant saturation magnetization

$$m_i = \frac{M_i}{M_s} \tag{5}$$

where M_i are the components of the magnetization field vector. The modulus of the magnetization vector is assumed to be constant and equal to the saturation magnetization. Therefore, it is more convenient to use the magnetization unit vector as the primary order parameter.

The total free energy density functional for phase-field micromagnetics modeling consists of contributions from magnetocrystalline anisotropy energy h_{aniso}, exchange energy h_{exch}, elastic energy $h_{elastic}$, and magnetostatic energy $h_{magnetostatic}$. The competition between these four energy terms defines the magnetic state of the material:

$$h = h_{aniso} + h_{exch} + h_{elastic} + h_{magnetostatic} \tag{6}$$

The magnetocrystalline anisotropy energy arises because the magnetization process depends on the crystallographic directions. Ignoring the higher order terms, the magnetocrystalline anisotropy energy for cubic symmetry is

$$h_{aniso} = K_1\left(m_1^2m_2^2 + m_1^2m_3^2 + m_2^2m_3^2\right) + K_2\left(m_1^2m_2^2m_3^2\right) \tag{7}$$

Here K_1 and K_2 are denoted as the first and second anisotropy constants. Depending on their sign and magnitude, Eq. (7) creates energy wells that favor certain magnetization directions.

The exchange energy or gradient energy is related to the inhomogeneous distribution of the magnetization and originates from a short-range interaction between magnetic moments while tending to keep them parallel. The mathematical expression is defined as the square of the spatial gradient of the magnetization directions:

$$h_{exch} = A_{exch}\left(m_{1,1}^2 + m_{1,2}^2 + m_{1,3}^2 + m_{2,1}^2 + m_{2,2}^2 + m_{2,3}^2 + m_{3,1}^2 + m_{3,2}^2 + m_{3,3}^2\right) \tag{8}$$

where A_{exch} is the exchange stiffness constant.

The elastic energy arises from deformations due to the effect of inhomogeneous magnetostriction. The compatibility of the strain field requires that the magnetostrictive strains be accompanied by elastic strains. The resultant elastic energy contains a positive term for pure elastic strains and a negative term for quasi-plastic magnetostrictive strains [29].

$$h_{elastic} = \frac{1}{2}C_{ijkl}\left(\varepsilon_{ij} - \varepsilon_{ij}^m\right)\left(\varepsilon_{kl} - \varepsilon_{kl}^m\right) = \frac{1}{2}C_{ijkl}e_{ij}e_{kl} \tag{9}$$

where C_{ijkl} is the fourth order elastic stiffness tensor, ε_{ij} is the pure