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2000 J. Phys.: Condens. Matter 12 2811

(http://iopscience.iop.org/0953-8984/12/12/319)

J. Phys.: Condens. Matter **12** (2000) 2811–2818. Printed in the UK PII: S0953-8984(00)07879-6

Local stiffness and stability of the ferromagnetic state in metals

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Received 16 September 1999, in final form 4 February 2000

Abstract. The concept of local stiffness associated with transverse local spin fluctuations is introduced and used to investigate the coupling between the magnetization of atomic planes in the ferromagnetic state of a metallic material. It is shown that the coupling strength in the surface region may differ significantly from that in the bulk, and the possibility of an antiferromagnetic alignment between surface and bulk magnetizations is pointed out. A comparison between calculated values of local and spin-wave stiffnesses is presented, and the stability of the ferromagnetic state against both short- and long-wavelength spin fluctuations is discussed.

1. Introduction

The exchange interaction between itinerant electrons may lead to the formation of local magnetic moments in metals. The nature of the coupling between these moments—its sign, strength, and range—depends on the electronic structure of the system, which is determined by its composition and atomic arrangement. The study of interaction between magnetic moments has been one of the main issues in metallic magnetism, and a great deal of attention has been concentrated on the subject, both theoretically and experimentally.

Information about the magnetic coupling in ferromagnets can be obtained from the behaviour of the magnetization M in the low-temperature T-region, where Bloch's law, $M(T)/M(0) \simeq 1 - CT^{3/2}$, is observed. The pre-factor C depends on the strength of the coupling. This is directly seen in the case of the simple Heisenberg Hamiltonian

$$H = -\sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

for localized spin systems, where J_{ij} is the exchange coupling between spins $\vec{S}_i$ and $\vec{S}_j$, on sites i and j, respectively. Within the random-phase approximation and for a homogeneous system with nearest-neighbour interactions only, we find for the bulk magnetization that

$$C = \zeta\left(\frac{3}{2}\right)\frac{g\mu_B}{M(0)^2}\left(\frac{k_B}{\pi S a^2 J}\right)^{3/2} \tag{1}$$

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0953-8984/00/122811+08$30.00

where ζ is the Riemann zeta function, g is the gyromagnetic factor, μ_B is the Bohr magneton, k_B is the Boltzmann constant, S is the spin magnitude, and a is the lattice parameter. Thus, the stronger the coupling J, the slower the reduction of M with temperature.

The sensitivity of the coupling with the atomic arrangement is well illustrated by the difference between the temperature dependences of the surface (M_S) and bulk (M_B) magnetizations of a ferromagnet. For sufficiently low T, it has been shown [1, 2] that both M_S and M_B follow Bloch's law, with different pre-factors C_S and C_B. Assuming that the magnetization and exchange interactions are uniform throughout the system, it is possible to show, using rather general arguments, that $C_S = 2C_B$ [1,2]. The factor 2 is purely geometrical and is a result of environmental differences between surface and bulk sites (at the surface, there are neighbouring sites on just one of its sides). However, for several systems, surface magnetization measurements with spin-polarized electrons yield other values for the ratio C_S/C_B [3–6]. This is a consequence of changes in the local electronic structure and magnetic interactions introduced by the surface in its proximity. Using localized Heisenberg models with nearest-neighbour interactions only, Mathon and Ahmad [7] have shown that changes in the coupling J_S between the surface plane and the rest of the system lead to a pseudo-$T^{3/2}$ law for the surface magnetization, with $C_S \neq 2C_B$. In fact, on the basis of this simple model and by treating the ratio between J_S and the bulk exchange interactions J_B as an adjustable parameter, Mathon and Ahmad [7] and Scholl *et al* [6] have succeeded in reproducing experimental $M_S(T)$ data for several permalloy systems. Such a phenomenological approach highlights the need for a proper calculation of the exchange interactions in metallic systems, on the basis of their electronic structure.

The above results have motivated the present study of the coupling between the magnetization of a given atomic plane and the rest of the system in itinerant-electron ferromagnets. Our initial aim was to investigate how this coupling changes as one goes from the bulk to the surface. The theory that we developed has naturally led to the definition of a local stiffness D_l of the system with respect to transverse local spin fluctuations, which proved useful to our investigation. We have also found that such a stiffness constant provides the basis for an additional stability criterion for the ferromagnetic state.

We arrange this paper as follows. In section 2 we develop the theory and introduce the concept of local stiffness. We then focus on a simple model and present in section 3 results on D_l for planes both in the bulk and at the surface. In section 4 we discuss the stability of the ferromagnetic state against both local and extended transverse spin fluctuations. Finally, in section 5, we present our conclusions.

2. Local stiffness constant

We consider the ferromagnetic state of an interacting itinerant-electron system, with its magnetization pointing in the $\hat{z}$-direction. We assume that the electron–electron interaction is described by an effective local exchange field, acting on the magnetic carriers, which is parallel to the local magnetization direction. In order to estimate the strength of the coupling between the magnetization M_l in a given atomic plane l and the rest of the system, we calculate the energy necessary to rotate the direction of M_l by an angle θ relative to $\hat{z}$, as represented in figure 1.

Such a quantity can be calculated from the thermodynamic potential [8–10]

$$\Omega = -\frac{1}{\beta}\int \mathrm{d}\omega \, \ln\{1 + \mathrm{e}^{\beta(\mu-\omega)}\}\left(-\frac{1}{\pi}\,\mathrm{Im}\,\mathrm{Tr}\sum_i [G_{ii}(\omega)]\right) \tag{2}$$

where $\beta = 1/k_BT$, μ is the chemical potential, $[G_{ii}(\omega)]$ is the Green function matrix for

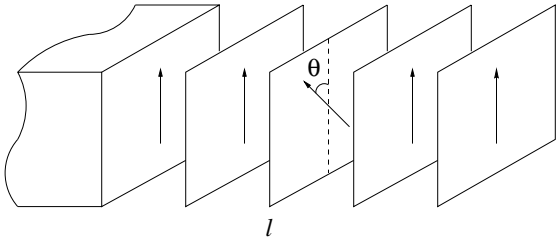

Figure 1. A schematic representation of a ferromagnet where the magnetization in plane l is rotated by an angle θ with respect to that of the rest of the system.

electrons with up and down spins at site i, and Tr stands for the trace over spin and atomic orbital indices. The rotation corresponds to a change in the direction of the local exchange field, with a consequent variation $V(\theta)$ of the local electron potential. Assuming that the atomic planes are parallel to the zx-plane, one has

$$[V(\theta)] = -\{[\cos(\theta) - 1]\sigma_z + \sin(\theta)\sigma_x\} \otimes [V_{ex}] \tag{3}$$

where σ_z and σ_x are the usual Pauli matrices, and $[V_{ex}]$ is a matrix in orbital indices representing the strength of the local exchange potential. The change $\Delta\Omega_l(\theta)$ can be written as

$$\Delta\Omega_l(\theta) = \frac{1}{\pi\beta}\sum_{\boldsymbol{q}}\int \mathrm{d}\omega\, \ln\{1 + \mathrm{e}^{\beta(\mu-\omega)}\}\,\mathrm{Im}\,\mathrm{Tr}\sum_{p}[\Delta G_{pp}(\boldsymbol{q},\omega,\theta)] \tag{4}$$

where $\boldsymbol{q}$ is a two-dimensional wave vector parallel to the planes and belonging to the two-dimensional Brillouin zone, and p is a plane index. It is easy to show that

$$\Delta G_{pp}(\theta) = [g_{pl}][T_{ll}(\theta)][g_{lp}] \tag{5}$$

where $[g] = [G(\theta = 0)]$ is the Green function of the ferromagnetic state,

$$[T_{ll}(\theta)] = [V(\theta)]\{[I] - [g_{ll}][V(\theta)]\}^{-1} \tag{6}$$

is the scattering matrix associated with $[V(\theta)]$, and $[I]$ represents the identity matrix in spin and orbital indices. To simplify the notation, we have omitted the arguments $\boldsymbol{q}$ and ω in the above two equations. It follows that

$$\Delta\Omega_l(\theta) = \frac{1}{\pi}\sum_{\boldsymbol{q}}\int \mathrm{d}\omega\; f(\omega)\,\mathrm{Im}\,\mathrm{Tr}\ln\{[I] - [g_{ll}(\boldsymbol{q},\omega)][V(\theta)]\} \tag{7}$$

which reduces to [10]

$$\Delta\Omega_l(\theta) = \frac{1}{\pi}\sum_{\boldsymbol{q}}\int \mathrm{d}\omega\; f(\omega)\,\mathrm{Im}\,\mathrm{tr}\ln\{[\mathcal{I}] + (\cos(\theta) - 1) \times [V_{ex}]([g_{ll}^{\uparrow}] - [g_{ll}^{\downarrow}] + 2[g_{ll}^{\uparrow}][V_{ex}][g_{ll}^{\downarrow}])\}. \tag{8}$$

Here tr stands for the trace over orbital indices only, $f(\omega)$ is the Fermi function, $[\mathcal{I}]$ is the identity matrix in orbital indices, and $[g_{ll}^{\sigma}(\boldsymbol{q},\omega)]$ is the propagator for an electron with spin σ in the ferromagnetic state.

For small values of θ, charge fluctuations can be ignored and the change in the thermodynamic potential can be written as $\Delta\Omega_l(\theta) \simeq D_l\theta^2$, where

$$D_l = -\frac{1}{2\pi}\sum_{\boldsymbol{q}}\int_{-\infty}^{\infty}\mathrm{d}\omega\; f(\omega)\,\mathrm{Im}\,\mathrm{tr}\{[V_{ex}]([g_{ll}^{\uparrow}] - [g_{ll}^{\downarrow}] + 2[V_{ex}][g_{ll}^{\uparrow}][g_{ll}^{\downarrow}])\}. \tag{9}$$

D_l represents a local stiffness of the system against local transverse spin fluctuations, and measures the strength of the coupling between the magnetization M_l and the rest of the system.

Information about the electronic structure is contained in the local one-electron propagators $g_{ll}^{\uparrow(\downarrow)}$ for $\uparrow$ ($\downarrow$) spins in plane l. D_l depends upon surface orientation and varies with l, approaching a bulk value D_B for $l \to \infty$. It is to be distinguished from the usual stiffness constant D_{sw} which enters the dispersion relation of long-wavelength spin waves (extended transverse spin fluctuations in the ferromagnetic state). Both D_l and D_{sw} are related to the exchange interactions between local magnetic moments, but they reflect the rigidity of the system to different types of excitations. The relation between D_l and the exchange couplings is most clearly seen in the case of the ferromagnetic Heisenberg model. The rotation of M_l with respect to the magnetization of the rest of the system leads to the definition of a local stiffness given by

$$D_l = \frac{1}{2} \sum_{i \in l,\, j \notin l} J_{ij} S_i S_j. \tag{10}$$

It is clear that the dependence of D_l upon l provides information on the variation of the exchange coupling across the system. In particular, it can be used to investigate the local stiffness in the bulk and at the surface. To illustrate this point, we present in the next section results on D_l for a simple model.

3. Calculation of D_l

We consider a single-band tight-binding model on a simple cubic lattice, with nearest-neighbour hoppings only. We keep just on-site electron–electron interaction U, and use the Hartree–Fock approximation. We assume that the one-electron self-consistent equations have a ferromagnetic solution and, in this case, the Hamiltonian for electrons with spin σ reads

$$H_\sigma = \sum_i (\epsilon_i - \sigma V_{ex}) c_{i\sigma}^\dagger c_{i\sigma} + \sum_{i,j} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} \tag{11}$$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) is the operator which creates (destroys) an electron with spin σ on site i, ϵ_i and t_{ij} are the site and hopping energies, and $V_{ex} = Um/2$. Here $m = n_\uparrow - n_\downarrow$ is the reduced magnetization, which we assume to be uniform throughout the system, with n_σ representing the number of electrons with spin σ on each site. The exchange splitting between the up- and down-spin bands is equal to $2V_{ex}$.

We assume that the atomic planes are perpendicular to the (010) direction. It is interesting to look at the dependence of D_l on the band filling, for fixed values of V_{ex}. We focus on two particular cases, corresponding to planes at the surface and in the bulk. The energy scale has been chosen such that $2t = 1$. Figure 2 presents results for D_S and D_B as functions of the Fermi energy E_F, for $V_{ex} = 0.5$ (a) and 1.0 (b). The curves are symmetric with respect to E_F, and $E_F = 0$ corresponds to band half-filling. We remark that on fixing the value of V_{ex} we are assuming that for each E_F the on-site interaction U is such that a ferromagnetic solution exists. Therefore, negative values of D_S and D_B mean that the ferromagnetic ground state is unstable against a local infinitesimal rotation of the surface and bulk plane magnetizations, respectively. Such instabilities suggest a tendency for a local antiferromagnetic alignment, and are expected to depend upon the Fermi energy. To make the comparison between D_S and D_B meaningful we have multiplied the former by 2, since the surface magnetization is coupled to planes on just one of its sides. We notice that the two curves have similar behaviour: both oscillate between positive and negative values as a function of E_F. However, the magnitudes of the two stiffnesses are not equal, which highlights the difference in strength between the inter-plane couplings at the surface and in the bulk. This difference results from changes in

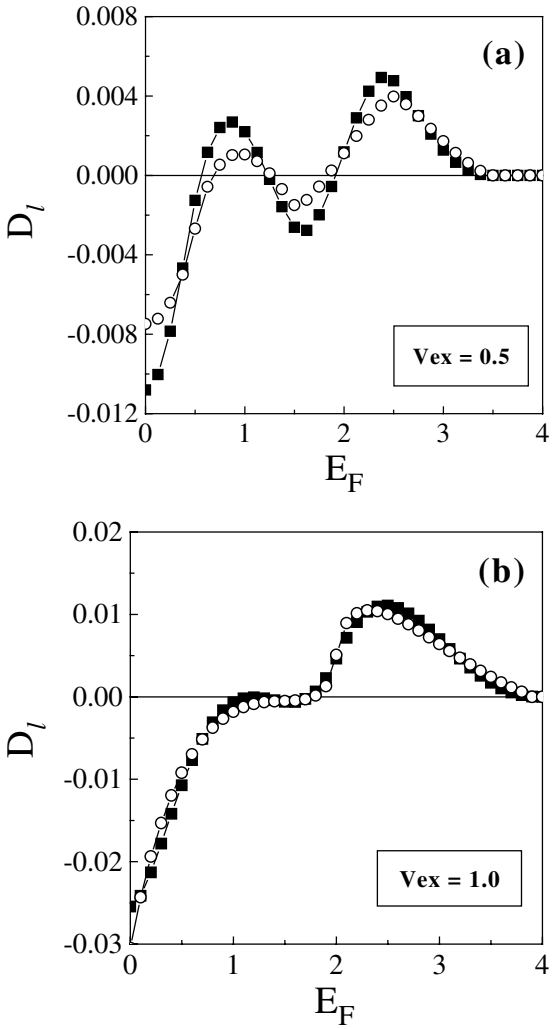

Figure 2. Calculated values of D_B (open circles) and $2D_S$ (filled squares) as functions of the Fermi energy E_F, for $V_{ex} = 0.5$ (a) and 1.0 (b). The energy scale is such that the hopping $t = 0.5$. The curves are symmetric with respect to E_F, and $E_F = 0$ corresponds to band half-filling.

the local electronic structure of the system near the surface. We notice in figure 2 that the coupling at the surface may be either weaker or stronger than that in the bulk.

Thus, the present calculations provide support to previous works on the temperature dependence of the surface magnetization of ferromagnets, which assumed either softening or strengthening of the surface exchange [6, 7, 11]. It should be stressed that a negative value of D_B indicates that the corresponding ferromagnetic solution of the one-electron self-consistent equation is not stable with respect to local transverse spin fluctuations. Indeed, the result $D_B < 0$ for $E_F = 0$ is in agreement with the fact that the ground state of the half-filled one-band model on a simple cubic lattice is expected to be antiferromagnetic, rather than ferromagnetic.

It is interesting to notice that for some values of E_F, for which D_B is positive, we may find negative values of D_S. This indicates the possibility of an antiferromagnetic alignment of the

surface magnetization with respect to a ferromagnetic bulk. In other words, the mere existence of a free surface may yield an antiferromagnetic coupling between the surface plane and the rest of the system. In real systems, however, other effects such as surface imperfections and anisotropies [12–15], may also strongly influence the surface magnetic ground state, and must be taken into account in a more comprehensive analysis. Appraisal of the relative importance of such effects in a particular system requires good experimental surface characterization [12].

4. The stability criterion for the ferromagnetic state

The stability of the ferromagnetic state against long-wavelength spin-wave excitation can be investigated by calculating the spin-wave stiffness constant D_{sw}. A strong positive value of D_{sw} indicates stability of the ferromagnetism. However, the stiffness constant D_B introduced above provides us with an additional stability condition for the ferromagnetic state. Clearly, the ferromagnetic state must be stable against both extended long-wavelength ($D_{sw} > 0$) and localized ($D_B > 0$) transverse spins fluctuations. But one could rightly argue over whether these two conditions are in fact independent. In other words: does stability against one of these fluctuations imply stability against the other? In order to answer these questions, we have evaluated D_{sw} and D_B for the same ferromagnetic state.

The spin-wave stiffness can be obtained from the ferromagnetic band-structure of the material. For the Hamiltonian in equation (10) and at $T = 0$, D_{sw} is given by [16]

$$D_{sw} = \frac{2V_{ex}}{3m}\left[\sum_{\varepsilon_k<\mu_+}\left(\frac{\nabla^2\varepsilon_k}{4V_{ex}} - \frac{|\nabla\varepsilon_k|^2}{4V_{ex}^2}\right) + \sum_{\varepsilon_k<\mu_-}\left(\frac{\nabla^2\varepsilon_k}{4V_{ex}} - \frac{|\nabla\varepsilon_k|^2}{4V_{ex}^2}\right)\right] \tag{12}$$

where ε_k is the paramagnetic band-structure, m is the magnetic moment, and $\mu_\pm = E_F \pm V_{ex}$.

Figure 3 shows D_B (full line) and D_{sw} (dashed line) as functions of E_F for V_{ex} equal to 0.5 (a) and 1.0 (b). Since the two constants have different dimensions, they have been normalized by their respective values at $E_F = 0$. For the two values of V_{ex}, the behaviour of D_{sw} with E_F is qualitatively the same, showing a single region of instability of the ferromagnetic state, a near-half-filled band. We notice that the width of this region increases with V_{ex}. As regards D_B, the change in the shape of the curve with V_{ex} is more pronounced and interesting. For $V_{ex} = 0.5$ we find two stability regions (with their counterparts for $E_F < 0$), the first of which is no longer present for $V_{ex} = 1.0$. However, the most significant point is the fact that, in the two cases, the regions of positive values of D_{sw} and D_B do not necessarily coincide. This means that the same system may be stable with respect to one type of spin fluctuation but not with respect to the other. It follows that the conditions for the stability of the ferromagnetic state are indeed more restrictive than those predicted solely by imposing $D_{sw} > 0$. It is certainly possible for the ferromagnetic state to be unstable with respect to different excitations, particularly if the stable ground state is antiferromagnetic, a spin-density wave, or has spiral structure. A more comprehensive analysis would involve investigating the stability against other spin perturbations, but this is much more laborious, particularly when surface effects are present.

5. Conclusions

In this paper we have investigated the coupling between the magnetizations of given atomic planes in the ferromagnetic state of metallic systems. We have introduced the concept of a local stiffness constant associated with local transverse spin fluctuations, which was used to study some surface effects on the exchange couplings. Results for a simple model

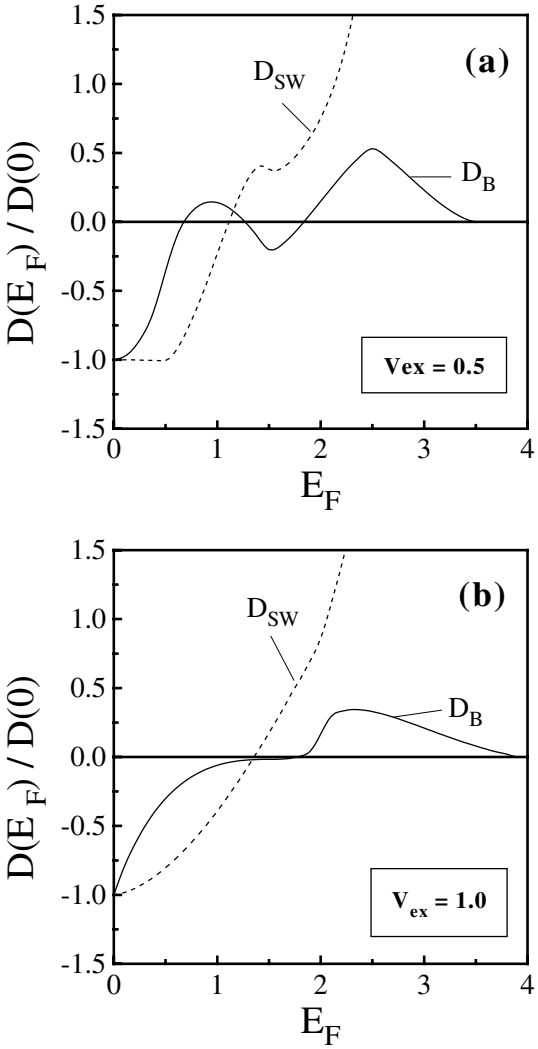

Figure 3. D_B (full line) and D_{sw} (dashed line) as functions of E_F for a ferromagnetic system for $V_{ex} = 0.5$ (a) and 1.0 (b). The energy scale is such that the hopping $t = 0.5$. Both constants are normalized by their respective values at $E_F = 0$.

indicate that the coupling between the surface plane and the rest of the system can be either stronger or weaker than that in the bulk. This result supports assumptions made in previous works [6,7,11] to explain observed relations between the temperature dependence of the surface and bulk magnetizations in several systems. Finally, we have proposed an additional stability criterion for the ferromagnetic state, which verifies its stiffness against localized transverse spin fluctuations. The criterion is relatively simple to examine, and may restrict the stability range obtained solely by spin-wave analysis. As far as ferromagnetic surface stability is concerned, in addition to the local transverse spin-fluctuation stiffness discussed here, other effects such as surface anisotropies, the existence of roughness, impurities, and lattice relaxations, as well as formation of magnetic domains, may be very important, and must be taken into account.

Acknowledgments

We thank D M Edwards and J Mathon for helpful discussions. This work was partially supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Brazil, and Fundação de Amparo a Pesquisa do Estado do Rio de Janeiro (FAPERJ).

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