Collective Electronic Excitations in Ferromagnetic Metals

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Abstract. Collective electronic excitations in the system of interacting conduction electrons of ferromagnetic metals (Fe, Co, Ni) are investigated. These conduction electrons stem from relatively narrow d-type bands and a suitable model to describe them is the Hubbard model. The tensor of the magnetic susceptibility is calculated within the Hubbard model. While the poles of the diagonal contribution represent the famous Stoner criterion for the stability of magnetic order the transverse susceptibility yields spin wave energies representing magnons. Magnetizations m as a function of temperature T and occupation number n, Curie temperatures T_C, and the temperature dependent exchange splitting ΔE_{ex} are numerically evaluated within reasonable agreement with experimental results.

1. Introduction

In ferromagnetic transition metals, e.g. Fe, Co, Ni both magnetism and the electric current are caused by the same electron group which stem from relatively narrow d-type conduction bands. Band ferromagnetism is a consequence of strong electron correlations where the correlation energy is the deviation of the exact ground state energy of the interacting electron system from the corresponding Hartree-Fock result and can only be approximately determined. Furthermore, correlation effects are not properly taken into account by standard band theory so that a full microscopic explanation of band ferromagnetism is still not available yet.

Even though density functional theory DFT is in principle an exact ground state theory its local density approximation LDA seems to underestimate electron correlation effects. The exchange splitting $\Delta E_{ex}\ (T = 0\ K)$ for example comes out far bigger than in the experiment. Also short range magnetic order in the paramagnetic phase is not reproduced in LDA in contrast to experimental results (e.g. Curie-Weiss law). The main shortcoming of LDA is its restriction to $T = 0\ K$. These difficulties could in principle be overcome by extending DFT to finite temperatures. An alternative strategy uses model Hamiltonians that are then approximately evaluated using many body techniques. A suitable model for band magnetism is the Hubbard model that is introduced in the following section. Collective electronic excitations in the interacting electron system are then investigated. One distinguishes between charge density waves (plasmons) and spin density waves (magnons). In this paper we concentrate on the latter which results in Section 3 into expressions for spin wave energies and the exchange splitting.

The exchange splitting is temperature dependent which is further investigated in Section 4 using a many body approach. From equations for the particle numbers $n_\uparrow$, $n_\downarrow$ results for the magnetization $m\ (T, n)$ and the Curie temperature $T_C\ (n)$ are derived. The results are compared with those of other methods.

2. The Model

Starting point for a theoretical description of ferromagnetic metals is the tight binding approximation which implies that both hopping integrals and Coulomb matrix elements are calculated from atomic wavefunctions. In the Hubbard approximation [1] only the dominant intraatomic matrix element is retained yielding the model Hamiltonian

$$H = \sum_{ij\sigma}\left(T_{ij} - \mu\ \delta_{ij}\right)\ a_{i\sigma}^{+}\ a_{j\sigma}\ + \frac{1}{2}\ U\ \sum_{i\sigma} n_{i\sigma}\ \ n_{i,-\sigma} \qquad (1)$$

Here $a_{i\sigma}^{+}$ denotes the creation operator for a σ-electron at lattice site R_i; $a_{i\sigma}$ is the corresponding annihilation operator. U is the intraatomic Coulomb matrix element mentioned above while the T_{ij} describe hopping integrals. Note that in the tight binding approximation the indices i, j are restricted to nearest neighbors only.

μ denotes the chemical potential that is both T and n-dependent. The temperature dependence follows from a statistical description of the thermodynamic properties of an ideal Fermi gas, i.e.

$$\mu\,(T)\ \cong\ \varepsilon_F\ \left(1 - \frac{\pi^2}{12}\ \left(\frac{k_B\ T}{\varepsilon_F}\right)^2\right) \qquad (2)$$

Note that for simple metals, e.g. Na, Cu, Au, etc at room temperature the ratio

$$\frac{k_B\,T}{\varepsilon_F}\ \sim\ 5\ \cdot\ 10^{-3}$$

So, the temperature dependence of μ becomes practically negligible. On the other hand, the n-dependence of μ is at low temperatures reasonably well described by the Stoner result

$$\mu\,(\,T = 0)\ =\ \varepsilon_F + \frac{1}{2}\ U\ n \qquad (3)$$

As a first approximation one may regard the d-band degeneracy as not particularly decisive and limit the calculation to narrow s-bands. The Coulomb interaction then obviously only acts if both electrons occupy the same Wigner-Seitz cell; these two electrons must then have opposite spins. The Hubbard model is thus the simplest model to describe both electronic and magnetic properties of the transition metals. It explains ferromagnetism on the basis of a spin-dependent band shift between the ↑ and ↓-density of states below the Curie temperature T_C [2]. This is further investigated in Fig 1 that schematically depicts the density of states for both ↑ and ↓-electrons according to the Stoner model.

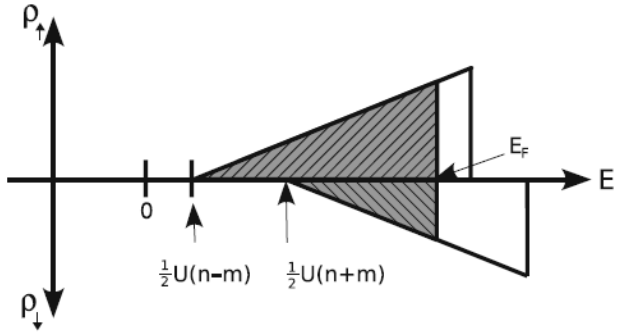

Fig 1: Schematic plot of the spin-dependent density of states $\rho_\uparrow$ and $\rho_\downarrow$ for a ferromagnetic metal according to the Stoner model. The figure is adopted from reference [3].

The bands are rigidly shifted against each other by an energy amount of $\Delta E = U\,m$; it will later be shown that ΔE defines the exchange splitting. The exchange splitting is temperature dependent and vanishes above T_C; this temperature dependence is further investigated in the next section. Furthermore, as both spin bands are filled up to the Fermi level ε_F it follows $n_\uparrow > n_\downarrow$ and thus a spontaneous magnetization $m = n_\uparrow - n_\downarrow > 0$ is observed. Despite its simplicity the Hubbard model remains a non-trivial many body problem and is in general not exactly solvable. Approximate solutions confirm a possible collective magnetic order under certain conditions for the external parameters, i.e. Coulomb coupling U/W where W denotes the band width, the lattice structure, and the band occupation n. M

Ulmke [4] numerically proved magnetic order within the model at finite temperatures in an extended parameter region using Quantum Monte Carlo calculations. W Nolting [5] points out that a correct inclusion of Fermi liquid properties is essential to obtain ferromagnetic solutions within the Hubbard model. The effect of electron correlations on the stability of magnetic order in the Hubbard model is also discussed in reference [6] where it is found that ferromagnetic order is generally suppressed by correlation effects. On the other hand, the Hartree-Fock approximation does not include the correlation energy and thus overestimates the strength of the ferromagnetic order.

Other interesting solutions discussed so far in the literature include
-The Mermin-Wagner theorem rules out ferromagnetic order at finite temperatures for dimensions $d \leq 2$ [7].
-For small band occupations n only a paramagnetic solution

$$n_{\uparrow} = n_{\downarrow} = \frac{1}{2} n$$

is observed. This agrees with the exact result of Kanamori [8].
-In the strong coupling limit $U \gg W$ a saturated ferromagnetic solution is expected for large band occupations n in agreement with the results of Nagaoka [9,10].
-In the zero bandwidth limit where $T_{ij} \rightarrow T_0 \ \delta_{ij}$ no ferromagnetic solutions are observed [11], the conventional explanation being that quasiparticle damping prevents any spontaneous magnetic order.
Current techniques used to improve the understanding of the microscopic origin of ferromagnetism in metals include the development of new mathematical methods, e.g. dynamic mean field theory or density matrix renormalizations [12,13] or alternatively new ab initio schemes [14]. Recent investigations deal with a possible linkage between magnetic order and structural phase transitions in Fe, Co, Ni [15]. The authors of ref [15] report a pressure induced suppression of magnetic order in Fe where the magnetic phase transition is accompanied by a structural phase transition from bcc ↔ hcp. The stability of the bcc-phase is due to the ferromagnetic order. Besides its application to band magnetism the Hubbard model is also successfully applied to describe metal insulator transitions and high temperature superconductivity [16, 17].

3. Spin Density Waves

Besides charge density waves another form of collective excitations in ferromagnetic metals is caused by the existence of the electron spin. The tensor of the magnetic susceptibility is determined from the Green functions involving the components of the spin operator S_i. The diagonal or longitudinal contribution $X_q^{zz}(E)$ yields information regarding the stability of the magnetic order. Within the Hubbard model it has the form

$$X_q^{zz}(E) = -\frac{\mu_0 \mu_B^2}{\hbar NV} \quad \frac{X_0(q,E)}{1-\frac{U}{2N\hbar} X_0(q,E)} \tag{4}$$

Here

$$X_0(q,E) = 2N\hbar \ \rho_0(\varepsilon_F) \tag{5}$$

denotes the temperature independent Pauli spin susceptibility; $\rho(\varepsilon_F)$ is the density of states at the Fermi level. Inserting Eq (5) into (4) yields

$$X_q^{zz}(E) = -\frac{\mu_0 \mu_B^2}{2V} \quad \frac{\rho_0(\varepsilon_F)}{1-U\,\rho_0(\varepsilon_F)} \tag{6}$$

The poles of the susceptibility represent the famous Stoner criterion

$$U \ \rho_0(\varepsilon_F) \geq 1 \tag{7}$$

regarding the stability of magnetic order. According to Eq (7) ferromagnetic order becomes energetically favourable if there is, firstly a large intraatomic Coulomb interaction U as this maximizes the gain in potential energy and secondly a large density of states at the Fermi level. In that case a lot of ↓-electrons can flip their spin and become ↑-electrons without increasing the kinetic energy of the system too much.

On the other hand, the transverse susceptibility $X_q^{+-}\ (E)$ is computed within the Stoner model as

$$X_q^{+-}(E) = \frac{\gamma}{N} \sum_k \frac{\langle n_{k+q,\downarrow}\rangle - \langle n_{k\uparrow}\rangle}{E - \Delta E_{\uparrow\downarrow}(k,q)} \quad (8)$$

The poles are identical to the spin flip excitation energies

$$\Delta E_{\uparrow\downarrow}(k,q) = \varepsilon(k+q) - \varepsilon(k) + U\,m \quad (9)$$

describing transitions between the two spin bands $E_\sigma(k)$. In the case of the Stoner model the quasiparticle energies $E_\sigma(k)$ are plotted as a function of wavevector k in Figure 2 below.

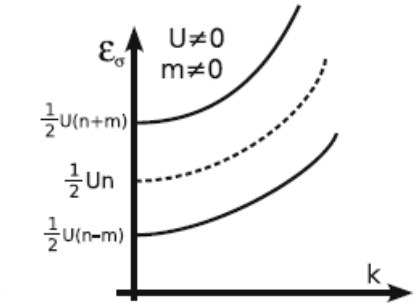

Figure 2: Quasiparticle energies $E_\sigma(k)$ as a function of wavevector k according to the Stoner model. Depicted is the most general case $U \neq 0, m \neq 0$. The figure is adopted from reference [3].

Particularly interesting is the case $U \neq 0, m \neq 0$ where the rigid energy difference between the two bands defines the exchange splitting

$$\Delta E_{ex} = E_\downarrow - E_\uparrow = U\,m \quad (10)$$

Note that the exchange splitting is temperature dependent. The $T = 0$-value can be used to estimate the Curie temperature T_C of the metal The parameter U is first fixed via the exchange splitting and the corresponding value is then inserted into the result from the strong coupling limit $U \gg W$, i.e.

$$k_B\ T_C \cong \frac{1}{4}\ U \quad (11)$$

The theoretically calculated values appear in the fifth column of Table 1 below and are compared with experimental values from the literature. Table 1 shows that our simple model calculation yields surprisingly accurate results for the Curie temperature T_C especially in the case of Ni and Co. Only for Fe the value for the Curie temperature is slightly overestimated due to the large exchange splitting at $T = 0\ K$. However, the proportionality $\Delta\ E_{ex} \sim m$ is not confirmed by experimental results.

metal	$m(T=0)$	$\Delta\ E_{ex}\ (T=0)$	T_C (exp. result)	T_C (theoret. result)
Fe	2.22 μ_B	2 eV	1043 K	1320 K
Ni	0.56 μ_B	0.35 eV	631 K	780K
Co	1.7 μ_B	1.5 eV	1388 K	1280K

Table 1: Experimental and theoretical values for magnetic properties of ferromagnetic metals

The Stoner model additionally suggests that the exchange splitting vanishes in the paramagnetic phase $T > T_C$. Especially in the case of Fe a persistent exchange splitting above T_C has been experimentally observed. The Stoner model also predicts a Pauli like susceptibility $X\ (T) = const$ in the paramagnetic phase with no indication of a Curie-Weiss behaviour again in contradiction to experimental evidence. One reason for the deficiencies of the Stoner model at finite temperatures is its suppression of spin waves. Spin waves in ferromagnetic metals are experimentally detected by inelastic neutron scattering. Expected is therefore at low temperatures $T \rightarrow 0K$ a deviation of the magnetization from saturation of the form

$$\frac{m\,(T)}{m_0} \sim 1 - C\,T^2 \qquad (12)$$

Eq (12) is, however, not reproduced by the Stoner model.

4. Results

Using the thermodynamic properties of an ideal Fermi gas the particle numbers $n_\uparrow$ and $n_\downarrow$ are calculated from the quasiparticle density of states $\rho_\sigma\ (E)$, i.e.

$$n_\sigma = \frac{N_\sigma}{N} = \int_{-\infty}^{+\infty} dE \quad f_-\,(E,T) \quad \rho_\sigma\ (E)$$

Here

$$f_-\ (E,T) = \frac{1}{e^{\beta(E-\mu)}+1}$$

denotes the Fermi function. As $N_\uparrow + N_\downarrow = N$ it follows $n_\uparrow + n_\downarrow = 1$. The magnetization is then defined as

$$m = n_\uparrow - n_\downarrow$$

and is plotted in Figure 3 below as a function of $k_B\,T$ with $n = const.$ Note that in the Stoner model Eq (11) applies. Figure 3 shows the typical Brillouin function behaviour for the magnetization $m\ (T)$ which shows that qualitatively the Stoner model describes the phase transition ferromagnetism ↔ paramagnetism reasonably well. The small kink in the magnetization curve close to T_C could be due to the fact that the particle dependence of the chemical potential according to Eq (3) is strictly only valid at small temperatures. Interesting is also the question in which region of n-values ferromagnetic solutions $m \neq 0$ can be expected in the Hubbard model. This is worked out in Figure 4.

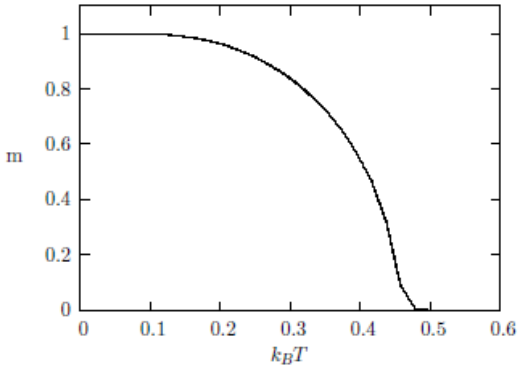

Figure 3: Magnetization m of a ferromagnetic metal as a function of $k_B\,T$ at fixed particle number n.

Note that for all band occupations n there is always a paramagnetic solution

$$n_\uparrow = n_\downarrow = \frac{1}{2}\ n$$

Additionally for particle numbers $n > 0.5$ (half- filled band) there is also a ferromagnetic solution with $m = n$ describing ferromagnetic saturation and similar findings are also reported for example in reference [18]. For small band occupations n a ferromagnetic solution is not possible as μ lies within the band and a solution is only obtained if $n_\uparrow = n_\downarrow$. This is consistent with the exact results of Kanamori [8].

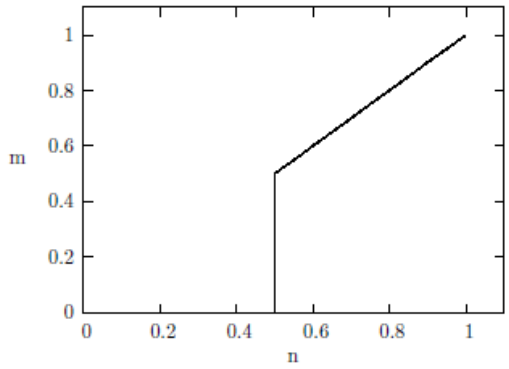

Figure 4: Magnetization m as a function of particle number n at fixed temperature $T = 0$.

The Curie temperature T_C depends on the strength of the ferromagnetic coupling which is reflected in the result

$$k_B\ T_C = \frac{W}{4\ \tanh^{-1}(W/U)} \qquad (13)$$

In the strong coupling limit Eq (13) then reduces to Eq (11) meaning $k_B\ T_C \sim U$. A similar increase of T_C with U has also been obtained by other authors and qualitatively agrees with experimental results [19].

5. Conclusions

In this work collective electronic excitations in ferromagnetic metals (Fe, Co, Ni) are investigated. Using the Hubbard model as a theoretical description the exchange splitting between the ↑ and ↓ density of states is calculated within an RPA-approximation from the tensor of the magnetic susceptibility. A numerical evaluation of the magnetization $m\ (T, n)$ shows that our model calculation describes the phase transition ferromagnetism ↔ paramagnetism reasonably well. Ferromagnetic solutions are only obtained for band fillings $n > 0.5$ describing a half-filled band. The Curie temperature T_C comes out surprisingly accurate, and its dependence on the intraatomic Coulomb interaction U qualitatively agrees with experimental results.

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