

ON THE SUPERCONDUCTIVITY OF A FERROMAGNETIC WITH A WEAK EXCHANGE INTERACTION

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Submitted to JETP editor March 3, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) 39, 384-392 (August, 1960)

We discuss the influence of s-d exchange on the condition for the existence of a superconducting state in a ferromagnet, and also on its characteristics such as the critical temperature, critical field, and the specific heat.

It was shown in a paper by the present authors¹ that the establishment of superconductivity in ferromagnetics of the type Fe, Co, and Ni is prevented by the shift in the Fermi spheres of the s-conduction electrons with opposite spins, a shift caused by their coupling with the d or f electrons of the interior shells which have no spin-saturation, i.e., caused by the (s-d)-exchange interaction. It was also shown that superconductivity can occur in ferromagnetics with a sufficiently weak (s-d)-exchange coupling ($\mu J \ll \hbar\omega$, where μ is the excess of d or f electrons with the predominant spin orientation per lattice site;* J is the energy parameter of the (s-d) exchange, which is approximately independent of the wave vector k (reference 2); ω is the average phonon frequency). The existence of a superconducting ferromagnet was apparently established by Matthias, Suhl, and Corenzwit³ in the (Ce, Gd)Ru₂ system. In this connection it is of interest to consider in more detail the influence of the above-mentioned shift on the characteristics of the superconducting state, and this is the aim of the present paper.†

1. THE GROUND STATE AND FREE ENERGY OF A FERROMAGNETIC SUPERCONDUCTOR

When a ferromagnetic superconductor has an excess of s electrons with spins oriented to the left due to the magnetizing effect of the d or the

* μ is the relative magnetization of the d or f electron per lattice site. This quantity is weakly temperature dependent in the low temperature region.

† We use in the following the Bardeen-Cooper-Schrieffer scheme.⁴ We could use also the more rigorous Bogolyubov scheme,⁵ but it is then necessary to modify the canonical transformations in order to take the Fermi shift into account.

f electrons, one can write the Hamiltonian of the conduction electrons, between which there are interactions induced by the phonons⁴ and the ferromagnons⁶ and also a Coulomb repulsion, in the form^{4,2}

$$\mathcal{H} = \sum_{k > k_F^+} \epsilon_k^+ n_k^+ + \sum_{k > k_F^-} \epsilon_k^- n_k^- + \sum_{k < k_F^+} |\epsilon_k^+| (1 - n_k^+) + \sum_{k < k_F^-} |\epsilon_k^-| (1 - n_k^-) - \sum_{kk'} V_{kk'} b_k^\dagger b_{k'}, \tag{1}$$

where $\epsilon_k^+ = E_k + 1/2 \mu J - E_F$ and $\epsilon_k^- = E_k - 1/2 \mu J - E_F$ are the energies, relative to the Fermi energy E_F , of electrons F with spins oriented to the right (+) and to the left (-), respectively; n_k^+ and n_k^- , k_F^+ and k_F^- are respectively the occupation numbers and maximum wave vectors at $T = 0$, when there is no interaction;

$$b_k = c_k^+ c_{-k}^+, \quad b_k = c_{-k}^- c_k^+$$

are the pair creation and annihilation operators, expressed in terms of the usual Fermi operators for electrons \bar{c} and \bar{c}^\dagger ; $V_{kk'}$ are the matrix elements of the interaction which will be assumed to be constant,⁴ equal to V within a narrow region $|\epsilon_k| = |E_k - E_F| < \hbar\omega$, and equal to zero outside that region. In order that superconductivity exist, it is necessary⁶ to have $V = V_p - V_c - V_{sd} = 0$ [V_p , V_c , and V_{sd} are respectively the matrix elements of the interelectronic interaction induced by the phonons, the quasi-Coulomb interaction, and the effective repulsion between conduction electrons induced by s-d exchange]. Since $V_{sd} \sim J^2$ (reference 7) and the influence of the shift in the Fermi sphere is a first-order effect in the parameter J (references 1 and 2), we assume in the case of weak (s-d) exchange that although the quantity V_{sd} decreases the constant V , it still leaves it positive.

Since in our case, apart from pairs of Fermi particles ($k+$, $-k-$), there must already exist in the ground state "left-hand singlets" (i.e., electrons with $-$ spin in the state $-k$ without electrons with $+$ spin in the state k), one must write the wave function in the form

$$\Psi = \prod_k [(1 - h_k)^{1/2} + h_k^{1/2} b_k^\dagger] \prod_{k' \neq k} c_{k'}^- \Phi_0, \quad (2)$$

where Φ_0 is the "vacuum" of the system, and h_k the probability for the production of a pair ($k+$, $-k-$).

According to (2) the expansions

$$\Psi = h_k^{1/2} \varphi_1 + (1 - h_k)^{1/2} \varphi_0, \quad (3)$$

occur for any given k if k corresponds to a state of basic pairs;

$$\Psi = (h_k h_{k'})^{1/2} \varphi_{11} + [h_k (1 - h_{k'})]^{1/2} \varphi_{10} + [(1 - h_k) h_{k'}]^{1/2} \varphi_{01} + [(1 - h_k) (1 - h_{k'})]^{1/2} \varphi_{00}, \quad (4)$$

if k and k' belong to states of basic pairs; and

$$\Psi = c_k^\pm \varphi_0, \quad (5)$$

if k belongs to a state of "left-hand singlets." Here, φ_1 is a state with a pair ($k+$, $-k-$), φ_0 the state when there is no such pair, φ_{11} the state with pairs ($k+$, $-k-$; $k'+$, $-k'-$), φ_{00} a state when there are no such pairs, φ_{01} the state with a pair in the k' state and no pair in the k state, and φ_{10} the state with a pair in k and without a pair in k' . If s_k^- is the probability that the state k is occupied by a left-hand singlet, the expansions (3) to (5) correspond respectively to probabilities $(1 - s_k^-)$, $(1 - s_k^-)(1 - s_{k'}^-)$, and s_k^- . By analogy with Bardeen, Cooper, and Schrieffer's paper⁴ we get from the condition that the energy be a minimum an expression for h_k :

$$h_k = 1/2 [1 - \varepsilon_k / (\varepsilon_k^2 + \varepsilon_0^2)^{1/2}], \quad (6)$$

where unlike in reference 4

$$\varepsilon_0 = V \sum_k [h_k (1 - h_k)]^{1/2} (1 - s_k^-) \quad (7)$$

(so that only states with $s_k^- \neq 1$ give a contribution to ε_0).

The expression for the average energy of the system will be of the form

$$\begin{aligned} W_0 = & - \sum_{\text{III}} \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \varepsilon_k - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\} \\ & + \sum_{\text{III}} s_k^- \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \frac{\mu J}{2} - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\} \\ & - \sum_{\text{II}} (1 - s_k^-) \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \frac{\mu J}{2} - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\} \\ & - \sum_{\text{I}} \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} + \varepsilon_k + \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\} \\ & + \sum_{\text{I}} s_k^- \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \frac{\mu J}{2} - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\}, \quad (8) \end{aligned}$$

where I, II, and III denote respectively the regions of summation with $k < k_F^+$, $k_F^+ < k < k_F^-$, and $k > k_F^-$. The region II has then clearly an energy width μJ and contains within it the Fermi surface E_F . Since

$$(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \varepsilon_k - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2} > 0$$

and even more strongly in I and III

$$(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \mu J/2 - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2} > 0$$

(because $|\varepsilon_k| > 1/2 \mu J$ in I and III), according to (8) there will be no left-hand singlets in I and III (i.e., $s_k^- = 0$), which is of great advantage. The absence of left-hand singlets in II, however, and the corresponding presence of such singlets in I (or III) is energetically unprofitable because of the inequality

$$\begin{aligned} \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \frac{\mu J}{2} - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\}_{\text{I}} & > \{(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} \\ & - \frac{\mu J}{2} - \varepsilon_0^2 [2(\varepsilon_k^2 + \varepsilon_0^2)]^{-1/2}\}_{\text{II}}, \quad (9) \end{aligned}$$

where the expression with index I refers to any state from the region I, and the index II to the region II. There corresponds thus to the ground state a distribution of pairs in the regions I and III, according to (6), and an occupation of region II by the left-hand singlets ($s_{k, \text{II}}^- = 1$), so that the summation in (7) extends only over regions I and III (with $s_k^- = 0$). It follows therefore from (7) and (6) that

$$[N(0)V]^{-1} = \int_{\mu J/2}^{\hbar\omega} (\varepsilon^2 + \varepsilon_0^2)^{-1/2} d\varepsilon, \quad (10)$$

where $N(0)$ is the density of states near the Fermi surface.

Using (10) we find instead of (7) and (8)

$$\begin{aligned} \varepsilon_0 = & [2 \sinh(1/N(0)V)]^{-1} \{[2\hbar\omega e^{-1/N(0)V} - \mu J] \\ & \times [2\hbar\omega e^{1/N(0)V} - \mu J]^{1/2}\}, \quad (11) \end{aligned}$$

$$W_0 = -N(0) [2\hbar\omega - \mu J e^{1/N(0)V}]^2 / 2 [e^{2/N(0)V} - 1]. \quad (12)$$

Equations (11) and (12) go over into Eqs. (2.40) and (2.42) of reference 4 when $\mu J = 0$. When μJ increases from 0 to $2\hbar\omega e^{-1/N(0)V}$ the quantity ε_0 decreases to zero and the energy of the ground state W_0 increases to $W_n = 0$, i.e., to the energy of the normal state. Superconductivity is thus possible only for weak (s-d) coupling, provided that

$$\mu J < 2\hbar\omega e^{-1/N(0)V}. \quad (13)$$

The quantity ε_0 determines the dispersion law for the excitations which can be formed in the case under consideration, firstly by the breaking up in I or III of a pair (k_1+ , $-k_1-$) during a transition $k_1+ \rightarrow k_2+$ (or respectively $-k_1- \rightarrow -k_2-$), where k_2 also belongs to the region I or III; secondly through the formation in I and III of an excited pair generated by the operator $[(1 - h_k)^{1/2} b_k^\dagger$

$-h_k^{1/2}$]; thirdly by the breaking up in region I or III of a pair $(k_1 +, -k_1 -)$ during a transition $k_1 + \rightarrow k_2 +$, where k_2 belongs to II. In the first case the excitation energy is of the form

$$\begin{aligned} W_{k_1 k_2} - W_0 &= \varepsilon_{k_2}^+ + \varepsilon_{k_1}^- - (\varepsilon_{k_2}^+ + \varepsilon_{k_2}^-) h_{k_2} - (\varepsilon_{k_1}^+ + \varepsilon_{k_1}^-) h_{k_1} \\ &+ \{ [h_{k_1} (1 - h_{k_1})]^{1/2} + [h_{k_2} (1 - h_{k_2})]^{1/2} \} V \sum_{I, III} [h_k (1 - h_k)]^{1/2} \\ &= (\varepsilon_{k_1}^2 + \varepsilon_0^2)^{1/2} + (\varepsilon_{k_2}^2 + \varepsilon_0^2)^{1/2} \end{aligned} \quad (14)$$

and its minimum value is $2 [\frac{1}{2} \mu J]^2 + \varepsilon_0^2]^{1/2}$. In the second case we find for the excitation energy

$$\begin{aligned} W_k - W_0 &= (\varepsilon_k^+ + \varepsilon_k^-) (1 - 2h_k) \\ &+ 4\varepsilon_0 [h_k (1 - h_k)]^{1/2} = 2 (\varepsilon_k^2 + \varepsilon_0^2)^{1/2} \end{aligned} \quad (15)$$

with the same minimum value. In the third case the excitation energy is of the form

$$\begin{aligned} W_{k_1 k_2} - W_0 &= \varepsilon_{k_1}^- + \varepsilon_{k_2}^+ - (\varepsilon_{k_1}^+ + \varepsilon_{k_1}^-) h_{k_1} \\ &+ 2 [h_{k_1} (1 - h_{k_1})]^{1/2} V \sum_{I, III} [h_k (1 - h_k)]^{1/2} = \varepsilon_{k_2} + (\varepsilon_{k_1}^2 + \varepsilon_0^2)^{1/2} \end{aligned} \quad (16)$$

with a minimum value $-\frac{1}{2} \mu J + [(\frac{1}{2} \mu J)^2 + \varepsilon_0^2]^{1/2} > 0$ (since the minimum of ε_{k_2} is $-\frac{1}{2} \mu J$, and the minimum of $|\varepsilon_{k_1}|$ is $\frac{1}{2} \mu J$) which vanishes only for $\varepsilon_0 = 0$. It follows thus that also in the case of a ferromagnetic superconductor ε_0 from (11) determines the magnitude of the energy gap for the elementary excitations of the system.

When $T > 0$ one must take into account, apart from the basic pairs and the left-hand singlets, also the excited pairs and right-hand singlets. One must thus take instead of (2) a wave function of the form

$$\begin{aligned} \Psi' &= \prod_{k(0)} [(1 - h_k)^{1/2} + h_k^{1/2} b_k^+] \prod_{k_1(\text{exc})} [(1 - h_{k_1})^{1/2} b_{k_1}^+ \\ &- h_{k_1}^{1/2}] \prod_{k_2(s^-)} c_{k_2}^- - \prod_{k_3(s^+)} c_{k_3}^+ + \Phi_0, \end{aligned} \quad (17)$$

where 0, exc, s^- , and s^+ refer to states occupied respectively by basic pairs, excited pairs, left-hand, and right-hand singlets. We introduce, apart from the probability s_k^- , the probabilities s_k^+ and p_k for finding in the state k respectively a right-hand singlet or an excited pair, and we express them in terms of the distribution functions f_k^+ and f_k^- of fermions with + and - spins:

$$s_k^+ = f_k^+ (1 - f_k^-), \quad s_k^- = f_k^- (1 - f_k^+), \quad p_k = f_k^- f_k^+. \quad (18)$$

(This connection follows from the fact that s_k^+ is the probability that the state with $k+$ is occupied while the state $-k-$ is vacant; s_k^- the probability that the state with $k-$ is occupied while the state $-k+$ is vacant, and finally p_k the probability that the state with $k+$ and the state with $-k-$ are simultaneously occupied.) We get then for the average energy, instead of (8),

$$\begin{aligned} W &= \sum_{k > k_F^+} \varepsilon_k^+ [f_k^+ + (1 - f_k^+ - f_k^-) h_k] + \sum_{k > k_F^-} \varepsilon_k^- [f_k^- \\ &+ (1 - f_k^- - f_k^-) h_k] + \sum_{k < k_F^+} |\varepsilon_k^+| [1 - f_k^+ - (1 - f_k^+ - f_k^-) h_k] \\ &+ \sum_{k < k_F^-} |\varepsilon_k^-| [1 - f_k^- - (1 - f_k^- - f_k^-) h_k] \\ &- V \sum_{kk'} [h_k (1 - h_k) h_{k'} (1 - h_{k'})]^{1/2} (1 - f_k^+ - f_k^-) (1 - f_{k'}^+ - f_{k'}^-). \end{aligned} \quad (19)$$

The usual expression for the entropy of fermions

$$\begin{aligned} S &= -k \sum_k [f_k^+ \ln f_k^+ + (1 - f_k^+) \ln (1 - f_k^+) + f_k^- \ln f_k^- \\ &+ (1 - f_k^-) \ln (1 - f_k^-)] \end{aligned} \quad (20)$$

(where k is Boltzmann's constant), together with the condition that the free energy, $F = W - TS$, is a minimum

$\partial F / \partial h_k = \partial F / \partial f_k^+ = \partial F / \partial f_k^- = 0$ while $1 - f_k^- - f_k^- \neq 0$, yields

$$\begin{aligned} f_k^+ &= [\exp \{ \beta [(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} + \mu J / 2] \} + 1]^{-1}, \\ f_k^- &= [\exp \{ \beta [(\varepsilon_k^2 + \varepsilon_0^2)^{1/2} - \mu J / 2] \} + 1]^{-1}, \\ h_k &= \frac{1}{2} [1 - \varepsilon_k / (\varepsilon_k^2 + \varepsilon_0^2)^{1/2}], \end{aligned} \quad (21)$$

where $\beta = (\kappa T)^{-1}$ and, in contradistinction to (7),

$$\varepsilon_0 = V \sum_k [h_k (1 - h_k)]^{1/2} (1 - f_k^+ - f_k^-). \quad (22)$$

In III, where $h_k < \frac{1}{2}$, the functions f_k^+ and f_k^- describe the distributions of electrons with + and - spin, respectively. In I, where $h_k > \frac{1}{2}$, these functions describe the corresponding hole distributions. Since the electron and hole distributions found here are symmetric with respect to $\varepsilon_k = 0$, it is necessary to put in II the functions $f_k^- = 1$ and $f_k^+ = 0$, for in that region $1 - f_k^+ - f_k^- = 0$. The summation in (22) and also in (7) extends thus only over the regions I and III.

2. CRITICAL TRANSITION TEMPERATURE

The following equation for ε_0 results from (21) and (22)

$$\frac{1}{N(0)V} = \int_{\mu J/2}^{\hbar\omega} \frac{d\varepsilon}{E} \frac{\sinh(\varepsilon/\kappa T)}{\cosh(\varepsilon/\kappa T) + \cosh(\mu J/2\kappa T)}, \quad (23)$$

where $E = (\varepsilon^2 + \varepsilon_0^2)^{1/2}$. Taking it into account that at the critical temperature T_c the quantity ε_0 tends to zero, we find from (23) the following equation for T_c

$$\frac{1}{N(0)V} = \int_{\mu J/2}^{\hbar\omega} \frac{d\varepsilon}{\varepsilon} \frac{\sinh(\varepsilon/\kappa T_c)}{\cosh(\varepsilon/\kappa T_c) + \cosh(\mu J/2\kappa T_c)}. \quad (24)$$

In the case $\mu J/2\kappa T_c \ll 1$, we can restrict ourselves to terms of first order in that quantity and we can find from (24) the following equation for T_c

$$1/N(0)V = \ln(1.14\hbar\omega/\kappa T_c) - \mu J/4\kappa T_c, \quad (25)$$

from which, in the same approximation, it follows that

$$\kappa T_c = 1.14\hbar\omega \exp\{-1/N(0)V\} - \mu J/4, \quad (26)$$

so that T_c decreases when μJ increases.

When we consider the case of large μJ , with $\mu J/2\kappa T_c \gg 1$, it is convenient to write (24) in the form

$$\frac{1}{N(0)V} = \ln \frac{2\hbar\omega}{\mu J} + 2 \sum_{m=1}^{\infty} (-1)^m \left[\text{Ei} \left(-\frac{m\hbar\omega}{\kappa T_c} \right) - \text{Ei} \left(-\frac{m\mu J}{2\kappa T_c} \right) \right] \cosh \frac{m\mu J}{2\kappa T_c}, \quad (27)$$

where Ei is the exponential integral. Using, moreover, an asymptotic expansion for Ei and restricting ourselves to terms linear in $2\kappa T_c/\mu J$ we get easily from (27)

$$\kappa T_c = \frac{\mu J}{2 \ln 2} \left(\ln \frac{2\hbar\omega}{\mu J} - \frac{1}{N(0)V} \right). \quad (28)$$

It follows from (28) that when $\mu J = 2\hbar\omega \exp\{-1/N(0)V\}$ the critical temperature tends to zero, which agrees with condition (13).

3. SPECIFIC HEAT OF A FERROMAGNETIC SUPERCONDUCTOR

Using (11), we can transform (20) and get for the electron specific heat in the superconducting state the following expression

$$C_{es} = T \frac{\partial S}{\partial T} = -2\kappa\beta^2 \sum_{k>k_F} \left[\left(E_k + \frac{\mu J}{2} \right) \frac{\partial f_k^+}{\partial \beta} + \left(E_k - \frac{\mu J}{2} \right) \frac{\partial f_k^-}{\partial \beta} \right] \quad (29)$$

Equation (29) can be transformed to

$$\begin{aligned} C_{es} = 2\kappa\beta^2 \sum_{k>k_F} \left\{ f_k^+ (1 - f_k^+) \left[\left(E_k + \frac{\mu J}{2} \right)^2 + \beta \left(E_k + \frac{\mu J}{2} \right) \right. \right. \\ \left. \left. \times \left(\frac{1}{2E_k} \frac{\partial \epsilon_0^2}{\partial \beta} + \frac{\partial}{\partial \beta} \frac{\mu J}{2} \right) \right] + f_k^- (1 - f_k^-) \left[\left(E_k - \frac{\mu J}{2} \right)^2 \right. \right. \\ \left. \left. + \beta \left(E_k - \frac{\mu J}{2} \right) \times \left(\frac{1}{2E_k} \frac{\partial \epsilon_0^2}{\partial \beta} - \frac{\partial}{\partial \beta} \frac{\mu J}{2} \right) \right] \right\}. \quad (30) \end{aligned}$$

Considering that at $T = T_c$ the quantity ϵ_0 tends to zero and that the distribution function (21) takes on its normal value

$$\begin{aligned} f_{k,n}^+ &= [\exp\{\beta(\epsilon_k + \mu J/2)\} + 1]^{-1}, \\ f_{k,n}^- &= [\exp\{\beta(\epsilon_k - \mu J/2)\} + 1]^{-1}, \end{aligned} \quad (31)$$

one gets easily from (30) an expression for the jump in the specific heat, accurate to terms of order $\mu J/\kappa T_c$,

$$(C_{es} - C_{en})_{T_c} = \kappa N(0) \beta_c^2 (1 - \mu J/4\kappa T_c) \partial \epsilon_0^2 / \partial \beta |_{T_c}, \quad (32)$$

where C_{en} is the specific heat of the normal state. In the same approximation, by differentiating (23)

with respect to β and using numerical integration, we can find for the quantity $\partial \epsilon_0^2 / \partial \beta |_{T_c}$ the following expression

$$\frac{\partial \epsilon_0^2}{\partial \beta} |_{T_c} = \frac{10.30}{\beta_c^3} \left(1 - 0.07 \frac{\mu J}{\kappa T_c} \right). \quad (33)$$

From (32) and (33) we get

$$(C_{es} - C_{en})_{T_c} = 10.30 \kappa^2 N(0) (1 - 0.32 \mu J / \kappa T_c), \quad (34)$$

from which it follows that the (s-d)-exchange interaction reduces the magnitude of the jump in the specific heat. Assuming (33) to be valid when $T \lesssim T_c$, we can easily find also the temperature dependence of ϵ_0 for $T \sim T_c$ and $\mu J/\kappa T_c < 1$:

$$\epsilon_0(T) = 3.209 \kappa T_c (1 - T/T_c)^{1/2} (1 - 0.035 \mu J / \kappa T_c), \quad (35)$$

which goes over into the corresponding expression (3.31) of reference 4 when $\mu J \rightarrow 0$.

4. CRITICAL MAGNETIC FIELD

It is well known that the critical magnetic field H_c can be determined by using the relation

$$H_c^2 / 8\pi = F_n - F_s, \quad (36)$$

where F_n and F_s are respectively the free energies of the normal and the superconducting states. To evaluate $F_s = W - TS$ we use (21) and (22), changing from a summation over the quasi-momentum to integration over the energy $d\epsilon$, to transform (19) to the form

$$\begin{aligned} W = 2N(0) \left\{ \int_{\mu J/2}^{\hbar\omega} \epsilon d\epsilon \int_{\mu J/2}^{\infty} \left[\frac{\mu J}{2} (f_k^+ - f_k^-) \right. \right. \\ \left. \left. - \frac{\epsilon^2}{2E} (e^{\beta(E - \mu J/2)} - 1) (f_k^- + f_k^+) \right] d\epsilon - \frac{\epsilon_0^2}{V} \right\}. \end{aligned} \quad (37)$$

Similarly we can use (21), change from summation to integration, integrate by parts, and obtain an expression for the entropy

$$\begin{aligned} TS = 2N(0) \left\{ \int_{\mu J/2}^{\infty} [(\epsilon^2/E + E) (f_k^+ + f_k^-) + (\mu J/2) (f_k^+ - f_k^-)] d\epsilon \right. \\ \left. - \kappa T (\mu J/2) \ln [2(1 + e^{-\beta\mu J})] \right\}. \end{aligned} \quad (38)$$

We find from (37) and (38), and also from (22) and (21), after some simple calculations

$$\begin{aligned} F_s = -N(0) \int_{\mu J/2}^{\infty} (2\epsilon^2 + \epsilon_0^2) E^{-1} (f_k^+ + f_k^-) d\epsilon + N(0) \{ (\hbar\omega)^2 [1 \\ + \sqrt{1 + (\epsilon_0/\hbar\omega)^2}] - (\mu J/2)^2 [1 - \sqrt{1 + (2\epsilon_0/\mu J)^2}] \} \\ + N(0) \kappa T \mu J \ln [2(1 + e^{-\beta\mu J})]. \end{aligned} \quad (39)$$

Substituting (39) and the equation $F_n = -\pi^2 N(0) \times (\kappa T)^2/3$ for the free energy of the normal state into (36), we get

$$H_c^2/8\pi = -N(0) \{ (\hbar\omega)^2 [1 - \sqrt{1 + (\epsilon_0/\hbar\omega)^2}] - (\mu J/2)^2 [1 - \sqrt{1 + (2\epsilon_0/\mu J)^2}] \} - N(0) \times T \mu J \ln [2(1 + e^{-\beta\mu J})] - \frac{\pi^2}{3} N(0) (\times T)^2 \left\{ 1 - \frac{3\beta^2}{\pi^2} \int_{\mu J/2}^{\infty} \frac{2\epsilon^2 + \epsilon_0^2}{E} (f_k^+ + f_k^-) d\epsilon \right\}. \quad (40)$$

From (40) it follows for $T = 0$ that

$$H_c^2/8\pi = N(0) \{ \epsilon_0^2(0)/2 + (\mu J/2)^2 [1 - \sqrt{1 + (2\epsilon_0(0)/\mu J)^2}] \}, \quad (41)$$

where H_0 is the critical field for $T = 0^\circ\text{K}$. In the case $\mu J \ll 2\hbar\omega \exp\{-1/N(0)V\}$, when $\epsilon_0(0) \gg \mu J$ according to (11), it follows from (41) that

$$H_0 \sim (4\pi N(0))^{1/2} [\epsilon_0(0) - \mu J/2]. \quad (42)$$

When $\mu J \rightarrow 2\hbar\omega \exp\{-1/N(0)V\}$, when $\epsilon_0(0) \rightarrow 0$ according to (11), it follows from (41) that $H_0 \rightarrow 0$.

Using (41) one can write Eq. (40) in the form

$$H_c^2/8\pi = H_0^2/8\pi - \alpha_1 T - \alpha_2 T^2, \quad (43)$$

$$\alpha_1 = N(0) \times \mu J [\ln 2 + \ln(1 + e^{-\beta\mu J})].$$

The (s-d) exchange leads thus not only to a decrease in the critical field but also to the appearance of a linear term $\alpha_1 T$ besides the usual quadratic dependence of H_c^2 on T (reference 4), which is described by the term $\alpha_2 T^2$.

5. POSSIBILITIES OF EXPERIMENTAL VERIFICATION OF THE THEORETICAL DEDUCTIONS: CONCLUSION

We mentioned in the foregoing that the existence of a ferromagnetic superconductor has only been established so far, apparently, in the (Ce, Gd) Ru_2 system.³ The possibility is, however, not excluded that an improvement in the techniques for obtaining low temperatures will enable us to observe a similar phenomenon also in other systems in which there is a tendency for an intersection of the curves of the critical temperature (T_c) and of the Curie point (Θ) vs. the concentration of one of the components [for instance, in the (Ce, Pr) Ru_2 system³]. On the other hand, it is clear from the foregoing theory that in the case of a superconducting ferromagnet the characteristics of the superconducting state differ from the usual ones by a number of features caused by the (s-d)-exchange coupling. An experimental study of these features would thus be not only of interest for the theory of superconductivity, but also for a verification of the (s-d)-exchange model of ferromagnetic metals and above all for an independent estimate of the (s-d)-exchange coupling parameter J . This parameter could, for instance, be determined by a study of such experimentally accessible quantities

as the jumps in the specific heat at T_c and in the critical temperatures and the specific heat of the normal state near T_c . One can, apart from this, according to (43), use the experimental determination of the coefficient α_1 in the temperature dependence of H_c^2 , and so on. Accordingly, experiments on a detailed study of the superconducting characteristics of the (Ce, Gd) Ru_2 system and also on a search for new superconducting ferromagnets, would be very worthwhile.

From what has been said it follows that the presence of a shift of the Fermi surface, caused by the fermion-magnetization effect, makes the existence of a superconducting state in a ferromagnet possible only when the (s-d)-exchange coupling is sufficiently weak, when condition (13) is satisfied and this shift reduces the energy gap, the critical temperature, the critical magnetic field, and the jump in the specific heat. The effective repulsion of the conduction electrons, induced by the (s-d)-exchange coupling,⁷ also acts in the same direction; the appropriate matrix element V_{sd} decreases thus the quantity V in all expressions obtained. Since, however, the quantity V_{sd} is proportional to J^2 , one can expect that when the (s-d)-exchange interaction is weak the dominant role will be played by the dependences on μJ obtained in the foregoing, in which this quantity enters linearly.

We note in conclusion that Anderson and Suhl⁸ have used the Kittel-Ruderman-Yosida^{9,10} representation for the interaction between conduction electrons and spin moments to conclude that a special state of spin-atomic ordered "cryptoferromagnetism" could possibly exist in the form of a domain structure with zero resultant moment. The scheme for calculations given in our paper could be generalized also for the case of dilute solutions. We shall present a detailed analysis of that case in another paper.

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Translated by D. ter Haar