Quantum Method for Calculating the Coercivities of Transition-metal Magnetic Systems

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A new method to estimate the coercivities of transition metal magnetic systems 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 interactions if the magnetization is in its easy axis. When a magnetic field is applied opposite to the spontaneous magnetization and if it is strong enough to overcome the excitation gap, a spin reversal transition will take place which defines the coercive field. Applications are given to some small magnetic microclusters.

Key words: coercivity, transition metal

1. Introduction

Historically, the classical method proposed by Stoner and Wohlfarth¹⁾ worked well for the coercivities of macromagnetic systems²⁾. Recently, since the size of the recording domain has become smaller and smaller and the structure more and more complicated, such a phenomenological theory seems to be inadequate due to large quantum fluctuations. Within the framework of the local magnetic moment model (LMMM), we recently proposed a quantum method for coercivity³⁾, and applied it to study various kinds of recording systems⁴⁾. Unfortunately, since the real recording system is usually made of transition metals which can not be well described by the LMMM, the present theory is still inadequate for transition metals. Although many efforts have been devoted to studying the anisotropies of transition-metal magnetic systems from the view of itinerant-electron magnetism (IEM)⁵⁾⁻⁸⁾, and the anisotropy is really *closely related* to the coercivity, a theory which can be used directly to estimate the coercivity still seems not existing. The present work is devoted to establishing the quantum theory of coercivity for transition-metal magnetic systems in IEM. Applications will be given to small magnetic microclusters, and the comparison with the classical theory for coercivity will be presented.

2. Formalism of the theory

The Hamiltonian describing a transition-metal magnetic material is given by:

$$H = \sum_{ij, \alpha\beta, \sigma} t_{ij}^{\alpha\beta} C_{i\alpha\sigma}^+ C_{j\beta\sigma} + \xi \sum_{i, \alpha\beta} \sum_{\sigma_1 \sigma_2} S_{\alpha\beta}^{\sigma_1 \sigma_2} C_{i\alpha\sigma_1}^+ C_{i\beta\sigma_2} + \sum_{i\alpha\sigma} (U n_i - J \sigma \mu_i / (2\mu_B^2)) C_{i\alpha\sigma}^+ C_{i\alpha\sigma}, \quad (1)$$

where the first one is the hopping term, the second one the spin-orbit (SO) interaction, and the last one the intra-atomically electron-electron (e-e) interaction presented under a Hartree-Fock approximation. Apply a unitary transformation

$$\begin{cases} C_{i\alpha\sigma}^+ = \sum_{\{l\}} P_{i\alpha\sigma, \{l\}}^* \tilde{C}_{\{l\}}^+ \\ C_{i\alpha\sigma} = \sum_{\{l\}} P_{i\alpha\sigma, \{l\}} \tilde{C}_{\{l\}} \end{cases} \quad (2)$$

to diagonalize the Hamiltonian, we find the electronic eigenstates (energy bands) of the system:

$$H = \sum_{\{l\}} E_{\{l\}} \tilde{C}_{\{l\}}^+ \tilde{C}_{\{l\}} \quad (3)$$

where $\{l\}$ denote a set of quantum number $\{j\beta\sigma'\}$. Thus, we have

$$\begin{aligned} n_i &= \sum_{\alpha\sigma\{l\}} |P_{i\alpha\sigma, \{l\}}|^2 \Theta(E_f - E_{\{l\}}) \\ \mu_i &= \sum_{\alpha\sigma\{l\}} \sigma |P_{i\alpha\sigma, \{l\}}|^2 \Theta(E_f - E_{\{l\}}) \end{aligned} \quad (4)$$

to determine n_i and μ_i self-consistently where E_f is the fermi energy.

We need to know the elementary excitations in such systems. The low-lying excitations should be small *transverse* variations of the magnetization which can be obtained by studying the transverse magnetic susceptibility function. Suppose the magnetization is in z direction and we have some transverse disturbing field (h_i^x, h_i^y) , then the perturbation energy is

$$\delta H = (\mu_B/2) \sum_i (h_i^+ S_i^- + h_i^- S_i^+) e^{i\omega t}$$

where

$$h_i^\pm = h_i^x \pm i h_i^y, \quad S_i^\pm = S_i^x \pm i S_i^y. \quad (5)$$

According to Kubo's linear response formulation⁹⁾, the induced magnetization can be measured by:

$$\begin{aligned} 2\mu_B \langle S_i^+ \rangle e^{i\omega t} &= \left[\sum_j \chi_{ij}^{+-}(\omega) h_j^+ + \sum_j \chi_{ij}^{++}(\omega) h_j^- \right] e^{i\omega t} \\ 2\mu_B \langle S_i^- \rangle e^{i\omega t} &= \left[\sum_j \chi_{ij}^{-+}(\omega) h_j^- + \sum_j \chi_{ij}^{--}(\omega) h_j^+ \right] e^{i\omega t} \end{aligned} \quad (6)$$

where the magnetic susceptibilities are defined by some spin correlation functions⁹⁾:

$$\chi_{ij}^{\pm\pm}(t) = -2\mu_B^2 \langle \langle S_i^{\pm}(t) | S_j^{\pm} \rangle \rangle. \quad (7)$$

Because of the many-particle effects introduced by the e-e interaction, the above correlation functions can not be evaluated exactly so that some approximations are necessary. Here we follow the arguments made by Friedel¹⁰⁾ to determine first the *Hartree-Fock* susceptibility in which the *dynamic correlation corrections* have been neglected. For example, one component of the *Hartree-Fock* susceptibility is:

$$\chi_{i,j}^{+-}(\omega) = 2\mu_B^2 \sum_{\alpha\beta\{l\}\{m\}} \frac{P_{i\alpha\uparrow,\{l\}}^* P_{j\beta\uparrow,\{l\}} P_{j\beta\downarrow,\{m\}}^* P_{i\alpha\downarrow,\{m\}}}{\hbar\omega - (E_{\{m\}} - E_{\{l\}})} [\Theta(E_f - E_{\{l\}}) - \Theta(E_f - E_{\{m\}})]. \quad (8)$$

According to Eq. (6), the normal excitation modes can be obtained by diagonalizing the following magnetic susceptibility matrix:

$$\begin{pmatrix} \chi_{i,j}^{+-}(\omega) & \chi_{i,j}^{++}(\omega) \\ \chi_{i,j}^{--}(\omega) & \chi_{i,j}^{+-}(\omega) \end{pmatrix}. \quad (9)$$

Then we can incorporate some dynamic correlations. According to Hamiltonian (1), the e-e interactions contribute effectively an internal magnetic field which is proportional to the magnetization

$$\tilde{h}_{eff} = (J/2\mu_B^2)\tilde{\mu}, \quad (10)$$

if the variations of the magnetization are small and the frequency ω is low enough, one can expect that the internal field can follow *instantaneously* any changes of the magnetization. So, the induced magnetization can be determined by the following equation¹⁰⁾:

$$\Delta\tilde{\mu} = \chi \cdot \tilde{h} = \chi_{HF} \cdot [\tilde{h} + (J/2\mu_B^2)\Delta\tilde{\mu}]. \quad (11)$$

Thus, we have

$$1/\chi(\omega) = 1/\chi_{HF}(\omega) + (J/2\mu_B^2) \quad (12)$$

to relate the magnetic susceptibility $\chi(\omega)$ which we need to the *Hartree-Fock* susceptibilities χ_{HF} which has been obtained from diagonalizing the matrix (9). Remembering that the poles of the magnetic susceptibility $\chi(\omega)$ correspond to the excitation energies, we have the following equation

$$(J/2\mu_B^2) \cdot \chi_{HF}(\omega) = 1 \quad (13)$$

to determine the elementary excitation spectrums.

Generally, there are three kinds of excitations shown schematically in Fig. 1 where $1/\chi(\omega)$ as the functions of ω has been illustrated. In case (a), there are no real solutions of $1/\chi(\omega) = 0$ so that the excitation energy, if can be defined, must be imaginary. Both case (b) and (c) correspond to two solutions $\pm\omega_0$ due to the symmetry properties of the χ function. However, a detailed analysis shows that (c) corresponds

to a positive excitation while (b) to a negative excitation. Thus, we can determine the elementary excitation spectrums $E_n(h) = \hbar\omega_n$ as a function of the external field.

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

The minimum of the elementary excitation is defined as the energy gap: $\Delta(h) = \text{Min}[E_n(h)]$. According to the arguments in Ref. 3, the gap of the elementary excitation, induced by the anisotropically SO interaction, is the order parameter to monitor the metastability of the state. If there are a positive gap in the elementary excitations, such a state must be metastable since any fluctuations will raise the energy. Otherwise, the state is unstable. So, the coercivity is determined with the condition $\Delta(h^{\text{coer}}) = 0$.

3. Applications

As the illustrations of the above-described method, we have studied an Iron triangular magnetic microcluster whose geometry is shown in the inset of Fig. 2(b). The parameters are selected to be: $U = 5.4\text{eV}$, $J = 0.76\text{eV}$, $\xi = 0.05\text{eV}$, and the hopping parameters are assumed to be the same as Ref. 8. The coercivity h_c along z axis and the spin polarization M have been shown in Fig. 2 as the functions of the bond length where d_0 is the bond length of bulk material.

It is clear that the coercivity of the microcluster depends on the bond length sensitively. In a definite spin polarization region, the coercivity increases as d decreases. This is easy to be understood. When the atoms approach each other, the electronic hopping become strong so as to make the geometrically anisotropic nature of z axis stronger, as the result, the coercivity increases. In the limit of $d \rightarrow \infty$, the mi-

crocluster becomes effectively three isolated atoms so that all the axis become equivalent. However, shrinking of the bond length may lead to a decrement of the spin polarization. Because of the finite size, such a jump might be very significant (see, from $3.0\mu_B$ to $2.333\mu_B$). Under a small spin polarization, the states which have contributions to the magnetism are certainly small. As the result, the coercivity of the system in this region is much smaller than those in the high-polarization region. It also should be noted that in a region near the spin polarization jump, the z axis is no longer a magnetic easy axis.

Fig. 2 Coercivity h_c along z axis and the spin polarization M as the functions of the bond length

We have compared the present method with the classically Stoner theory for coercivity. In the latter case, the anisotropy is assumed to be $D(\vec{S} \cdot \vec{z}^0)^2$ where $\vec{z}^0$ is the magnetic easy-axis¹¹⁾. With the help of a standard variation analysis, one obtains the coercivity $h^c = 2SD$ in this approach. So, if one have got the magnetic anisotropy energy (K) which is the energy difference between the hard axis state and the easy axis state, one can use $h^c = 2K/S$ to estimate the coercivity in a Stoner approach. Comparison shows that the coercivity calculated by the present approach is *much smaller* than those estimated by the Stoner theory. For example, for $d/d_0 = 1$, the coercivity is estimated to be $592KG$ (According to the calculations in Ref. 8) by a Stoner-type theory, while it is only $300KG$ in the present quantum theory (see Fig. 2(a)). This is because (1) the uniaxial anisotropy model is not strictly valid for the present systems especially due to the mixing effect between spin-up and spin-down states, and (2) the classical approach fails to consider

more appropriate elementary excitation modes which may possess lower energy. Thus, the quantum theory of coercivity developed here can be used to capture more quantum effects which has been neglected by a classical theory.

4. Conclusions

To summarize, the quantum theory of coercivity has been established for transition-metal magnetic systems within the framework of itinerant electron magnetism. Some small magnetic microclusters have been studied as the illustration of the method. It is shown that more quantum effects can be captured in the new method compared to the original classical one. The coercivity is sensitive to the lattice length of the microclusters.

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