Metallic ferromagnetism without exchange splitting

J. E. Hirsch
Department of Physics, University of California, San Diego, La Jolla, California 92093-0319
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In the band theory of ferromagnetism there is a relative shift in the position of majority and minority spin bands due to the self-consistent field due to opposite spin electrons. In the simplest realization, the Stoner model, the majority and minority spin bands are rigidly shifted with respect to each other. Here we consider models at the opposite extreme, where there is no overall shift of the energy bands. Instead, upon spin polarization one of the bands broadens relative to the other. Ferromagnetism is driven by the resulting gain in kinetic energy. A signature of this class of mechanisms is that a transfer of spectral weight in optical absorption from high to low frequencies occurs upon spin polarization. We show that such models arise from generalized tight binding models that include off-diagonal matrix elements of the Coulomb interaction. For certain parameter ranges it is also found that reentrant ferromagnetism occurs. We examine properties of these models at zero and finite temperatures, and discuss their possible relevance to real materials. [S0163-1829(99)04209-5]

I. INTRODUCTION

In the Stoner model for metallic ferromagnetism,[1] a rigid shift in the positions of the majority and minority spin bands occurs. The difference in band energy between minority and majority spin electrons is

$$\epsilon_{k\downarrow} - \epsilon_{k\uparrow} = \Delta = Um. \tag{1}$$

Here Δ is the exchange splitting, m the magnetization, and U the exchange interaction. In deriving the Stoner model from a tight binding model, U corresponds to an on-site interaction, because the shift in Eq. (1) is momentum independent. The Stoner model has been widely used for the description of metallic ferromagnets.[2]

More elaborate approaches using spin-density-functional theory derive the band structure of majority and minority spins taking into account electron-electron interactions within the local spin-density approximation.[3] The result is exchange-split bands where the exchange splitting is in general not constant across the Brillouin zone. Nevertheless, here again there is always an overall shift in energy of majority and minority spin bands with respect to each other.

In this paper we consider models where there is no overall shift in the relative position of the energy bands. Can ferromagnetism still occur? The answer is yes, if there is instead a change in the relative bandwidth of majority and minority spin electrons upon spin polarization. Figure 1 contrasts the situation in the Stoner model and in the models considered in this paper. The gain of energy in the ferromagnetic state arises from an effective mass reduction, or equivalently a bandwidth expansion, of the majority spin electrons, leading to a lowering of kinetic energy.

The possible role of effective mass reduction in driving the transition to ferromagnetism was first proposed in connection with the ''double exchange'' mechanism to describe manganese oxides.[4] More generally, we have suggested in the context of describing metallic ferromagnetism with a single band generalized tight binding model,[5,6] that effective mass reduction upon spin polarization may be an essential feature of *all* itinerant ferromagnets. It was pointed out[5] that the anomalous drop in resistivity and the negative magnetoresistance that is usually found in ferromagnets could be explained as arising from a change in effective mass rather than a change in scattering time as usually assumed.[7] Clear

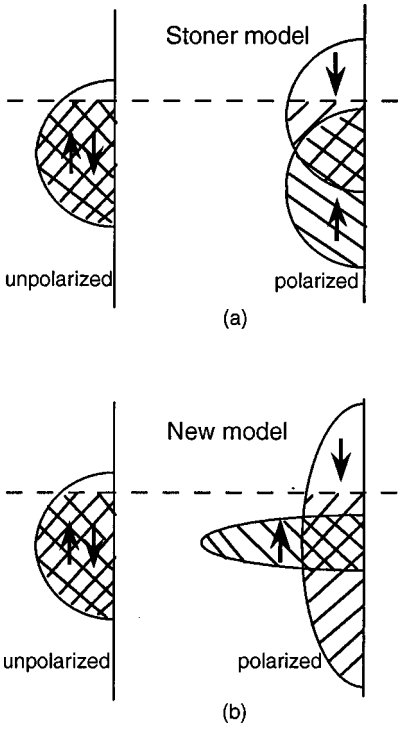

FIG. 1. Density of states for up and down electrons in the unpolarized (left) and magnetic (right) states (schematic). The dashed line indicates the position of the Fermi level. (a) Stoner model: as the temperature is lowered, the bands for up and down spin electrons rigidly shift with respect to each other. (b) Model considered in this paper: as the temperature is lowered, the bands change their width relative to each other, without relative displacement of their centers. For band filling above one half, as shown in the figure, the broader band corresponds to minority spin carriers; for band filling below one half, the situation is reversed.

experimental evidence for this phenomenon has been found in optical properties of the colossal magnetoresistance manganites[8] as well as of some rare-earth hexaborides[9] and magnetic semiconductors.[10] Whether this is a universal phenomenon that occurs in all metallic ferromagnets remains an open question.

In the model that we considered originally,[5] ferromagnetism arises from the combined effect of exchange energy and band broadening as the system orders. Such may well be the situation applicable to real materials. However, to better understand the various theoretical possibilities it is of interest to ask whether ferromagnetism could be driven just by band broadening in the absence of exchange energy. As we show in this paper, this is indeed possible, if the bandwidth for one spin expands while the one for the other spin contracts. In contrast, in the model considered originally where exchange energy was also present, both minority and majority spin bands could expand upon spin polarization. Still, both in the original model and in the ones considered here a signature of this physics is that an overall shift in optical spectral weight from high to low frequencies takes place as spin polarization develops. Application of this model to describe the shift in optical spectral weight observed in EuB_6 is discussed elsewhere.[11]

II. THE MODELS

We consider a single band of width D, with density of states $g(\epsilon)$ symmetric around the origin:

$$\int_{-D/2}^{D/2} d\epsilon g(\epsilon)\epsilon = 0. \tag{2}$$

The magnetization m and number of electrons n per site are given by

$$m = \int_{-D/2}^{D/2} d\epsilon g(\epsilon)\{f[\epsilon_\uparrow(\epsilon)] - f[\epsilon_\downarrow(\epsilon)]\} \tag{3a}$$

$$n = \int_{-D/2}^{D/2} d\epsilon g(\epsilon)\{f[\epsilon_\uparrow(\epsilon)] + f[\epsilon_\downarrow(\epsilon)]\} \tag{3b}$$

with f the Fermi function. In the class of models under consideration here the quasiparticle energies are given by

$$\epsilon_\sigma(\epsilon) = (1 - 2j_1 I_\sigma - 2j_2 I_{-\sigma})\epsilon - \sigma D\left[\frac{k}{2}m + h\right] - \mu. \tag{4}$$

Here, $h = H/D$, with H an external magnetic field (in units of energy), and the bond charge I_σ is given by

$$I_\sigma = \int_{-D/2}^{D/2} d\epsilon g(\epsilon)\left(\frac{-\epsilon}{D/2}\right) f[\epsilon_\sigma(\epsilon)]. \tag{5}$$

The ''exchange'' interaction k gives an overall rigid shift in the band energies, as in the Stoner model. The new features here arise from the interactions j_1 and j_2 that give rise to band narrowing that depends on the bond-charge occupation for each spin I_σ, which in turn will be a function of temperature and magnetization.

III. RELATION TO TIGHT BINDING MODELS

Consider a tight binding model with on-site, nearest-neighbor, exchange and pair hopping interactions[12–14,6]

$$H = -t\sum_{\langle ij\rangle,\sigma}(c_{i\uparrow}^\dagger c_{j\sigma} + \text{H.c.}) + U\sum_i n_{i\uparrow}n_{i\downarrow} + V\sum_{\langle ij\rangle} n_i n_j + J\sum_{\langle ij\rangle,\sigma,\sigma'} c_{i\sigma}^\dagger c_{j\sigma'}^\dagger c_{i\sigma'}c_{j\sigma} + J'\sum_{\langle ij\rangle}(c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger c_{j\downarrow}c_{j\uparrow} + \text{H.c.}) \tag{6}$$

The interactions arise from the various matrix elements of the Coulomb interaction between orbitals at nearest-neighbor sites. A mean field decoupling leads to the quasiparticle energies

$$\epsilon_\sigma(\epsilon_k) = \left[1 - I_\sigma\left(\frac{J}{t} - \frac{V}{t}\right) - I_{-\sigma}\left(\frac{J}{t} + \frac{J'}{t}\right)\right]\epsilon_k - \sigma\frac{(U + zJ)}{2}m \tag{7}$$

with

$$I_\sigma = \langle c_{i\sigma}^\dagger c_{j\sigma}\rangle \tag{8a}$$

$$m = \langle c_{i\uparrow}^\dagger c_{i\uparrow}\rangle - \langle c_{i\downarrow}^\dagger c_{i\downarrow}\rangle \tag{8b}$$

the bond charge and magnetization parameters (i, j nearest neighbors). The bandwidth and band energies are

$$D = 2zt, \tag{9a}$$

$$\epsilon_k = -t\sum_\delta e^{ik\delta}, \tag{9b}$$

with z the number of nearest neighbors to a site and δ a vector connecting a site to its nearest neighbors. The interactions in Eq. (4) are then

$$j_1 = \frac{zJ}{D} - \frac{zV}{D}, \tag{10a}$$

$$j_2 = \frac{zJ}{D} + \frac{zJ'}{D}, \tag{10b}$$

$$k = \frac{U}{D} + \frac{zJ}{D}. \tag{10c}$$

In connection with itinerant ferromagnetism we have considered in the past the model with U and J only,[5] and also the one with U, J, and $J' = J$.[15] In terms of the notation in Eq. (4), the first case corresponds to

$$j_1 = j_2 = j, \tag{11a}$$

$$k = u + j, \tag{11b}$$

with $j = zJ/D, u = U/D$. In this particularly simple case the hopping amplitude gets renormalized equally by bond charge density of equal and opposite spin. When one includes the pair hopping term J', and in particular for $J' = J$, we have

$$j_1 = j, \tag{12a}$$

$$j_2 = 2j, \tag{12b}$$

$$k = u + j, \tag{12c}$$

so that in this case more of the hopping renormalization is due to electrons of opposite spin. As seen in Ref. 15, this case favors more the existence of ferromagnetism near the top and bottom of the band compared to the first case.

Note that according to Eq. (10) the band renormalization parameter j_1 from the same-spin electrons should be expected to be smaller than that of opposite spin electrons j_2 due to the effects of nearest-neighbor repulsion and of pair hopping. In this paper we will focus on that situation.

IV. GROUND STATE PROPERTIES

We parametrize the interactions in Eq. (4) as

$$j_1 = aj, \tag{13a}$$

$$j_2 = j, \tag{13b}$$

with $0 \leqslant a \leqslant 1$. The case studied previously with only exchange interaction J corresponds to $a = 1$, and the case with pair hopping $J' = J$ corresponds to $a = 1/2$. If the system is not completely polarized the chemical potential is determined by the condition

$$\epsilon_\uparrow(\epsilon_{F\uparrow}) = \epsilon_\downarrow(\epsilon_{F\downarrow}) \tag{14}$$

with

$$n_\sigma = \int_{-D/2}^{\epsilon_{F\sigma}} d\epsilon g(\epsilon) \tag{15}$$

and

$$n = n_\uparrow + n_\downarrow, \tag{16a}$$

$$m = n_\uparrow - n_\downarrow, \tag{16b}$$

the total occupation and magnetization, respectively. We assume for simplicity a flat density of states

$$g(\epsilon) = \frac{1}{D}, -\frac{D}{2} \leqslant \epsilon \leqslant \frac{D}{2}. \tag{17}$$

We have seen earlier that the properties of these type of models do not depend strongly on energy variation of the density of states, unlike in the Stoner model.

The occupations of up and down spin bands at $T = 0$ are given by

$$n_\sigma = \frac{\epsilon_{F\sigma}}{D} + \frac{1}{2} \tag{18}$$

so that

$$\frac{\epsilon_{F\sigma}}{D} = \frac{n + \sigma m - 1}{2} \tag{19}$$

and the bond charge densities are given by

$$I_\sigma = \frac{1 - (1 - n - \sigma m)^2}{4} = n_\sigma(1 - n_\sigma) \tag{20}$$

so that the condition Eq. (14) is

$$(j_1 - j_2)(1 - n)^2 - (j_1 + j_2)\frac{1 - m^2 - (1 - n)^2}{2} = k - 1 \tag{21}$$

and for the parametrization Eq. (13) we obtain

$$j = \frac{2(1 - k)}{(1 + a)(1 - m^2) - (3a - 1)(1 - n)^2}. \tag{22}$$

The conditions for onset of ferromagnetism and for full spin polarization result from setting $m = 0$ and $m = n$, respectively, in Eq. (22).

The resulting phase diagrams for various values of a are shown in Fig. 2. For all cases, partially polarized ferromagnetic regions occur predominant around the 1/2-filled band. Note that for exactly half filling no full polarization ocurs for $k < 1$. Also, note that for small values of a, spin polarization is easier near the edges of the band, and for large values of a near the half-filled band. For the value $a = 1/3$ (not shown), the boundary for onset of spin polarization is independent of n, at $j/(1 - k) = 1.5$.

However, the phase diagrams in Fig. 2 need to be supplemented by the condition that the coefficients of ϵ in Eq. (4) need to remain positive, i.e.,

$$1 - 2j_1 I_\sigma - 2j_2 I_{-\sigma} > 0 \tag{23}$$

which may not be satisfied for some values of k. While this condition is always satisfied at the points where onset of ferromagnetism occurs, it is not so for partial or full spin polarization. For $k = 0$, condition (23) is equivalent to the constraint

$$n \leqslant 1 - m \tag{24}$$

for $n \leqslant 1$, and the corresponding symmetric one for $n > 1$. In particular, condition (24) implies that full polarization in the absence of exchange, i.e., $k = 0$, can only be achieved for $n \leqslant 1/2$ or $n \geqslant 3/2$. Partial polarization of magnitude $m = cn, 0 < c < 1$, can be achieved only in the range $n < 1/(1 + c)$ and the corresponding symmetric one for $n > 1$.

For nonzero exchange k, full polarization can be achieved for any n if the condition

$$\frac{k}{1 - a} \geqslant \frac{1}{2} \tag{25}$$

holds, which in particular is true if $k \geqslant 0.5$ for any a. (By contrast, the Stoner model requires $k > 1$ to give rise to ferromagnetism.) Otherwise, full polarization is only achieved in the range

$$n \leqslant \frac{1/2}{1 - k/(1 - a)} \tag{26}$$

and the corresponding symmetric one above half filling. Note that as a approaches unity, condition (26) will be satisfied for any filling for arbitrarily small exchange k.

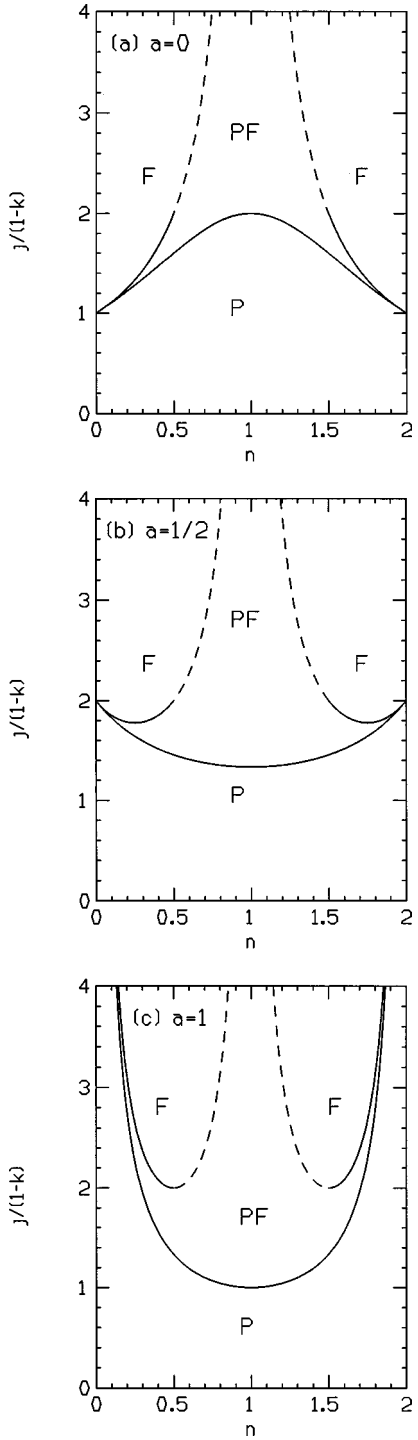

FIG. 2. Ground state phase diagrams. $a=j_1/j_2$. $j=j_2$. P, PF, and F denote paramagnetic, partially ferromagnetic, and fully polarized ferromagnetic regions, respectively. In the absence of exchange ($k=0$), full polarization cannot be achieved for band fillings between 1/4 and 3/4; that portion of the phase boundary is indicated by a dashed line. As k increases the region where full polarization can be achieved increases and for $k\geqslant 0.5$ it covers the entire phase diagram except for the point $n=1$.

Note also that the case $a=1$ is a very singular point. For exchange $k=0$, there is no ferromagnetism in the model as no change in relative occupation of up and down spin bands can occur. On the other hand, for any $k\neq 0$ the criteria for onset and full polarization can be satisfied for suitable j for any value of occupation n.

V. FINITE TEMPERATURES

Equation (5) for the bond charge yields

$$I_\uparrow - I_\downarrow = \int_{-D/2}^{D/2} d\epsilon g(\epsilon)\left(\frac{-\epsilon}{D/2}\right)\{f[\epsilon_\uparrow(\epsilon)]-f[\epsilon_\downarrow(\epsilon)]\} \tag{27}$$

and using

$$j_1 I_\uparrow + j_2 I_\downarrow = \frac{j_1+j_2}{2}(I_\uparrow + I_\downarrow) + \frac{j_1-j_2}{2}(I_\uparrow - I_\downarrow) \tag{28}$$

we obtain on expanding the Fermi functions on the right-hand side of Eq. (27)

$$I_\uparrow - I_\downarrow = (I_\uparrow - I_\downarrow)(j_2-j_1)D\int_{-D/2}^{D/2} d\epsilon g(\epsilon)\left(\frac{-\epsilon}{D/2}\right)^2\left(-\frac{\partial f}{\partial \epsilon_\sigma}\right) \tag{29}$$

in the absence of exchange, $k=0$. Thus the equation that determines the critical temperature is found by cancelling $(I_\uparrow - I_\downarrow)$ on both sides of this equation:

$$1=(j_2-j_1)D\int_{-D/2}^{D/2} d\epsilon g(\epsilon)\left(\frac{-\epsilon}{D/2}\right)^2\left(-\frac{\partial f}{\partial \epsilon_\sigma}\right). \tag{30}$$

More generally, in the presence of exchange we consider also Eq. (3a) for the magnetization, and obtain the set of equations

$$I_\uparrow - I_\downarrow = (j_2-j_1)(I_\uparrow - I_\downarrow)G_2 + kmG_1, \tag{31a}$$

$$m=(j_2-j_1)(I_\uparrow - I_\downarrow)G_1 + kmG_0 \tag{31b}$$

with

$$G_l = D\int_{-D/2}^{D/2} d\epsilon g(\epsilon)\left(\frac{-\epsilon}{D/2}\right)^l\left(-\frac{\partial f}{\partial \epsilon_\sigma}\right) \tag{32}$$

and the condition for the critical temperature is obtained by setting to zero the determinant of the coupled Eqs. (31), yielding

$$[1-(j_2-j_1)G_2][1-kG_0]-(j_2-j_1]kG_1^2=0. \tag{33}$$

We focus here on ferromagnetism without exchange, so that the T_c equation is given by Eq. (30), or

$$1=(j_2-j_1)G_2. \tag{34}$$

For infinite temperature G_2 is zero, and as T decreases it becomes positive. At a critical temperature, that increases when the difference (j_2-j_1) increases, Eq. (34) will be satisfied. The situation is analogous to the usual Stoner model where the critical temperature is determined by the equation

$$1=kG_0. \tag{35}$$

At low temperatures we can approximate the Fermi function derivative in Eq. (34) as a δ function, yielding

$$1=\frac{4(j_2-j_1)}{1-2(j_1+j_2)I_\sigma}\left(\frac{\epsilon_{F\sigma}}{D}\right)^2 \tag{36}$$

with $(\epsilon_{F\sigma}/D)$ given by Eq. (19). As $T\to 0, I_\sigma$ is given by Eq. (20) and the T_c equation (36) reduces to the condition Eq. (22) for onset of ferromagnetism $(m=0)$ for the case $k=0$. One can also obtain an approximate analytic form for T_c in weak coupling by expanding G_2 in Eq. (34) to one higher order than used to obtain Eq. (36). This is in contrast to the case $j_1=j_2$, where the lowest order equation analogous to Eq. (36),

$$1=\frac{k}{1-2j(I_\uparrow+I_\downarrow)} \tag{37}$$

directly yields T_c at low temperatures on expanding I_σ as

$$I_\sigma=I_\sigma^0-\frac{\pi^2}{3}\left(\frac{k_BT}{D}\right)^2\frac{1}{1-2(j_1+j_2)I_\sigma^0} \tag{38}$$

with $I_\sigma^0=I_\sigma(T=0)$.

In fact, the integral G_2 [Eq. (32)] will not be monotonically decreasing as the temperature increases, as G_0 is. For example, for a half-filled band the Fermi function derivative as $T\to 0$ approaches a δ function at zero energy, and the integral G_2 will vanish due to the extra factors of ϵ. As the temperature is increased from zero, G_2 will increase until a critical temperature is reached where Eq. (34) is satisfied. At a higher temperature G_2 will start decreasing until a second temperature is reached where Eq. (34) is satisfied. Thus we can expect to find reentrant ferromagnetism in this model for intermediate values of the effective coupling (j_2-j_1).

The magnetic susceptibility above T_c is obtained by taking the derivative of the magnetization Eq. (3a) with respect to the external magnetic field. For the case $j_1\neq j_2$ the magnetic field dependence of the bond charge needs to be taken into account. We obtain for $\chi=dm/dh$

$$\chi=2\frac{G_0-(j_2-j_1)(G_0G_2-G_1^2)}{(1-kG_0)[1-(j_2-j_1)G_2)]-k(j_2-j_1)G_1^2} \tag{39}$$

with G_l given by Eq. (32). In particular, G_0 is the magnetic susceptibility per spin in the absence of interactions, and Eq. (39) reduces to the usual RPA form if $j_1=j_2$.[5] More generally, the susceptibility Eq. (39) diverges as $1/(T-T_c)$ as T approaches T_c given by the solution of Eq. (33). Parametrization of the susceptibility as

$$\chi(T)=\frac{p_{\rm eff}^2(T)}{3(T-T_c)} \tag{40}$$

defines the effective moment $p_{\rm eff}$, which is temperature-independent within the Curie-Weiss law, which is often seen experimentally.

VI. NUMERICAL RESULTS

We solve the mean field equations numerically for various cases. Figure 3 shows the magnetization versus temperature for $n=0.5, a=0$ (so that $j_1=0$) and various values of $j_2=j$ in the absence of exchange $(k=0)$. In this case the phase diagram Fig. 2(a) shows that onset of magnetization at zero temperature occurs for $j=1.6$ and full polarization for $j=2$. However, note that at finite temperature ferromagnetism also occurs for $j<1.6$ as discussed in the previous section. For $j=1.6$ the magnetization approaches zero as $T\to 0$, and for $j<1.6$ reentrant ferromagnetism occurs. For $j=2$ the system becomes fully polarized as $T\to 0$, and for $j>2$ the model becomes unphysical because the condition Eq. (23) fails to be satisfied as the temperature is lowered. Note that for these cases the shape of the magnetization curve is very different than what is obtained from the Stoner model or from molecular field theory.

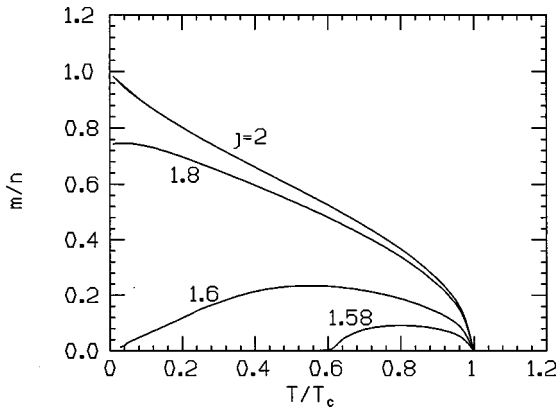

FIG. 3. Magnetization versus temperature for band filling $n=0.5, k=0, j1=0$. The numbers next to the curves give the values of $j_2=j$. For $j<1.6$ reentrant ferromagnetism occurs in this case.

The effective bandwidth for spin σ, D_σ^*, or equivalently the effective mass m_σ^*, are given by the relation

$$\frac{D}{D_\sigma^*}=\frac{m_\sigma^*}{m}=1-2j_1I_\sigma-2j_2I_{-\sigma} \tag{41}$$

with D and m the (spin-independent) bare bandwidth and mass, respectively. Figure 4 shows the temperature dependence. For the majority spins, the effective mass increases as the temperature decreases, and then decreases as T is lowered below T_c. For the minority spins, m^* increases more sharply as T is lowered below T_c.

For small values of j, however, the effective mass for the majority spins also increases as the temperature is lowered further, and becomes eventually larger than its value at T_c. One may ask why it is still advantageous to spin polarize, when the energy will no longer be lower than what it was in the unpolarized state at T_c. The answer is of course that at this lower temperature if the system was unpolarized the bandwidth for both up and down spin electrons would be much smaller, giving rise to a larger energy than in the polarized state (see Fig. 1 of Ref. 5b for an illustration of this effect).

The behavior of the effective moment, Eq. (40), versus temperature for this case is shown in Fig. 5. For small j the temperature dependence is strong and it increases as T approaches T_c, for larger j it decreases somewhat as T approaches T_c.

Results for a case where the band renormalization for the same spin j_1 is not zero are shown in Fig. 6. The case $a=0.5$ shown corresponds to parameters $J'=J$ and $V=0$ in the tight binding model. Here the effective moment is much

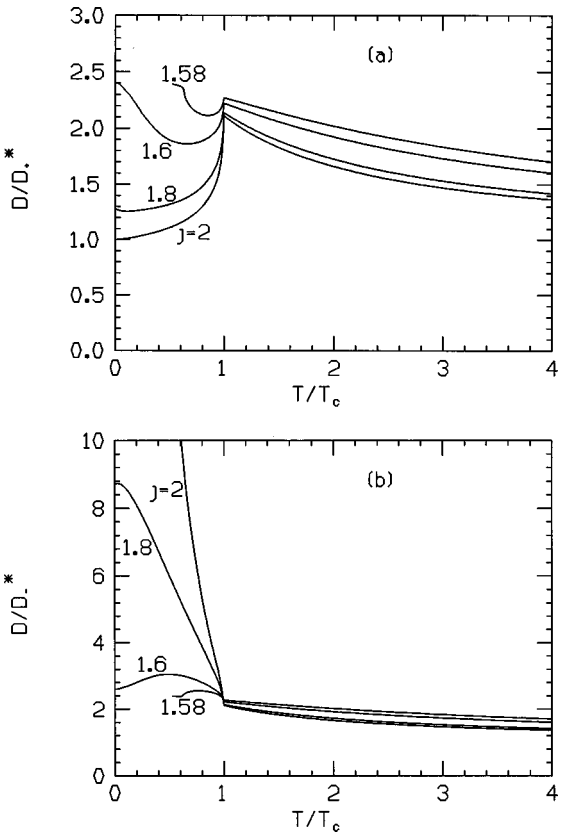

FIG. 4. Effective mass ratio, or inverse of bandwidth, versus temperature for the parameters of Fig. 3. (a) Majority spins; (b) minority spins. The numbers next to the curves give the values of $j_2=j$.

less temperature dependent and no reentrant behavior is found for this set of parameters.

Figure 7 shows the effect of exchange on the behavior of the magnetization and effective moment. With increasing k the magnetization curve becomes steeper and resembles more the conventional behavior. The effective moment is rather constant with temperature for these cases corresponding to a Curie-Weiss law for the susceptibility. As discussed in Ref. 5, for the pure Stoner model (exchange only) a strong temperature dependence of the effective moment is found.

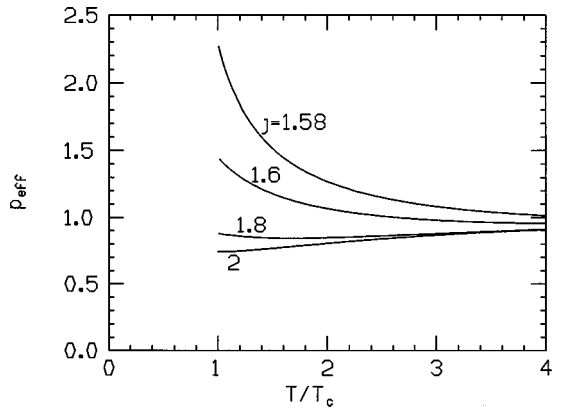

FIG. 5. Effective moment [Eq. (40)] versus temperature for the parameters of Fig. 3.

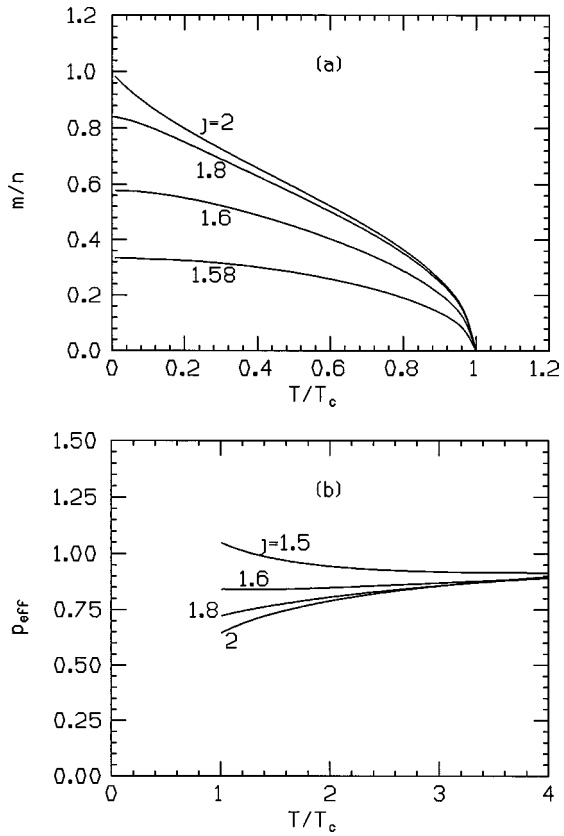

FIG. 6. (a) Magnetization versus temperature and (b) effective moment versus temperature for $n=0.5, k=0, a=j_1/j_2=0.5$. The values of $j_2=j$ are given next to the curves.

VII. OPTICAL ABSORPTION, MAGNETORESISTANCE, AND PHOTOEMISSION

The Drude formula for the real part of the optical conductivity is

$$\sigma_1(\omega)=\frac{ne^2}{m^*}\frac{\tau}{1+\omega^2\tau^2} \tag{42}$$

which describes optical absorption through intraband processes. The conductivity sum rule that results from this formula is

$$\int_0^{\omega_m} d\omega\sigma_1(\omega)=\frac{\pi}{2}\frac{ne^2}{m^*}. \tag{43}$$

The cutoff ω_m is to exclude transitions to other bands, which are processes not described by the expression Eq. (42). We assume that $\omega_m\tau\gg 1$ in order to derive Eq. (43) from Eq. (42). Ordinarily, if no change in effective mass occurs upon spin polarization, the optical absorption given by these formulas will not depend on spin polarization (assuming a constant n). While one may expect changes in optical absorption as function of temperature due to changes in the relaxation time τ, no changes would be expected as function of magnetic field at fixed temperature.

In contrast, in the models considered here we have

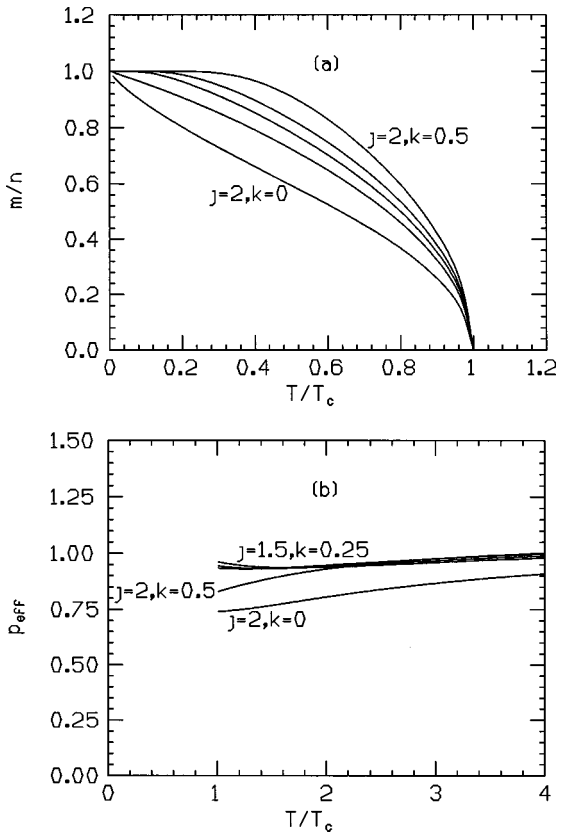

FIG. 7. Effect of exchange splitting on the behavior of (a) magnetization and (b) effective moment versus temperature. For (a), the curves of increasing steepness correspond to the cases (i) $j/(1-k)=2,k=0$; (ii) $j/(1-k)=2,k=0.25$; (iii) $j/(1-k)=2,k=0.5$; (iv) $j/(1-k)=4,k=0.5$.

$$\sigma_1(\omega)=\frac{e^2\tau}{1+\omega^2\tau^2}\left(\frac{n_\uparrow}{m_\uparrow^*}+\frac{n_\downarrow}{m_\downarrow^*}\right) \tag{44}$$

and

$$\int_0^{\omega_m} d\omega\,\sigma_1(\omega)=\frac{\pi}{2}e^2\left(\frac{n_\uparrow}{m_\uparrow^*}+\frac{n_\downarrow}{m_\downarrow^*}\right) \tag{45}$$

and changes in optical absorption will occur both as a function of temperature and of magnetic field due to changes in the degree of spin polarization and corresponding changes in the effective masses. We assume that the effective mass is proportional to the inverse bandwidth, so that

$$\frac{m}{m_\sigma^*}=1-2j_1I_\sigma-2j_2I_{-\sigma}. \tag{46}$$

Figure 8 shows the typical behavior expected in low frequency optical absorption, for parameters $j_1=0$, $j_2=2$, $n=0.5$, $k=0$ as an example. The optical absorption decreases for T above T_c as the temperature is lowered, and increases again rapidly below T_c. When a magnetic field is applied the optical absorption increases at all temperatures, with the largest increase at T_c. Figure 9(a) shows the optical weight versus temperature illustrating this behavior. In Fig. 9(b) optical weights for majority and minority spin electrons are shown separately; when a magnetic field is applied, the

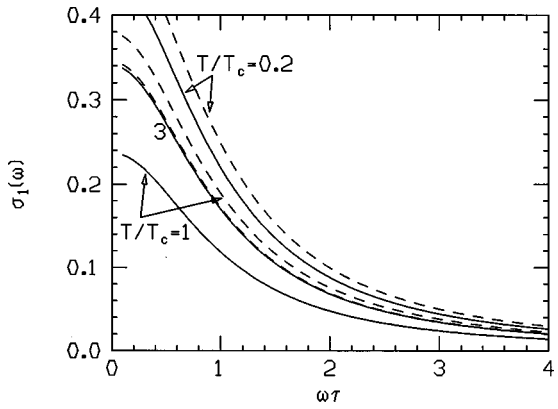

FIG. 8. Optical conductivity (arbitrary units) versus frequency for the parameters of Fig. 3. The full lines give the optical conductivities at temperatures $T/T_c=0.2$, 1, and 3, and the dashed lines the corresponding ones in the presence of a magnetic field $h=H/D=0.05$. The effect of the magnetic field is largest at T_c, and becomes very small for $T/T_c=3$.

optical absorption of majority spins increases and that of minority spins decreases but less strongly, giving rise to the change seen in Fig. 9(a).

The dc conductivity is given by

$$\sigma=\frac{ne^2\tau}{m^*} \tag{47}$$

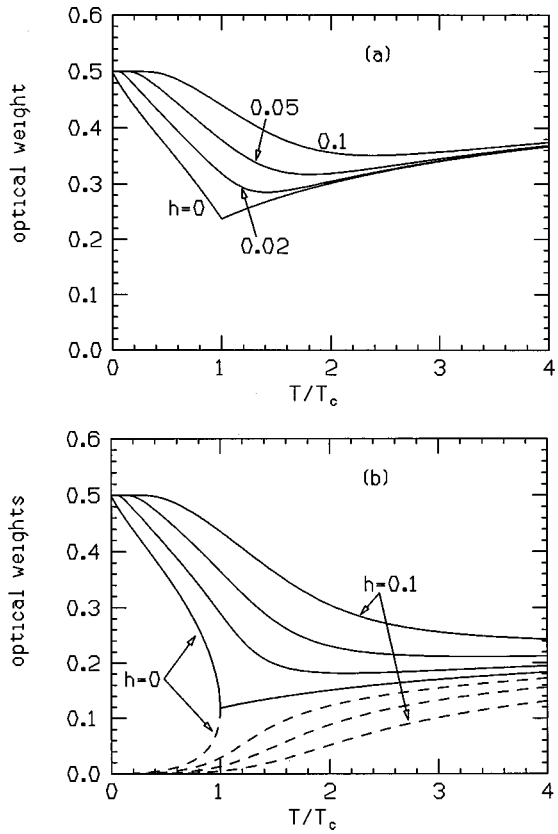

FIG. 9. Optical weight n/m^* given by the low-frequency integral of the conductivity versus temperature in the presence of a magnetic field $h=H/D$ (numbers next to the lines). (a) Total, (b) for spin up (full lines) and spin down (dashed lines) respectively. The parameters are those of Fig. 3.

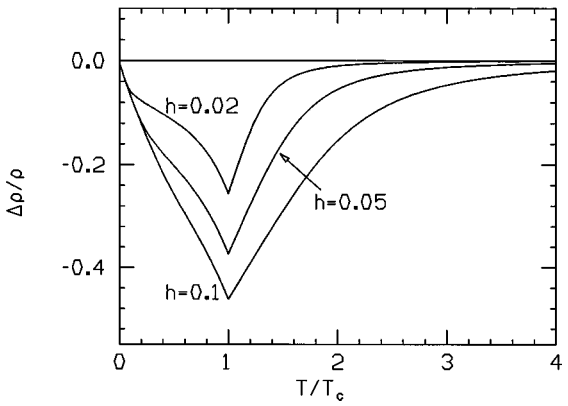

FIG. 10. Magnetoresistance versus temperature for the parameters of Fig. 3.

and the magnetoresistance in our model will be determined by the change in effective mass, or bandwidth, with spin polarization. Assuming a constant relaxation time the magnetoresistance is given by

$$\frac{\Delta\rho}{\rho}\equiv\frac{\rho(H)-\rho(0)}{\rho(0)}$$

$$=\frac{\sum_{\sigma} n_{\sigma}(H)[1-2j_1 I_{\sigma}(H)-2j_2 I_{-\sigma}(H)]}{\sum_{\sigma} n_{\sigma}(0)[1-2j_1 I_{\sigma}(0)-2j_2 I_{-\sigma}(0)]}-1 \quad (48)$$

and is shown in Fig. 10 for one case. It is maximum at T_c and remains large well above T_c. Similar results are found for other parameters in the model.

Note that under the assumption that there is no significant change in the relaxation time with spin polarization, there should be a definite relation between the optical weight

$$W(T,H)\equiv\frac{2}{\pi e^2}\int_0^{\omega_m} d\omega\,\sigma_1(\omega) \quad (49)$$

and the magnetoresistance. For example, the quantity

$$f(T,H)=\frac{\Delta\rho/\rho(0)}{\Delta W/W(H)} \quad (50)$$

should be a constant independent of temperature and magnetic field. Deviations from constant behavior would indicate a dependence of the relaxation time on spin polarization.

Another important experiment that can shed light on the nature of the ferromagnetic transition is angle-resolved photoemission. For example, some results on manganites have recently been reported.[16] In the models discussed here the quasiparticle dispersion changes with magnetization, and hence one would expect characteristic signatures in the photoemission spectrum as function of temperature or magnetic field. Of particular interest would be to obtain high resolution angle- *and* spin-resolved photoemission spectra of ferromagnetic metals.[17] Figure 11 shows an example of predicted angle-resolved photoemission spectrum for our model for one case without exchange: for majority spins, the Fermi velocity increases as the system polarizes and the quasiparticle peak disperses faster, and the opposite occurs for minority spins. For the non-spin-resolved spectrum [Fig. 11(c)], two peaks develop as the system polarizes, one of which disperses slower and the other one faster than in the unpolarized case. However, for other parameters in the model, in the presence of exchange, one finds that both majority and minority spins disperse faster in the ferromagnetic state, reflecting the gain in kinetic energy upon spin polarization.

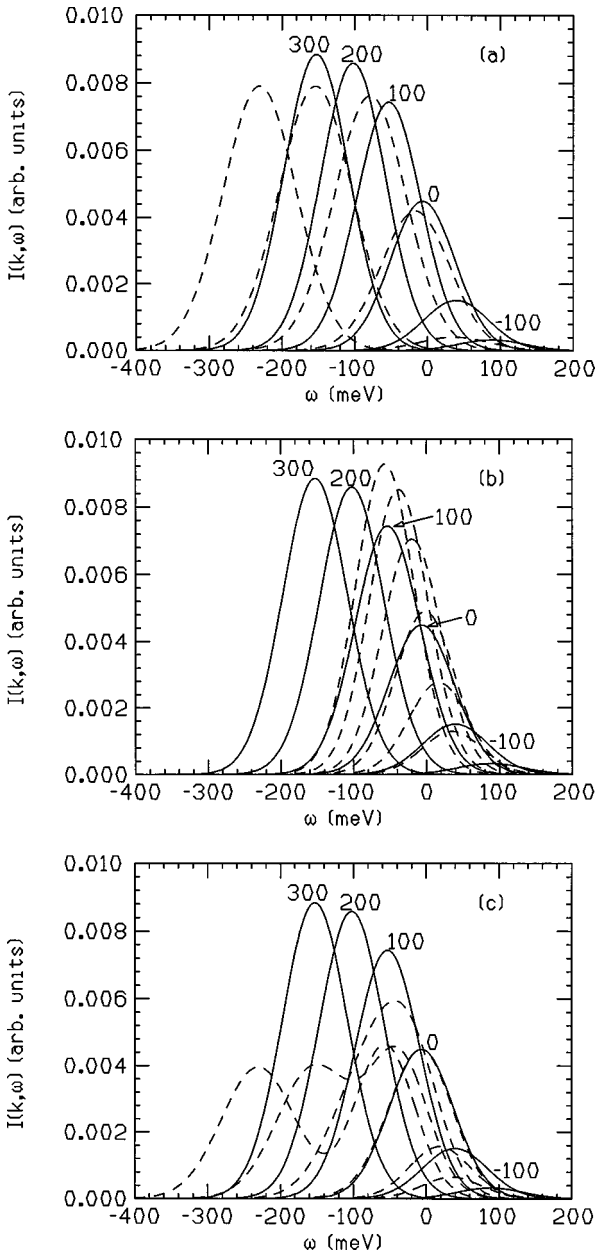

FIG. 11. Angle-resolved photoemission spectra for the parameters of Fig. 3, giving rise to $T_c/D=0.081$. We assume a bandwidth $D=0.3$ eV, so $T_c=280$ K. Full lines correspond to temperature $T/T_c=1.2$, dashed lines to $T/T_c=0.8$. The numbers next to the full lines give the values of $\epsilon_k-\mu$ (in meV). (a) Majority spins; below T_c, the Fermi velocity increases and the peaks disperse faster. (b) Minority spins: the Fermi velocity decreases upon ordering and the peaks disperse slower. (c) Non-spin-resolved: two peaks appear below T_c, one disperses faster and one slower than the peaks above T_c. Similar results would be found at fixed temperature under application of a magnetic field. In particular, application of a magnetic field $h=H/D=0.03$ at $T/T_c=1.2$ gives results almost identical to those shown in the figure for $T/T_c=0.8$.

VIII. SUMMARY AND DISCUSSION

We have considered a model for ferromagnetism where the magnetic order arises from a modification of the width of the bands upon spin polarization instead of the usual exchange splitting. Thus, ferromagnetism arises from a gain in kinetic rather than potential energy. This represents the opposite extreme of the conventional understanding of ferromagnetism as arising from a gain in potential energy (exchange energy) despite an associated cost in kinetic energy.[18] More generally, the models considered here also include an exchange splitting parameter, in the presence of which ferromagnetism arises from a combination of gains in potential and kinetic energy.

As we have seen, these models can be derived from a single band tight binding model that includes off-diagonal matrix elements of the Coulomb interaction. The terms that give rise to kinetic energy gain upon spin polarization originate in nearest neighbor matrix elements conventionally termed ''exchange'' and ''pair hopping.'' Physically, they represent ''bond-charge repulsion,''[13,14] i.e., the repulsive Coulomb energy of electrons at the interstitial region between neighboring atoms rather than at the atoms themselves. In the present context the use of the term ''exchange'' or ''Heisenberg exchange'' for such matrix elements is somewhat misleading, because the driving force for ferromagnetism in fact is direct Coulomb repulsion of electrons in the bonds. The model discussed in this paper with exchange parameter $k=0$ describes ferromagnetism without exchange splitting, even though the main interaction is the parameter J which is conventionally called an exchange integral.

It is generally believed that band degeneracy is essential to metallic ferromagnetism.[18] It is certainly true that ferromagnetism usually arises in systems that have atoms with valence electrons in degenerate orbitals. Nevertheless, it is difficult to see how atomic orbital degeneracy could be essential for example in the case of Ni and Ni-Cu alloys, which have a small fraction of a d hole per atom and negligible polar fluctuations. Furthermore, it should be kept in mind that all energy bands in metals are in fact nondegenerate, except at sets of states of measure zero in the Brillouin zone (points or lines). Hence we believe that an approach that focuses on a single nondegenerate band, as in this paper, is sensible, whether that band arises from degenerate or nondegenerate atomic orbitals. On transforming from Bloch to Wannier states for that band and computing the matrix elements of the Coulomb interaction with the Wannier states, the matrix elements discussed in Sec. III (as well as others) result. The question of what is the magnitude of these matrix elements in particular materials is a difficult one. Within the point of view of this paper (and our previous work[5,19]), band degeneracy may be important in determining the magnitude of these matrix elements, but not in determining the structure of the theory.

The model considered here naturally gives rise to partial spin polarization, even with a constant density of states, unlike the Stoner model. In the absence of exchange splitting, we found that ferromagnetism does occur, however, full spin polarization can only be achieved in a limited range of parameter space (far from the half-filled band). As the exchange splitting increases from zero, full polarization can be achieved over an increasing range of band filling. The condition for onset of spin polarization may become more or less stringent as the band filling increases depending on the ratio of the same-spin to opposite-spin band renormalization parameters j_1/j_2.

The magnetization versus temperature showed unconventional behavior in the absence of exchange, with the curves being substantially less steep. An interesting feature is that reentrant ferromagnetism can occur in certain parameter ranges. The magnetic susceptibility versus temperature showed Curie-Weiss behavior for a wide range of parameters, although deviations can also occur.

Reentrant ferromagnetism, and in particular a situation where the magnetic order *increases* as the temperature increases, is somewhat counterintuitive but by no means unphysical. In fact, experimental observations of such behavior have been reported in Y_2Ni_7 (Ref. 20) and in $ThFe_3$.[21] Theoretical models that have been found to exhibit such behavior are Ising models with random ferromagnetic and antiferromagnetic exchange,[22] and even the Stoner model for special forms of the density of states.[23] However, in the Stoner model such behavior is always associated with first order transitions. In the models discussed here, the reentrant behavior can be easily understood. Consider a case where the magnetism has disappeared at sufficiently low temperatures. As the temperature is raised, the contribution of entropy to the free energy of the system increases relative to that of energy. Now in our model (in the absence of exchange), by spin polarizing one of the bands will narrow and as a consequence the entropy contribution from electrons in that band will increase, so that this effect added to the energy gain from the electrons in the broadened band can lead to an overall decrease of the free energy upon spin polarization at higher temperatures. As the temperature increases further there will be a point where entropy from both bands will favor a vanishing of the magnetization as in the usual cases.

The most characteristic feature of the models discussed here, however, both in the absence and in the presence of exchange splitting, arises in optical properties. A transfer of optical spectral weight from high frequencies to low frequencies always occurs upon spin polarization in the presence of the interactions j_1 and j_2 that modify the bandwidth. Furthermore, above T_c these interactions cause spectral weight to be transfered from low to high frequencies as the temperature is lowered. These phenomena have been observed in manganites[8] and hexaborides[9] as a function of temperature. Within the models considered here, the same effects should be seen as a function of magnetic field. In fact, recent optical experiments on Gd-doped Si in the presence of large magnetic fields show just such a remarkable behavior.[10] A systematic search for these effects in other ferromagnets may reveal that they are a universal signature of metallic ferromagnetism, which would lend support to the models discussed here. The magnitude of the effects however, just as the magnitude of the magnetoresistance, is likely to vary widely from material to material. As discussed in Sec. VII, there should be a definite relation between the magnitude of the magnetoresistance and the change in optical absorption with magnetic field, if indeed the dominant effect is the lowering of effective mass with spin polarization, as suggested

by our model, rather than a change in the relaxation time. Further discussion of these issues and the relation of our model with the double exchange model[4] is given elsewhere.[11]

Unfortunately, the model discussed here does not in itself contain the physics of the high-energy degrees of freedom from where the extra optical spectral weight that appears at low frequencies gets transfered from. It would be of great interest to have a model that would describe this high-energy physics and give rise to the Hamiltonian considered here as an effective Hamiltonian for its low energy degrees of freedom.

Finally, photoemission experiments, especially if angle and spin resolved, should be able to provide essential clues on the validity of the model discussed here. An increase in the Fermi velocity of electrons of at least one spin orientation as function of increasing magnetic field or decreasing temperature (below T_c) would be expected to also be a universal feature of metallic ferromagnets if the models discussed here are applicable.

There has recently been substantial interest in reanalyzing the problem of metallic ferromagnetism,[24,25] and there seems to be a consensus that this old problem is still not well understood despite the practical successes of spin-density-functional theory.[3] In particular, recent work has suggested that peaks in the density of states are the dominant mechanism giving rise to ferromagnetism.[24,25] It has also been suggested[25] that because ferromagnetism is a strong coupling problem it is not really possible to pinpoint the single ultimate cause of metallic ferromagnetism, whether peaks in the density of states, band degeneracy, or particular electron-electron interactions. However, the work discussed here and previously[5,6,11] suggests otherwise. We expect metallic ferromagnetism to be always accompanied by the transfer of optical spectral weight discussed above, as well as by a change in the quasiparticle dispersion. Establishing experimentally that these phenomena do not occur would prove the invalidity of the model discussed here. Concerning band degeneracy, if ferromagnetism is found in a system where conduction clearly occurs through nondegenerate bands, such as metallic hydrogen,[26] it would establish that band degeneracy is not essential and lend further support to the model discussed here.

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