PHYSICAL REVIEW VOLUME 143, NUMBER 1 MARCH 1966

Itinerant Antiferromagnetism*

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(Received 8 September 1965)

The itinerantly antiferromagnetic or spin-density-wave state is examined for an electron gas and a more realistic band model. The paramagnetic electron gas, which is unstable to the formation of an antiferromagnetic state in the Hartree-Fock approximation, is shown to be stable when either static Fermi-Thomas or dynamic screening is included. A mathematical model is constructed for the spin-density-wave state below its transition temperature and its experimental consequences examined. A criterion for itinerant antiferromagnetism in a band model is derived and shown to be consistent with the antiferromagnetism of Cr. A crude model is employed to compute qualitatively the thermodynamic and electromagnetic properties of an itinerant antiferromagnet like Cr. In particular, the specific heat, spin susceptibility, collective modes, and electromagnetic absorption are evaluated.

1. INTRODUCTION

FOR the past few years there has been contention about whether the ground state of a high-density electron gas is paramagnetic or antiferromagnetic and whether the antiferromagnetism of Cr is related to the Hartree-Fock instability of a free-electron gas. This paper discusses these problems.

As Overhauser[1] has stressed, when the Coulomb interactions in an electron gas are treated in the Hartree-Fock approximation, the antiferromagnetic (spin-density-wave) state has lower energy than the paramagnetic state. This conclusion has also been corroborated by Fukuda, Iwamoto, and Sawada.[2,3] However, the interesting question of whether the screened electron gas is unstable toward antiferromagnetism has not been examined.[4]

We investigate the question in Sec. 2 by examining the stability of spin density waves in the paramagnetic state. We show that when Fermi-Thomas static screening is taken into account, the Overhauser instability disappears. We also show, albeit crudely, that the more realistic dynamically screened Coulomb potential eliminates the instability. The crudeness of the latter demonstration lies in the fact that we do not allow for modification of the dynamic screening from the random-phase approximation.

In Sec. 3, we briefly investigate the properties of a hypothetical electron gas antiferromagnet assuming that screening can be ignored. The resulting system is shown to exhibit a small specific heat at low temperatures and a discontinuity in the specific heat at the transition temperature. The state does not exhibit optical absorption.

In Sec. 4, the stability of a more realistic band model is investigated. The band structure introduces many complications. To cope with them umklapp processes in the screened exchange interaction are neglected and a static local screened potential is employed. A semiquantitative criterion for antiferromagnetism is derived. Whether a metal is antiferromagnetic or not depends almost exclusively on its band structure and in particular on its Fermi surface. The Bloch states and screening length are relatively unimportant. The criterion predicts that Cr is antiferromagnetic by the Lomer[5] two-band mechanism. It seems highly unlikely that any one-band model could fulfill this criterion.

A very crude two-band model representative of Cr is chosen in Sec. 5 to calculate some of the thermodynamic and electromagnetic properties of an itinerant antiferromagnet. A pairing of electrons with different spins in different bands is responsible. The specific heat, spin susceptibility, collective modes, and electromagnetic absorption are shown to agree with existing experiments. Some features of the crude model are dependent on its unrealistic Fermi surface and would undoubtedly be altered in a more realistic picture.

2. STABILITY OF THE FREE-ELECTRON GAS

In this section, we shall investigate the stability of a paramagnetic electron gas with respect to the formation of an antiferromagnetic state. As usual, the electrons are assumed to move in a uniform background of positive charge. The Hamiltonian is

$$H=\sum_\sigma \int d^3r\, \psi_\sigma^\dagger(\mathbf{r}t)[-(\nabla^2/2m)-nv_0]\psi_\sigma(\mathbf{r}t)$$
$$+\tfrac{1}{2}\sum_{\sigma\sigma'}\int d^3r d^3r'\, \psi_\sigma^\dagger(\mathbf{r}t)\psi_{\sigma'}^\dagger(\mathbf{r}'t)$$
$$\times v(\mathbf{r}-\mathbf{r}')\psi_{\sigma'}(\mathbf{r}'t)\psi_\sigma(\mathbf{r}t)\,, \quad (2.1)$$

with $v(\mathbf{r})$ the Coulomb potential between electrons and $nv_0=n\int d^3r\,(e^2/\mathbf{r})$ the interaction energy of the electrons

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* Based in part upon a portion of the doctoral thesis submitted by P. A. Fedders to the Physics Department of Harvard University, 1965. Supported in part by the National Science Foundation.

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[1] A. W. Overhauser, Phys. Rev. **128**, 1437 (1962).

[2] F. Iwamoto and K. Sawada, Phys. Rev. **126**, 887 (1962).

[3] N. Fukuda, F. Iwamoto, and K. Sawada, Phys. Rev. **135**, A932 (1964); see Appendix.

[4] There appears to be a widespread but erroneous notion that the latter authors investigated this question.

[5] W. M. Lomer, Proc. Phys. Soc. (London) **80**, 489 (1962).

with the uniform background. The operator $\psi_\sigma(\mathbf{r}t)$ destroys an electron of spin σ at the space-time point $(\mathbf{r},t)$. We have set $\hbar=1$.

Since the susceptibility is a measure of the response of the system to a spin density disturbance, we may use it to examine spin density fluctuations and thus the differential stability of the system. The growth of uniform spin density fluctuations in the paramagnetic state indicates it is not even metastable to the ferromagnetic state; the growth of spin density fluctuations with a finite wave number indicates the corresponding instability to antiferromagnetism. Conversely, the decay of spin density fluctuations indicates stability to small deviations and thus, at worst, metastability. We shall show in this section that the paramagnetic state of an electron gas is unstable to the formation of spin density waves in the Hartree-Fock approximation but at least metastable in a better approximation which takes screening into account.

We examine these spin density fluctuations by calculating

$$\chi_{ij}(12)=\langle T(M_i(1)M_j(2))\rangle \tag{2.2}$$

whose arguments, n, stand for space-time coordinates $(\mathbf{r}_n,t_n)$ of the magnetization density (μ is the electron moment)

$$M_i(1)=\mu\sum_{\alpha,\beta}\psi_\alpha{}^\dagger(1)\sigma_{\alpha\beta}{}^{(i)}\psi_\beta(1).$$

The symbol $T(\ )$ indicates that the bracketed operators are to be time ordered, and the angular brackets, $\langle X\rangle$, that the expectation value $\mathrm{Tr}\rho X$ is to be taken over a thermal equilibrium ensemble ρ. In the paramagnetic state $\chi_{ij}(12)$ is proportional to δ_{ij}; we can therefore consider any of the equal elements, say χ_{zz}.

The actual nonlocal frequency-dependent susceptibility is given by

$$\chi(\mathbf{r}\mathbf{r}',\omega)=i\int_0^\infty dt\, e^{i\omega t}\langle[M(\mathbf{r}t),M(\mathbf{r}'0)]\rangle.$$

It can be obtained from (2.2) by the algorithm

$$\chi(\mathbf{r}\mathbf{r}',\omega)=\beta\lim_{\epsilon\to 0^+}\lim_{\omega_\nu\to\omega+i\epsilon}\int_0^1 ds\int_0^1 ds'\, e^{\beta\omega_\nu(s-s')}\chi(\mathbf{r}s;\mathbf{r}'s').$$

In this expression, as usual, $\beta=1/k_BT$, $s=it/\beta$ is an imaginary dimensionless time, and $\omega_\nu=\pi\nu/-i\beta$, where ν is an even integer. The notation is discussed further in Appendix A.

For a system of interacting particles which move about through space, no equation can be derived directly for χ. Instead one finds an integral equation for what we may call the "fractional susceptibility" or the correlation function for electron-hole scattering L,

$$L(11',22')=\tfrac{1}{2}\langle T(\sum_{\alpha\beta}\psi_\alpha{}^\dagger(1')\sigma_{\alpha\beta}{}^{(z)}\psi_\beta(1)\sum_{\gamma\delta}\psi_\gamma{}^\dagger(2')\sigma_{\gamma\delta}{}^{(z)}\psi_\delta(2))\rangle. \tag{2.3}$$

χ can be extracted by setting $1'=1$, $2'=2$ and multiplying by $2\mu^2$.

Our method for examining the metastability of the paramagnetic state is to examine the analytic behavior of the Fourier transform of L. More specifically, if we take an electron gas, which has translational invariance, and set $t_1'=t_1$, $t_2'=t_2$, L can be written in momentum space as

$$\begin{aligned}\langle\mathbf{p}|L(\mathbf{q}\omega)|\mathbf{p}'\rangle &= \lim_{\omega_\nu\to\omega+i\epsilon}\beta\int_0^1 ds\, e^{\beta\omega_\nu s}\tfrac{1}{2}\langle T(\sum_{\alpha\beta}\psi_\alpha{}^\dagger(\mathbf{p}-\tfrac{1}{2}\mathbf{q},s)\sigma_{\alpha\beta}{}^{(z)}\\ &\times\psi_\beta(\mathbf{p}+\tfrac{1}{2}\mathbf{q},s)\sum_{\gamma\delta}\psi_\gamma{}^\dagger(\mathbf{p}'+\tfrac{1}{2}\mathbf{q},0)\sigma_{\gamma\delta}{}^{(z)}\psi_\delta(\mathbf{p}'-\tfrac{1}{2}\mathbf{q},0))\rangle,\end{aligned} \tag{2.4}$$

and the total susceptibility as the integral of this fractional susceptibility

$$\chi(\mathbf{q}\omega)=2\mu^2\int\frac{d^3p}{(2\pi)^3}\frac{d^3p'}{(2\pi)^3}\langle\mathbf{p}|L(\mathbf{q}\omega)|\mathbf{p}'\rangle. \tag{2.5}$$

$\langle\mathbf{p}|L(\mathbf{q}\omega)|\mathbf{p}'\rangle$ is the electron-hole pair correlation function between electron-hole pairs of total momentum $\mathbf{q}$, opposite spin, and relative momenta $\mathbf{p}$ and $\mathbf{p}'$. The statement that $L(\omega)$ has a pole in the upper half ω plane means that $L(t)$ grows exponentially. In other words, the paramagnetic state is differentially unstable to the formation of spin density waves of wave number $\mathbf{q}$.

The equation for L is deduced in Appendix A using Green's-function techniques. The problem divides itself naturally into two parts: first, an exact integral equation for L in terms of an exact interaction kernel J between the electrons and holes,

$$\begin{aligned}L(11',22')&=L_0(11',22')+\int d3d3'd4d4'\\ &\times L_0(11',33')J(33',44')L(44',22'),\end{aligned} \tag{2.6}$$

where $\int dn$ is shorthand for $\int d\mathbf{r}_n dt_n$ and L_0 is the free $(v=0)$ electron-hole pair correlation function; second, an iterative expansion determining the exact electron-hole interaction kernel J. A consistent and straightforward method of characterizing successive approximations for J or L with necessary conservation and symmetry properties[6] can be based on approximations to the free energy. Associated with each approximation to the free energy of the high-density electron gas to some order in the dimensionless parameter $r_s=me^2/\alpha p_F$ [p_F is the Fermi momentum; $\alpha=(4/9\pi)^{1/3}$] is a unique approximation for J which is self-consistent to that order. It is this approximation which we refer to when we call an equation for L exact to a given order in r_s.

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[6] G. Baym and L. P. Kadanoff, Phys. Rev. **124**, 287 (1961); G. Baym, *ibid.* **127**, 1391 (1962).

The first such approximation is the Hartree-Fock approximation, exact to order $1/r_s$. To this order, J is given by the unscreened Coulomb potential. The second such approximation is a dynamically screened approximation which is exact to order $\ln r_s$. It is in this approximation that we examine the instability.

The equation for L to a certain order in r_s involves the Coulomb potential v and the single-particle Green's function or propagator. There exists a well-defined propagator to the correct order in r_s which is also uniquely dictated by the requirement of self-consistency with the free energy to that order. For the $1/r_s$ approximation, the propagator contains the familiar Hartree-Fock single-particle energy

$$E_{\mathrm{HF}}(p)=\frac{p^2}{2m}-\frac{e^2p_F}{2\pi}\left[2+\frac{p_F{}^2-p^2}{pp_F}\ln\left|\frac{p+p_F}{p-p_F}\right|\right]. \quad (2.7)$$

In practice, the errors resulting from the use of the free particle or Hartree energy

$$E_0(p)=p^2/2m \quad (2.8)$$

play a negligible numerical role in our consideration to order $1/r_s$ or $\ln r_s$.

Rather than talk in terms of the fractional susceptibility L (the wave function or Green's function for electron-hole scattering), it is sometimes more convenient to restate the problem in terms of the quantity T (the scattering matrix for electron-hole scattering) related to L by the equation

$$\int d2d2'T(11',22')L_0(22',33')=\int d2d2\,J(11',22')L(22',33'). \quad (2.9)$$

T, which satisfies the equation

$$T(11',22')=J(11',22')+\int d3d3'd4d4'\times J(11',44')L_0(44',33')T(33',22'), \quad (2.10)$$

is a "generalized pseudopotential" for electron-hole scattering. When we find the Fourier transform of T has a pole with the imaginary part of ω greater than zero, L will also have this pole and the paramagnetic state will be unstable.[7]

The Hartree-Fock Approximation (for Arbitrary Temperature)

From (A15), (A17), and (A22) we see that in the Hartree-Fock approximation the electron-hole pair correlation function satisfies the equation

$$[\omega-\Delta E(\mathbf{p},\mathbf{q})]\langle\mathbf{p}|L(\mathbf{q}\omega)|\mathbf{p}'\rangle+\mathcal{P}(\mathbf{p},\mathbf{q})\int\frac{d^3p''}{(2\pi)^3}v(\mathbf{p}-\mathbf{p}'')\langle\mathbf{p}''|L(\mathbf{q}\omega)|\mathbf{p}'\rangle=\mathcal{P}(\mathbf{p},\mathbf{q})(2\pi)^3\delta(\mathbf{p}-\mathbf{p}'). \quad (2.11)$$

L is a function of only one time difference in this approximation. For convenience the abbreviated notation

$$\Delta E(\mathbf{p},\mathbf{q})=E(\mathbf{p}+\tfrac{1}{2}\mathbf{q})-E(\mathbf{p}-\tfrac{1}{2}\mathbf{q}),$$
$$\mathcal{P}(\mathbf{p},\mathbf{q})=f[E(\mathbf{p}+\tfrac{1}{2}\mathbf{q})-\mu]-f[E(\mathbf{p}-\tfrac{1}{2}\mathbf{q})-\mu]$$

is introduced where μ is the chemical potential and f is the Fermi factor $f(x)=[e^{\beta x}+1]^{-1}$.

Equation (2.11) is just the Schrödinger-like equation for the electron-hole pair with momentum $\mathbf{q}$ and opposite spin created by $\mathrm{tr}(\psi^\dagger(\mathbf{p}+\tfrac{1}{2}\mathbf{q})\sigma^{(z)}\psi(\mathbf{p}-\tfrac{1}{2}\mathbf{q}))$. The interpretation is straightforward. ω is the time derivative, $\Delta E(\mathbf{p},\mathbf{q})$ is the energy of the noninteracting electron-hole pair (measured from the Fermi energy), and the remaining term on the left is the interaction term between the electron-hole pair.

Equivalently, T is the solution to the equation[8]

$$\langle\mathbf{p}|T(\mathbf{q}\omega)|\mathbf{p}\rangle=v(\mathbf{p}-\mathbf{p}')-\int\frac{d^3p''}{(2\pi)^3}v(\mathbf{p}-\mathbf{p}'')\frac{\mathcal{P}(\mathbf{p}'',\mathbf{q})}{\Delta E(\mathbf{p}'',\mathbf{q})-\omega}\langle\mathbf{p}''|T(\mathbf{q}\omega)|\mathbf{p}'\rangle \quad (2.12)$$

in the Hartree-Fock approximation. It represents the process where an electron-hole pair of total momentum $\mathbf{q}$ and relative momentum $\mathbf{p}$ scatter into an electron-hole pair of total momentum $\mathbf{q}$ and relative momentum $\mathbf{p}'$. The poles in ω of the scattering matrix give the energies of the intermediate states and their decay (or growth) rates.

We shall not dwell on the fact that $\langle\mathbf{p}|T(\mathbf{q}\omega)|\mathbf{p}'\rangle$ has a pole above the real axis when $\mathbf{q}\approx 2p_F$ and the temperature is below a certain critical temperature since this is merely a restatement of Overhauser's (or Fukuda's) conclusions. We merely remark that the temperature at which the instability first occurs and the wave number $\mathbf{q}$ at which this happens are the critical temperature and wave number of the ensuing antiferromagnetic state at the critical temperature. Below this temperature it is nonsense to discuss the paramagnetic state since it is unstable (not even metastable) with respect to the formation of a static spin density wave with wave number near $2p_F$. The proof that T always has such a pole for the unscreened Coulomb potential is given in Appendix B. The temperature at which the pole first occurs depends on r_s.

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[7] This should not be confused with the t approximation for particle scattering frequently employed at low densities.

[8] While there clearly exists a variational principle for this equation, whose stationary value is the susceptibility, we have not found it tractable analytically. We therefore refrain from discussing it, referring the reader, if interested, to the doctoral dissertation of P. A. Fedders, Appendix B.

Effects of Screening

We have proved that the paramagnetic Hartree-Fock electron gas is unstable with respect to formation of an itinerant antiferromagnetic state below a certain critical temperature. However, the Hartree-Fock approximation is quite often unrealistic in dealing with Coulomb interaction problems. The long-range nature of the potential gives rise to an effective screened potential which, in general, includes both static and dynamic effects.

Before introducing the equation for T in a dynamically screened approximation which is exact to order $\ln r_s$, let us first consider only static screening. Heuristically, static screening can be included by introducing a Yukawa or screened Coulomb potential of the form

$$v(r)=(e^2/r)\exp(-\xi r)\,, \tag{2.13}$$

where ξ is the screening momentum or inverse screening length. One usually uses the Fermi-Thomas momentum ξ_{FT} for its value. We shall first show that there is a screening length below which the antiferromagnetic instability does not persist and shall put an upper bound ξ_C on the inverse length. Furthermore, for all r_s, ξ_{FT} exceeds ξ_C so that an electron gas with static Fermi-Thomas screening is always stable. The proof follows.

If there is a pole in T, then, in the neighborhood of that pole, the finite inhomogeneous term v in (2.12) can be neglected.

$$\phi(\mathbf{p})=-\int\frac{d^3p''}{(2\pi)^3}v(\mathbf{p}-\mathbf{p}'')\frac{\mathcal{P}(\mathbf{p}'',\mathbf{q})}{\Delta E(\mathbf{p}'',\mathbf{q})-\omega}\phi(\mathbf{p}'')\,,$$

where $\phi=\mathrm{Res}T$. For this equation to be satisfied, there must be some value of $\mathbf{p}$, in particular, the $\mathbf{p}$ for which $\mathrm{Res}T$ has its maximum value, for which

$$|R(\mathbf{p})|=\left|\int\frac{d^3p''}{(2\pi)^3}v(\mathbf{p}-\mathbf{p}'')\frac{\mathcal{P}(\mathbf{p}'',\mathbf{q})}{\Delta E(\mathbf{p}'',\mathbf{q})-\omega}\right| \tag{2.14}$$

is greater than one. Let us therefore see when $|R(\mathbf{p})|$ can be largest, letting $\omega=0$, since that is the most favorable circumstance for the instability. By examining R as a function of $\mathbf{p}$ and $\mathbf{q}$, one may show that R has a maximum at $\mathbf{p}=0$ and $\mathbf{q}=2p_F$ for sufficiently small ξ.

At the point $\omega=0$, $\mathbf{q}=2p_F$, and $\mathbf{p}=0$, R is given by

$$R=\alpha r_s/2\pi\{\ln^2(2p_F/\xi)-\mathcal{L}_2(-1)+\tfrac{1}{2}\mathcal{L}_2(-(\xi/2p_F)^2)\} \tag{2.15}$$

for $\xi\leq 2p_F$, where $\mathcal{L}_2$ is the dilogarithm $\mathcal{L}_2(x)=\sum x^n/n^2$. ξ_C is determined by setting $R=1$. From Table I we can see that $\xi_{\mathrm{FT}}>\xi_C$ for all values of r_s up to 6. For larger r_s, ξ_C is bounded by $\frac{1}{2}\xi_{\mathrm{FT}}$. Recall that $R\leq 1$ is a sufficient but not a necessary condition for stability. Note further that the statically screened electron gas is most stable for small r_s, the region where the equation is most valid. As a final comment, note that the first Fredholm determinant, which would replace the maximum value of R by its mean value, would not give a poor estimate of the requirement for instability when the potential has a short range. It is this observation which will enable us to make statements about the dynamically screened interaction.

TABLE I. Upper bound on inverse screening length for antiferromagnetism.

r_s	1	2	3	4	5	6
ξ_{FT}/k_F	0.82	1.15	1.41	1.63	1.82	2.00
ξ_c/k_F	0.06	0.20	0.34	0.46	0.57	0.67

As is well known, Fermi-Thomas screening is the zero-frequency limit of the dynamic interparticle screening. We now take into account the dynamically screened time-dependent interparticle potential. In momentum space, the latter is equal to the Coulomb potential $v(\mathbf{p})$ divided by the dynamic dielectric constant $\epsilon(\mathbf{p}\omega)$. It will be shown that there is still no spin density instability in an electron gas when the dynamic nature of the screening is taken into account.

In Appendix A the expected equation for T to order $\ln r_s$ is derived. In terms of Feynman diagrams the equation replaces each Coulomb potential "rung" of the "ladder" equation by a dynamically screened potential "rung," $v(\mathbf{p})/\epsilon_{\mathrm{RPA}}(\mathbf{p}\omega)$; the Coulomb interaction acquires the dynamic screening appropriate to the random-phase approximation (RPA). The increased screening from the imaginary part of the frequency-dependent dielectric constant and the frequency region where the real part is negative compensate for the reduction in the real part of the screening at low frequencies. A more mathematical description will follow. Because of the complexity of the problem, only the zero-temperature limit is studied.

From (A18), (A19), and (A22), we see that the equation for T corresponding to (2.12) but with dynamic screening reads

$$\langle\mathbf{p}_0\omega_0|T(\mathbf{q}\omega)|\mathbf{p}_0'\omega_0'\rangle=V_s(\mathbf{p}_0-\mathbf{p}_0',\omega_0-\omega_0')-\int_{-\infty}^{\infty}\frac{d\omega_0''}{2\pi i}\int\frac{d^3p_0''}{(2\pi)^3}V_s(\mathbf{p}_0-\mathbf{p}_0'',\omega_0-\omega_0'')$$
$$\times\frac{\langle\mathbf{p}_0''\omega_0''|T(\mathbf{q}\omega)|\mathbf{p}_0'\omega_0'\rangle}{[E(\mathbf{p}_0''+\frac{1}{2}\mathbf{q})-\mu-(\omega_0''+\frac{1}{2}\omega)][E(\mathbf{p}_0''-\frac{1}{2}\mathbf{q})-\mu-(\omega_0''-\frac{1}{2}\omega)]}\,. \tag{2.16}$$

the singularities in ω in the propagators and V_s being displaced in the usual fashion. We have introduced

$$V_s(\mathbf{p},\omega)\equiv V(\mathbf{p})/\epsilon_{\mathrm{RPA}}(\mathbf{p}\omega).$$

When we employ a Fredholm expansion

$$\begin{aligned}\langle\mathbf{p}_0\omega_0|T(\mathbf{q}\omega)|\mathbf{p}_0'\omega_0'\rangle\\ &=V_s(\mathbf{p}_0-\mathbf{p}_0',\,\omega_0-\omega_0')+\frac{N(\mathbf{p}_0\omega_0,\mathbf{q}\omega,\mathbf{p}_0'\omega_0')}{1-D(\mathbf{q}\omega)},\end{aligned}$$

the first approximation gives back the static screening result we referred to earlier; the denominator is not more than $\frac{1}{2}$ for any $\mathbf{q}$ or ω. A rough estimate of the next order gives a small correction. Thus the paramagnetic electron gas remains differentially stable with dynamic screening for all r_s for which the high-density approximations are tenable.

Our argument does not rule out the possibility that the paramagnetic state is metastable to the spin density wave state. However, we are unaware of any mathematical or physical argument for expecting the screened case to differ from the unscreened case in this respect. Stated alternatively, we have not ruled out a first-order transition to a state in which a modified screening resulting from the itinerantly antiferromagnetic spin wave sustains itself. We merely assert that this is not the case for the prototype transition proposed by Overhauser, and no one has proposed any reason for believing that it should occur and be fundamental in more accurate theories.

3. PROPERTIES OF AN ELECTRON-GAS ANTIFERROMAGNET

In the previous section we proved that an electron gas has no antiferromagnetic state. The spin-density instability in the paramagnetic state of Hartree-Fock electron gas vanishes when one uses an approximation which correctly includes screening. In spite of this, it is instructive to study the properties of an itinerant antiferromagnetic electron gas in the Hartree-Fock approximation. In this section, we briefly investigate the consequences of this invalid approximation. We exhibit the energy spectrum and discuss the specific heat and absorption coefficient. For the reader who is not interested in the properties of a nonexistent state, this section may be omitted.

In order to compute the properties of the antiferromagnet, it is nost convenient to use the Green's-function formalism. Appendix A contains most of the necessary information. We recall [Eq. (A8)] that the equation of motion for the matrix Green's function in the Hartree-Fock approximation reads

$$\begin{aligned}\mathcal{G}(11')=\{(\partial/\partial s)-\beta[(\nabla^2/2m)+\mu]\}\delta(11')\\ +\beta v(11')\mathcal{G}(11').\quad(3.1)\end{aligned}$$

$\mathcal{G}$ is a 2×2 matrix in the spin indices with components $G_{\pm\pm}(11')$.

The general solution to these equations takes the form

$$\begin{aligned}G_{\alpha\beta}(\mathbf{r}t,\mathbf{r}'t')\\ =\sum\nolimits_n\exp[-\tfrac{1}{2}\mathbf{Q}_n\cdot(\mathbf{r}+\mathbf{r}')]f_{n,\alpha\beta}(\mathbf{r}-\mathbf{r}',\,t-t').\quad(3.2)\end{aligned}$$

The paramagnetic solution is the one for which $f_{n,\alpha\beta}(\mathbf{r}t)=\delta_{n,0}\delta_{\alpha,\beta}h(\mathbf{r}t)$, where $\mathbf{Q}_0=0$. Following Overhauser,[1] we search for a more general solution in which $f_{\pm1,\alpha\beta}(\mathbf{r}t)=\sigma_{\alpha\beta}{}^{\pm}\bar{h}(\mathbf{r}t)$ are also nonvanishing and $\mathbf{Q}_{\pm1}=\pm\mathbf{Q}$. This solution represents a circularly polarized spin wave along the quantization axis.[9] To derive the needed expressions, the following definitions of the Fourier transforms are used:

$$\begin{aligned}G_{\pm\mp}(12)=\int\frac{d^3k}{(2\pi)^3}G_{\pm\mp}(\mathbf{k},\,t_1-t_2)e^{i(\mathbf{k}\mp\frac{1}{2}\mathbf{Q})\cdot\mathbf{r}_1}e^{-i(\mathbf{k}\pm\frac{1}{2}\mathbf{Q})\cdot\mathbf{r}_2}\\ =e^{\mp i\frac{1}{2}\mathbf{Q}\cdot(\mathbf{r}_1+\mathbf{r}_2)}\bar{h}(\mathbf{r}_1-\mathbf{r}_2,\,t_1-t_2)\quad(3.3)\end{aligned}$$

$$\begin{aligned}G_{\pm\pm}(12)=\int\frac{d^3k}{(2\pi)^3}G_{\pm\pm}(\mathbf{k},\,t_1-t_2)e^{i(\mathbf{k}\mp\frac{1}{2}\mathbf{Q})\cdot(\mathbf{r}_1-\mathbf{r}_2)}\\ =h(\mathbf{r}_1-\mathbf{r}_2,\,t_1-t_2).\quad(3.4)\end{aligned}$$

Equation (3.1), the Hartree-Fock equation, takes the form

$$\beta\mathcal{G}(\mathbf{k},\omega_\nu)=-\begin{pmatrix}\omega_\nu-\epsilon_+(\mathbf{k}) & -g(\mathbf{k})\\ -g(\mathbf{k}) & \omega_\nu-\epsilon_-(\mathbf{k})\end{pmatrix},\quad(3.5)$$

where

$$\epsilon_\pm(\mathbf{k})=[(\mathbf{k}\mp\tfrac{1}{2}\mathbf{Q})^2/2m]-\mu,\quad(3.6)$$

$$g(\mathbf{k})=\sum_\nu\int\frac{d^3k'}{(2\pi)^3}v(\mathbf{k}-\mathbf{k}')G_{+-}(\mathbf{k}',\omega_\nu),\quad(3.7)$$

and energies are measured from the Fermi level. To find $\mathcal{G}$, we invert (3.5),

$$\begin{aligned}\beta\mathcal{G}(\mathbf{k},\omega_\nu)=-\begin{pmatrix}\omega_\nu-\epsilon_-(\mathbf{k}) & g(\mathbf{k})\\ g(\mathbf{k}) & \omega_\nu-\epsilon_+(\mathbf{k})\end{pmatrix}\\ [(\omega_\nu-\omega_+(\mathbf{k}))(\omega_\nu-\omega_-(\mathbf{k}))]^{-1},\quad(3.8)\end{aligned}$$

$$\begin{aligned}\omega_\pm(\mathbf{k})=\tfrac{1}{2}(\epsilon_+(\mathbf{k})+\epsilon_-(\mathbf{k}))\\ \mp[\tfrac{1}{4}(\epsilon_+(\mathbf{k})-\epsilon_-(\mathbf{k}))^2+g^2(\mathbf{k})]^{1/2}.\quad(3.9)\end{aligned}$$

As in Sec. 2, we have neglected the Fock part of the single-particle energies.

The essential feature of the electron gas which gives rise to the spin density instability in the Hartree-Fock approximation is the very large weighting of low momentum states by the bare Coulomb potential. Since the actual Coulomb potential is difficult to use and since the system is unphysical anyway, we employ the Overhauser idealization. The interparticle potential v in the antiferromagnetic state is taken to be a constant $\bar{v}$ in

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[9] Overhauser's equations have been cast in the Green's-function formalism by A. K. Rajagopal, Ph.D. dissertation, Harvard University, 1964 (unpublished).

momentum space inside of a cylinder of radius R and length L. Outside of this cylinder v is taken to be zero. The direction of Q defines the Z axis. R and L are less than k_F. Thus, in calculations, we shall take v to be a delta function in configuration space and use the cutoffs R and L in momentum space.

Assuming, as Overhauser did, that $\omega_+(\mathbf{k})\leq 0$ and $\omega_-(\mathbf{k})\geq 0$ for all $\mathbf{k}$ in the region of integration, we recover the gap energy at zero temperature from (3.7) and (3.8):

$$g_0=QL[4m\sinh(4\pi^2Q/m\bar{v}R^2)]^{-1}. \tag{3.10}$$

The energy spectrum and gap equation for the electron-gap antiferromagnet are quite similar to those for a superconductor. Thus we expect the specific heat from the ordered electrons to be exponentially small at low temperatures and to exhibit a discontinuity at the transition temperature. The model indeed predicts this, although we shall not compute C_v here.

The fact that the energy spectrum has a gap is responsible for the exponentially small specific heat at low temperatures. However, this is modified by several mechanisms. First, because the Fermi surfaces are complicated and the model applies to only a portion of the surface, only part has a decreased specific heat. Second, there can be a mechanism for repopulation which can lead to an appreciable C_v at low temperatures. So while we expect that an electron-gas antiferromagnet would have a specific heat somewhat smaller than the usual electronic and lattice specific heat, it would not necessarily be an easily observable difference.

The presence of a discontinuity in the specific heat at the transition temperature can be inferred from the similarity of this system with a superconductor. Because of this and the fact that the calculation is similar to one later in this paper for a two-band model, we shall not do it here. It turns out that the jump in C_v is only a few percent of the lattice specific heat at the proposed transition temperature and the anomaly is only a few degrees wide. Thus, if it did exist, it would be difficult to find experimentally.

With essentially the model which we have described, Overhauser[10] has tried to explain the optical absorption experiments of El Naby.[11] That is, Overhauser claims that this model predicts the absorption of light at frequencies $\omega>2g$.

Hopfield[12] has pointed out that Overhauser's calculation violates conservation of momentum. The absorption coefficient has a factor of the matrix element of the momentum between the initial and final states. However, since the electron gas possesses full translational invariance, the total momentum $\mathbf{P}$ is conserved and thus commutes with the Hamiltonian H,

$$0=\langle f|[\mathbf{P},H]|i\rangle=(E_i-E_f)\langle f|\mathbf{P}|i\rangle.$$

When $E_i\neq E_f$, the matrix element must be zero. Therefore, there can be no absorption for finite $\omega=E_f-E_i$, since infrared ($\mathbf{k}=0$) absorption involves this matrix element.

More precisely, if we introduce $\chi_{\mathrm{jj}}''(\mathbf{k}\omega)$, the absorptive response function[13]

$$\chi_{\mathrm{jj}}''(\mathbf{k}\omega)=\frac{1}{2}\int d^3r\, e^{-i\mathbf{k}\cdot\mathbf{r}}\int dt\, e^{i\omega t}\langle[j(\mathbf{r}t),j(\mathbf{0},0)]\rangle,$$

and neglect the ionic motion, so that the current is given by $\mathbf{j}_e=\mathbf{j}=(e/m)\mathbf{p}$ (where $\mathbf{p}$ is the electronic momentum density) and if the electron momentum is conserved, Hopfield's argument then says $\chi_{\mathrm{jj}}''(\mathbf{k}\omega)$ vanishes as $\mathbf{k}\to 0$ except at $\omega=0$. When $\mathbf{k}$ approaches zero, all the absorption takes place at $\omega=0$ exhausting the f-sum rule.

The response when an electric field is applied to the system is described in terms of the $(\mathbf{j},\mathbf{j})$ correlation function or the two-particle Green's function $\mathcal{G}_2$,[14]

$$\mathcal{G}_2(11',22')=\langle T(\psi(1)\psi(2)\psi^\dagger(2')\psi^\dagger(1'))\rangle.$$

Overhauser's calculation sets

$$\mathcal{G}_2(11',22')=\mathcal{G}(11')\mathcal{G}(22')-\mathcal{G}(12')\mathcal{G}(21'). \tag{3.11}$$

This approximation is inconsistent with momentum conservation and does not predict a collective spin-density mode. To avoid this pitfall, one must calculate $\mathcal{G}_2$ as one calculated G_2 in Sec. 2, from a conserving approximation. This requires inclusion of the interaction between electron-hole pairs which lead to a collective mode of the system. (The mode describes a slow variation of the gap or coherent electron-hole pairs.)

Thus the additional interactions Overhauser neglects are the parallel in the condensed state of the interactions in the paramagnetic state which give rise to the spin-density wave in the first place. The antiferromagnetic mode resulting from a better calculation is analogous to the collective mode in a neutral superconductor. To obtain either, one must use a conserving approximation; (3.11), or its analog for the superconductor, violates the conservation law and is not sufficient.

We have calculated the absorption in the "ladder" approximation which is exact to order $1/r_s$ and have shown explicitly that in the corrected calculation the absorption vanishes as k approaches zero for finite ω.[15] Whether a coupling to a lattice mode or some other feature suppresses the additional terms we have calculated remains to be seen. Short of such a demonstration there appears to be no compelling reason for supposing that optical absorption in K could be explained by an antiferromagnetic (spin-density wave) state.

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[10] A. W. Overhauser, Phys. Rev. Letters **13**, 190 (1964).

[11] M. H. El Naby, Z. Physik **174**, 267 (1963).

[12] J. J. Hopfield, Phys. Rev. **139**, A419 (1965).

[13] See L. P. Kadanoff and P. C. Martin, Ann. Phys. (N. Y.) **24**, 219 (1963).

[14] The transport coefficients are given in terms of Green's functions by P. C. Martin and J. Schwinger, Phys. Rev. **115**, 1342 (1959).

[15] We refer the reader interested in details to P. A. Fedders, Ph.D. dissertation, Harvard University, 1965 (unpublished).

4. STABILITY OF A BAND MODEL

From our study of the free-electron gas we conclude that the itinerant antiferromagnetic behavior of Cr is not due to the long-range nature of the Coulomb potential as implied by Overhauser.[1] On the other hand, Lomer[5] has suggested that it is the exchange coupling of different sheets of the Fermi surface which is responsible for these properties. In this section, we shall derive a criterion for the existence of an itinerant antiferromagnetic state in a real solid, including screened interband and intraband interactions. This criterion is consistent with antiferromagnetism in Cr by the Lomer interband coupling but not with antiferromagnetism in a metal, like K, with a free-electron Fermi surface.

Formally, the precedure is much the same as in Sec. 2, except that Bloch states and real band energies are used instead of plane waves and free-particle energies. The details are more difficult, however, because of umklapp and interband processes, and because the energies of the states and the matrix elements for these processes are not known exactly. Many approximations have therefore been made. We neglect umklapp processes in the exchange interaction and employ a static local screened potential. A rough examination indicates that the matrix elements are probably not crucial and that the stability criterion depends almost entirely on the band energies. Since we believe that itinerant antiferromagnetism occurs because of interband transitions, this case is stressed in what follows.

The stability of the paramagnetic state is again investigated through the T matrix. The Hamiltonian for this system is

$$H=\sum_{\sigma}\int d^3r\,\psi_\sigma^\dagger(\mathbf{r}t)[-\nabla^2/2m+w(\mathbf{r})]\psi_\sigma(\mathbf{r}t)$$
$$+\tfrac{1}{2}\sum_{\sigma\sigma'}\int d^3r d^3r'\,\psi_{\sigma'}^\dagger(\mathbf{r}t)\psi_{\sigma'}^\dagger(\mathbf{r}'t)$$
$$\times v(\mathbf{r}-\mathbf{r}')\psi_{\sigma'}(\mathbf{r}'t)\psi_\sigma(\mathbf{r}t)\,, \quad (4.1)$$

where $w(\mathbf{r})$ is the periodic potential of the lattice and core electrons. The Bloch functions $b_n(\mathbf{k},\mathbf{r})$ used are the solutions to the Hartree-Fock equation with energies $E_n(\mathbf{k})$,

$$[-\nabla^2/2m+V(\mathbf{r})]b_n(\mathbf{k},\mathbf{r})=E_n(\mathbf{k})b_n(\mathbf{k},\mathbf{r})\,, \quad (4.2)$$

where $V(\mathbf{r})$ is the Hartree-Fock periodic potential. The functions $b_n(\mathbf{k},\mathbf{r})$ are labeled by band indices (n) and wave vectors $(\mathbf{k})$ reduced to the first Brillouin zone (B.Z.). All lower case momenta will be taken to belong to the first B.Z. unless otherwise stated. The normalization is

$$\int d^3r\, b_n(\mathbf{k},\mathbf{r})b_{n'}^*(\mathbf{k}',\mathbf{r})=\delta_{n,n'}\delta_{\mathbf{k},\mathbf{k}'}\,, \quad (4.3)$$

where the integral is over all space. $b_n(\mathbf{k},\mathbf{r})$ can also be written as

$$b_n(\mathbf{k},\mathbf{r})=u_n(\mathbf{k},\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}\,, \quad (4.4)$$

where u is the periodic part of b.

From (A17), (A19), and (A25), we see that the Bloch transform of T satisfies the equation

$$\langle\mathbf{k}_1n_1;\mathbf{k}_2n_2|T(\omega)|\mathbf{k}_3n_3;\mathbf{k}_4n_4\rangle=\langle\mathbf{k}_1n_1;\mathbf{k}_2n_2|v|\mathbf{k}_3n_3;\mathbf{k}_4n_4\rangle$$
$$-\frac{1}{\Omega}\sum_{n_0n_0'}\sum_{\mathbf{k}_0\mathbf{k}_0'}\langle\mathbf{k}_1n_1;\mathbf{k}_0n_0|v|\mathbf{k}_3n_3;\mathbf{k}_0'n_0'\rangle\frac{f_{n_0'}(\mathbf{k}_0')-f_{n_0}(\mathbf{k}_0)}{E_{n_0'}(\mathbf{k}_0')-E_{n_0}(\mathbf{k}_0)-\omega}\langle\mathbf{k}_0'n_0';\mathbf{k}_2n_2|T(\omega)|\mathbf{k}_0n_0;\mathbf{k}_4n_4\rangle\,, \quad (4.5)$$

where the Bloch transform of the potential is defined by

$$\langle\mathbf{k}_1n_1;\mathbf{k}_2n_2|v|\mathbf{k}_3n_3;\mathbf{k}_4n_4\rangle=\Omega\int d^3r_1d^3r_2b_{n_1}^*(\mathbf{k}_1,\mathbf{r}_1)b_{n_2}^*(\mathbf{k}_2,\mathbf{r}_2)v(\mathbf{r}_1-\mathbf{r}_2)b_{n_3}(\mathbf{k}_3,\mathbf{r}_2)b_{n_4}(\mathbf{k}_4,\mathbf{r}_1) \quad (4.6)$$

and f is the Fermi factor $f_n(\mathbf{k})=[\exp\beta(E_n(\mathbf{k})-\mu)+1]^{-1}$. Because of the complexity of the problem and its apparent secondary importance, dynamic screening has not been considered. However, screening is included in a static approximation by using a screened potential $v(r)$.

After some manipulations, (4.6) can be brought into the form

$$\langle\mathbf{k}_1n_1;\mathbf{k}_2n_2|v|\mathbf{k}_3n_3;\mathbf{k}_4n_4\rangle=\sum_{\mathbf{K}}v([\mathbf{k}_1-\mathbf{k}_4]_Z+\mathbf{K})(\mathbf{k}_1n_1|\mathbf{k}_4+[\mathbf{k}_1-\mathbf{k}_4]_R-\mathbf{K}n_4)$$
$$\times(\mathbf{k}_2n_2|\mathbf{k}_3+[\mathbf{k}_2-\mathbf{k}_3]_R+\mathbf{K}n_3)\delta_{[\mathbf{k}_3-\mathbf{k}_2]_Z,[\mathbf{k}_1-\mathbf{k}_4]_Z}\,, \quad (4.7)$$

where the sum over $\mathbf{K}$ is over all reciprocal lattice vectors and

$$(\mathbf{k}n|\mathbf{k}'+\mathbf{K}n')=\int d^3r\,u_n(\mathbf{k},\mathbf{r})u_{n'}^*(\mathbf{k}'+\mathbf{K},\,\mathbf{r})=\int d^3r\,b_n(\mathbf{k},\mathbf{r})b_{n'}^*(\mathbf{k}',\mathbf{r})e^{i(\mathbf{k}'+\mathbf{K}-\mathbf{k})\cdot\mathbf{r}}. \quad (4.8)$$

For a sum of wave vectors each in the first B. Z., the notation

$$\mathbf{k}_1+\mathbf{k}_2=[\mathbf{k}_1+\mathbf{k}_2]_Z+[\mathbf{k}_1+\mathbf{k}_2]_R \tag{4.9}$$

has been introduced, where $[\mathbf{k}_1+\mathbf{k}_2]_Z$ is the reduced part (belonging to the first B. Z.) and $[\mathbf{k}_1+\mathbf{k}_2]_R$ is a reciprocal lattice vector.

We can take advantage of the momentum-conserving Kronecker delta in (4.7) by relabeling $\mathbf{k}_1$, $\mathbf{k}_2$, $\mathbf{k}_3$, and $\mathbf{k}_4$ as follows:

$$\mathbf{k}_1=\mathbf{k}+\tfrac{1}{2}\mathbf{q},\quad \mathbf{k}_2=\mathbf{k}'-\tfrac{1}{2}\mathbf{q},\quad \mathbf{k}_3=\mathbf{k}-\tfrac{1}{2}\mathbf{q},\quad \mathbf{k}_4=\mathbf{k}'+\tfrac{1}{2}\mathbf{q}. \tag{4.10}$$

Umklapp processes may be neglected, since (1) the potential behaves like $4\pi e^2/|\mathbf{p}+\mathbf{K}|^2+\xi^2$, where ξ is of the order of the Fermi-Thomas momentum, and is largest for $\mathbf{K}=0$; and (2) the matrix elements in (4.8) are primarily of interest for $n=n'$ and for $|\mathbf{k}-\mathbf{k}'|$ small compared to any reciprocal lattice vector.

We may now therefore simplify our notation defining

$$(\mathbf{k}n|\mathbf{k}'n')=\int d^3r\, b_n(\mathbf{k},\mathbf{r})b_{n'}^*(\mathbf{k}',\mathbf{r})e^{i([\mathbf{k}'-\mathbf{k}]_Z)\cdot\mathbf{r}} \tag{4.11}$$

and $v(\mathbf{k}-\mathbf{k}')$ as $v([\mathbf{k}-\mathbf{k}']_Z)$. Then (4.5) becomes

$$\begin{aligned}\langle\mathbf{k}+\tfrac{1}{2}\mathbf{q}n_1;\mathbf{k}'-\tfrac{1}{2}\mathbf{q}n_2|T(\omega)|\mathbf{k}-\tfrac{1}{2}\mathbf{q}n_3;\mathbf{k}'+\tfrac{1}{2}\mathbf{q}n_4\rangle&=v(\mathbf{k}-\mathbf{k}')(\mathbf{k}+\tfrac{1}{2}\mathbf{q}n_1|\mathbf{k}'+\tfrac{1}{2}\mathbf{q}n_4)(\mathbf{k}'-\tfrac{1}{2}\mathbf{q}n_2|\mathbf{k}-\tfrac{1}{2}\mathbf{q}n_3)\\ &-\frac{1}{\Omega}\sum_{\mathbf{k}''}\sum_{n_0n_0'}v(\mathbf{k}-\mathbf{k}'')(\mathbf{k}+\tfrac{1}{2}\mathbf{q}n_1|\mathbf{k}''+\tfrac{1}{2}\mathbf{q}n_0')(\mathbf{k}''-\tfrac{1}{2}\mathbf{q}n_0|\mathbf{k}-\tfrac{1}{2}\mathbf{q}n_3)\frac{f_{n_0'}(\mathbf{k}''+\frac{1}{2}\mathbf{q})-f_{n_0}(\mathbf{k}''-\frac{1}{2}\mathbf{q})}{E_{n_0'}(\mathbf{k}''+\frac{1}{2}\mathbf{q})-E_{n_0}(\mathbf{k}''-\frac{1}{2}\mathbf{q})-\omega}\\ &\times\langle\mathbf{k}''+\tfrac{1}{2}\mathbf{q}n_0';\mathbf{k}'-\tfrac{1}{2}\mathbf{q}n_2|T(\omega)|\mathbf{k}''-\tfrac{1}{2}\mathbf{q}n_0;\mathbf{k}'+\tfrac{1}{2}\mathbf{q}n_4\rangle.\end{aligned} \tag{4.12}$$

We assume the crystal has inversion symmetry so that all of the matrix elements $(\mathbf{k}n|\mathbf{k}'n')$ are real. We also set $\omega=0$, since if T has a pole at $\omega=0$ for some temperature, it should have a pole for imaginary ω at slightly lower temperatures.

As in Sec. 2, it is necessary to examine the kernel of the equation for T which here contains the positive definite matrix

$$\Gamma_{nn'}(\mathbf{k}'',\mathbf{q})=\Gamma_{n'n}(\mathbf{k}'',-\mathbf{q})=-\frac{f_n(\mathbf{k}''+\frac{1}{2}\mathbf{q})-f_{n'}(\mathbf{k}''-\frac{1}{2}\mathbf{q})}{E_n(\mathbf{k}''+\frac{1}{2}\mathbf{q})-E_{n'}(\mathbf{k}''-\frac{1}{2}\mathbf{q})} \tag{4.13}$$

describing the phase space and energy denominator of the interaction between two bands. For simplicity, let us assume (as appears to be the case in Cr) that there is only one pair of bands, a and b, for which Γ has a small energy denominator over a non-negligible portion of phase space. Then, in (4.12), we may keep only the two terms with $n_0'=a$, $n_0=b$ and $n_0'=b$, $n_0=a$ in the sum.

Since the matrix elements between the different bands tend to vanish, the only large components of the kernel are the terms with $n_1=n_0'$ in one band and $n_2=n_0$ in the other. This eliminates the band sum making the problem identical with that of Sec. 2. The instability will occur at that $\mathbf{q}_0$ at which

$$R=1/\Omega\textstyle\sum_{\mathbf{k}''}v(0)\gamma^2\Gamma_{ab}(\mathbf{k}'',\mathbf{q}_0) \tag{4.14}$$

first becomes larger than one. In this expression, we have inserted the quantity γ, the average of $(\mathbf{k}+\frac{1}{2}\mathbf{q}n|\mathbf{k}''+\frac{1}{2}\mathbf{q}n)$ over the region where the kernel is unusually large. γ is less than one but is probably not small. Because of the many approximations, the instability may occur when R is somewhat larger than one.

We see here, as argued by Lomer, that the presence of two nearby bands, whose energies are close to each other over an appreciable part of the sum in (4.14), can lead to an instability in T. In Fig. 1, two hypothetical bands are drawn in the (100) plane of reciprocal space with the occupied part of each shaded. These bands represent Cr. The occupied region of band a is seen to be similar in shape to the unoccupied region of band b. If band b is shifted by $\mathbf{q}=\mathbf{Q}$, the energy denominator in (4.13) is small over an appreciable region, namely, the area of contact indicated by cross section in Fig. 2. δ is an average distance between the two surfaces. From the illustrations and from estimates of the band structure of Cr (see Lomer), the region where the energy denominator is small has dimensions which are small compared to a reciprocal lattice vector.

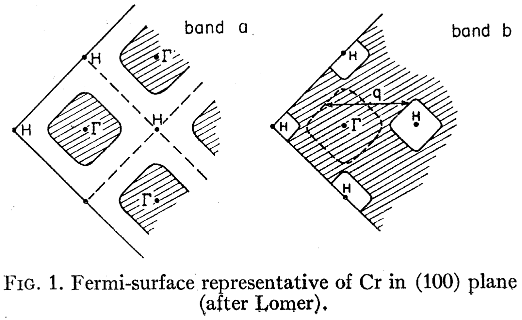

FIG. 1. Fermi-surface representative of Cr in (100) plane (after Lomer).

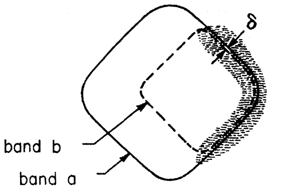

FIG. 2. Fermi surfaces displaced by q.

Since only a small volume near the Fermi surface will contribute significantly to the sum (4.14), the criterion can be expressed in terms of the Fermi velocities. (4.15) is approximately

$$R \approx \frac{e^2\gamma^2\pi A}{2\xi_{\mathrm{FT}}{}^2 a^3 E_F(v_a+v_b)} \ln(\pi/2\delta), \qquad (4.15)$$

where v_a and v_b are the Fermi velocities of the two bands, δ is an average separation of the two surfaces as shown in Fig. 2, A is the area of close contact of the two surfaces (the area shaded in Fig. 2), and a is half the cube edge in lattice space. $v(0)$ has been set equal to $4\pi e^2/\xi^2{}_{\mathrm{FT}}$.

In the above analysis, we have assumed that only one pair of bands will lead to a large R. While there is nothing to guarantee this in general, it is true for Cr. If it were not true, the analysis would still be the same although the matrix inversion would be more complicated. The formula for the one-band itinerant antiferromagnet with constant matrix elements is obtained by setting $a=b$. Cr satisfies the criterion for antiferromagnetism by interband transitions. While not impossible, it is highly unlikely that this type of antiferromagnetism can arise from transitions within a single band. The Fermi surface of the band would have to be almost cubical to gain the required phase space for low-energy transitions.

5. PROPERTIES OF AN ITINERANT ANTIFERROMAGNET

Having demonstrated that Cr is most probably unstable to the formation of spin-density waves, we now wish to investigate some of the thermodynamic and electromagnetic properties of the ensuing itinerant antiferromagnetic (spin-density wave) state. From our preceding analysis, a model which has particular reference to Cr is chosen. However, the model is not sufficiently detailed to apply specifically to a certain metal. The results, therefore, apply equally roughly to any metal which has the type of instability (due to interband transitions) discussed.

For mathematical simplicity, we assume that the energies of the two relevant bands (those that are responsible for the instability) as measured from the Fermi surface are given by

$$\begin{aligned} \epsilon_a(\mathbf{k}) &= E_a(\mathbf{k}) - E_F = v_a(k-k_c), \\ \epsilon_b(\mathbf{k}) &= E_b(\mathbf{k}+\mathbf{Q}) - E_F = -v_b(k-k_c). \end{aligned} \qquad (5.1)$$

v_a and v_b are the Fermi velocities of the two bands and k_c is some wave vector in the first B.Z.—a sort of average Fermi momentum. $\mathbf{Q}$ is any one of the wave vectors which gives the greatest instability.

Of course, we know that this is not a very good approximation, but it is necessary in order to calculate some properties of the system. The results should be qualitatively and semiquantitatively correct, although behavior depending on the details of the energy bands will be lost. In a real transition metal, the surfaces of constant energy are not spheres and the Fermi surfaces of the two bands are not congruent. In this sense, our approximation will tend to overestimate the area of close contact of the Fermi surfaces. The approximation that the energies are linear in k is not serious, since only the energy structure near the Fermi surfaces is important.

Thus our crude model is given by (5.1) with two other simplifications. First, we approximate the first B.Z. by a sphere of radius K, since the properties of the state should be relatively insensitive to the shape of the zone. Second, as earlier, matrix elements of the form $(\mathbf{k}a|\mathbf{k}'a)$ will be replaced by γ. In a real metal, this was justified by the fact that the volume of close contact of the Fermi surfaces had dimensions which were small compared to any reciprocal lattice vector. This is now distorted because we have taken the Fermi surfaces to be congruent spheres. The idea of approximating the matrix elements in this manner is still valid, however. Finally, we note that in all integrations, the important region will be near k_c. We could have put some cutoff or damping factor into the matrix elements, but this would complicate the model without appreciably altering the results.

We recall from (4.14) that the condition for the instability of the paramagnetic system was that R be greater than one. With our model, R is given by

$$R = -\frac{v(0)\gamma^2}{2\pi^2}\int_0^K k^2 dk \, \frac{f(v_a(k-k_c)) - f(-v_b(k-k_c))}{(v_a+v_b)(k-k_c)}. \qquad (5.2)$$

The critical temperature or transition temperature, denoted by T_N or $\beta_N = 1/k_B T_N$, is determined by setting $R=1$. For temperatures above T_N, the system is paramagnetic. Below T_N, the paramagnetic state is unstable to the formation of an antiferromagnetic state which we shall investigate shortly. Performing the integration in (5.2), which is similar to the corresponding integral for a superconductor, yields, for $R=1$,

$$1 = \lambda[\ln(2\beta_N \bar{v}\bar{k}_c\gamma_e/\pi) + (K^2+2Kk_c-6k_c^2)/2k_c^2 + O(1/\beta_N E_F)],$$

where we have used the experimental fact that $\beta_N E_F \gg 1$. The average velocities v and $\bar{v}$, a dimensionless measure of the potential λ, and a momentum $\bar{k}_c$ have been defined as follows:

$$\bar{v} = (v_a v_b)^{1/2}, \qquad v = \tfrac{1}{2}(v_a+v_b),$$

$$\lambda = \gamma^2 v(0) k_c^2/2\pi^2 v, \quad \ln\gamma_e = 0.577\cdots \quad \text{(Euler's constant)}, \qquad (5.3)$$

$\bar{k}_c=[k_c(K-k_c)]^{1/2}$.

Solving for T_N yields

$$k_BT_N=(2\gamma_e\bar{v}\bar{k}_cf/\pi)\exp(-1/\lambda),$$
$$\ln f=(K^2+2Kk_c-6k_c^2)/4k_c^2. \tag{5.4}$$

Of course, it is senseless to predict T_N from our crude model because of the exponential dependence on some of the quantities involved. However, (5.4) is useful in expressing other quantities in terms of T_N and using the experimental value of $T_N=(312\pm1)$°K for Cr.

To deduce any properties of the system below T_N, we must find the correlation functions for a stable itinerant antiferromagnetic state. As we shall see, there are many similarities between these functions in the antiferromagnetic state and superconducting states.[16]

From Appendix A we recall the inverted equation of motion for the one-particle Green's function $\mathcal{G}$ in the Hartree-Fock approximation,

$$\mathcal{G}^{-1}(11')=\{\partial/\partial s-\beta[\nabla^2/2m+\mu-V_H(1)]\}\delta(11')$$
$$+\beta v(11')\mathcal{G}(11'), \tag{5.5}$$

where $\mathcal{G}$ is a 2×2 matrix in spin space, V_H is the periodic potential in the Hartree approximation, and v is a statically screened Coulomb potential. We seek a self-consistent solution to (5.5) with a linearly polarized spin wave in the y direction, mixing bands a and b. The relevant part of $\mathcal{G}$ is then given by

$$\begin{aligned}G_{\alpha\beta}(\mathbf{rr}',\omega_\nu)=\textstyle\sum_{\mathbf{k}}\{&[f_a(\mathbf{k}\omega_\nu)b_a(\mathbf{k},\mathbf{r})b_a^*(\mathbf{k},\mathbf{r}')\\&+f_b(\mathbf{k}\omega_\nu)b_b(\mathbf{k}+\mathbf{Q},\,\mathbf{r})b_b^*(\mathbf{k}+\mathbf{Q},\,\mathbf{r}')]\delta_{\alpha\beta}\\&+\bar{f}(\mathbf{k},\omega_\nu)[b_a(\mathbf{k},\mathbf{r})b_b^*(\mathbf{k}+\mathbf{Q},\,\mathbf{r}')\\&\qquad+b_b(\mathbf{k}+\mathbf{Q},\,\mathbf{r})b_a^*(\mathbf{k},\mathbf{r}')]\sigma_{\alpha\beta}^{(1)}\}.\end{aligned} \tag{5.6}$$

We may therefore introduce

$$\mathcal{G}(\mathbf{k},\omega_\nu)=\int d^3rd^3r'\begin{pmatrix}b_a^*(\mathbf{k},\mathbf{r}) & 0\\ 0 & b_b^*(\mathbf{k}+\mathbf{Q},\,\mathbf{r})\end{pmatrix}$$
$$\times\mathcal{G}(\mathbf{rr}',\omega_\nu)\begin{pmatrix}b_a(\mathbf{k},\mathbf{r}') & 0\\ 0 & b_b(\mathbf{k}+\mathbf{Q},\,\mathbf{r}')\end{pmatrix}. \tag{5.7}$$

This, of course, is not the most general kind of pairing possible. To be thorough, one should consider the possibilities of pairing between all momenta, spin directions, and all bands. In other words, the state described by (5.6) will have lower free energy than the paramagnetic state and eliminate the instability found earlier, but it could conceivably be unstable to other deformations.

Since the expectation values of operators in a thermal ensemble of these states obey the usual fluctuation-dissipation theorem, we can impose the usual thermal boundary conditions on the Green's functions. Then, Bloch transforming (5.5) in the manner stated in (5.7), we find

$$\mathcal{G}^{-1}(\mathbf{k},\omega_\nu)=-\beta\begin{pmatrix}\omega_\nu-\epsilon_a(\mathbf{k}) & g(\mathbf{k})\\ g(\mathbf{k}) & \omega_\nu-\epsilon_b(\mathbf{k})\end{pmatrix}, \tag{5.8}$$

$$g(\mathbf{k})=\sum_\nu\int\frac{d^3k'}{(2\pi)^3}v(\mathbf{k}-\mathbf{k}')G_{+-}(\mathbf{k}'\omega_\nu)$$
$$\times(\mathbf{k}a|\mathbf{k}'a)(\mathbf{k}b|\mathbf{k}'b). \tag{5.9}$$

$$\beta\mathcal{G}(\mathbf{k},\omega_\nu)=-\begin{pmatrix}\omega_\nu-\epsilon_b(\mathbf{k}) & g(\mathbf{k})\\ g(\mathbf{k}) & \omega_\nu-\epsilon_a(\mathbf{k})\end{pmatrix}$$
$$\times[(\omega_\nu-\omega_1(\mathbf{k}))(\omega_\nu-\omega_2(\mathbf{k}))]^{-1}, \tag{5.10}$$

where

$$\omega_{1,2}(\mathbf{k})=\tfrac{1}{2}(\epsilon_a(\mathbf{k})+\epsilon_b(\mathbf{k}))$$
$$\mp[\tfrac{1}{4}(\epsilon_a(\mathbf{k})-\epsilon_\beta(\mathbf{k}))^2+g^2(\mathbf{k})]^{1/2}, \tag{5.11}$$

in which the minus sign goes with ω_1 and the plus with ω_2.

The value of the gap g is now computed at zero temperature and near the transition temperature. In accordance with our model, the matrix elements in (5.9) are approximated by γ and the potential is taken to be slowly varying. From (5.8) through (5.11) we see that the gap equation is

$$1=\int\frac{d^3k}{(2\pi)^3}\gamma^2v(0)\tfrac{1}{2}[f(\omega_1(\mathbf{k}))-f(\omega_2(\mathbf{k}))]$$
$$\times[v^2(k-k_c)^2+g^2]^{-1/2},$$

where the summation has been performed.

At zero temperature we note that the Fermi factors are step functions and $\omega_1<0$, $\omega_2>0$ for all k. The integral is easily done if we neglect terms of order g/E_F compared to one. Using (5.4) for T_N, we find

$$g_0=g(T=0)=2v\bar{k}_cfe^{-1/\lambda}=\pi vk_BT_N/\gamma_e\bar{v}. \tag{5.12}$$

For Cr, this predicts that g_0 is about 2.6×10^{-2} eV.

To determine the behavior of the gap for temperatures near the transition temperature, it is more convenient to start from (5.9) and (5.10) without performing the sum. Since g vanishes at T_N, we can carry out an expansion of the denominator of (5.10) in powers of g obtaining

$$1=-\frac{1}{\beta}\sum_\nu\int\frac{d^3k}{(2\pi)^3}v(0)\gamma^2\left\{\frac{1}{(\omega_\nu-\epsilon_a(\mathbf{k}))(\omega_\nu-\epsilon_b(\mathbf{k}))}\right.$$
$$\left.+\frac{g^2}{[(\omega_\nu-\epsilon_a(\mathbf{k}))(\omega_\nu-\epsilon_b(\mathbf{k}-)]^2}+\cdots\right\}.$$

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[16] Many of the equations in this section are similar mathematically to those in the theory of superconductivity. The calculation of the gap and specific heat are similar to those done in A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzaloshinski, *Methods of Quantum Field Theory in Statistical Mechanics*, translated by R. A. Silverman (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963), Chap. 7. The collective mode equations closely resemble those in L. P. Kadanoff and P. C. Martin, Phys. Rev. 124, 670 (1961).

The first term is similar to the integral done in obtaining T_N. For the second term, it is easiest to integrate before summing and to neglect terms of order k_BT_N/E_F with respect to one. We find, on combining results,

$$g=\pi k_BT_N(8v^2/7\bar{v}^2\zeta(3))^{1/2}[1-(T/T_N)]^{1/2},\quad T_N-T\ll T<T_N, \tag{5.13}$$

$\zeta(3)$ is the Riemann zeta function of argument three.

The gap varies with temperature as in a superconductor. Neutron-diffraction experiments[17] seem to verify that this is the general shape of the gap although they give a small tail which goes past T_N. This tail is only 3% of the total intensity for single crystals of Cr but becomes significantly larger for more finely granulated samples. It is not present in this theory. The $(1-T/T_N)^{1/2}$ behavior of the gap is characteristic of an effective field approximation. The exponent is probably not $\frac{1}{2}$ in the exact theory.

Any of the thermodynamic derivatives characterizing the antiferromagnetic state can be found from the thermodynamic potential $\Omega=\langle H-\mu N\rangle$, which can be expressed in terms of the interaction Hamiltonian by

$$\Omega=\Omega_0+\int_0^\lambda d\lambda/\lambda\langle H_{\text{int}}\rangle. \tag{5.14}$$

λ is the coupling strength and Ω_0 is the thermodynamic potential of the noninteracting system. We shall compute only that part of Ω due to bands a and b. In terms of the Green's functions, $\langle H_{\text{int}}\rangle$ is easily obtained from the spectral function A.

$$\langle H_{\text{int}}\rangle_{ab}=\int\frac{d^3k}{(2\pi)^3}\int\frac{d\omega}{2\pi}\{(\omega+\epsilon_a(\mathbf{k}))A_{++}(\mathbf{k}\omega)+(\omega+\epsilon_b(\mathbf{k}))A_{--}(\mathbf{k}\omega)\}f(\omega), \tag{5.15}$$

$$A_{\pm\pm}(\mathbf{k}\omega)=\beta[G_{\pm\pm}(\mathbf{k},\omega+i\epsilon)-G_{\pm\pm}(\mathbf{k},\omega-i\epsilon)].$$

Since our model is far too crude to calculate accurate values of Ω for the paramagnetic state, we only compute the contribution to Ω due to the difference of the antiferromagnetic state Ω_{AF} and the paramagnetic state Ω_{P}. The contribution to H_{int} is

$$\langle H_{\text{int}}\rangle_{\text{AF}}-\langle H_{\text{int}}\rangle_{\text{P}}=-k_c^2g^2/\pi^2v\lambda. \tag{5.16}$$

Changing the variable of integration from λ to g in (5.14) gives us the addition to the free energy of the system

$$F_{\text{AF}}-F_P=-\frac{k_c^2\bar{v}^2}{2\pi^2v^2\pi}\left(\frac{1}{k_BT}\right)^2\frac{7}{8}\zeta(3)g^4. \tag{5.17}$$

Substituting the value for g from (5.13) and taking the appropriate derivative yields the specific heat of the system at T_N:

$$(C_V)_{\text{AF}}-(C_V)_{\text{P}}=8vk_c^2k_B^2T_N/7\zeta(3)\bar{v}^2. \tag{5.18}$$

This is the addition to the specific heat per unit volume from the antiferromagnetic state to first order. To find C_V just below T_N one needs the term to order g^4 in the expression for T_N. The value of the discontinuity in the specific heat at T_N is compatible with the value given by experiment.[18,19] The discontinuity is only a few percent of the total. We have not computed the contribution to C_V at lower temperatures. It would be very difficult to compare to experiment, in any case, because of the contributions from the other bands.

As one would expect, the antiferromagnetic state has a collective mode in the form of a dynamic spin-density wave which has a soundlike dispersion relation. The mode manifests itself in the form of a pole in the spin-density correlation function and the fractional susceptibility along the z direction.

From (A15), we take the equation for the fractional susceptibility as a matrix in the spin indices 1 and 1′:

$$\mathcal{L}(11';2)=\mathcal{L}^0(11';2)-\int d4d4'v(44')\mathcal{G}(14)\mathcal{L}(44';2)\mathcal{G}(4'1'), \tag{5.19}$$

where

$$L_{\alpha\alpha'}{}^0(11';2)=-\sum_{\beta\beta'}G_{\alpha\beta'}(12)\tau_{\beta\beta'}{}^{(z)}G_{\beta\alpha'}(21').$$

The spin-density correlation function is given by

$$\sum_{\alpha\alpha'}\sigma_{\alpha\alpha'}{}^{(z)}L_{\alpha\alpha'}(11;2)=\langle T[(n_+(1)-n_-(1))(n_+(2)-n_-(2))]\rangle. \tag{5.20}$$

After Bloch transforming, taking the potential to be short ranged, restricting to two bands, and using our approximation on the matrix elements, we obtain, for the transform of $L(1;2)$,

$$\begin{aligned}\mathcal{L}(\mathbf{k},\omega_\nu)=-\sum_{\nu0}\int\frac{d^3k_0}{(2\pi)^3}\mathcal{G}(\mathbf{k}_0+\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}+\tfrac{1}{2}\omega_\nu)\sigma^{(z)}\\ \times\mathcal{G}(\mathbf{k}_0-\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}-\tfrac{1}{2}\omega_\nu)-\beta\sum_{\nu0}\int\frac{d^3k_0}{(2\pi)^3}v(0)\gamma^2\\ \times\mathcal{G}(\mathbf{k}_0+\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}+\tfrac{1}{2}\omega_\nu)\mathcal{L}(\mathbf{k},\omega_{\nu0})\\ \times\mathcal{G}(\mathbf{k}_0-\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}-\tfrac{1}{2}\omega_\nu).\end{aligned} \tag{5.21}$$

Equation (5.21) is a 2×2 algebraic matrix equation. To solve for $\mathcal{L}$, we need the convolutions

$$\begin{aligned}B_{ij}=\lim_{\omega_\nu\to\omega+i\epsilon}\beta\sum_{\nu0}\int\frac{d^3k_0}{(2\pi)^3}v(0)\gamma^2G_{\{i\}}(\mathbf{k}_0+\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}+\tfrac{1}{2}\omega_\nu)\\ \times G_{\{j\}}(\mathbf{k}_0-\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}-\tfrac{1}{2}\omega_\nu),\\ \{0\}=+-\text{ or }-+,\quad\{+\}=++,\quad\{-\}=--.\end{aligned} \tag{5.22}$$

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[17] G. E. Bacon, Acta Cryst. 14, 823 (1961).

[18] R. H. Beaumont, H. Chihara, and J. A. Morrison, Phil. Mag. 5, 188 (1960).

[19] We refer the reader interested in the details of this and other calculations in this section to the doctoral dissertation submitted by P. A. Fedders to the Physics Department of Harvard University, 1965 (unpublished).

These convolutions have been evaluated in the low-frequency, low-wave-number limit to second order in ω/g and vq/g. At finite temperatures, the B's are very complicated. Thus, to look for the collective modes, we first examine the zero-temperature limit. In this limit, the B's are relatively simple. Their values are

$$B_{\pm\pm}-B_{00}=-2B_{00}=\lambda\,,$$
$$B_{+0,0+}=-B_{0-,-0}=(\lambda/4g)[\pm\omega+(4g^2/vk_c)\ln(2v\bar{k}_c/g)]$$

(where the + sign goes with the left-hand pair of subscripts and the − sign with the right),

$$\begin{aligned}1+B_{\pm\mp}-B_{00}=(\lambda/4g^2)[\omega^2-c^2k^2\\ \pm\omega(2g^2/vk_c)\ln(2v\bar{k}_c/g)]\,,\qquad(5.23)\\ c^2=\tfrac{1}{3}v_av_b=\tfrac{1}{3}\bar{v}^2.\end{aligned}$$

Because of the equality of many of the B's in this limit, $L_{++}=L_{--}$. L_{++} is given by

$$\gamma^2v(0)L_{++}[1+\gamma^2v(0)L_{++}]^{-1}=\lambda c^2k^2[\omega^2-c^2k^2]^{-1}\,,\qquad(5.24)$$

where c is the velocity of the collective mode. Thus, at zero temperature, the two-band itinerant antiferromagnet possesses a collective mode with a sound-like dispersion law, if ω and ck are less than the gap energy g. The collective mode dominates the response, and it may be observable in neutron-diffraction experiments in spite of the small total moment. This result is hardly surprising. At vanishing temperatures and long wavelengths, the spin waves in an itinerant antiferromagnet are like those of an insulating antiferromagnet just as in the parallel ferromagnetic examples.

Equation (5.24) is valid only for $T\ll T_N$; at temperatures above absolute zero, the coefficients B become much more complicated and have an imaginary part which is of order $e^{-\beta g}$ compared to the real terms. These terms represent the Landau damping of the thermally excited quasiparticles. For temperatures near T_N, the collective mode is essentially damped out. The damping will also be increased by the low-lying excitations of a more realistic model. A similar phenomenon occurs in the itinerant antiferromagnet.

Since there is a gap energy g keeping the spins antiferromagnetic, the magnetic susceptibility should be depressed below the transition temperature. As the temperature approaches the transition temperature, we expect the susceptibility to rise quite sharply since the gap is diminishing rapidly. Above T_N, we expect the susceptibility to be relatively constant. Again, more complicated surfaces will make the effect less dramatic.

We now investigate these questions quantitatively, recognizing that the static susceptibility is given by $\lim_{k\to0}\lim_{\omega\to0}\chi(k\omega)$. In order to do the required integrals, we shall assume that $v_a=v_b=v$. This should not affect the results greatly, since nothing in the theory depends on v_a-v_b. From (A12), we see that

$$\chi_{zz}=2\mu^2\int\frac{d^3k}{(2\pi)^3}\left\{\frac{g^2}{E^3}\tanh\tfrac{1}{2}\beta E-\frac{g^2}{E^2}\frac{\partial}{\partial E}(\tanh\tfrac{1}{2}\beta E)\right\}$$
$$+2\mu^2\int\frac{d^3k}{(2\pi)^3}\frac{\partial}{\partial E}(\tanh\tfrac{1}{2}\beta E)\,,\qquad(5.25)$$

where

$$E^2=\epsilon^2+g^2\,,$$
$$\epsilon=v(k-k_c)\,.\qquad(5.26)$$

We shall compute χ_{zz} only in the interesting region near T_N. In this region, the first part of (5.25) is small. The second integral is most easily done by computing $\chi(T_N)$ and $\chi(T)-\chi(T_N)$ to obtain $\chi(T)$. At T_N the gap vanishes, and χ is given by

$$\chi_{zz}(T_N)=\chi_{xx}(T_N)=2\mu^2k_c^2/\pi^2v\,.\qquad(5.27)$$

For $T_N-T\ll T<T_N$,

$$\chi_{zz}(T)=\chi_{xx}(T)=\chi_{zz}(T_N)\{1-2[1-(T/T_N)]\}\,.\qquad(5.28)$$

Also, χ_{yy} is given by

$$\chi_{yy}=2\mu^2\int\frac{d^3k}{(2\pi)^3}\frac{\partial}{\partial E}(\tanh\tfrac{1}{2}\beta E)\,.\qquad(5.29)$$

Near the transition temperature $\chi_{yy}=\chi_{zz}$, while at near-zero temperature χ_{yy} vanishes as $e^{-\beta g}$.

Above T_N our model predicts a constant susceptibility to the order we are calculating. It is useless to calculate to the next order, since presumably the chemical potential changes to order T^2. The rest of the bands should contribute only a slowly varying amount to the total susceptibility and thus a kink should be observed in χ. Lingelback[20] has made measurements on pure Cr which agree with our analysis. However, our results are not compatible with measurements contradicting those of Lingelback made by Collings, Hedgcock, and Seddiqi.[21] Because of the contributions from the other bands, further quantitative checks cannot be made.

We finally calculate the absorption coefficient for the two-band model at long wavelengths. For the longitudinal conductivity, we find an absorption edge at $\omega=2g$. We only compute that part of the absorption due to band a and b and neglect umklapp processes. Since there is no momentum conservation and the collective mode is insignificant, we find that the long-wavelength complex conductivity is

$$\frac{1}{k^2}\lim_{\omega_\nu\to\omega+i\epsilon}8\pi e^2\beta\sum_{\nu0}\int\frac{d^3k_0}{(2\pi)^3}$$
$$\times\mathrm{tr}\{\mathcal{G}(\mathbf{k}_0+\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}+\tfrac{1}{2}\omega_\nu)\mathcal{G}(\mathbf{k}_0-\tfrac{1}{2}\mathbf{k},\,\omega_{\nu0}-\tfrac{1}{2}\omega\,)\}\,.\qquad(5.30)$$

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[20] R. Lingelback, Z. Phys. Chem. **14**, 1 (1958).

[21] E. W. Collings, F. T. Hedgcock, and A. Seddiqi, Phil. Mag. **6**, 1955 (1961).

This has an imaginary part, which, when $v_a = v_b$, reduces to

$$\frac{8\pi e^2 g^2 k_c^2 v}{3\pi\omega^3}\left[\frac{\omega^2}{4} - g^2\right]^{-1/2} \tanh\frac{\beta\omega}{4} \tag{5.31}$$

for $\omega > 2g$ and vanishes for $0 < \omega < 2g$.

ACKNOWLEDGMENT

The authors wish to thank Professor Henry Ehrenreich for many helpful discussions.

APPENDIX A

In deriving equations for the correlation functions, we use the Hamiltonian (4.1). The external field w can be set equal to $-nv_0$ to obtain results for the free-electron gas. In addition, an external nonlocal magnetic field $\mathcal{H}$ is introduced in order to generate the desired correlation functions. After obtaining these, $\mathcal{H}$ is set equal to zero. The magnetic field is introduced in a formal way by defining the single-particle Green's function as

$$G_{\alpha\beta}(12) = \langle T(\psi_\alpha(1)\psi_\beta^\dagger(2))\rangle = \frac{\operatorname{tr}(T[\rho S \psi_\alpha(1)\psi_\beta^\dagger(2)])}{\operatorname{tr}(T[\rho S])}, \tag{A1}$$

$$S = \exp\left[\beta\int_0^1 ds \int_0^1 ds' \int d^3r d^3r' \times \sum_{\alpha\alpha'} \psi_\alpha^\dagger(\mathbf{r}s)\mathcal{H}_{\alpha\alpha'}(\mathbf{r}s,\mathbf{r}'s')\psi_{\alpha'}(\mathbf{r}'s')\right], \tag{A2}$$

where $\rho = \exp[-\beta(H-\mu N)]$ is the density matrix and $s = it/\beta$. G is Fourier transformed in time according to

$$G(\mathbf{r}\mathbf{r}',\omega_\nu) = \int_0^1 ds \int_0^1 ds' e^{\beta\omega_\nu(s-s')} G(\mathbf{r}s,\mathbf{r}'s'), \tag{A3}$$

where $\omega_\nu = \pi\nu/-i\beta$ and ν is an odd integer.

In order to obtain self-consistent approximations[6] to a given order in r_s, one can start from the potential Φ which is related to the free energy by $F = -\beta\Phi$. To the order to which we shall work, Φ is given diagrammatically in Figs. 3 and 4, where dashed lines stand for $-\frac{1}{2}\beta v$, solid lines for G, and there is a factor of $(-1)^N$ where N is the number of closed fermion loops. Figure 3 contains the Hartree-Fock terms which contribute to F to order $1/r_s$. The infinite set of "ring" diagrams, those diagrams which represent the most divergent terms to each order in v, contribute to order $\ln r_s$. Figure 4 is an example of one of the infinite set.

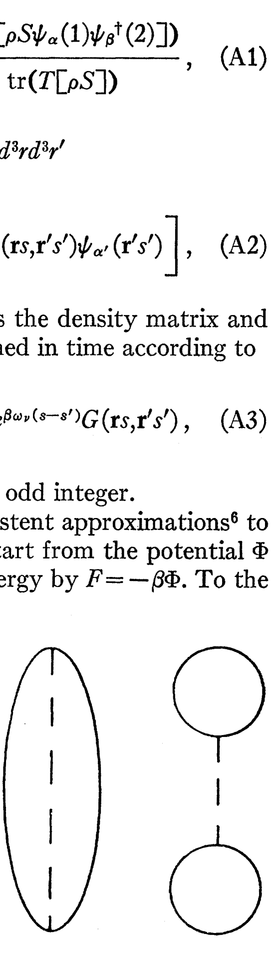

FIG. 3. Feynman diagrams to order r_s for Φ.

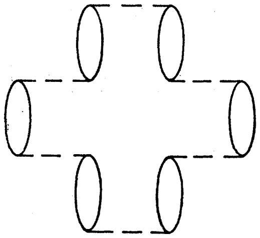

FIG. 4. Feynman diagrams to order $\ln r_s$ for Φ.

It is convenient to write the Green's functions as 2×2 matrices in spin space, denoted by $\mathcal{G}(11')$ where the arguments 1 and 1′ of matrix quantities denoted by script letters refer to their space-time-spin coordinates. In relations involving matrix quantities, the notation

$$\int d1 = \int d^3r_1 \int ds_1 \sum_{\sigma_1}, \tag{A4}$$

$$v(11') = v(\mathbf{r}_1 - \mathbf{r}_{1'})\delta(s_1 - s_{1'}), \tag{A5}$$

$$\delta(11') = \delta(\mathbf{r}_1 - \mathbf{r}_{1'})\delta(s_1 - s_{1'})\delta_{\sigma_1\sigma_{1'}} \tag{A6}$$

is used.

The equation of motion for $\mathcal{G}$ reads

$$\mathcal{G}^{-1}(11') = [\partial/\partial s - \beta(\nabla^2/2m + \mu - w(1))]\delta(11') + \beta\mathcal{V}(11') - \beta\mathcal{H}(11'), \tag{A7}$$

$$\beta\mathcal{V}(11') = \delta\Phi/\delta\mathcal{G}(1'1), \tag{A8}$$

where Φ is taken to the desired order. The correlation functions are formed by taking functional derivatives

$$\mathcal{L}(11',22') = -\delta\mathcal{G}(11')/\delta\beta\mathcal{H}(2'2). \tag{A9}$$

Since $\mathcal{G}$ is a multiple of the unit matrix in the paramagnetic state when $\mathcal{H} = 0$, we have

$$L(11',22') = \tfrac{1}{2}[\sum(\mathcal{L}(11',22')\sigma_{11'}^{(z)}\sigma_{22'}^{(z)})]_{\mathcal{H}=0}, \tag{A10}$$

where the sum is over all spins.

From (A7) and (A9), we find the equation for $\mathcal{L}$,

$$\mathcal{L}(11',22') = -\mathcal{G}(12')\mathcal{G}(21') - \int d3d3'd4d4' \times \mathcal{G}(13')\mathcal{G}(31')\mathcal{J}(33',44')\mathcal{L}(44',22'), \tag{A11}$$

where

$$\beta\mathcal{J}(33',44') = \frac{\delta}{\delta\mathcal{G}(44')}\frac{\delta}{\delta\mathcal{G}(3'3)}\Phi. \tag{A12}$$

In the Hartree-Fock approximation

$$\beta\mathcal{V}(11')=-\beta\delta(11')\int d2v(12)\mathcal{G}(22)+\beta v(11')\mathcal{G}(11'),\quad\text{(A13)}$$

$$\mathcal{J}(33',44')=-\beta\delta(33')\delta(44')v(34)+\beta\delta(34')\delta(3'4)v(33).\quad\text{(A14)}$$

When we set $\mathcal{H}=0$, $\mathrm{tr}(\mathcal{G}(22')\sigma^{(z)})$, and $\mathrm{tr}_4\mathcal{L}(44,22')$, both vanish. Thus the first term in (A14) will not contribute to L. Multiplying (A11) by $\frac{1}{2}\sigma_{11'}{}^{(z)}\sigma_{22'}{}^{(z)}$ and summing yields

$$L(11',22')=L_0(11',22')+\int d3d3'd4d4'\times L_0(11',33')J(33',44')L(44',22'),\quad\text{(A15)}$$

where

$$L_0(11',22')=-G(12')G(21').\quad\text{(A16)}$$

In the Hartree-Fock approximation, J is given by

$$J(33',44')=\beta\delta(34')\delta(3'4)v(33'),\quad\text{(A17)}$$

and, in the approximation to order $\ln r_s$ by

$$J(33',44')=\beta\delta(34')\delta(3'4)V_s(33'),\quad\text{(A18)}$$

where V_s is the dynamically screened potential equal to v/ϵ, where ϵ is the RPA dielectric constant.

The T matrix can be defined by the equation

$$T(11',22')=J(11',22')+\int d3d3'd4d4'\times J(11',33')L_0(33',44')T(44',22'),\quad\text{(A19)}$$

and is related to L through the equation

$$\int d2d2'T(11',22')L_0(22',44')=\int d2d2'J(11',22')L(22',44').\quad\text{(A20)}$$

For the free-electron gas, we set $w(1)=-nv_0$ as in Sec. 2. Since this system is spatially invariant,

$$G(\mathbf{r}\mathbf{r}',\omega_\nu)=\int\frac{d^3k}{(2\pi)^3}e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}G(\mathbf{k},\omega_\nu).\quad\text{(A21)}$$

The functions of four space-time points are defined as

$$\langle\mathbf{p}_0\omega_0|A(\mathbf{p}\omega)|\mathbf{p}_0'\omega_0'\rangle=\int d^3r_0d^3r_0'd^3rds_0ds_0'dsA(11',22')\times e^{-i\mathbf{p}_0\cdot\mathbf{r}_0}e^{\beta\omega_0s_0}e^{-i\mathbf{p}\cdot\mathbf{r}}e^{\beta\omega s}e^{-i\mathbf{p}_0'\cdot\mathbf{r}_0'}e^{\beta\omega_0's_0'},\quad\text{(A22)}$$

where

$$\begin{aligned}\mathbf{r}&=\tfrac{1}{2}(\mathbf{r}_1+\mathbf{r}_{1'}-\mathbf{r}_2-\mathbf{r}_{2'}), & s&=\tfrac{1}{2}(s_1+s_{1'}-s_2-s_{2'}),\\ \mathbf{r}_0&=\mathbf{r}_1-\mathbf{r}_{1'}, & s_0&=s_1-s_{1'},\\ \mathbf{r}_0'&=\mathbf{r}_2-\mathbf{r}_{2'}, & s_0'&=s_2-s_{2'}.\end{aligned}\quad\text{(A23)}$$

For Bloch states, w is the periodic potential and G is Bloch transformed according to

$$G_n(\mathbf{k},\omega_\nu)=\int d^3rd^3r'b_n^*(\mathbf{k},\mathbf{r})G(\mathbf{r}\mathbf{r}',\omega_\nu)b_n(\mathbf{k},\mathbf{r}').\quad\text{(A24)}$$

The Bloch transform of a four-point function $A(\mathbf{r}_1\mathbf{r}_3;\mathbf{r}_2\mathbf{r}_4)$ in which $\mathbf{r}_1$ and $\mathbf{r}_2$ are the particle coordinates, and $\mathbf{r}_3$ and $\mathbf{r}_4$ the hole coordinates, is

$$\langle\mathbf{k}_1n_1;\mathbf{k}_2n_2|A|\mathbf{k}_3n_3;\mathbf{k}_4n_4\rangle=\Omega\int d^3r_1d^3r_2d^3r_3d^3r_4b_{n_1}^*(\mathbf{k}_1,\mathbf{r}_1)b_{n_2}^*(\mathbf{k}_2,\mathbf{r}_2)\times A(\mathbf{r}_1\mathbf{r}_3;\mathbf{r}_2\mathbf{r}_4)b_{n_3}(\mathbf{k}_3,\mathbf{r}_3)b_{n_4}(\mathbf{k}_4,\mathbf{r}_4).\quad\text{(A25)}$$

APPENDIX B

In order to prove that T contains a pole for the unscreened Coulomb potential, we examine the quantity

$$Q(\mathbf{p},\mathbf{p}')=\left[\frac{-\mathcal{P}(\mathbf{p},\mathbf{q})}{\Delta E(\mathbf{p},\mathbf{q})}\right]^{1/2}\langle\mathbf{p}|T(\mathbf{q},0)|\mathbf{p}'\rangle\left[\frac{-\mathcal{P}(\mathbf{p}',\mathbf{q})}{\Delta E(\mathbf{p}',\mathbf{q})}\right]^{1/2}\quad\text{(B1)}$$

with $\mathbf{q}=2p_F$ in the z direction. From (2.13) we see that Q satisfies the integral equation

$$Q(\mathbf{p},\mathbf{p}')=K(\mathbf{p},\mathbf{p}')+\int\frac{d^3p''}{(2\pi)^3}K(\mathbf{p},\mathbf{p}'')Q(\mathbf{p}'',\mathbf{p}'),\quad\text{(B2)}$$

where K is the real, symmetric kernel

$$K(\mathbf{p},\mathbf{p}')=\left[\frac{-\mathcal{P}(\mathbf{p},\mathbf{q})}{\Delta E(\mathbf{p},\mathbf{q})}\right]^{1/2}v(\mathbf{p}-\mathbf{p}')\left[\frac{-\mathcal{P}(\mathbf{p}',\mathbf{q})}{\Delta E(\mathbf{p}',\mathbf{q})}\right]^{1/2}.\quad\text{(B3)}$$

In order to show that Q (or T) has a pole, it is sufficient to show that the homogeneous equation

$$\phi(\mathbf{p})=\int\frac{d^3p''}{(2\pi)^3}K(\mathbf{p},\mathbf{p}'')\phi(\mathbf{p}')\quad\text{(B4)}$$

is satisfied at some temperature. This temperature is the transition temperature. We shall express (B4) as

$$\phi=K\phi\quad\text{(B4')}$$

for convenience.

Now let ψ_n and E_n be the eigenfunctions and eigenvalues of the equation $K\psi_n=E_n\psi_n$. If, at zero temperature, we can find a function ϕ such that $\phi K\phi=\lambda>1$, then at least one of the eigenvalues E_{n_0} must be greater than zero, since

$$\phi=\sum_n a_n\psi_n,\quad \sum_n|a_n|^2=1,$$
$$\phi K\phi=\lambda=\sum_n|a_n|^2E_n>1.$$

Since K effectively decreases continuously with increasing temperature, we expect the eigenvalues to decrease continuously as the temperature increases. Thus, if $E_{n_0}(T=0)>1$, there will be a temperature T_0 at which $E_{n_0}(T_0)=1$. At this temperature $\psi_{n_0}(T_0)$ satisfies (B4′).

If we take ϕ as

$$\begin{aligned}\phi(\mathbf{p})&=[\ln(2/\mu)]^{-1/2}\frac{1}{p}\left[\frac{-\mathcal{P}(\mathbf{p},\mathbf{q})}{\Delta E(\mathbf{p},\mathbf{q})}\frac{8\pi^2}{m}\right]^{1/2},\quad \mu p_F\leq p\leq e\mu p_F\\&=0,\quad p\leq\mu p_F,\quad p\geq e\mu p_F,\end{aligned}\tag{B5}$$

it is easily seen that $\phi K\phi$ is proportional to $\ln\mu$ for sufficiently small μ. This completes the proof.

PHYSICAL REVIEW VOLUME 143, NUMBER 1 MARCH 1966

Quantum Theory of Magnetic-Resonance Line Shape in a Rigid Lattice*

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(Received 4 October 1965)

The autocorrelation function of the transversal magnetic moment is investigated in the presence of dipole-dipole interaction. For that purpose an integrodifferential equation is derived which is exact in all orders of the perturbation. The resulting equation is simplified with the aid of a linearity and Gaussian assumption and is solved numerically. Good agreement is thereby found with the experimental situation. In particular, it is shown that the physical nature of the oscillations found in the free-induction-decay experiments of Lowe and Norberg can be understood as a precession around an average internal magnetic field.

I. INTRODUCTION

IN the calculations of the resonance linewidth in paramagnetic systems with dipole-dipole interaction one in general makes use of the Gaussian assumption for the line shape.[1,2] This has clearly the advantage that the method of moments can be applied in a simple way. On the other hand, it is experimentally demonstrated by Lowe and Norberg[3] using the free-induction-decay method, that the decay curves are of an oscillatory nature (in contrast to the monotonic decrease of a Gaussian function).

The autocorrelation function $\Phi(t)$ of the transversal magnetic moment, which essentially determines the decay curves, has also been evaluated by Lowe and Norberg. Their calculation is based on a power-series expansion of the form

$$\Phi(t)=\sum_{n=0}^{\infty}t^nF_n(t),\tag{1}$$

in lowest order of which the Ising and exchange parts of the dipole-dipole interaction are assumed to commute. Because of the mathematical complications only the terms up to $n=4$ have been calculated. Remarkable agreement is thereby found with the experimental situation. It is, however, not clear from the theoretical point of view whether the higher order terms can be neglected.

The present paper is devoted to the study of the autocorrelation function within the framework of a formalism which has been developed previously[4] and which is based on a many-particle treatment of the spin system. From the present treatment the physical mechanism of the oscillatory behavior in time can, at least qualitatively, be understood.

We first shall be concerned with a derivation of an integrodifferential equation for the autocorrelation function with the use of a power-series expansion in terms of the Ising part of the interaction Hamiltonian. Subsequently, the resulting equation is simplified with the aid of a linearity condition. Finally, it is solved numerically and a comparison is made with experiment.

II. FORMULATION OF THE PROBLEM

In this section we briefly recall some details for further reference. Consider for definiteness a crystal of identical paramagnetic ions with spin $\frac{1}{2}$ which is placed in a large external magnetic field **H**. The field **H** is supposed to be along the z axis. Furthermore, let us confine ourselves to the situation that we may neglect

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* Work supported in part by the U. S. Atomic Energy Commission.

† On leave from the University of Nijmegen, The Netherlands.

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