$FeBr_2$, $(FeCl_2)_I$, and $(FeCl_2)_{II}$. The magnetic phase diagram of $FeBr_2$, which is shown in Fig. 2, exhibits changes in the slopes of the transition lines at the tricritical point, as reported at 1 atm by Fert *et al.*[2] For the two structural phases of $FeCl_2$, Fig. 3, a small change in slopes has been observed, but it might be associated with inhomogeneities of the internal magnetic field.

$FeCl_2$ provides a system in which the tricritical point can be studied in two different environments without change in composition: In the low-pressure phase $(FeCl_2)_I$ the tricritical temperature decreases with applied pressure, but in the high-pressure phase $(FeCl_2)_{II}$ it increases as it does in $FeBr_2$.

The zero-pressure values for threshold field, Néel temperature, tricritical temperature, and field and their pressure derivatives are given in Table I. The 1-atm values are in satisfactory agreement with those given by previous experiments.[2]

The variations of exchange and anisotropy can be deduced from these and other measurements such as antiferromagnetic resonance[6] or perpendicular susceptibility[7] under pressure. By relating these variations to the tricritical lines the validity of theories of phase transitions can be tested. Elastic and inelastic neutron scattering studies in $(FeCl_2)_{II}$ and compressibility measurements are now in progress in order to use volume rather than pressure basis. A complete discussion of these results will be reported later.

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Persistence of the Stoner Splitting in Metallic Ferromagnets above T_c †

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Recent neutron-diffraction experiments by Mook, Lynn, and Nicklow, as well as previous photoemission experiments by Pierce and Spicer, provide evidence that the Stoner band splitting in nickel might not go to zero at T_c, as it does in conventional band theory of magnetism. It will be shown that the persistence of such splitting in the paramagnetic regime can be explained quite naturally within the framework of the random-phase–approximation theory of spin fluctuations.

Recently Mook, Lynn, and Nicklow observed using inelastic neutron diffraction[1] the rapid reduction of the spin-wave scattering intensity on entering the stoner continuum in nickel as a function of temperature. They found that the lower edge of the Stoner continuum did not appear to come down as they raised the temperature through the Curie temperature, as Stoner-Hartree-Fock theory would predict. This result lends support to the photoemission results of Pierce and Spicer,[2] which also imply that no

significant changes occur in the band structure of nickel as one passes through the Curie temperature. We shall see that the occurrence of Stoner splitting in the paramagnetic regime is predicted quite naturally by the random-phase-approximation (RPA) expression for the self-energy of the one-electron Green's function.

Brinkman and Engelsberg write down the following expression for this self-energy for a ferromagnet in the vicinity of the Curie temperature:

$$M(\vec{k},\omega) = (U^2/N)\sum_{\vec{k}'}\int S(\vec{k}',\omega')[\omega - \omega' - \epsilon(\vec{k}-\vec{k}')]^{-1}\, d\omega', \tag{1}$$

where $S(\vec{k},\omega)$ is the Fourier transform of the spin-spin correlation function, U is the exchange energy, and $\epsilon(\vec{k})$ is the one-electron energy of the system.[3] This is what they call the "boson term." They also calculate a "fermion term" which they show to be insignificant in the calculation of ReM for a nonzero Curie temperature. Since $S(\vec{k}',\omega')$ is a sharply peaked function of ω' (i.e., its width is much less than the bandwidth), we may to a good approximation write

$$\mathrm{Re}M(\vec{k},\omega) = (U^2/N)\mathrm{P}\sum_{\vec{k}'}\{S(\vec{k}')/[\omega - \epsilon(\vec{k}-\vec{k}')]\}, \tag{2}$$

where P signifies taking the principal part and where

$$S(\vec{k}') = \int d\omega'\, S(\vec{k}',\omega').$$

Brinkman and Engelsberg found that the resulting effective mass did not diverge at the Curie temperature, but it led to a divergent specific heat. We shall see shortly that the situation is actually even more interesting. Right at T_c there are two solutions to

$$\omega - \epsilon(\vec{k}) - \mathrm{Re}M(\vec{k},\omega) = 0 \tag{3}$$

for each $\vec{k}$, implying that the Stoner splitting remains when $T = T_c$, even for weak exchange interaction.

To show this, let us calculate Re$M(\vec{k},\omega)$ for $\Delta\omega = \omega - \epsilon(\vec{k})$ very small in magnitude since if the RPA is valid anywhere it should be valid for small U and hence small splitting. In this limit, Eq. (1) becomes

$$\mathrm{Re}M(\vec{k},\omega) - \frac{\Omega}{(2\pi)^3}U^2\mathrm{P}\int_{R_1} d^3k' \frac{S(\vec{k})}{\Delta\omega + \vec{v}_{\vec{k}}\cdot\vec{k}'} + \frac{\Omega}{(2\pi)^3}U^2\mathrm{P}\int_{R_2} d^3k' \frac{S(\vec{k}')}{\epsilon(\vec{k}) - \epsilon(\vec{k}-\vec{k}')}, \tag{4}$$

where R_1 is a region at the middle of the Brillouin zone of radius $\bar{k}$ much smaller than the radius of the Brillouin zone but much greater than $|\Delta\omega|/|v_{\vec{k}}|$, where $v_{\vec{k}}$ is $\nabla_{\vec{k}}\epsilon(\vec{k})$ and Ω is the unit cell volume. The last term is independent of $\Delta\omega$; the first term, assuming that $S(\vec{k}')$ is the Ornstein-Zernike function $(k^2 + k_c^2)^{-1}$, where k_c is the reciprocal of the correlation length, is proportional to

$$\int_0^{\bar{k}} \frac{k'\,dk'}{k'^2 + k_c^2} \ln\left|\frac{\Delta\omega + |\vec{v}_k|k'}{\Delta\omega - |\vec{v}_k|k'}\right|. \tag{5}$$

Since $\bar{k} \gg \Delta\omega/|\vec{v}_k|$, $\bar{k}$ can be taken to be infinite and the integral can be evaluated exactly by contour integration for small k_c to yield

$$\tfrac{1}{2}\pi[\pi - 2\arctan(k_c v_k/2|\Delta\omega|)]\,\mathrm{sgn}(\Delta\omega). \tag{6}$$

Thus, for $k_c = 0$, Eq. (3) has two solutions, no matter how weak the interaction is. When we are away from T_c and $k_c \neq 0$, the same two solutions will certainly exist if they both occur for values of $\Delta\omega$ for which $\Delta\omega \gg \frac{1}{2}k_c v_k$, and thus there is a range of temperatures above T_c for which there is band splitting (see Fig. 1). Since when k_c becomes so large that $\frac{1}{2}k_c v_k$ is much greater than $\Delta\omega$, Eq. (6) becomes zero, there will clearly exist a maximum value of k_c (and hence a temperature above T_c) beyond which only one solution to Eq. (3) will exist.

Using the full expression for the self-energy (i.e., including both "fermion" and "boson" terms),[3] the imaginary part is found to be given by

$$\mathrm{Im}M(\vec{k},\omega) = (\pi/N)U^2\sum_{\vec{k}'}\int d\omega'\,\delta(\omega - \omega' - \epsilon(\vec{k}-\vec{k}'))\,\mathrm{Im}\chi(\vec{k}',\omega')\left[\tanh\left(\frac{\omega'}{2kT}\right) + \coth\left(\frac{(\omega-\omega')}{2kT}\right)\right], \tag{7}$$

(all energies measured from the Fermi energy). If we use the standard RPA expression for χ, we obtain an integral which is very much like the one done by Quinn and Ferrell[4] for the imaginary part of the self-energy of an electron gas, but with the Coulomb interaction replaced by U. If ω is taken to be very close to the Fermi surface

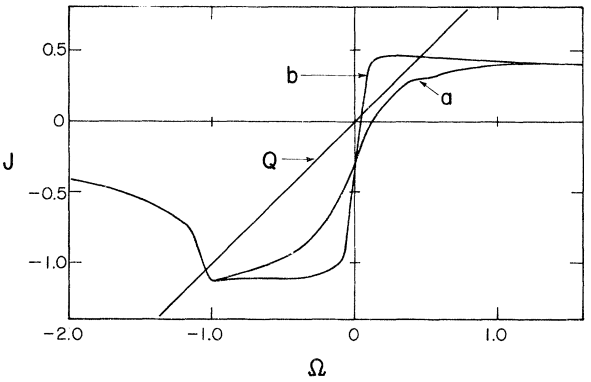

FIG. 1. Plots of the dimensionless quantities $J=M(\vec{k},\omega)/\epsilon(\vec{k})$ and $Q=[\epsilon(\vec{k})/U]^2[\Delta\omega/\epsilon(\vec{k})]$ versus $\Omega=\Delta\omega/\epsilon(\vec{k})$ for parabolic bands for $\epsilon(\vec{k})/U=1$. We approximate the Brillouin zone boundary by cutting off our integrals over k at a value k_1. These plots are for $k=0.6\,k_1$. Curve a, $X_0=k_c/k_1=0.1$; curve b, $X_0=0.01$.

[as occurs if the splitting is very small and $\epsilon(\vec{k})$ lies near the Fermi surface] and if we use parabolic bands, ImM goes as $(k/k_F-1)^2$, as shown by Quinn and Ferrell.[4] Thus, since ImM is small there will be two distinct peaks in the spectral density for $\epsilon(\vec{k})$ near the Fermi level.[5]

Actually, as seen in Fig. 1, when $k_c\neq 0$, there appear three solutions to Eq. (3) instead of two. Although it is not difficult to understand two solutions physically, it is more difficult to understand three. Perhaps it is spurious and shows a breakdown of perturbation theory. Perhaps it is real and comes about because when the range of the correlations is not infinite (i.e., above T_c), there exist large regions where there is very little fluctuating spin polarization and this leads to the central resonance in the spectral density [i.e., the middle solution to Eq. (3)]. The middle

$$G(\vec{k},\omega)=\tfrac{1}{2}[\omega-\epsilon(\vec{k})-Um_0]^{-1}+\tfrac{1}{2}[\omega-\epsilon(\vec{k})+Um_0]^{-1}. \tag{10}$$

This is the physically expected result when the spin correlations are infinitely long range. The reason for obtaining an unphysical result from the supposedly better approximation of including M in the G used to find M is that to be consistent, it is apparently necessary to renormalize the vertex function in addition to the Green's function, which we have not done.[7]

If we try to apply the RRPA to paramagnetic metals, which are nearly ferromagnetic (for which the second-order perturbation expansion of the mass operator is likely to be valid), we find that in addition to the term given in Eq. (1), there is the "fermion" term[3] considered by Doniach and Engelsberg[8] which leads to the effective mass of an electron becoming large at T_c. If we include this term, expanding it to first order in ω, and defining the Fermi energy to be zero, we see from Eq. (6) and Ref. 8 that to find the solutions to Eq. (3) for $\vec{k}$ on the Fermi surface, near T_c we must solve the equation

$$A_0\ln(k_1/k_c)\Delta\omega=f(\Delta\omega), \tag{11}$$

where

$$f(\Delta\omega)=U^2B\left(1-\frac{2}{\pi}\arctan\frac{k_c|v_k|}{2|\Delta\omega|}\right), \tag{12}$$

solution can be made to occur at $\Delta\omega=0$ if we include the second (i.e., constant) term in Eq. (4) in the definition of $\Delta\omega$. This constant term is the standard second-order perturbation-theory shift of the one-electron energy $\epsilon(\vec{k})$ of $\Delta\omega$. The first term in Eq. (4) represents the Stoner-like splitting. Let us call this the renormalized RPA or RRPA.

If we do this, however, why not include the full self-energy, including the first term in Eq. (4), in the Green's function used to calculate the self-energy, as this would appear to be a more consistent approximation? Such an approximation was originally used by Izuyama in discussing the wave vector dependence of the susceptibility near T_c.[6] If we do this and perform the resulting integrals for $T=T_c$ for small $\Delta\omega$, we find that now there is only one solution for weak interactions. We can understand why this may be so if we consider this same approximation in the hypothetical limit in which $S(\vec{k}')$ is a δ function (i.e., in real space the correlation function is constant). We then find that

$$M(\vec{k},\omega)=U^2m_0^2/[\Delta\omega-M(\vec{k},\omega)], \tag{8}$$

where m_0 is the square root of

$$N^{-1}\textstyle\sum_{\vec{k}}S(\vec{k}).$$

When we solve for M, and using M find the spectral density, we find that it is proportional to

$$[(Um_0)^2-(\tfrac{1}{2}\Delta\omega)^2]^{1/2} \tag{9}$$

for $|\Delta\omega|<2Um_0$ and 0 for $\Delta\omega>2Um_0$. If as done previously we do not include M in the Green's function used to calculate the self-energy, we then find that in the limit that $S(\vec{k})$ becomes a δ function in $\vec{k}$, the Green's function for electrons of either spin up or spin down becomes

where A_0 and B are constants. Since the initial slope of $f(\omega)$ is given by

$$\partial f/\partial(\Delta\omega) = (4/\pi)BU^2(2/k_c v_k), \tag{13}$$

there clearly exists a sufficiently small value of k_c or a sufficiently large value of U (but still $\ll$ the bandwidth) so that the curves $f(\Delta\omega)$ versus $\Delta\omega$ and $\Delta\omega A_0 \ln(k_1/k_c)$ versus $\Delta\omega$ intersect at the three points mentioned earlier. Since the magnitude of the splitting is now proportional to

$$f[\ln(k/k_c)]^{-1}, \tag{14}$$

however, the band splitting will reduce to zero when $k_c = 0$. Thus, for very nearly ferromagnetic metals, we should see the appearance of a small band splitting near $T = 0$ which gets smaller as we approach absolute zero temperature.

Although it is not entirely clear that second-order mass-operator perturbation theory should be correct for interactions not small compared to the bandwidth (as occur in nickel), since as seen earlier this approximation gives physically correct results in the limit in which $S(\vec{k})$ becomes a δ function, we expect that it may also be good for correlations that fall off as $1/r$ as occurs at T_c in Ornstein-Zernike theory. (The result that in the presence of correlations correct results can often be obtained in perturbation theory in a regime where perturbation theory would not ordinarily be expected to be valid is the basis of several theories of liquid metals.[9]) Then, let us use the free-electron gas model of nickel of Lowde and Windsor[10] to find the band splitting. We find by procedures illustrated by Fig. 1 that in the RPA, the solutions to Eq. (3) occur at $\Delta\omega = -1.02\epsilon(\vec{k})$ and $0.5\epsilon(\vec{k})$, and in the RRPA they occur at $\Delta\omega = -0.81\epsilon(\vec{k})$ and $0.75\epsilon(\vec{k})$ for $k_c = 0$. If $\epsilon(\vec{k})$ is taken to be the Fermi energy, this splitting is of the order of the zero-temperature Stoner splitting. Since as we go below T_c we expect the Stoner splitting not to decrease, the behavior observed in Ref. 1 is consistent with the present theory.

In order to make contact with the results of Ref. 1, we will now try to calculate the susceptibility. Using an approach like that used by Izuyama,[6] we write

$$\chi(\vec{q},\omega) = \chi_0(\vec{q},\omega)/[1 - \bar{U}(\vec{q})\chi_0(\vec{q},\omega)],$$

where $\bar{U}(\vec{q})$ is U plus an interatomic exchange interaction $J(\vec{q})$ [$J(\vec{q})$ is defined so that $J(0) = 0$, i.e., $J(0)$ is absorbed into U]. Here[6]

$$\chi_0(\vec{q}, i\omega_\nu) = \sum_{\nu',\vec{k}} G(\vec{k}+\vec{q}, i\omega_\nu + i\omega_{\nu'}) G(\vec{k}, i\omega_{\nu'}).$$

Using the RRPA result for M (at T_c where $k_c = 0$), and hence G [i.e., keeping only the first term in Eq. (6)] for the case of $k_c = 0$, we find that in the weak-interaction limit (where RPA is expected to be valid) there are no spin-wave poles in χ for sufficiently small q and ω. It has been assumed that T is sufficiently small for ImM to be neglected. It is difficult to estimate how small q and ω must be, but this value of q appears to be considerably greater than the observed minimum values of q where spin waves have been observed.[1] In this regime χ becomes like the RPA paramagnetic state χ, and thus the correlation function has Ornstein-Zernike form for small[11] q as assumed in the beginning of this Letter. For larger q, there are spin-wave poles in χ. [They would be unstable modes had $J(\vec{q})$ not been included but this is a well-known defect of the single-band model[12]; had a multiband model been used we would not need $J(\vec{q})$ for spin waves to be well defined.] Thus, the exact point of intersection with the continuum cannot be predicted in this model because it is too crude [i.e., $J(\vec{q})$ is not known], but the Stoner-like splitting in the paramagnetic phase does not vary too much with increasing k_c (and hence with increasing temperature), at least for the parameters appropriate to nickel until the splitting disappears suddenly at a sufficiently large value of k_c. It is not expected that the spin-wave poles will do more to the self-energy above T_c than below—i.e., cause small broadening and shifts in the energies which should not affect the conclusions about band splitting for $T > T_c$.

Below T_c straightforward application of RRPA as done previously gives split bands of each spin of the form $\epsilon(\vec{k}) - \frac{1}{2}\sigma U m - \sigma\Delta$, where Δ reduces to the first integral in Eq. (6) when $m = 0$; m is the magnetization, and σ is the spin of the electron considered. Thus, the paramagnetic spin-split bands go smoothly into the ferromagnetic split bands. Well below T_c, Δ should approach zero as T approaches zero because the magnetic fluctuations do.

It should be pointed out that this problem could have been treated as that of an electron moving in a Hartree-Fock effective field but with the exchange interaction a disordered potential. Using the methods of Ref. 9, we obtain the "boson part" (in the language of Ref. 3) of the self-energy to lowest order in U.

If the Stoner splitting does not decrease as we go above T_c, the question is: "What causes magnetism in metals?" This will be the subject of

future work.

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Lifetimes in the Ground-State Band of ^{184}Hg

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The ground-band level sequence in ^{184}Hg has been identified to spin 10^+ and possibly to 14^+. Lifetimes for the 2^+, 4^+, and 6^+ states have been measured. The results imply the onset of a permanent deformation above the 2^+ level.

Isotope-shift measurements on the odd-mass mercury isotopes reveal a sudden increase in nuclear volume between $A = 187$ and $A = 185$.[1,2] This suggests that a dramatic shape change occurs in the Hg isotopes which might be associated either with a transition from a spherical to a deformed shape or from a spherical shape to a spherical shell or bubble nucleus.[3-5] The ground-state band of ^{186}Hg has been identified by Proetel *et al.*[6] They found the first 2^+ state at 405 keV, similar to the situation for the heavier Hg isotopes, and it seems unlikely that ^{186}Hg is deformed in its ground state. Hornshøj *et al.*[5] have studied the α decay of ^{188}Pb and they conclude that the 2^+ level in ^{184}Hg must be higher than 300 keV to account for the absence of fine structure in the α spectrum.

In a series of experiments at the Chalk River MP tandem Van de Graaff we have identified the ground-state band of ^{184}Hg by applying the techniques of in-beam γ-ray spectroscopy to the reaction ^{156}Gd(^{32}S, $4n$)^{184}Hg. The yield was found to peak at about 156 MeV bombarding energy, about as expected for the evaporation of four nucleons from the compound nucleus. To corroborate the mass assignment, recoils from the target were stopped downstream and their γ rays were counted with a large-volume Ge(Li) detector shielded from direct target radiation. Essentially all of the γ rays seen were from the known $A = 184$ decay chain.[7] The Z identification was made by showing that the cascade γ rays from the ground-state band were in coincidence with Hg K x rays. γ-γ coincidences were used to establish the coincidence relationships in the cascade. The angular distributions were those expected for a stretched sequence of $E2$ transitions. The sequence of γ-ray energies is unusual in that both the $4^+ \rightarrow 2^+$ and $6^+ \rightarrow 4^+$ transitions are lower in energy than the $2^+ \rightarrow 0^+$. This ordering is clear from the relative intensities in Table I and is confirmed by decay curves measured by the recoil-distance