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Magnetic instability criterion for spin–lattice systems

Takahiro Shimada *, Kenji Ouchi, Ichiro Ikeda, Yoshiyuki Ishii, Takayuki Kitamura

Department of Mechanical Engineering and Science, Kyoto University, Nishikyo-ku, Kyoto 615-8540, Japan

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ABSTRACT

It is of vital importance to determine the critical magnetic and/or mechanical conditions under which the magnetic spin order becomes unstable in order to understand the microscopic nature of magnetic instabilities in ferromagnetic materials, often in conjunction with structural lattice instabilities, e.g., magnetic phase transitions and domain switching, as the source of diverse functionalities or the cause of critical failure of magnetic devices. Here, we propose an analytical method based on state-of-the-art spin–lattice modeling of ferromagnets to enable rigorous descriptions of magnetic instabilities in arbitrary atomic systems under a finite magnetic field and/or mechanical loading. The present theory yields, as an instability criterion, the condition that the minimum eigenvalue of the Hessian matrix of potential energy with respect to atomic coordinate and magnetic moment must be zero. In addition, the corresponding eigenvector represents the magnetic behavior of the spin moment at the instability, which is successfully validated by applying the criterion to magnetization switching in ferromagnetic Fe under an external magnetic field. Our approach thus provides a novel insight into the cause of magnetic instabilities and allows us to address complicated magnetic instability issues in practical situations.

1. Introduction

Magnetic transition metals, such as iron, and their compounds and alloys have been drawing continuous attention because of their diverse magnetic properties and technological applications including nonvolatile magnetic random-access memory (MRAM), magnetic sensors and motors, and promising spintronics devices [1–5]. The behavior of the spin moment in these materials is regarded as a central issue in this research field. Magnetic instabilities, *viz.*, a rapid or catastrophic change in magnetic ordering or reversal of magnetic moment with respect to the external magnetic field and/or mechanical loading, e.g., magnetic phase transitions and domain switching, essentially characterize the magnetic behavior of materials and lead to diverse functionalities or critical malfunction of devices. Magnetic instabilities are often observed in conjunction with structural lattice instabilities [6–10], e.g., a ferromagnetic-to-antiferromagnetic phase transition with a structural change from a body-centered cubic (bcc) to face-centered cubic (fcc) iron [6,9], which indicates that the strong coupling between the spin and lattice degrees of freedom (DOFs) plays an important role. This coupling is particularly important in magnetic materials at the nanoscale, where a novel magnetic phase transition occurs because of the characteristic atomic arrangement, due to the nontrivial effect of surfaces or interfaces and structural low-dimensionality [11–15]. Thus, understanding the nature of magnetic instabilities at the nanoscale is both scientifically interesting and technologically important for magnetic materials.

Quantum mechanical approaches, such as first-principles density-functional theory, provide reliable information about the magnetic behavior of materials. However, the huge computational cost restricts model ensembles to an extremely small number of atoms. Instead, very recently, attempts have been made to construct computationally efficient models of interatomic interactions to simultaneously describe spin moments and spin–lattice interactions, and several spin–lattice models have been proposed via the quantum mechanical base by explicitly including the intrinsic spin DOFs into semi-empirical atomic models with many-body interactions [16–20]. Although these models enable large-scale spin–lattice dynamics simulations [18,21], the absence of any theory to describe magnetic instabilities makes it difficult to capture the onset of magnetic instability events, which generally occur from only a local site in such a large-scale system. Therefore, an analytical criterion for magnetic instabilities is essential to detect such local instability events.

As for mechanical (structural) instabilities, several criteria, such as the lattice stability criteria and phonon soft modes, were proposed for crystal lattices [22–27]. We previously proposed a criterion to rigorously describe the onset of mechanical instabilities and

* Corresponding author.
E-mail address: shimada@me.kyoto-u.ac.jp (T. Shimada).

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the deformation mode at the instabilities in arbitrary atomic structures by explicitly taking into account the delicate balance between the potential energy and work done by an external load and/or constraint with respect to all the DOFs of atoms [28–32]. The advantage of our theory is its rigor in dealing with arbitrary structures without any limitation or assumption and its flexibility for other systems with different degrees of freedom [33–35]. For example, we successfully extended our theory to instabilities in dielectrics and ferroelectrics where electric (charge) degrees of freedom must be taken into account [35]. Thus, it should also be possible to develop a criterion for magnetic instabilities by extending our theory to magnetic systems through the incorporation of the spin and lattice degrees of freedom into the formulation.

In this paper, we propose a criterion for magnetic instabilities in an atomic system by explicitly including the spin and lattice degrees of freedom, and validate the proposed theory by applying the criterion to magnetization switching in ferromagnetic iron under an external magnetic field. This paper is organized as follows. Section 2 outlines the theory for magnetic instabilities and formulates it for a spin–lattice system. Section 3 describes the simulation procedure and results for the validation of the present theory. Section 4 presents our conclusion.

2. Proposal of magnetic instability criterion

2.1. Theory of magnetic instabilities for a spin–lattice system

Here, we consider a spin–lattice system consisting of N atoms with magnetic moments. The potential energy of the system, U, can be described by the atoms' coordinates, $\boldsymbol{R}$, and magnetic moments, $\boldsymbol{M}$,

$$U = U(\boldsymbol{R}, \boldsymbol{M}), \tag{1}$$

where

$$\boldsymbol{R} = {}^t(r_x^1, r_y^1, r_z^1, r_x^2, r_y^2, r_z^2, \ldots, r_x^N, r_y^N, r_z^N), \tag{2}$$

$$\boldsymbol{M} = {}^t(m_x^1, m_y^1, m_z^1, m_x^2, m_y^2, m_z^2, \ldots, m_x^N, m_y^N, m_z^N). \tag{3}$$

Here, r_i^α and m_i^α denote the atomic coordinate and the magnetic moment of atom α (=1,…,N) in the i (=x, y, z) direction. The irreducible number of lattice (atomic) degrees of freedom (DOFs) in the system is $I_R = 3N - 6$ because the DOFs for the rigid body translation (3) and rotation (3) are subtracted from the total DOFs of atoms ($3N$) [34]. On the other hand, the number of spin DOFs is $I_M = 3N$. Therefore, the number of lattice and spin DOFs is $I = I_R + I_M = 6N - 6$. Here, an arbitrary deformation and/or perturbation of the magnetic moment of the system can be represented by a change in the following I-dimensional vector $\boldsymbol{X}$ consisting of all DOFs:

$$\begin{aligned}\boldsymbol{X} &= {}^t(r_z^2, r_x^3, r_y^3, r_x^4, r_y^4, r_z^4, \ldots, r_x^N, r_y^N, r_z^N, m_x^1, m_y^1, m_z^1, m_x^2, m_y^2, m_z^2, \ldots, m_x^N, m_y^N, m_z^N)\\ &= {}^t(X_1, \ldots, X_i, \ldots, X_I).\end{aligned} \tag{4}$$

When the system is at equilibrium ($\boldsymbol{X} = \boldsymbol{X}_0$) under a static external load and/or magnetic field, the total energy of the system, Π, consists of the potential energy, U, the work done by an external force, V, and the work done by an external magnetic field, W, and is given by

$$\Pi = U + V + W. \tag{5}$$

The total energy of the system in terms of an infinitesimal deformation and/or perturbation of the magnetic moment, $\Delta\boldsymbol{X}$, $\Pi(\boldsymbol{X}_0 + \Delta\boldsymbol{X})$, can be described by the Taylor's series expansion of the total energy, $\Pi(\boldsymbol{X}_0)$, by $\Delta\boldsymbol{X}$, and is given by

$$\begin{aligned}\Pi(\boldsymbol{X}_0 + \Delta\boldsymbol{X}) = \Pi(\boldsymbol{X}_0) &+ \sum_{k=1}^{I} \left.\frac{\partial \Pi}{\partial X_k}\right|_{\boldsymbol{X}=\boldsymbol{X}_0} \Delta X_k\\ &+ \frac{1}{2}\sum_{k=1}^{I}\sum_{l=1}^{I} \left.\frac{\partial^2 \Pi}{\partial X_k \partial X_l}\right|_{\boldsymbol{X}=\boldsymbol{X}_0} \Delta X_k \Delta X_l + \cdots\end{aligned} \tag{6}$$

The second term on the right-hand side (the first derivative of total energy) is zero because the system is at equilibrium. Owing to the static loading, the external load is constant and thus the work is proportional to the displacement of atoms. As a result, we get [28,29]

$$\left.\frac{\partial^2 V}{\partial X_k \partial X_l}\right|_{\boldsymbol{X}=\boldsymbol{X}_0} = 0. \tag{7}$$

On the other hand, the second derivative of the work done by external magnetic field is not always zero as we will show in the next section. Ignoring the higher-order terms, the change in total energy, $\Delta\Pi$, can be rewritten in terms of an infinitesimal change in the system, $\Delta\boldsymbol{X}$:

$$\Delta\Pi \equiv \Pi(\boldsymbol{X}_0 + \Delta\boldsymbol{X}) - \Pi(\boldsymbol{X}_0) = \frac{1}{2}{}^t\Delta\boldsymbol{X}\mathbf{A}\Delta\boldsymbol{X}, \tag{8}$$

where $\mathbf{A}$ is the $I \times I$ Hessian matrix whose components are given by the second derivative of the total energy with respect to the DOFs,

$$A_{kl} \equiv \left.\frac{\partial^2 \Pi}{\partial X_k \partial X_l}\right|_{\boldsymbol{X}=\boldsymbol{X}_0} = \left.\frac{\partial^2 U}{\partial X_k \partial X_l}\right|_{\boldsymbol{X}=\boldsymbol{X}_0} + \left.\frac{\partial^2 W}{\partial X_k \partial X_l}\right|_{\boldsymbol{X}=\boldsymbol{X}_0} \quad (k, l = 1, \ldots, I). \tag{9}$$

Denoting the eigenvalues of matrix $\mathbf{A}$ by η_i ($\eta_1 \leqslant \cdots \leqslant \eta_i \leqslant \cdots \leqslant \eta_I$), the matrix is diagonalized using the eigenvector, $\boldsymbol{p}_i$

$$\boldsymbol{P}^{-1}\mathbf{A}\boldsymbol{P} = {}^t\boldsymbol{P}\mathbf{A}\boldsymbol{P} = \begin{pmatrix} \eta_1 & & O \\ & \ddots & \\ O & & \eta_I \end{pmatrix}, \tag{10}$$

where $\boldsymbol{P} = (\boldsymbol{p}_1 \ldots \boldsymbol{p}_I)$. Here, the eigenvector $\boldsymbol{p}_i$ is normalized. Introducing

$$\Delta\mathbf{Q} \equiv \boldsymbol{P}^{-1}\Delta\boldsymbol{X} = {}^t(\Delta Q_1, \ldots, \Delta Q_I), \tag{11}$$

the total energy change in Eq. (8) becomes

$$\Delta\Pi = \frac{1}{2}{}^t(\boldsymbol{P}\Delta\mathbf{Q})\mathbf{A}(\boldsymbol{P}\Delta\mathbf{Q}) = \frac{1}{2}{}^t\Delta\mathbf{Q}({}^t\boldsymbol{P}\mathbf{A}\boldsymbol{P})\Delta\mathbf{Q} = \frac{1}{2}\sum_{i=1}^{I}\eta_i(\Delta Q_i)^2. \tag{12}$$

In a similar discussion with the previous work [28,29], the critical condition for instability, $\Delta\Pi = 0$, appears when the minimum eigenvalue reaches zero, $\eta_1 = 0$, and the corresponding eigenvector $\boldsymbol{p}_1$ at $\eta_1 = 0$ represents the change in magnetic moment and the deformation of atoms at the instability, i.e., the instability mode vector.

2.2. Formulation of the Hessian matrix on the basis of the spin–lattice dynamics model

A spin–lattice dynamics (SLD) model is proposed to simulate the behavior of atoms and spins in magnetic materials [18,21]. In SLD, each magnetic atom is treated as a classical particle with intrinsic spins, and the time evolution of the system is analyzed with the coordinates of the atoms, $\boldsymbol{R}$, and the unit vectors of the magnetic moments. Here, the unit vector of the magnetic moment of atom α, $\boldsymbol{e}^\alpha$, is expressed in terms of θ^α and ϕ^α,

$$e_x^\alpha = \sin\theta^\alpha \cos\phi^\alpha, \quad e_y^\alpha = \sin\theta^\alpha \sin\phi^\alpha, \quad e_z^\alpha = \cos\theta^\alpha. \tag{13}$$

The vector of DOFs, $\boldsymbol{X}$, is now rewritten as