

Atomistic theory of the critical field for intrinsic spin reversal in transition metals

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(Received 25 February 1998; revised manuscript received 1 July 1998)

A method to estimate the critical field for intrinsic spin reversal in transition metals is proposed within the framework of itinerant-electron magnetism. By studying the transverse magnetic susceptibility under a Hartree-Fock approximation, the dispersion relations of the spin wave excitations are obtained, which possess a positive gap induced by the anisotropically coupled spin-orbit interaction when the magnetization is along the easy axis. When a magnetic field applied opposite to the magnetization is strong enough to overcome the excitation gap, a spin reversal transition takes place which defines the critical field. To illustrate the present method, transition-metal magnetic monolayers and microclusters have been examined. Comparisons show that the method can be applied to capture effects missed in the classical Stoner treatment.

[S0163-1829(99)10101-2]

I. INTRODUCTION

The phenomenon of magnetic hysteresis has been known for more than a century, and its application in magnetic recording has resulted in great economic achievements. An important quantity in a recording system is the critical field at which intrinsic spin reversal (ISR) takes place.¹ To determine this field, Stoner and Wolfarth established a classical treatment — they first proposed a phenomenological model including the anisotropy energy, then used a second order variation method to determine the critical field.² This idea had been proven useful in presenting a qualitative description of macroscopic magnetic systems.³ In recent decades, in order to enlarge the memory density, the size of each recording unit has become smaller and smaller and the structure more and more complicated. Therefore, the Stoner-like theory^{2,3} is becoming inadequate for the following reasons. First, to use Stoner-like theory, one has to find out the anisotropy energy function with respect to the spin direction. However, this is only possible for systems with very high symmetry (for example, the magnetic thin film). Second, Stoner's approach assumes that the spin reversal is a uniform mode and the magnetization maintains unchanged in the reversal process. Again, this requirement is not generally satisfied, for example, in inhomogeneous systems such as microclusters and magnetic multilayers. Finally, all quantum effects have been neglected in Stoner-like theory.

Recently, based on the local magnetic moment model, we proposed a quantum method to calculate the critical field for ISR in a magnetic system.⁴ The key point is to study the quantum spin wave excitations rather than varying the spin moments classically. It is found that the anisotropy induces a gap to the elementary excitations which can help the system to maintain metastability. Thus, only when a negative field is strong enough to overcome the gap, does the spin turn over,

which in turn, determines the critical field.⁴ This approach has been applied to study various kinds of inhomogeneous systems⁵ and had been extended to incorporate thermal fluctuations.⁶ However, since practical recording systems are usually made of transition metals which cannot be well described by the local magnetic moment model, the theory cannot be applied naively to study such systems.

On the other hand, the itinerant electron model has been very successful in the description of the magnetism of transition metals. Many efforts have been devoted to the anisotropies of ultrathin magnetic films and microclusters both from first-principles calculations⁷⁻⁹ and model calculations.¹⁰⁻¹⁴ The spin-orbit (SO) interaction is believed to be mainly responsible for the anisotropies in metallic systems, and because of the reduced symmetry and lacking neighbors, in some cases, a large anisotropy is expected. Although there have been many authors investigating the anisotropies and the anisotropy is really *closely related* to the critical fields for ISR, a theory which can be used directly to estimate the critical field still does not exist to our knowledge.

In the present work, we establish the atomistic theory of the critical field for ISR in transition-metal magnetic systems. We assume that the magnetic systems concerned are small enough so that no domain wall motion needs to be considered. As a reasonable approximation, the Hubbard model with SO interaction is used to describe a transition metal as in Refs. 10–14 while the magnetostatic interaction¹⁵ is neglected in the present paper. The low-lying spin-wave excitations are obtained under a Hartree-Fock mean-field approximation with the SO interaction treated self-consistently rather than perturbatively.¹² When the magnetization is pointing along the easy axis, the anisotropic SO interaction induces a positive excitation gap even though a field is applied opposite to the magnetization. The field which is strong enough to overcome the gap is the critical field for ISR, since

the previous spin state cannot maintain metastability due to large fluctuations at this field.⁴ Free-standing transition-metal monolayers and small magnetic microclusters have been studied to illustrate the main ideas of the present method. Since the possible hybridization effects with the substrates have been neglected in the present idealized models, the calculated results seem less realistic. However, the aim of the present work is to describe how a quantum-mechanical calculation can be performed to study the spin wave excitations so as to determine the critical field for ISR in itinerant-electron systems. Special attention will be paid to the comparisons with the classical Stoner theory.

This paper is outlined as follows. The next section is devoted to the description of the theory and Sec. III applies the theory to monolayers and microclusters. Finally, the conclusions are summarized in the last section.

II. THEORY

The Hamiltonian describing a transition-metal magnetic system should contain the following terms:

$$H = \sum_{ij} \sum_{\alpha, \beta, \sigma} t_{ij}^{\alpha\beta} C_{i\alpha\sigma}^+ C_{j\beta\sigma} + \frac{1}{2} \sum_i \left[U \hat{n}_i \hat{n}_i - \frac{J}{\mu_B} \hat{\mu}_i \hat{\mu}_i \right] + \xi \sum_{i, \alpha\beta} \sum_{\sigma_1 \sigma_2} S_{\alpha\beta}^{\sigma_1 \sigma_2} C_{i\alpha\sigma_1}^+ C_{i\beta\sigma_2} - \sum_i h \mu_B \sigma C_{i\alpha\sigma}^+ C_{i\alpha\sigma}. \quad (1)$$

The first term is the electronic hopping interaction where $\alpha\beta$ denote the $3d(xy, yz, zx, x^2 - y^2, 3z^2 - r^2)$ and $4s$ wave channels, and σ is the spin index. The second term describes the Hubbard-like intra-atomic electron-electron ($e-e$) interactions, where $U = U_{\uparrow\uparrow} + U_{\downarrow\downarrow}$ is the direct Coulomb interaction and $J = U_{\uparrow\downarrow} - U_{\downarrow\uparrow}$ the exchange interaction,¹³ and \hat{n}_i and $\hat{\mu}_i$ are defined by $\hat{n}_i = \sum_{\alpha\sigma} C_{i\alpha\sigma}^+ C_{i\alpha\sigma}$, $\hat{\mu}_i = \mu_B \sum_{\alpha\sigma} \sigma C_{i\alpha\sigma}^+ C_{i\alpha\sigma}$, respectively. The third term is the SO interaction which has been given explicitly in Ref. 11. Finally, an external field is applied along the magnetized direction. In a Hartee-Fock approximation (HFA), the $e-e$ interaction can be decoupled as

$$H_{ee} \approx -\frac{1}{2} \sum_i \left(U n_i^2 - \frac{J}{\mu_B} \mu_i^2 \right) + \sum_{i\alpha\sigma} \left(U n_i - \frac{J\sigma}{\mu_B} \mu_i \right) C_{i\alpha\sigma}^+ C_{i\alpha\sigma}, \quad (2)$$

where $n_i = \langle \hat{n}_i \rangle$ and $\mu_i = \langle \hat{\mu}_i \rangle$ should be determined self-consistently.

Diagonalizing Hamiltonian (1) under the HFA (2) with the help of the following transformation:

$$\begin{cases} C_{i\alpha\sigma}^+ = \sum_{\{l\}} P_{i\alpha\sigma, \{l\}}^* \tilde{C}_{\{l\}}^+, \\ C_{i\alpha\sigma} = \sum_{\{l\}} P_{i\alpha\sigma, \{l\}} \tilde{C}_{\{l\}}, \end{cases} \quad (3)$$

we can find the eigenvalues (energy bands) of the system:

$$H = -\frac{1}{2} \sum_i \left(U n_i^2 - \frac{J}{\mu_B} \mu_i^2 \right) + \sum_{\{l\}} E_{\{l\}} \tilde{C}_{\{l\}}^+ \tilde{C}_{\{l\}}, \quad (4)$$

where $\{l\}$ denote a set of quantum number $\{j\beta\sigma'\}$, and $E_{\{l\}}$ are the eigenvalues of the Hamiltonian matrix with $\{P_{\{m\}, \{l\}}, m=1, \dots\}$ as corresponding eigenvectors. Thus the self-consistent equations should be

$$\begin{cases} n_i = \sum_{\alpha\sigma} \sum_{\{l\}} |P_{i\alpha\sigma, \{l\}}|^2 \Theta(E_f - E_{\{l\}}), \\ \mu_i = \sum_{\alpha\sigma} \sum_{\{l\}} \sigma |P_{i\alpha\sigma, \{l\}}|^2 \Theta(E_f - E_{\{l\}}) \end{cases} \quad (5)$$

in which E_f is the Fermi energy.

What are the *elementary excitations* in such systems? Similar to the Heisenberg model, the *elementary excitations* (or the spin waves) should be small *transverse* variations of the magnetization, which can be determined by studying the transverse magnetic susceptibility function.^{16,17} Suppose some disturbing field (h^x, h^y) is applied perpendicular to the magnetization, the perturbation energy is

$$\begin{aligned} \delta H &= 2\mu_B \sum_i (h_i^x S_i^x + h_i^y S_i^y) e^{i\omega t} \\ &= \mu_B \sum_i (h_i^+ S_i^- + h_i^- S_i^+) e^{i\omega t}, \end{aligned} \quad (6)$$

where

$$\begin{cases} h_j^\pm = h_j^x \pm i h_j^y, \\ S_j^\pm = S_j^x \pm i S_j^y \end{cases} \quad (7)$$

and the spin operators are defined by

$$\begin{cases} S_i^+ = \sum_{\alpha} C_{i\alpha\uparrow}^+ C_{i\alpha\downarrow}, \\ S_i^- = \sum_{\alpha} C_{i\alpha\downarrow}^+ C_{i\alpha\uparrow}. \end{cases} \quad (8)$$

According to the Kubo's formulas,¹⁸ the induced magnetization is given by

$$\begin{cases} 2\mu_B \langle S_i^+ \rangle e^{i\omega t} = \left[\sum_j \chi_{ij}^{+-}(\omega) h_j^+ + \sum_j \chi_{ij}^{++}(\omega) h_j^- \right] e^{i\omega t}, \\ 2\mu_B \langle S_i^- \rangle e^{i\omega t} = \left[\sum_j \chi_{ij}^{-+}(\omega) h_j^- + \sum_j \chi_{ij}^{--}(\omega) h_j^+ \right] e^{i\omega t}, \end{cases} \quad (9)$$

where the magnetic susceptibilities are defined by some spin correlation functions:¹⁸

$$\chi_{ij}^{\pm\pm}(t) = -2\mu_B^2 \langle \langle S_i^\pm(t) | S_j^\pm \rangle \rangle = 2\mu_B^2 i\hbar \langle 0 | [S_i^\pm(t), S_j^\pm] | 0 \rangle, \quad (10)$$

$$\chi_{ij}^{\pm\pm}(\omega) = \int_{-\infty}^{\infty} \chi_{ij}^{\pm\pm}(t) e^{i\omega t} dt. \quad (11)$$

According to the standard equations of motion for the Green's functions, we have the following equations to evaluate those spin correlation functions:

$$\omega \langle \langle S_i^\pm | S_j^\pm \rangle \rangle_\omega = \langle 0 | [S_i^\pm, S_j^\pm] | 0 \rangle + \langle \langle [S_i^\pm, H] | S_j^\pm \rangle \rangle_\omega. \quad (12)$$

Unfortunately, because of the many-particles effect introduced by the e - e interaction, the correlation functions cannot

be calculated exactly so that some approximations (say, the random phase approximation¹⁷) are necessary. Here we follow essentially the arguments made by Friedel¹⁶ to first determine the *Hartree-Fock* susceptibility in which the *dynamic correlation corrections* are neglected. By putting Hamiltonian (1) under the HFA (2) into Eq. (12) directly, the *Hartree-Fock* susceptibility functions are found exactly as follows:

$$\chi_{i,j}^{+-0}(\omega) = -2\mu_B^2 \sum_{\alpha\beta} \sum_{\{l\}\{m\}} \frac{P_{i\alpha\downarrow,\{l\}}^* P_{j\beta\uparrow,\{l\}} P_{j\beta\downarrow,\{m\}}^* P_{i\alpha\downarrow,\{m\}}}{\omega - (E_{\{m\}} - E_{\{l\}})} [\Theta(E_f - E_{\{l\}}) - \Theta(E_f - E_{\{m\}})], \quad (13)$$

$$\chi_{i,j}^{-+0}(\omega) = -2\mu_B^2 \sum_{\alpha\beta} \sum_{\{l\}\{m\}} \frac{P_{i\alpha\downarrow,\{l\}}^* P_{j\beta\downarrow,\{l\}} P_{j\beta\uparrow,\{m\}}^* P_{i\alpha\downarrow,\{m\}}}{\omega - (E_{\{m\}} - E_{\{l\}})} [\Theta(E_f - E_{\{l\}}) - \Theta(E_f - E_{\{m\}})], \quad (14)$$

$$\chi_{i,j}^{++0}(\omega) = -2\mu_B^2 \sum_{\alpha\beta} \sum_{\{l\}\{m\}} \frac{P_{i\alpha\uparrow,\{l\}}^* P_{j\beta\downarrow,\{l\}} P_{j\beta\uparrow,\{m\}}^* P_{i\alpha\downarrow,\{m\}}}{\omega - (E_{\{m\}} - E_{\{l\}})} [\Theta(E_f - E_{\{l\}}) - \Theta(E_f - E_{\{m\}})], \quad (15)$$

$$\chi_{i,j}^{--0}(\omega) = -2\mu_B^2 \sum_{\alpha\beta} \sum_{\{l\}\{m\}} \frac{P_{i\alpha\downarrow,\{l\}}^* P_{j\beta\uparrow,\{l\}} P_{j\beta\downarrow,\{m\}}^* P_{i\alpha\uparrow,\{m\}}}{\omega - (E_{\{m\}} - E_{\{l\}})} [\Theta(E_f - E_{\{l\}}) - \Theta(E_f - E_{\{m\}})]. \quad (16)$$

Substituting the above functions into Eq. (9), we find that the normal modes of the elementary excitations can be obtained by diagonalizing the following Hermitian magnetic susceptibility matrix:

$$\begin{pmatrix} \chi_{i,j}^{+-0}(\omega) & \chi_{i,j}^{++0}(\omega) \\ \chi_{i,j}^{-+0}(\omega) & \chi_{i,j}^{--0}(\omega) \end{pmatrix}. \quad (17)$$

Thus, Eq. (9) becomes

$$2\mu_B \langle \tilde{S}_n^\pm \rangle e^{i\omega t} = \chi_n^{\pm 0}(\omega) \tilde{h}_n^\pm e^{i\omega t}, \quad (18)$$

where

$$\tilde{S}_n^\pm = \sum_i (Q_{in}^{\pm\pm})^* S_i^\pm + \sum_i (Q_{in}^{\pm\mp})^* S_i^\mp, \quad (19)$$

$$\tilde{h}_n^\pm = \sum_i (Q_{in}^{\pm\pm})^* h_i^\pm + \sum_i (Q_{in}^{\pm\mp})^* h_i^\mp, \quad (20)$$

in which $\chi_n^{\pm 0}(\omega)$ are the eigenvalues of the matrix (17), and matrices $\{Q_{in}^{\pm\pm}, Q_{in}^{\pm\mp}\}$ contain the corresponding eigenvectors.

Now we try to incorporate the *dynamic correlation corrections*. According to Hamiltonian (1), the e - e interactions contribute effectively an internal magnetic field which is proportional to the magnetization:

$$\tilde{h}_{\text{eff}}^\pm = \frac{J}{\mu_B} \tilde{\mu}. \quad (21)$$

Since the variations of the magnetization are small and the frequency is low (we only consider the low-lying excitations), we expect that *Friedel's argument*¹⁶ is valid, namely

the internal field can follow *instantaneously* any change of the magnetization. Thus, the transversely induced magnetization can be determined by

$$\begin{aligned} \Delta \vec{\mu} e^{i\omega t} &= \chi(\omega) \cdot \vec{h} e^{i\omega t} = \chi^0(\omega) (\vec{h} + \vec{h}_{\text{eff}}) e^{i\omega t} \\ &= \chi^0(\omega) \left(\vec{h} + \frac{J}{\mu_B} \Delta \vec{\mu} \right) e^{i\omega t}, \end{aligned} \quad (22)$$

where $\Delta \vec{\mu}$ denotes every normal excitation mode obtained in Eq. (18), and $\chi^0(\omega)$ the corresponding Hartree-Fock susceptibility. So, based on this argument, the magnetic susceptibility containing the dynamic correlation can be calculated by

$$\frac{1}{\chi_n^\pm(\omega)} = \frac{1}{\chi_n^{\pm 0}(\omega)} - \frac{J}{\mu_B^2}. \quad (23)$$

Since the poles of the magnetic susceptibility function correspond to the elementary excitation energies, we thus have the following equations:

$$\frac{J}{\mu_B} \chi_n^{\pm 0}(\omega) = 1 \quad (24)$$

to fix the elementary excitation spectrum $\omega_n(h)$ as a function of the external field.

One may check that whenever Eq. (24) has a solution $\omega = \omega_0$, there must exist another solution $\omega = -\omega_0$. Which one is the excitation energy? Generally, there are three different cases shown schematically in Fig. 1. In case (a), there are no real solutions of $1/\chi(\omega) = 0$ so that the excitation energy, if at all definable, must be imaginary. Both cases (b) and (c) have two real solutions, however, by comparing to

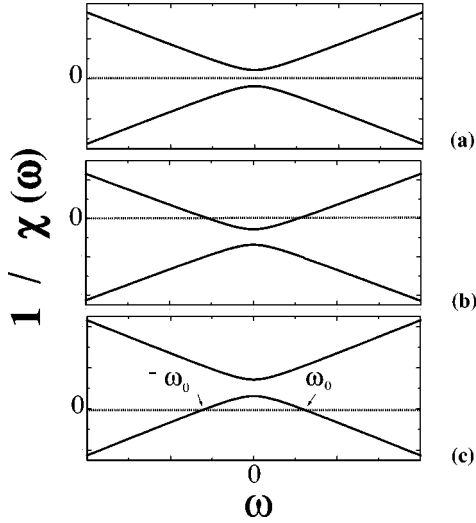


FIG. 1. Schematic illustration of three cases of the excitation energy: (a) imaginary, (b) negative, (c) positive.

the Lehmann representation²¹ of the magnetic susceptibility function, we find that (c) corresponds to a positive excitation while (b) to a negative excitation.

The minimum excitation energy is the energy gap which is a function of the external field: $\Delta(h) = \text{Min}[\hbar\omega_n(h)]$. According to Ref. 4, the gap of the elementary excitation is the order parameter which indicates the metastability of a given state: if the gap is positive, such a state is either stable or metastable because any fluctuation must raise the energy; on the other hand, when an external field is applied opposite to the magnetization and it is strong enough to make the gap negative or imaginary, the state must be unstable since some ‘‘soft modes’’ of the excitations will destroy the present spin

configuration. In the latter case, a spin reversal transition will take place. Thus the critical field h^c is determined by the condition

$$\Delta(h^c) = 0. \quad (25)$$

For a system with translation-invariant symmetry, a Fourier transformation is helpful to greatly simplify the above formulas. In the case that there is only one atom per unit cell, there is no charge transfer and the magnetization should be uniform. Thus, the Hamiltonian (1) under the HFA becomes

$$H = \sum_{\mathbf{k}} \sum_{\alpha\beta;\sigma\sigma'} \left[\left(\epsilon_{\alpha\beta}(\mathbf{k}) - \frac{J\mu\sigma}{\mu_B} - h\mu_B\sigma \right) \delta_{\sigma\sigma'} + \xi S_{\alpha\beta}^{\sigma\sigma'}(\mathbf{k}) \right] C_{\alpha\sigma}^+(\mathbf{k}) C_{\beta\sigma'}(\mathbf{k}) + N \frac{J\mu^2}{2\mu_B^2}, \quad (26)$$

and the self-consistent equation is

$$\mu = \frac{1}{N} \sum_{\mathbf{k}} \sum_{\alpha\sigma} \sum_{\beta\sigma'} \sigma |P_{\alpha\sigma,\beta\sigma'}(\mathbf{k})|^2 \Theta[E_f - E_{\beta\sigma'}(\mathbf{k})], \quad (27)$$

where matrix $P_{\alpha\sigma,\beta\sigma'}(\mathbf{k})$ is used to diagonalize the Hamiltonian matrix in subspace of \mathbf{k} whose form has been shown in Eq. (26). The effect of the direct Coulomb interaction U has been absorbed into the hopping parameters.

The magnetic susceptibility matrix (17) now turns to a collection of N independent 2×2 submatrices in the subspace of \mathbf{q} :

$$\begin{pmatrix} \chi^{+-0}(\omega, \mathbf{q}), & \chi^{++0}(\omega, \mathbf{q}) \\ \chi^{-+0}(\omega, \mathbf{q}), & \chi^{--0}(\omega, \mathbf{q}) \end{pmatrix}, \quad (28)$$

where

$$\chi^{+-0}(\omega, \mathbf{q}) = -\frac{2\mu_B^2}{N} \sum_{\alpha\beta} \sum_{\{l\}\{m\}} \sum_{\mathbf{k}} \frac{P_{\alpha\uparrow,\{l\}}^*(\mathbf{k}-\mathbf{q}) P_{\beta\uparrow,\{l\}}(\mathbf{k}-\mathbf{q}) P_{\beta\downarrow,\{m\}}^*(\mathbf{k}) P_{\alpha\downarrow,\{m\}}(\mathbf{k})}{\omega - [E_{\{m\}}(\mathbf{k}) - E_{\{l\}}(\mathbf{k}-\mathbf{q})]} \times \{ \Theta[E_f - E_{\{l\}}(\mathbf{k}-\mathbf{q})] - \Theta[E_f - E_{\{m\}}(\mathbf{k})] \}, \quad (29)$$

and the other matrix elements are given by similar expressions. After diagonalizing sub-matrices (28), one can use Eq. (24) to get the spin wave dispersion relations $\omega(\mathbf{q}, h)$ and the spin excitation gap: $\Delta(h) = \text{min}[\hbar\omega(\mathbf{q}, h)]$, and use Eq. (25) to determine the critical field for ISR.

In order to compare with the classical Stoner theory, the important equations in such a theory will be summarized here. Based on the perturbation theory, the anisotropy energy of a magnetic thin film is found to be¹²

$$H = U_0 - K_2 \cos^2 \theta - \vec{h} \cdot \vec{M} + o(\xi^4), \quad (30)$$

where $\theta=0$ corresponds to the perpendicular axis. Here, K_2 is the energy difference between the in-plane polarization and the perpendicular polarization states which can be calculated according to Eq. (4). If $K_2 > 0$ which means that the z axis is the easy axis, the critical field is

$$\tilde{h}^c = 2K_2/M. \quad (31)$$

On the other hand, if the z axis is the hard axis ($K_2 < 0$), fourth order contributions should be considered to calculate the critical field.

The uniaxial anisotropy model (30) has also been assumed for microcluster systems.²² The critical fields can thus be calculated similarly by Eq. (31) in Stoner theory.

III. APPLICATIONS

A. Transition-metal monolayers

Experimentally, it has been shown that the coercive fields of magnetic thin films might be much larger than those of the bulk materials, making them candidates for high-density recording systems. To illustrate the present approach, we have

TABLE I. Critical fields h^c of free standing Fe monolayers with lattice constants matching different substrates, compared with \tilde{h}^c which is estimated with Stoner theory.

Matching substrates	Ag(001)	Ag(111)	Pd(001)	Pd(111)
easy axis	z	z	z	y
$h^c \mu_B$ (meV)	0.718	0.0712	0.360	0.0193
K_2 (meV)	1.171	0.116	0.585	-0.0333
M (μ_B)	3.200	2.897	3.044	2.812
$\tilde{h}^c \mu_B$ (meV)	0.781	0.0800	0.384	$o(\xi^4)$

examined the critical fields of Fe, Co, and Ni monolayers with lattice structures matching various substrates.

In our calculations, the Slater-Koster hopping parameters¹⁹ are taken from Ref. 20, and a crystal-field parameter $\delta=0.5$ eV, which is the energy difference between the orbitals pointing outside the plane ($yz, zx, 3z^2-r^2$) and those lying in the plane (xy, x^2-y^2), is adopted according to Ref. 12 in order to take account of the reduced symmetry in monolayers compared to the bulk system. Five $3d$ and one $4s$ orbitals are considered so that the s - d hybridization effects are incorporated. By comparing to the spontaneous magnetization obtained by the first principles calculations for Fe, Co, Ni monolayers with lattice structures matching the Ag(001) substrate,²³ we fix the exchange parameter J to 0.24 eV (for Fe), 0.28 eV (for Co), and 0.36 eV (for Ni), respectively (see the spin polarization listed in Tables I–III). The SO interaction parameter ξ is chosen to be 0.05 eV, and over 10^4 \mathbf{k} points in the first irreducible Brillouin zone are considered in the numerical calculations. Using the above parameters, the critical fields for ISR in Fe, Co, and Ni monolayers with lattice structures matching Ag(001), Ag(111), Pd(001), and Pd(111) substrates are calculated and the results are listed in Tables I–III, together with the calculated anisotropy energy K_2 , the spin polarization M and the critical fields \tilde{h}^c estimated by the Stoner theory according to Eq. (31).

It is shown that the easy axes of Fe monolayers are perpendicular to the plane except for Fe/Pd(111), while those for Co and Ni series are all lying in the plane (y axis refers to an axis connecting two nearest neighbor sites in the plane). The calculated anisotropy energies and easy axes for Fe and Ni monolayers matching Ag(001) substrate are qualitatively consistent with the first principles calculations by Gay and Ritcher.⁷

From Tables I–III it is very interesting to find that (1) when the z axis is the easy axis, h^c is in fairly good agree-

TABLE II. Critical fields h^c of free standing Co monolayers with lattice constants matching different substrates, compared with \tilde{h}^c which is estimated with Stoner theory.

Matching substrates	Ag(001)	Ag(111)	Pd(001)	Pd(111)
easy axis	y	y	y	y
$h^c \mu_B$ (meV)	0.522	1.041	0.499	1.039
K_2 (meV)	-2.846	-4.093	-3.557	-4.053
M (μ_B)	2.160	1.873	1.996	1.800
$\tilde{h}^c \mu_B$ (meV)	$o(\xi^4)$	$o(\xi^4)$	$o(\xi^4)$	$o(\xi^4)$

TABLE III. Critical fields h^c of free standing Ni monolayers with lattice constants matching different substrates, compared with \tilde{h}^c which is estimated with Stoner theory.

Matching substrates	Ag(001)	Ag(111)	Pd(001)	Pd(111)
easy axis	y	y	y	y
$h^c \mu_B$ (meV)	0.245	0.445	0.125	0.106
K_2 (meV)	-1.936	-1.740	-1.256	-0.898
M (μ_B)	1.032	0.781	0.945	0.731
$\tilde{h}^c \mu_B$ (meV)	$o(\xi^4)$	$o(\xi^4)$	$o(\xi^4)$	$o(\xi^4)$

ment with \tilde{h}^c , although it is always a little smaller than \tilde{h}^c , (2) when the z axis is the hard axis, it is expected that the critical fields should be very small [$o(\xi^4)$] since the anisotropy within the x - y plane is at least the fourth order in ξ . However, we see from Tables II,III that the in-plane critical fields of Co and Ni series are not very small. Similar results can also be found in the first-principle calculations by Gay and Ritcher,⁷ where the in-plane anisotropy of Ni monolayer has the same order as the perpendicular anisotropy of Fe monolayer. This fact indicates that although the in-plane coercivity carries a factor of ξ^4 , its value is not necessary very small because of distinct band structures in different materials.

Some discussions are helpful to understand the results listed in Table I–III. According to Eq. (19), suppose the spin wave mode with the lowest excitation energy is of the following form:

$$\begin{aligned} \tilde{S}_{\mathbf{q}}^+ e^{i\omega t} &= A S_{\mathbf{q}}^+ e^{i\omega t} + B S_{\mathbf{q}}^- e^{i\omega t} \\ &= (A+B) S_{\mathbf{q}}^x e^{i\omega t} + i(A-B) S_{\mathbf{q}}^y e^{i\omega t}. \end{aligned} \quad (32)$$

Generally, the motion of spins will be an ellipsoidal polarization mode. However, two special cases are very interesting: (1) $B=0$, circular polarization and (2) $A=B$, linear polarization. In the case that the z axis is the easy axis, the lowest excitation mode is indeed a *nearly* circular polarization (in this case, we do find that B is very small and $\mathbf{q} \rightarrow 0$), so that the simple argument leading to Eq. (31) is reasonable enough to gain the main results although some quantitative corrections may exist according to our theory. Those corrections are due to high-order contributions, self-consistent treatment of the magnetization, and quantum fluctuations. On the other hand, when the easy-axis lies in the x - y plane, the lowest excitation mode is now a *nearly* linear polarization (in this case, B has the same order as A and $\mathbf{q} \rightarrow 0$) which describes the spin variations in the plane. In this case, if one can determine the energy function with respect to the spin direction correct to the fourth order in ξ , it is still possible to use a Stoner-like theory to analyze the critical fields. However, much more spin directions should be considered to fix the total energy function — for example, \hat{x} , \hat{z} , $\hat{x}+\hat{z}$, and $\hat{x}+\hat{y}$ spin directions should be considered in order to fix the anisotropy energy functions for a square monolayer up to fourth order.⁷ However, by using our theory, only one spin direction (the easy axis) needs to be examined, and all the high-order contributions and quantum effects have been incorporated automatically into the final results.

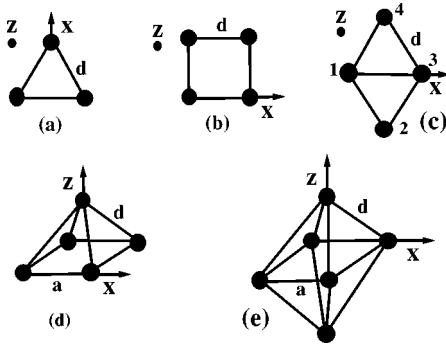


FIG. 2. Structures of Fe microclusters studied in this paper, z and x axes are the easy axes and the hard axes of the structures, respectively. In structures (d) and (e), we have $a=2d/\sqrt{3}$.

B. Magnetic microclusters

Recently, transition-metal magnetic microclusters have attracted much attention both theoretically^{13,22,24} and experimentally.²⁵ Pastor *et al.* have studied the structure and size dependence of the magnetic properties including the anisotropy energy for magnetic microclusters, by using the Haydock, Heine, and Kelly's recursion method based on the same Hubbard Hamiltonian.¹³ In this subsection, we will examine the microclusters as another illustration of the present method.

Structures shown in Fig. 2 for Fe microclusters have been studied with the help of the theoretical formulas developed in the first part of Sec. II. Taking the parameters from Ref. 13, we have calculated the critical fields h^c of those structures and listed the results in Table IV together with \tilde{h}^c estimated by the Stoner theory for comparison. The anisotropy energies leading to \tilde{h}^c are consistent with Ref. 13 although we are using the exact diagonalization method rather than the recursion method. We have also performed a test study for an isolated Fe atom, and found that the critical field is *indeed* zero.

It is very interesting to find that while in some cases h^c is very close to \tilde{h}^c [see structures (c) and (d)], in other cases, they are completely different. In fact, even for the same structure, things may be different if some other factors are changed. In Fig. 3, we show the critical fields h^c and the spin polarization M of structure (a) with respect to variation of the bond length, together with \tilde{h}^c for comparison. It is shown that the critical field of the microcluster depends sensitively on the bond length. In a definite spin polarization region, the critical field increases as d decreases. This can be explained

TABLE IV. Critical fields h^c of small Fe clusters whose structures are shown in Fig. 2 with interatomic bond length $d/d_0=1$, compared with \tilde{h}^c estimated with Stoner theory.

Structures	$h^c \mu_B$ (meV)	$\tilde{h}^c \mu_B$ (meV)	easy axis
(a)	1.727	2.711	z
(b)	2.038	3.623	z
(c)	0.184	0.185	z
(d)	0.580	0.592	z
(e)	1.656	3.226	z

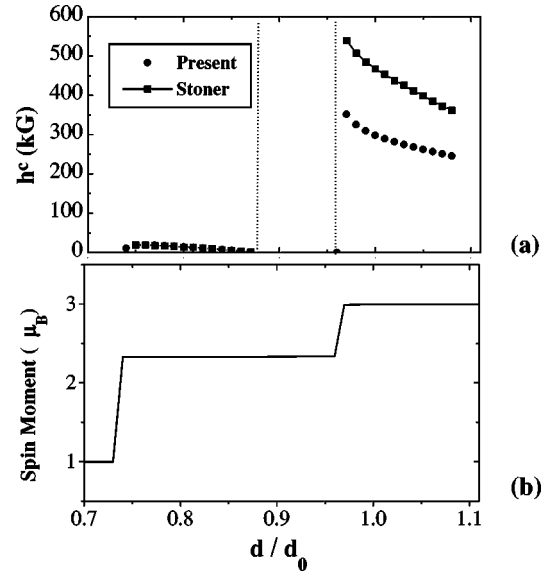


FIG. 3. (a) Critical fields h^c (along the z axis) of structure (a) as a function of the interatomic bond length where d_0 is the bond length of the bulk material, compared with those estimated with the classical Stoner theory. In the middle part of the figure, the z axis is not the easy axis of the microcluster so that the critical fields are not shown. (b) Spin polarization of structure (a) as a function of the interatomic bond length.

straightforwardly. When the atoms approach each other, the electronic hopping become strong so as to make the geometrically anisotropic nature of the z axis stronger, as a result, the critical field increases. In the limit of $d \rightarrow \infty$, the microcluster becomes effectively three isolated atoms so that all the axis become equivalent and the critical field approaches zero. As the atoms come close to each other, the effective "energy bandwidth" is enlarged leading to a sharp decrease of the spin polarization. It should also be noted that in a region near the spin polarization jump, the z axis is no longer a magnetic easy axis. The most interesting thing is that in the segment of $M \sim 3.0\mu_B$, the critical fields h^c are much smaller than those estimated by the Stoner theory \tilde{h}^c , while in the segment of $M \sim 2.33\mu_B$, the critical fields are almost exactly the same as those estimated by the Stoner theory (see Fig. 3).

How can we account for these differences? Let us examine structures (d) and (e) in the case of $d/d_0=1$. In Fig. 4, we show the critical fields of these two structures as functions of

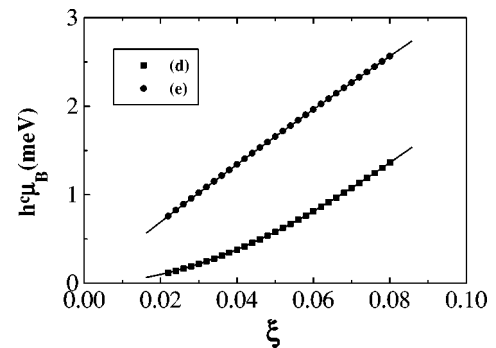


FIG. 4. Critical fields h^c of structures (d) and (e) as a function of ξ .

the SO interaction parameter ξ . It is interesting to see that the two structures have distinct ξ dependence. By fitting to polynomials, we find that for structure (d) $h^c = 4.8 \times 10^{-6} - 5.6 \times 10^{-4} |\xi| + 0.28 \xi^2 + \dots$, while for structure (e) $h^c = 9.2 \times 10^{-8} + 0.036 |\xi| - 0.064 \xi^2 + \dots$. It is thus shown that the first order contribution dominates in structure (e) while it can be neglected in structure (d). Similar first order contributions are also found in structure (a) ($M \sim 3.0$ segment) and structure (b). In such cases, the critical fields are certainly not consistent with the results coming from Eq. (31).

Basically, according to the nondegenerate perturbation theory, there are no first order contributions to the anisotropy energy since the diagonal elements are zero in the SO interaction. Things are different if degenerate energy levels exist at the Fermi surface. In this case, based on the present Hamiltonian (1), the SO interaction opens these degenerate levels so as to make the low-energy level occupied and the high-energy level unoccupied. The contribution to the anisotropy is thus first order in ξ . However, the present Hamiltonian (1), although widely adopted in the literature,¹²⁻¹⁴ is an oversimplified one in which the intra-atomic $e-e$ interaction U and J are assumed to be the same for different orbitals. If we adopt a more realistic $e-e$ interaction form

$$H_{ee} = \frac{1}{2} \sum_{i\alpha\beta} \left[U_{\alpha\beta} \hat{n}_{i\alpha} \hat{n}_{i\beta} - \frac{J_{\alpha\beta}}{\mu_B} \hat{\mu}_{i\alpha} \hat{\mu}_{i\beta} \right] \quad (33)$$

in the Hamiltonian, things may be different for such contributions coming from degenerate energy levels. A detailed work on this problem is now in process and will be presented elsewhere.²⁶

In microclusters, the spin reversal modes are not necessarily uniform because of the highly reduced symmetry. For example, for structure (c), we have the lowest spin excitation mode as

$$\begin{aligned} \tilde{S}_0^+ e^{i\omega t} = & a_1 (S_1^+ + S_3^+) e^{i\omega t} + a_2 (S_2^+ + S_4^+) e^{i\omega t} \\ & + a_1 (S_1^- + S_3^-) e^{i\omega t} + a_2 (S_2^- + S_4^-) e^{i\omega t}, \quad (34) \end{aligned}$$

with $a_1 = 0.351, a_2 = 0.356$. This effect, although not very significant for the present example, might be serious for other microclusters. In the latter cases, the classical Stoner treatment is questionable. Clearly, the present theory is helpful to obtain reasonable results.

IV. CONCLUSIONS

To summarize, we have established an atomistic theory to determine the critical field for intrinsic spin reversal in transition-metal magnetic systems within the framework of itinerant-electron magnetism. The Hubbard model with spin-orbit interaction has been used to describe a general transition-metal magnetic system, and the energy levels (energy bands) have been obtained by diagonalizing the Hamiltonian under a Hartree-Fock approximation. The spin-orbit interaction has been treated self-consistently on the same footing as the hopping and the electron-electron interacting terms, rather than perturbatively. By studying the transverse magnetic susceptibility function based on *Friedel's* arguments, the elementary excitation spectra are found to possess a positive gap if the spin is pointing along the easy axis. The spin excitation gap is the order parameter which indicates the metastability of a given state. When an external field is applied opposite to the spin direction and its strength is strong enough to overcome the gap, the present spin state is no longer metastable so that the spin reversal transition takes place which defines the critical field.

Fe, Co, and Ni monolayers with lattice structures matching Ag(001), Ag(111), Pd(001), and Pd(111) substrates have been examined. Comparison with the classical Stoner theory shows that quantitative corrections can be obtained from this theory, and less spin directions need to be considered in our theory than in the Stoner theory.

Applications to small microclusters have also been given. We show that the spin reversal mode needs not to be uniform, so that the Stoner theory is inadequate to estimate the critical fields of intrinsic spin reversal in small microclusters. However, by using the present theory, the critical field of intrinsic spin reversal in a transition-metal magnetic system can always be examined.

ACKNOWLEDGMENTS

This work was supported by the Japan Science and Technology Corporation. We are very grateful to Dr. Marcel Sluiter, Dr. Zhiqiang Li, and Dr. Keivan Esfarjani for helpful discussions. The authors would like to express their sincere thanks to the Materials Information Science Group of the Institute for Materials Research, Tohoku University, for their continuous support of the supercomputing facility.

¹In an ideal single-domain system, the critical field for intrinsic spin reversal is actually the coercive field. However, in this paper, we would rather not call this field the coercive field since this is not guaranteed in general.

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- ²⁶This problem is not serious in monolayers. Even when two degenerate energy levels exist at the Fermi surface (each level contains half an electron), the contribution, when divided by the total \mathbf{k} points adopted in the calculation, is very small. In our calculation, we have avoided such problems by choosing appropriate \mathbf{k} points.