Band Theory of Ferromagnetism, Antiferromagnetism, and Spin Waves

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Intra-atomic exchange (Hund's rule mechanism) and Heisenberg nearest-neighbor exchange are examined for their role in the ferromagnetism of metals with degenerate bands. The equations of motion of spin waves valid near $T=0°K$ are derived, and a number of branches are found. When the equations are solved in the approximation that $k_F \ll$ size of the Brillouin zone, the spin waves all have positive energy, and have wave vectors not exceeding $0.9k_F$ in magnitude. When the electrons approximately half-fill the Brillouin zone, it is possible for some spin waves to have negative energy, and for an antiferromagnetic state to become stable. This is not proved, but indicated on the basis of an earlier calculation that also showed that ferromagnetism is less stable against antiferromagnetism in face-centered cubic materials than in either the simple, or body-centered cubic structures.

THE direct exchange mechanism may be characterized by the vector-model interaction,

$$-J_{i,t;j,t'}S_{i,t}\cdot S_{j,t'} \tag{1}$$

connecting an electron on the Wannier site at R_i in band t with another electron at R_j in band t'. Due to the short range of the exchange integral, this interaction is greatest when $R_i=R_j$ (Hund's rule coupling), and there may also be some nonnegligible nearest-neighbor exchange. But there is considerable diver-

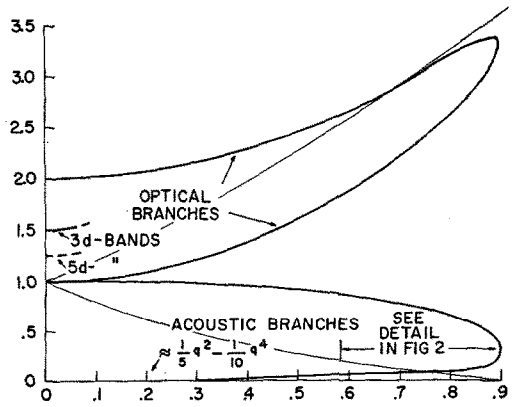

FIG. 1. Solutions of Eq. (3) in the case of two d bands, and $\Delta \geq \epsilon_F$. The vertical axis gives $\hbar\omega(q)/\Delta$, and the horizontal q/k_F. The shaded area indicates the continuum of scattering solutions to the equation. For three or five d bands, the optical spectra are changed as indicated, but the acoustic spectrum remains the same. It is assumed that umklapp may be neglected, and $j_{qr} \doteq j_{or}$.

gence of opinion as to the sign and even magnitude of the latter,[1,2] whereas the Hund's rule mechanism is of the right sign and strength to favor the creation of a net spin on a Wannier site, just as on the atom.

Opposing this creation of local magnetization is the band energy, or "kinetic energy" as we may denote it, which is minimized by filling every state below the Fermi level by two electrons, spin up and down; i.e., the ordinary paramagnetic Fermi sea. Also opposing it is the direct Coulomb interaction, which is not minimized unless the electrons (or holes) are kept apart, and which also serves to eliminate density fluctuations in the charged medium. In the random phase approximation, the equations of motion of the density fluctuations (plasmons) and of the spin waves defined by

$$S^{\dagger}_{q}=\sum_{q,k,t} g_{q,k,t}c^{\dagger}_{k+q,t,\uparrow}c_{k,t,\downarrow} \tag{2}$$

may be calculated independently, and they do not in-

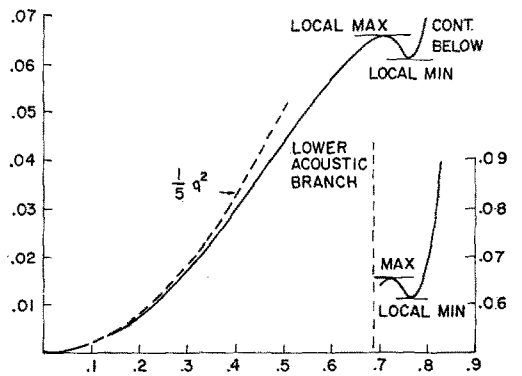

FIG. 2. Lower acoustic branch of Fig. 1 with expanded vertical scale. Detail of entry into continuum in lower right-hand corner. Formula valid for small q is $\hbar\omega/\Delta=(\frac{1}{5})(q/k_F)^2-(\frac{1}{10})(q/k_F)^4$.

terfere with one another. Details of the transformation of spin operators to Wannier operators, and finally to Bloch operators, are given elsewhere[2]; the result is an integral equation for the spin-wave energy $\hbar\omega_r(q)$, in terms of the one-electron energy $\epsilon(k)$, the Fermi function $f(\epsilon)$, and the "exchange matrix eigenvalues" j_{qr}:

$$1=\frac{j_{qr}}{N}\sum\frac{f(\epsilon_k)-f(\epsilon_{k+q}+\Delta)}{\Delta+\epsilon(k+q)-\epsilon(k)-\hbar\omega_r(q)}. \tag{3}$$

The lowest "acoustic" spin-wave branch is labeled $r=0$, the higher branches by $r=1, 2, \cdots$. The Stoner energy gap parameter is,

$$\Delta=j_{00}M/M_{\max}, \tag{4}$$

[1] E. Lieb and D. Mattis, Phys. Rev. **125**, 164 (1962). One of the principal objects of that paper was to prove how unreliable the Hartree–Fock scheme could be in the theory of ferromagnetism, and to show that a strong repulsive interaction was by no means a sufficient condition for ferromagnetism.

[2] D. Mattis, Phys. Rev. **132**, 2521 (1963). Includes the present results, and more detailed calculations and numerical results.

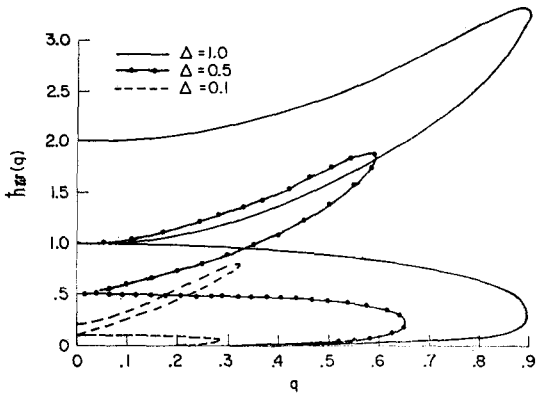

FIG. 3. Spectra for various values of Δ/ϵ_F. Continua cannot be shown, but q_{max} varies approximately linearly with Δ.

where M is the magnetization, and M_{max} the ideal maximum magnetization if all the electrons (holes) had spin up. Finally, j_{qr} are the eigenvalues of the t, t' matrix:

$$j_{q;t,t'}=(1/N)\sum_{i,j}J_{i,t;j,t'}\exp[iq\cdot(R_i-R_j)], \qquad (5)$$

which is the Fourier transform of the exchange matrix. The solutions at $q=0$ may be found exactly

$$\hbar\omega(0)=0, \Delta, \Delta(1-j_{0r}/j_{00}), \qquad (6)$$

and for other values of the wavevector are given in the figures, assuming $j_{qr}=j_{0r}$.

A note on the physical mechanism is in order. Besides producing the plasmon mode, the Coulomb repulsion has the effect of keeping electrons (holes) separated, and therefore prevents them from benefiting from (1); the net effect of the Coulomb interaction is therefore to *decrease* (4) from its optimum value in the Hartree-Fock approximation.[3] In the ideal electron approximation of plane waves and a single nondegenerate band, there can be no ferromagnetism—as first shown by Wigner.[4] A recent theorem[1] both confirms and enhances this result for ferromagnetism. It is believed that Slater first seriously advanced the mechanism of intraatomic exchange, or Hund's rule, as an explanation for ferromagnetism.[5] This is a very particular consequence of the Coulomb interaction, and bands arising from orbitally degenerate atomic states, such as the *d* bands, are required, as well as a sufficiently high density of states near the Fermi level. Antiferromagnetism can also be understood on this basis. For when the de Broglie wavelength of an electron at the Fermi surface is no longer large compared to the intercellular distance, when umklapp becomes important in the solution of Eq. (3), then almost by inspection of this equation it is clear that the spin waves need not all have positive energy solutions and the ferromagnetic state will no longer be stable against some antiferromagnetic spiral configuration.

We have not investigated antiferromagnetism in the full detail which it deserves. But there exists a calculation by Tachiki and Nagamiya[6] which is very similar to results we have previously obtained on the basis of the similar *indirect exchange theory* (i.e., Ruderman-Kittel interaction),[7] which shows the onset of antiferromagnetism when the Fermi surface approximately half fills the Brillouin zone.

What we also found in the indirect exchange theory was that as the number of electrons per atom was increased to this value, the face-centered cubic structure was the first of the three cubic structures to become antiferromagnetic, and by a good margin.[7] This result finds a happy echo in recent neutron diffraction experiments[8] on iron; the archetype ferromagnetic (usually bcc) has been found to be actually antiferromagnetic in its fcc γ phase, in accord with these simple notions.

In Figs. 1–3 we show the results of numerical solution of the integral Eq. (3). The "shaded area" to which reference is made in the caption of Fig. 1 is the area enclosed in the < shaped curve,[2] but the shading did not reproduce in the engraving process.

[3] J. Hubbard (to be published): effect of correlations.
[4] E. Wigner, Trans. Faraday Soc. **205,** 678 (1938).
[5] J. Slater, Phys. Rev. **49,** 537 and 931 (1936).
[6] M. Tachiki and T. Nagamiya, Phys. Letters **3,** 214 (1963).
[7] D. Mattis and W. Donath, Phys. Rev. **128,** 1618 (1962). This paper, reprinted together with extensive lattice sums of the Ruderman-Kittel function in the three principal cubic structures, is available in limited quantities as IBM Research Report R.C. 945 by Mattis, Anthony, and Horwitz. Negative spin-wave energies appear at $n=0.23$ for s.c. and bcc and already at $n=0.16$ for fcc; $n=$electrons per band per atom.
[8] S. C. Abrahams *et al.*, Phys. Rev. **127,** 2052 (1962).