Antiferromagnetic superconductors

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The properties of antiferromagnetic superconductors are discussed. There is an emphasis on their anomalous behavior: the gapless nature of the superconductivity, the nonmonotonic temperature dependence of the upper critical field, the possible appearance of self-induced vortices at domain walls, and the unusual spin-wave spectrum. Possible phases in which superconductivity coexists with weak ferromagnetism are analyzed.

TABLE OF CONTENTS

1. INTRODUCTION

The 1977 discovery of a new group of ternary compounds with a regular lattice of rare earth (RE) atoms, with the general formulas $RERh_4B_4$ and $REMo_6S_8$, provided a powerful stimulus for experimental and theoretical research on the problem of the coexistence of superconductivity and magnetism. Many ternary compounds have proved to be superconductors in which a transition to a magnetically ordered state occurs at temperatures below T_c, the superconducting transition temperature.

The transition to the ferromagnetic phase may go through a peculiar coexistence phase in which superconductivity gives rise to an inhomogeneous magnetic structure of a domain type with a wave vector $Q \sim (a\xi_0)^{-1/2}$, where a is the magnetic rigidity, which is of the order of the interatomic distance, and ξ_0 is the superconducting coherence length. Long-period magnetic structures of this sort have been observed in neutron scattering experiments in $HoMo_6S_8$ (Ref. 1), $ErRh_4B_4$ (Ref. 2), and, recently, $HoMo_6Se_8$ (Ref. 3). A further lowering of the temperature in $HoMo_6S_8$ and $ErRh_4B_4$ leads to a disruption of the superconductivity and of the inhomogeneous magnetic order which it induced; the transition to a normal ferromagnetic phase is a first-order transition. The temperature range of coexistence of superconductivity and magnetism in $HoMo_6S_8$ and $ErRh_4B_4$ is extremely narrow, while in $HoMo_6Se_8$ the inhomogeneous magnetic phase of coexistence persists down to extemely low temperatures. It is this inhomogeneous type of magnetic ordering and the distinctive behavior of ferromagnetic superconductors in a field near the Curie point which are primarily responsible for the interest in experimental and theoretical research on magnetic superconductors. This research direction is reviewed in Ref. 4.

Most of the magnetic superconductors which have been produced so far are antiferromagnetic substances. These are the ternary borides $RERh_4B_4$ with RE = Nd (Ref. 5), Tm (Ref. 6), or Sm (Ref. 7); the body-centered tetragonal (bct) phase of $ErRh_4B_4$ (Refs. 8 and 9); and the ternary chalcogenides $REMo_6S_8$ with RE = Tb, Dy, Er (Refs. 10 and 11), Gd (Ref. 11), and Nd (Ref. 12), and $ErMo_6Se_8$ (Ref. 13). It turns out that the onset of an antiferromagnetic order below the Néel point, $T_N < T_c$, does not disrupt the superconducting state,[1)] while in a situation with $T_c \lesssim T_N$ antiferromagnetism does not prevent superconducting pairing. These facts caused no surprise, since the weak mutual effects of superconductivity and antiferromagnetism were predicted theoretically back in 1963 by Baltensperger and Strassler.[14] Nevertheless, antiferromagnetic superconductors exhibit many anomalous features: gapless superconductivity, a nonmonotonic temperature dependence of the upper critical field H_{c2}, an unusual spin-wave spectrum, and the possible appearance of new phases of coexistence in weak ferromagnetic materials. These questions constitute the subject of the present review. We will be focusing on compounds with localized magnetic moments. For crystals with collectivized magnetic moments, in contrast, we will simply present the few experimental facts which are available and offer some brief comments. These compounds are covered in more detail in the review by Machida.[61]

2. MUTUAL EFFECTS OF SUPERCONDUCTIVITY AND ANTIFERROMAGNETISM

The reason for the weak mutual effects of superconductivity and antiferromagnetism is that there is no average

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magnetization in an antiferromagnetic substance (in contrast with ferromagnetism), and the magnetic moment changes direction over scale lengths small in comparison with the superconducting correlation length. Superconductivity has a very weak effect on this order, since Cooper pairing causes almost no change in the electron spin susceptibility at the wave vectors $|G|\sim a^{-1}$ characteristic of antiferromagnetic order. Here the exchange interaction remains the same [within a relative error $(a/\xi_0)\sim T_c/\mathscr{E}_F$ as in normal antiferromagnetic materials. The short-wave part of the electromagnetic interaction also remains essentially the same upon the appearance of superconductivity within $(a^3/\lambda_L^2\xi_0)$, where λ_L is the London screening depth. The reason is that effective screening of the field in a superconductor is possible only over distances of the order of or greater than λ_L.

The effect of antiferromagnetism on superconductivity turns out to be stronger. Two basic mechanisms for this effect can be identified:

a) The spin splitting of electronic levels by the exchange field set up by localized moments and the resulting appearance of a gap on a small part of the Fermi surface. Such a gap reduces the total density of electron states.[14,15] Another important point is that the exchange field makes the superconductivity gapless (in a pure compound); this effect is usually basically responsible for the suppression of superconductivity at low temperatures, $T<T_N\ll T_c$.

b) The magnetic scattering of electrons[16] by spin fluctuations above the Néel point and by spin waves below the Néel point.[17,18]

2.1. Effect of the exchange field

We first consider the effect of the exchange field on the superconductivity in pure superconductors with an electron mean free time $\tau\gg T_c^{-1}$. For simplicity we consider a cubic antiferromagnetic material with two sublattices and a single electron band,[19] $\varepsilon(\mathbf{k})$. The exchange field $h(\mathbf{r})$, which oscillates in space, is then characterized by a wave vector $\mathbf{Q}$ which is equal to half a reciprocal-lattice vector: $\mathbf{Q}=(\pi/a, \pi/a, \pi/a)$, where a is the distance beween localized moments.

The Hamiltonian of the system of electrons in the BCS model is

$$\mathscr{H}=\sum_{k,\sigma}\left[\varepsilon(k)c^+_{k,\sigma}c_{k,\sigma}+\sigma h_Q c^+_{k+Q,\sigma}c_{k,\sigma}\right]+\Delta\sum_k(c_{k,-}c_{-k,+}+\text{c.c.})+\frac{\Delta^2}{g},$$

$$\Delta=g\sum_k\langle c_{k,+}c_{-k,-}\rangle, \qquad (1)$$

where $\sigma=+,-$ specifies the spin projection onto the z axis, which is the direction of the exchange field, h_Q is a Fourier component of the exchange field, and g is a parameter of the electron-phonon interaction. The superconducting order parameter Δ is taken to be spatially uniform, since to incorporate its dependence on the coordinates within a unit cell would be to go beyond the accuracy of the model with which we are dealing.[2)]

In the absence of superconductivity, Hamiltonian (1) is easily diagonalized and takes the form

$$\mathscr{H}_0=\sum_{k,n,\sigma}E_{n,k}a^+_{n,k\sigma}a_{n,k\sigma},$$

$$E_{\pm1,k}=\varepsilon_k\pm(\delta_k^2+h_Q^2)^{1/2},$$

$$\varepsilon_k=\frac{1}{2}\left[\varepsilon\left(k+\frac{Q}{2}\right)+\varepsilon\left(k-\frac{Q}{2}\right)\right],$$

$$\delta_k=\frac{1}{2}\left[\varepsilon\left(k+\frac{Q}{2}\right)-\varepsilon\left(k-\frac{Q}{2}\right)\right]; \qquad (2)$$

here the wave vector k lies in a new Brillouin zone, corresponding to an antiferromagnetic material and the operators a_k are related to the operators c_k by a canonical Bogolyubov transformation:

$$\left.\begin{aligned}a_{1,k\sigma}&=u_kc_{k,\sigma}-\sigma v_kc_{k+Q,\sigma},\\ a_{-1,k\sigma}&=\sigma u_kc_{k,\sigma}+u_kc_{k+Q,\sigma},\\ \left.\begin{matrix}u_k^2\\ v_k^2\end{matrix}\right\}&=\frac{1}{2}\pm\frac{\delta_k}{2(\delta_k^2+h_Q^2)^{1/2}}.\end{aligned}\right\} \qquad (3)$$

In terms of the new operators, Hamiltonian (1) can be written

$$\mathscr{H}=\sum_{n,k}E_{n,k}a^+_{n,k\sigma}a_{n,k\sigma}+[\Delta_g(k)a_{n,-k,-}a_{n,k,+}+\text{c.c.}]+\frac{\Delta^2}{g}, \qquad (4)$$

where $\Delta_g(k)$ plays the role of a superconducting gap and is determined by the equations

$$\Delta_g(k)=\Delta(u_k^2-v_k^2)=\frac{\Delta\delta_k}{(\delta_k^2+h_Q^2)^{1/2}}, \qquad (5)$$

$$\Delta_g(k)=g(u_k^2-v_k^2)\sum_{h',k'}\langle a_{n',k',+}a_{n',-k',-}\rangle(u_{k'}^2-v_{k'}^2). \qquad (6)$$

In addition to the terms described above, Hamiltonian (4) also contains terms which are not diagonal in the indices n, with coefficients u_kv_k. Such terms can be ignored under the conditions $\Delta\ll h\ll v_FQ$; i.e., the superconducting part of Hamiltonian (4) is written with an accuracy h/v_FQ, $\Delta/h\ll1$.

It can be seen from (4) and (5) that the quasiparticle spectrum is determined by the expression

$$\tilde{E}_n(k)=(E_{n,k}^2+\Delta_g^2(k))^{1/2}. \qquad (7)$$

The critical temperature of the superconducting transition found from self-consistency equation (6) in the limit $\Delta\to0$, is given by the expression[14] (we are assuming the case $T_c<T_N$)

$$T_c=1.14\omega_D\exp\left(-\frac{1}{\tilde{\lambda}}\right),$$

$$\tilde{\lambda}=g\sum_{k,n=1,2}\delta(E_{n,k}-\mathscr{E}_F)\frac{\delta_k^2}{\delta_k^2+h_Q^2}. \qquad (8)$$

It follows from (8) that the exchange field affects the pairing constant because of the change in the state density and because of the modulations of the electron wave functions. In the case of a spherical Fermi surface, both of these effects are small, or the order of[3)] $h_Q/\mathscr{E}_F$. The change in the order parameter Δ at $T\ll T_c$ is more significant, $\delta\Delta/\Delta\approx(h/v_FQ)\ln(h/\Delta_0)$. The appearance of a logarithmic factor

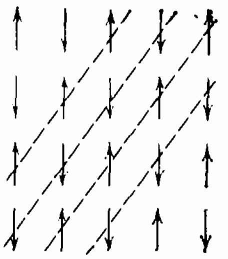

FIG. 1. Schematic diagram of an antiferromagnetic structure. Electrons moving along the directions indicated by the dashed lines experience the effect of a strong static exchange field.

here is a consequence of the gapless nature of the quasiparticle spectrum at $h \gg \Delta$.

According to (5) and (7), there is no gap in an antiferromagnetic superconductor with $h \gg \Delta$ on that band of the Fermi surface which is oriented perpendicular to the antiferromagnetism wave vector **Q**. For these momentum directions, the electrons move in a constant exchange field, and at $h \gg \Delta$ this field destroys the corresponding Cooper pairs (Fig. 1). The width of the gapless band is determined by the small parameter $h/v_F Q$. The electron state density $\rho(E)$ under the condition $E \ll \Delta$ (and $h \gg \Delta$) and the electron heat capacity are given by

$$\rho(E) = \frac{\pi h_Q E}{v_F Q \Delta} N(0), \quad \frac{C_e(T)}{\gamma} = \frac{h_Q T^2}{v_F Q \Delta}, \tag{9}$$

where γ is the coefficient of the linear term in the electron heat capacity of the normal phase. For ordinary antiferromagnetic ternary compounds, the quantity $h_Q/v_F Q$ is about 10^{-2}, and it would be difficult to observe the corresponding effects.[4)] The gapless band disappears under the condition $h < \Delta$, and the expression for the quasiparticle spectrum, which holds for arbitrary relations among Δ, h, and $v_F Q$, is

$$E^2_{n,k} = \varepsilon_k^2 + \delta_k^2 + \Delta^2 \pm 2\left[\varepsilon_k^2(\delta_k^2 + h_Q^2) + \Delta^2 h_Q^2\right]^{1/2}. \tag{10}$$

Spectrum (10) was derived in Ref. 23 through an exact diagonalization of Hamiltonian (1) for a helicoidal exchange field. An antiferromagnetic material with two sublattices is a particular case of a helicoid. Expression (10) reproduces results (5), (7), and (9) in the limit $h \gg \Delta$ and predicts $\Delta(k) = \Delta$ at $h \ll \Delta$.

What happens to the superconducting characteristics of an antiferromagnetic substance as the concentration of nonmagnetic impurities increases? Such impurities intensify the destructive effect of the exchange field on the superconductivity and in the sense are analogous to magnetic impurities. This circumstance was first pointed out by Morozov[24] (see also the more recent studies of Refs. 15, 20, 25–28). The effect of impurities depends strongly on the parameter $h_Q \tau$, where τ is the electron mean free time. In the case of a low impurity concentration, $h_Q \tau \gg 1$, the impurities destroy the superconductivity, as do magnetic impurities[30] with the inverse time for scattering with spin flip, $\tau^{-1} \sim h_Q/(\mathscr{E}_F \tau)$ (it is this case which corresponds to the study in Ref. 24). The gapless band remains, and the state density in (9) increases by a factor of[29] $1 + (\tau\Delta)^{-1}$. This result was derived by Panyukov for a domain structure, but it also applies, aside from the numerical coefficient of $(\tau\Delta)$, to antiferromagnetic materials. Under the condition $h_Q \tau \ll 1$, the gapless band disappears, since the motion of electrons becomes completely diffuse, and they do not sense the anisotropy of the antiferromagnetic structure. The effect of impurity scattering under these conditions becomes independent of τ and analogous to the situation in a superconductor with magnetic impurities,[16] with the inverse time of magnetic scattering, $\tau_s^{-1} \approx h_Q^2/v_F Q \approx T_N$. Correspondingly, under the condition $\tau_s \Delta > 1$ there is a gap in the spectrum of the system,

$$\Delta_g = \Delta\left[1 - (\Delta\tau_s)^{-2/3}\right]^{3/2}, \tag{11}$$

and the order parameter Δ is suppressed in the case $\tau_s \Delta \gg 1$ by an amount $\delta\Delta/\Delta \approx H_Q^2/v_F Q \Delta \approx T_N/T_c$ (Refs. 24 and 20). In the case $(\tau_s \Delta) < 1$, the superconductivity becomes gapless; the state density is nonzero even at $E = 0$, as in the Abrikosov-Gor'kov situation for a superconductor with magnetic impurities[5)] (Ref. 16). As the parameter $\tau_s \Delta$ is reduced further, the superconductivity disappears.

It was mentioned in Refs. 25–28 that the introduction of nonmagnetic impurities in an antiferromagnetic material with $h > \Delta$ in a one-dimensional system gives rise to localized states with an energy lying in the superconducting gap. We note, however, that these results apply only to compounds with a one-dimensional electron spectrum, in which both the magnetic gap and the superconducting gap appear over the entire Fermi surface. In a three-dimensional system, in contrast, the gap in the spectrum disappears at $h > \Delta$ even in the absence of impurities [see (10)].

In estimating the effect of the antiferromagnetic exchange field above we used the relation $h_Q^2/v_F Q \approx h_Q^2 N(0) \approx T_N$, under the assumption that the contribution of the exchange interaction to the Néel temperature T_N is no smaller in order of magnitude than the contribution from the magnetic dipole-dipole interaction. This condition always holds in ternary compounds (more on this below).

This estimate is valid only at temperatures below T_N, where the exchange field $h_Q(T)$ agrees in order of magnitude with the exchange field at $T = 0$. In the region near T_N, the field h_Q is weak, and we have $h_Q^2 N(0) \approx (T_N - T)$. In this temperature region, however, there is actually a substantial magnetic scattering by spin waves, to which we now turn.

2.2. Magnetic scattering

As follows from the Abrikosov-Gor'kov theory,[16] the inverse time for magnetic scattering, τ_s^{-1}, is $\tau_s^{-1} \approx h_Q^2 N(0) \approx T_N$ for uncorrelated magnetic moments (above T_N), when the concentration of these moments is of the same order as the electron concentration. In the general case, in the static approximation, the expression for τ_s^{-1} in the superconducting phase, in the limit of a dirty superconductor ($\tau T_c \ll 1$) is[17]

$$\tau_s^{-1} = N(0)(g_J - 1)^2 \sum_{\mathbf{q}} g(q) I^2(q) \langle \hat{\mathbf{J}}_{\mathbf{q}} \hat{\mathbf{J}}_{-\mathbf{q}} \rangle, \tag{12}$$

where the operator $\hat{\mathbf{J}}$ represents the angular momentum, I is the exchange integral, and g_I is the Landé factor. The weight function $g(q)$ has the same meaning (and the same q dependence) as the difference between the paramagnetic susceptibilities $\chi_n(q)$ and $\chi_s(q)$ in the normal and superconducting phases: $[\chi_n(q) - \chi_s(q)]/\chi_n(0) = \pi/(2q\xi_0)$ at $q \gg \xi_0^{-1}$. The function $g(q)$ in (12) describes a decrease in T_c due to magnetic scattering, while the difference $(\chi_s - \chi_n)$ describes the decrease in the superconducting order parameter at $T \ll T_c$. The exchange field h in an antiferromagnetic material is related to the exchange integral I by

$$\mathbf{h} = I(\mathbf{q} = \mathbf{Q})(g_I - 1)\langle \hat{J} \rangle. \tag{13}$$

The correlation function $\langle \hat{\mathbf{J}}_\mathbf{q} \hat{\mathbf{J}}_{-\mathbf{q}} \rangle$ from (12) must satisfy the sum rule

$$\sum_\mathbf{q} \langle \hat{\mathbf{J}}_\mathbf{q} \hat{\mathbf{J}}_{-\mathbf{q}} \rangle = J(J+1) \sum_\mathbf{q} 1, \tag{14}$$

where the summation over $\mathbf{q}$ is carried out over the first Brillouin zone of the magnetic lattice. As the temperature approaches T_N from above, the maximum of $\langle \hat{\mathbf{J}}_\mathbf{q} \hat{\mathbf{J}}_{-\mathbf{q}} \rangle$ shifts into the region of large $q \sim Q$, where the factor $g(q)$ is small. According to sum rule (14), the contribution from long-wave fluctuations should decrease in this case, leading to a decrease in τ_s^{-1}. The onset of an antiferromagnetic order, which weakens magnetic fluctuations with small values of q, thus leads to an increase in the superconducting order parameter.[17] This conclusion holds for any dirty superconductor with $h_Q \tau \ll 1$ and for pure compounds with a roughly spherical Fermi surface. In pure compounds ($h_Q \tau > 1$) with a more complicated Fermi surface, however, e.g., one which has plane regions which can be brought into coincidence by a translation $\mathbf{Q}$, the quantity τ_s^{-1} may increase near[18] T_N.

In dirty superconductors, expression (12) can be used for a complete description of an antiferromagnetic superconducting phase at temperatures $T < T_N$ if the spin-wave frequencies are low in comparison with Δ (here the condition $T_N \ll T_c$ must hold; more on this below). Expression (12) then describes the effect of the average exchange field (of the component with $q = Q$), examined previously, and the effect of scattering by spin waves (the other components of q). In the order of magnitude, τ_s^{-1} is equal to T_N in any case, but the role of the exchange field increases with decreasing temperature. For compounds with a roughly spherical Fermi surface, the onset of antiferromagnetic order leads to an overall intensification of the superconductivity at temperatures $T < T_N$ in comparison with the region $T > T_N$.

These estimates of τ_s^{-1} explain the fact that the condition $T_c > T_N$ holds in nearly all existing antiferromagnetic superconductors. Certain pseudoternary systems are exceptions to this rule (Section 4). In addition, superconductivity at $T_c \approx 0.8$ K was recently observed[75] in the antiferromagnetic compound $Tb_2Mo_3Si_4$, with $T_N \approx 19$ K. The superconducting transition turned out to be extremely blurred, and the critical temperature varied from sample to sample (a typical value was $\Delta T_c \approx 0.2$–0.3 K). This is the first case in which T_N is much higher than T_c. In principle, such a situation could occur in an antiferromagnetic superconductor with an easy-axis magnetic anisotropy or a large effective moment, $J \gg 1$ (the effective moment in $Tb_2Mo_3Si_4$ is[75] $J \approx 9$). The scattering by spin waves is strongly suppressed in the first case by the gap in the spin-wave spectrum, while in the second case it is suppressed in proportion to the parameter $1/J \ll 1$, because of the small value of the fluctuations of the moment. In both of these cases, the inequality $\tau_s^{-1} \ll T_N$ can hold; in such a case, the effect of the antiferromagnetic exchange field may prove to be important. In a pure superconductor, the effect of this field on T_c is small, proportional to the parameter $h/\mathscr{E}_F$. We could also expect the condition $T_N \gg T_c$ to hold. The addition of nonmagnetic impurities to such compounds will strengthen the effect of the antiferromagnetic exchange field and should result in a destruction of superconductivity under the condition $\tau^{-1} \gtrsim T_c(\mathscr{E}_F/h) \sim h(T_c/T_N)$. The pronounced blurring of the superconducting transition in[75] $Tb_2Mo_3Si_4$ agrees with these arguments. It would be interesting to study the decrease in T_c with decreasing electron mean free path in $Tb_2Mo_3Si_4$.

3. UPPER CRITICAL FIELD H_{c2} IN ANTIFERROMAGNETIC SUPERCONDUCTORS

The onset of antiferromagnetic order below T_N is quite obvious in the temperature dependence of the upper critical field H_{c2}. Figure 2 shows some typical curves of $H_{c2}(T)$, from which we see that there is no universal behavior of $H_{c2}(T)$.

In the presence of an external magnetic field H, a static exchange field $h = h_0 \chi_m H/n\mu$ arises in an antiferromagnetic material (in the normal state); here χ_m is the susceptibility of the magnetic subsystem, and μ is the effective saturation magnetic moment of the RE ions in the given crystal. The field $H_{c2}(T)$ thus depends on (a) magnetic scattering, (b) the exchange field (because of the paramagnetic effect, and (c) the induction B by virtue of the ordinary orbital effect).

The orbital effect itself leads to a smooth, monotonic increase in H_{c2} during cooling, but the effects of the exchange field and of magnetic scattering may disrupt this monotonic behavior. The susceptibility χ_m reaches a maximum at $T = T_N$, and below T_N it is constant when the field H is directed perpendicular to the antiferromagnetism vector $\mathbf{L}$, or it decreases in the case in which the field is parallel to this vector. Consequently, the exchange field reduces H_{c2} to the greatest extent near T_N, causing a decrease in the slope of the curve at $T \approx T_N$. The effect of magnetic scattering in compounds with an approximately spherical Fermi surface is the opposite; it weakens below T_N, causing an additional increase in H_{c2} here. The resultant curve $H_{c2}(T)$ depends on which of these two mechanisms is dominant.

In compounds with weak magnetic scattering, $\tau_s^{-1} \approx T_N \ll T_c$ (this situation prevails in $NdMo_6S_8$, $TmRh_4B_4$, and bct $ErRh_4B_4$), we can find a complete description of the dependence $H_{c2}(T)$, since in this case the only substantial effects are that of the exchange field and the orbital effect.

Under the assumption that the transition to the superconducting state is a second-order transition, we can write the following relation for the upper critical field in a dirty

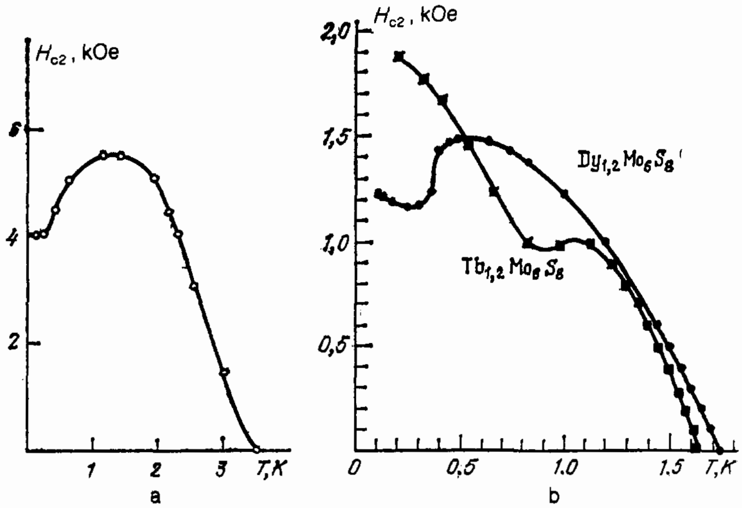

FIG. 2. Temperature dependence of the upper critical field in the antiferromagnetic superconductors (a) $NdMo_6S_8$ (Ref. 12) and (b) $Tb_{1.2}Mo_6S_8$ and $Dy_{1.2}Mo_6S_8$ (Ref. 11).

superconductor in the case $T_N < T < T_c$ (Ref. 31):

$$\ln\frac{T}{T_c}+\mathrm{Re}\left\{\psi\left[\frac{1}{2}+\frac{DeH}{2\pi T}(1+4\pi\chi_m)+\frac{ih_0\chi_m H}{2\pi T n\mu}\right]-\psi\left(\frac{1}{2}\right)\right\}=0, \qquad (15)$$

where D is the diffusion coefficient, and it is very important to note that χ_m is the susceptibility of the magnetic system along the field direction.

Near T_c, the behavior of the critical field is determined exclusively by the orbital effect, and we can easily determine the diffusion coefficient D from the slope of the $H_{c2}(T)$ curve. Also using the experimental value of H_{c2} at temperatures $T \ll T_c$ we can find the exchange field h_0 and plot the entire curve of $H_{c2}(T)$.

The anisotropy effects cause the susceptibility χ_m to depend on the field orientation even above T_N; as a result, the anisotropy is extremely significant both above and below T_N.

For the antiferromagnetic substance bct $ErRh_4B_4$, the corresponding dependence $H_{c2}(T)$ for three field directions and the values of the magnetization at $H = H_{c2}(T)$ are shown in Fig. 3 (Ref. 32). Figure 3a demonstrates the very strong anisotropy of H_{c2}, a consequence of the fact that the susceptibility χ_m differs by a factor of about seven along the [100] and [001] axes. It follows from measurements of the conductivity[32] that the very high residual resistance in this superconductor allows us to regard it as dirty.

After determining the diffusion coefficient D and the exchange field $h_0 \approx 20$ K from the dependence $H_{c2}(T)$ along the [001] axis, we can describe the behavior of the field $H_{c2}(T)$ along the [100] easy axis. In the temperature interval $0.5T_c < T < T_c$ there is good agreement with the experimental data. At $T < 0.5T_c$, however, the calculated critical field is lower than the experimental field. There is a good reason for this discrepancy: In a strong exchange field at these temperatures, there should be a first-order transition to a superconducting state.[31] It would be interesting to verify this conclusion experimentally in the case of bct $ErRh_4B_4$. In allowing for the possibility of a first-order transition, we can explain the experimental results of Ref. 32 without resorting to the assumption of an extremely strong spin-orbit scattering in this compound, as was done in Ref. 32.

Nearly all the antiferromagnetic superconductors which have been found so far are polycrystalline. This circumstance turns out to be of fundamental importance for a determination of H_{c2}: When the size of the grains exceeds ξ_0, the transition to the superconducting state near T_N is deter-

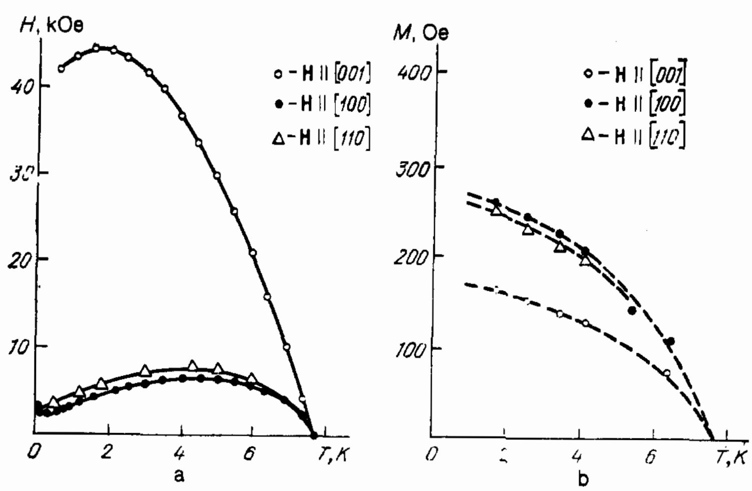

FIG. 3. a—Temperature dependence of the upper critical field in the antiferromagnetic single crystal of bct $ErRh_4B_4$ along three different crystallographic directions.[32]; b—magnetic moment at $H = H_{c2}$ for the same field orientations as in part a (Ref. 32).

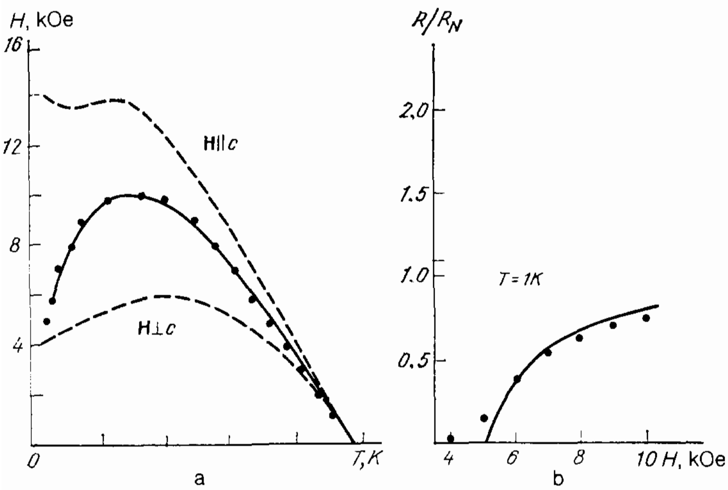

FIG. 4. Temperature dependence of the upper critical field H_{c2}, determined from the point at which the resistance was decreased by half, in polycrystalline $TmRh_4B_4$ (the division marks along the abscissa axis in part a are separated by 2 K; the points are experimental).[6] Dashed lines—Data from the measurements in Ref. 72 of the critical fields in a $TmRh_4B_4$ single crystal for various orientations of the magnetic field with respect to the c tetragonal axis.

mined in each grain by the particular field H_{c2} of that grain, which depends on the orientation of the grain[34,35] [expression (15), where χ_m is the magnetic susceptibility of the grain along the field direction]. The transition in the resistance is therefore highly blurred in principle, and the vanishing of the resistance should be determined by percolation effects.[33] The theory derived in Refs. 34 and 35 makes it possible not only to find the cure $H_{c2}(T)$ (in the approximation of a strong magnetic anisotropy of the easy-axis or easy-plane type) but also to calculate the dependence of the resistance of the sample on the magnetic field H {in the approximation of an effective medium,[33] we would have $R(H) = R_N[1 - 3C(H)]$, where R_N is the resistance in the normal state, and $C(H)$ is the concentration of the superconducting grains}.[6)]

Experimentally, H_{c2} is usually determined at the point at which the resistance has decreased by half. Figure 4a shows the results of corresponding calculations for H_{c2} (under the assumption of a pronounced easy-plane magnetic anisotropy), along with experimental data on $TmRh_4B_4$ polycrystalline samples.[6] The exchange field h_0 here is found to be $h_0 \approx 10$ K, giving us $\tau_s^{-1} < 0.1$ K. Figure 4b shows the theoretical and experimental dependence of the resistance on the field. Single crystals of $TmRh_4B_4$ have recently been successfully grown.[72] A preliminary study[72] of these crystals has revealed a sharp transition in a magnetic field and a pronounced anisotropy of H_{c2} corresponding to an easy-plane magnetic anisotropy. The results of the measurements[72] of H_{c2} are shown by the dashed lines in Fig. 4a.

For compounds with $\tau_s^{-1} \sim T_c$, magnetic scattering plays a fundamental role in the determination of H_{c2}. When the antiferromagnetic order appears, this scattering may either increase or decrease, as we have already mentioned, and it is not possible to predict the behavior of $H_{c2}(T)$. Figure 5 shows $H_{c2}(T)$ for[7] $SmRh_4B_4$. We see that the Néel temperature is the point of a change in slope of the $H_{c2}(T)$ curve. The temperature dependence of the Josephson critical current of $SmRh_4B_4$ junctions was measured in Ref. 36. The behavior found there is similar to the temperature dependence of the upper critical field. It can thus be definitely concluded that the superconducting order parameter increases below T_N because of a weakening of the magnetic scattering.

The important role played by exchange scattering in $SmRh_4B_4$ was demonstrated clearly in experiments[37] on irradiation of this compound. The observation of an exceedingly strong dependence of the superconducting transition temperature on the radiation dose was reported in Ref. 37. The apparent reason for this unusual suppression of superconductivity by ordinary defects is that by causing distortions of the crystal lattice the radiation locally increases the exchange interaction of electrons with magnetic ions[7)] (Ref. 37). We are thus dealing with a sort of superconductivity suppression due to an intensification of magnetic scattering with increasing number of defects. This suppression mechanism is extremely effective in $SmRh_4B_4$, where we have $\tau_s^{-1} \approx T_c$, but in e.g., $ErRh_4B_4$, where we have $\tau_s^{-1} \ll T_c$, it should play only a minor role. Indeed, the experimental data of Ref. 37 show that the decrease in T_c caused by irradiation in this compound is far smaller than that in $SmRh_4B_4$. The intensification of magnetic scattering with increasing disorder also explains the decrease in T_c with decreasing ratio of the residual resistance in various samples of sputtered $SmRh_4B_4$ films.[63]

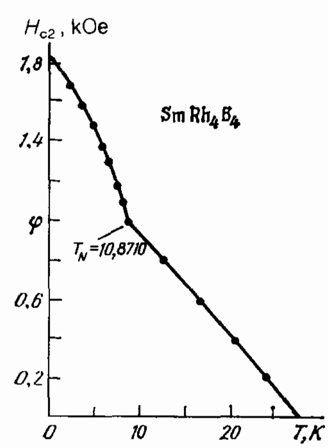

FIG. 5. Behavior of the upper critical field $H_{c2}(T)$ in $SmRh_4B_4$ (Ref. 7).

4. LOWER CRITICAL FIELD H_{c1} IN ANTIFERROMAGNETIC SUPERCONDUCTORS

The magnitude of the magnetization which arises in an antiferromagnetic material in a magnetic field is determined by the magnetic susceptibility $\chi_m(T)$ of the antiferromagnetic superconductor. When we incorporate the magnetization in the London equation describing the field distribution in the superconductor, we find a renormalization of the London penetration depth: $\lambda_L \to \lambda_L^{eff} = \lambda_L[1-4\pi\chi_m(T)]^{1/2}$. This effective decrease in the London depth, however, has essentially no effect on the lower critical field H_{c1}, for the appearance of vortices in antiferromagnetic superconductors (Ref. 4, for example):

$$H_{c1} = \frac{\Phi_0}{4\pi\lambda_L^2}\ln\frac{\lambda_L^{eff}}{\xi}, \qquad (16)$$

where $\Phi = \pi\hbar c/e$ is the quantum of magnetic flux. This expression differs from the usual expression in that λ_L has been replaced by its renormalized value only in the logarithm.

The antiferromagnetic ordering has essentially no effect on the structure of a vortex, except in the case in which the external magnetic field is directed along the antiferromagnetism vector, and a spin-flop transition involving a reversal of magnetic sublattices can occur. As Krzyszton has shown,[68,69] the nonuniformity of the magnetic field distribution in the vortex phase makes possible a spin-flop transition near the vortex core. The structure of the vortex becomes very nonuniform as a result. A sharp change in the magnetization at the spin-flop transition and also during metamagnetic transitions[66] can cause an abrupt change in the magnetization as a function of the field (if the field of the spin-flop transition is stronger than H_{c1}).

We have been discussing homogeneous antiferromagnetic superconductors. In real antiferromagnetic materials there are always defects in the magnetic structure, in particular, domain walls.[70] Domain walls may substantially reduce the critical field for the appearance of vortices at the walls, and under certain conditions there may be a spontaneous creation of vortices in the absence of an external magnetic field.[67]

We consider an antiferromagnetic structure of layers with magnetization vectors $\mathbf{M}_1$ and $\mathbf{M}_2 = -\mathbf{M}_1$ which alternate along the x axis (the vectors are directed along the z axis). A magnetic order of precisely this type has been observed in all the antiferromagnetic superconductors which have been studied by neutron diffraction. Figure 6 shows a domain wall in this structure. This wall separates two regions of the crystal with different signs of the antiferromagnetism vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$. The energy ($E_{VL}$) of the interaction of the magnetic field $B_z(x,y)$ which is localized at a domain wall of a superconducting vortex with local magnetic moments is written in the form

$$E_{VL} = 2\sum_{n=0}^{\infty}\sum_{m=-\infty}^{\infty}(-1)^n B_z\left(\frac{a}{2}+na,\ ma\right)M, \qquad (17)$$

where $M = |\mathbf{M}_1| = |\mathbf{M}_2| = L/2$. The vortex is directed along the z axis and is centered at $x = y = 0$; its magnetic field is $B = B_z = (\Phi_0/2\pi\lambda_L^2)K_0(\rho/\lambda_L)$ (Ref. 71, for example), where K_0 is the modified Bessel function and ρ is the coordinate in the x, y plane. Substituting this expression for the vortex field into (17), and switching from a summation to an integration, we find $E_{VL} = \Phi_0 aL/2\lambda_L$. The total energy of a vortex at the domain wall is therefore given by

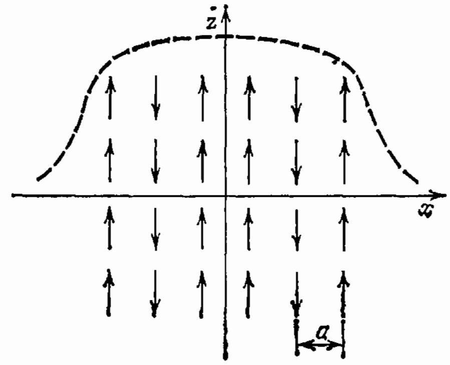

FIG. 6. Schematic diagram of a domain wall in an antiferromagnetic material (the x, z plane). The distance beween the magnetic atoms along the x direction is a. The dashes show the magnetic field of a vortex.

$$E_V = \left(\frac{\Phi_0}{4\pi\lambda_L}\right)^2\ln\frac{\lambda_L}{\xi} \pm \Phi_0\frac{aL}{2\lambda_L} - \Phi_0 H. \qquad (18)$$

The first term here corresponds to the intrinsic energy of the vortex; the last two terms describe the energy of the interaction of the vortex field with the localized magnetic moments and with the external field, respectively. The $\pm$ sign in (18) stems from the two possible orientations of the domain wall: The minus sign applies when the net moment of the wall is directed along the field H, while the plus sign applies when the net moment is directed opposite the field. The lower critical field for the appearance of a vortex at the domain wall, H_{c1}^*, is therefore

$$H_{c1}^* = H_{c1} - \frac{a}{2\lambda_L}L. \qquad (19)$$

If the condition $L > H_{c1}\cdot 2\lambda_L/a$ holds, vortices should appear at the domain wall even in the absence of an external field. In antiferromagnetic superconductors of the $REMo_6S_8$ type, we have $a \sim 5$ Å, $L(T=0) \approx 1-5$ kOe, and $H_{c1} \sim 10$ Oe (correspondingly, $\lambda_L \sim 3\cdot 10^{-5}$). Consequently, the condition for the appearance of self-induced vortices is $L > 10$ kOe. Since the quantity $L(0)$ is a few oersteds, self-induced vortices could be observed only in even dirtier compounds with H_{c1} of the order of several oersteds. Spontaneous creation of vortices at domain walls may have been observed in the experiments of Ref. 56, where a sharp increase in the captured flux was observed in bct $ErRh_4B_4$ as the temperature was lowered below the Néel point. An alternative explanation for this phenomenon, based on the assumption of weak ferromagnetism in this compound, is given in Section 7.

5. PSEUDOTERNARY COMPOUNDS

In addition to the stoichiometric ternary compounds, many alloys of these compounds have now been produced. It is possible to replace one rare earth element by another and also to replace the nonmagnetic atoms. Studies of pseudoternary compounds of this type make it possible to trace the

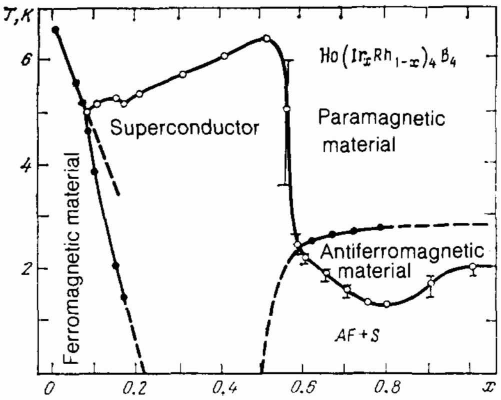

FIG. 7. Phase diagram of the pseudoternary compound $Ho(Ir_xRh_{1-x})_4B_4$ (Ref. 40). Filled circles—temperatures of the magnetic transition; open circles—temperatures of the superconducting transition.

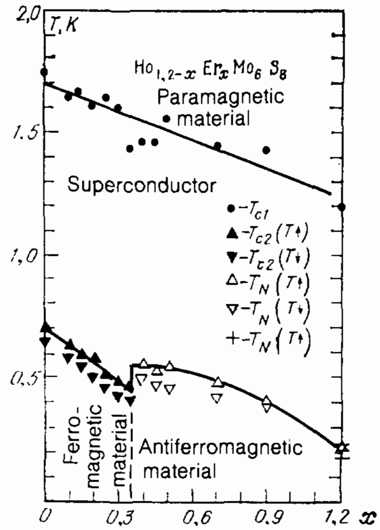

FIG. 8. Phase diagram of the compound $Ho_{1.2-x}Er_xMo_6S_8$ (Ref. 38).

transition from one type of magnetic order to another. In the compound $Ho_{1.2}Mo_6S_8$, for example, there is a transition to a ferromagnetic state with a disruption of the superconductivity at low temperatures,[1] while in $Tb_{1.2}Mo_6S_8$ the magnetic moments become ordered in an antiferromagnetic fashion without disrupting the superconductivity.[10] In the $Ho_{1.2-x}Tb_xMo_6S_8$, on the other hand, the transition from one type of magnetic order to another has been followed.[38]

A study of the pseudoternary compound $Ho(Ir_xRh_{1-x})_4B_4$ over the concentration range $x > 0.6$ revealed the appearance of an antiferromagnetic order at a temperature slightly above the superconducting transition temperature.[39] The phase diagram of this system is shown in Fig. 7. A transition to an antiferromagnetic state with $T_N > T_c$ also occurs in $Tb(Ir_xRh_{1-x})_4B_4$ at $x > 0.2$ (Ref. 40). In all these cases, in accordance with the discussion of the role of magnetic scattering in Section 2, the Néel temperature is only slightly above the critical temperature.

A complex magnetic order also occurs in the pseudoternary compound[62] $Dy(Ru_xRh_{1-x})_4B_4$. At concentrations $x \lesssim 0.35$, coexistence of superconductivity and a long-range magnetic order of an antiferromagnetic type has been observed. According to data on neutron scattering,[64] the antiferromagnetism vector is modulated with a wave vector $q = 0.133$ Å. A "complex" antiferromagnetic order of this type has also been observed through neutron scattering in the superconducting antiferromagnetic substances $NdRh_4B_4$ (Ref. 5) and $TmRh_4B_4$ (Ref. 65). The reason for the onset of modulation of the antiferromagnetic order in these compounds is not yet clear. A possibility is a slight ferromagnetism (Section 6). Evidence for the hypothesis of a slight ferromagnetism comes from the detection, in neutron-scattering experiments,[64] of antiferromagnetic order in addition to a ferromagnetic component in nonsuperconducting $DyRh_4B_4$; i.e., the long-range magnetic order in $DyRh_4B_4$ is complex, reminiscent of the situation in weak ferromagnetic materials.

An extremely unusual sequence of phase transitions has been observed[38] in the pseudoternary system $Ho_{1.2-x}Er_xMo_6S_8$ at concentrations from $x = 0.2$ to $x = 0.35$. As we have already mentioned, the compound $HoMo_6S_8$ is a superconducting ferromagnetic substance,[1] in which a long-period oscillatory magnetic structure[4] arises near $T_M \approx 0.7$ K as the temperature is lowered. A further lowering of the temperature disrupts the superconductivity, and below $T_{c2} \approx 0.6$ K the compound becomes a normal ferromagnetic substance. The compound $ErMo_6S_8$ is a typical antiferromagnetic superconductor. With increasing x in $Ho_{1.2-x}Er_xMo_6S_8$, there should accordingly be a replacement of ferromagnetic order by an antiferromagnetic one. The corresponding phase diagram, obtained in Ref. 38, is shown in Fig. 8. A remarkable circumstance, however, is that at concentrations $0.2 < x < 0.35$, i.e., when there is a transition to a normal ferromagnetic state in the system as the temperature is lowered, a transition back from the normal ferromagnetic state to a superconducting state is observed[38] at a temperature below $T_{c3} \approx 100$ mK. This transition occurs over a long time, of the order of several hours. Brossard *et al.*[38] attribute this restoration of superconductivity to a change in the nature of the order at T_{c3}, from ferromagnetic to antiferromagnetic. Unfortunately, at the moment we have no information of any sort on the nature of the magnetic order in the low-temperature superconducting state.

There is an alternative possibility for explaining this phenomenon. In principle, magnetic holmium and erbium ions could differ in the sign of the exchange integral for their interaction with electrons. As a result, the total exchange field would be $h = h_0 - |h_1|$ where h_0 and h_1 are the exchange fields exerted on electrons by the holmium and erbium ions, and where we have $h_1 < 0$. There would thus be a possible cancellation of the exchange fields of the different magnetic ions[41]: a sort of analog of the Jaccarino-Peter effect[42] (i.e., the paramagnetic effect of the external magnetic field is cancelled by the exchange field of the magnetic ions). According to this interpretation, the ordering of erbium ions below 100 mK reduces the exchange field to a level at which in the superconducting state there is either a domain phase[4] or a vortex state (Section 5). Measurements of the upper critical field might be of assistance in testing this hypothesis. The addition of magnetic ions with an exchange integral I of the other sign should raise the upper critical field near T_M, where this field is determined primarily by the paramagnetic effect.

In antiferromagnetic superconductors, there is the further possibility of an increase in the critical fields upon the addition of an impurity of magnetic ions with an exchange integral of the opposite sign. With increasing impurity concentration, the increase in the critical field may give way to a decrease because of an "overcancellation." This change in the critical fields upon the introduction of a magnetic impurity should be seen most clearly at low temperatures $T \ll T_c$.

We also wish to point out that the addition of a small amount of a magnetic impurity with a very strong exchange interaction $I_1 \gg I_0$ (I_0 is the exchange integral of the host compound), might substantially increase the magnetic-scattering parameter, causing τ_s^{-1} to approach T_c, without causing any important change in the exchange field ($h_0 \gg h_1$). In this connection it would be interesting to carry out a corresponding study with $HoMo_6Se_8$, where the domain phase of the coexistence of superconductivity and magnetism is stable down to $T = 0$. The addition of a magnetic impurity might weaken the Cooper pairing and change the characteristics of the coexistence phase (e.g., reduce its wave vector). At a sufficiently high concentration of the magnetic impurity there is the possibility of a transition from an inhomogeneous magnetic superconducting state to a ferromagnetic normal state.

The Jaccarino-Peter effect is manifested in an extremely unusual way in the behavior of the compound $Eu_{0.75}Sn_{0.25}Mo_6S_{7.2}$ in an external magnetic field.[43] The strong orbital critical field in this compound makes it essential to take into account the paramagnetic effect, while the presence of localized Eu magnetic moments leads to the possibility of a cancellation of this paramagnetic effect. In a magnetic field, the Eu moments orient along the field; by virtue of the exchange interaction with electrons, the result is a destruction of superconductivity. A further increase in the magnetic field has only a slight effect on the exchange field of the Eu atoms, since their moment tends toward saturation. One needs to keep in mind, however, the direct effect of the field on the electron spin; the total effective field acting on the spins, $h = \mu_B H + I\langle J\rangle$, decreases in the case $I < 0$. The result is a curious situation: Superconductivity is restored when the field is increased. Superconductivity is destroyed again at fields at which the orbital effect becomes important or there is an "overcancellation" of the total exchange field. The resulting phase diagram of this compound is extremely unusual and is shown in Fig.9. In the absence of a magnetic field, at low temperatures, an antiferromagnetic order of europium ions with $T_N \approx 0.5$ K apparently occurs.[43] The initial ternary compound $EuMo_6S_8$ goes superconducting only at pressures above 13 kbar (Ref. 44).

An even more surprising effect was discovered just recently in the heavy-fermion compound $CePb_3$ (Ref. 73). In the absence of a magnetic field, an antiferromagnetic transition occurs in this compound at $T_N \approx 1$ K, and there is no superconductivity down to extremely low temperatures. A strong magnetic field $H \sim 140$ kOe suppresses the antiferromagnetic transition and—strange as it may seem—induces superconductivity at $T < 0.2$ K. Lin *et al.*[73] attribute this behavior to a "freezing" of the magnetic scattering in the field, and the destructive effect of the magnetic field on superconductivity is offset by the exchange field of the Ce atoms ($I < 0$) by virtue of the Jaccarino-Peter effect. The orbital effect, on the other hand, is strongly suppressed in this case because of the large effective electron mass,[73] $m^* > 200\, m_0$ (m_0 is the mass of a free electron).

6. POSSIBLE PHASES OF COEXISTENCE IN SUPERCONDUCTING WEAK FERROMAGNETIC MATERIALS

While the moments of the magnetic lattices in ordinary antiferromagnetic materials cancel out exactly, the situation in a weak ferromagnetic material is different: The relativistic interaction distorts the original antiferromagnetic structure slightly, with the result that a weak ferromagnetic moment, of the order of 10^{-2}–10^{-3} of the nominal value, arises.[45] In the case of, say, a two-sublattice antiferromagnetic material the onset of weak ferromagnetism is described in the free-energy functional by an additional Dzyaloshinskiĭ invariant $\mathbf{D}[\mathbf{M}_1\mathbf{M}_2]$, where $\mathbf{D}$ is a vector, and $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations. A term of this sort in the energy is at a minimum when the magnetic sublattices are oriented at right angles to each other, but this tendency disrupts the collinearity of the original antiferromagnetic structure, which is maintained by the strong exchange interaction. As a compromise, the sublattices become tilted, and a slight ferromagnetic moment $M \sim \beta L$ arises, where L is the antiferromagnetic moment, and we have $\beta \ll 1$ because of the relative weakness of the relativistic interaction responsible for the weak ferromagnetism.

A weak ferromagnetism is rather common among antiferromagnetic substances, so that there is every reason to expect to find it also in antiferromagnetic superconductors.

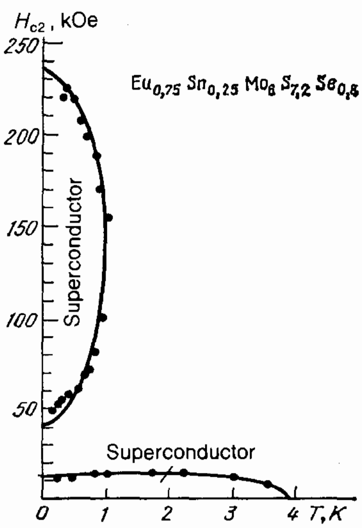

FIG. 9. *H, T* phase diagram of the compound[43] $Eu_{0.75}Sn_{0.25}Mo_6S_{7.2}Se_{0.8}$. The unusual shape of this diagram is a consequence of the cancellation of the paramagnetic effect by the exchange field produced by Eu atoms.

Perhaps a promising candidate is the body-centered tetragonal (bct) phase of $ErRh_4B_4$ (Ref. 46).

In the case of ferromagnetic superconductors, essentially the only possible phase in which a superconductivity would coexist with a long-range magnetic order is a phase with a domain magnetic structure,[4] whose parameters are determined for the most part by the exchange interaction of the conduction electrons and localized moments. In contrast with this situation, other coexistence phases are also possible for weak ferromagnetic substances: a phase with spontaneous vortices and a Meissner superconducting phase,[46] which were predicted earlier for ferromagnetic superconductors in the model of a purely electromagnetic interaction.[47–50]

Our analysis of the possible phases depending on the parameters of the system of electrons and localized moments is based on the free-energy functional corresponding to the symmetry (bct) of the $ErRh_4B_4$ phase,[46] but it reflects the particular features of all weak ferromagnetic materials, and the conclusions are of a general nature.

We assume that the antiferromagnetism vector **L** is directed along the y axis, while the ferromagnetism vector is directed along the x axis. The magnetic functional is the

$$F_m = \int d^3r \left\{ n\theta_{ex} \left[\frac{1}{2} A l^2 + \frac{1}{4} C (\mathbf{l}^2) + \frac{b}{2} \mathbf{m}^2 + \beta m_x l_y + a^2 (\nabla \mathbf{l})^2 \right] + \frac{1}{8\pi} B^2 - \mathbf{B}\mathbf{M} + 2\pi \mathbf{M}^2 \right\}, \quad (20)$$

where **l** and **m** are the vectors **L** and **M** normalized to the maximum magnetization, $M(0) = \mu n$ (n is the concentration of magnetic atoms); i.e., we have $l = L/M(0)$ and $\mathbf{m} = \mathbf{M}/M(0)$. The energy parameter θ_{ex} is of the order of T_N; the magnetic induction is $\mathbf{B} = \mathrm{curl}\,\mathbf{A}$; the parameters b and C are of order unity; and we have $\beta \ll 1$.

In the absence of superconductivity, (16) describes a weak ferromagnetic material with a magnetization $m = \beta l/b$, which appears along with l at the point T_N. Because of the small value of β, $\sim 10^{-2}$–10^{-3}, the ferromagnetic moment may not disrupt superconductivity. The reason is that the constant exchange field is lower than T_c, and the maximum induction $B_{max} = 4\pi M$, does not exceed the upper critical field H_{c2}. The interaction of superconductivity and magnetism in a weak ferromagnetic material near T_N can be described by perturbation theory. Below T_N, the interaction of superconductivity with the exchange field can always be described by perturbation theory, but the magnetic induction may in general exceed H_{c1}, and, if so, its effect on superconductivity will not be small. In such a situation, a phase with spontaneous vortices—a VS phase (more on this below)—becomes possible.

Superconducting pairing screens the long-wave part of the exchange and electromagnetic interactions, while having essentially no effect on their short-wave parts with $q > \xi_0^{-1}$. We therefore include in the functional F_{int}, which describes the interaction of superconductivity and magnetism, the difference between the energies of the long-range interactions in the superconducting and normal phases:

$$F_{int} = \sum_q \left[\left(\frac{1}{8\pi} \mathbf{B_q B_{-q}} - \mathbf{B_q M_{-q}} + 2\pi \mathbf{M_q M_{-q}} \right) n^{-1} + \frac{1}{2} Q_s(q) \mathbf{A_q A_{-q}} + \theta_{ex} \frac{\chi_n(q) - \chi_s(q)}{\chi_n(0)} \mathbf{m_q m_{-q}} \right],$$

$$\mathbf{M_q} = n\mu \mathbf{m_q}, \quad (21)$$

where $Q_s(q)$ is the superconducting electromagnetic kernel [in the London limit ($q_1 \ll \xi_0^{-1}$) we have $Q_s \sim \lambda_L^{-2}$ and $Q_s \sim 1/(\lambda_L^2 q\xi_0)$ at $q \gg \xi_0^{-1}$].

The complete functional of the system is $F = F_m + F_s + F_{int}$, where F_s is the superconducting functional (in the absence of localized moments). It has the usual form and is unimportant to the discussion below.

Minimizing the complete functional with respect to B and m, we find the free energy fuctional as a function of the antiferromagnetic moment $l_q \equiv l_{y,q}$ for the transverse structure:

$$F(l_q) = \theta_{ex} \sum_q \left\{ \frac{1}{2} A - \frac{\beta^2}{2B} + a^2 q^2 + \frac{\beta^2 F(q)}{2b[b + F(q)]} \right\} \mathbf{l_q l_{-q}},$$

$$F(q) = 1 - \frac{\chi_s(q)}{\chi_n(0)} + \frac{4\pi\theta_{em} Q_s(q)}{\theta_{ex}[q^2 + 4\pi Q_s(q)]}, \quad (22)$$

where $\theta_{em} = 2\pi\mu^2 n$ is the energy parameter of the magnetic dipole interaction of the localized moments. Like θ_{ex}, it is of the order of 1 K in ternary compounds.

We first consider the case in which β is not very small, and the condition $ab/\beta \ll \xi_0$ holds. Below T_N, a sinusoidal transverse structure $l_y \sim \sin Qr$ with a wave vector $Q \sim (a^2\xi_0)^{-1/3} \beta^{2/3}$ then appears if $\theta_{em}/\theta_{ex} (Q\lambda_L)^2 \ll 1$.

During cooling l and m increase, and the magnetic structure converts into the domain structure shown in Fig. 10a. The directions of **m** in neighboring domains are opposite (the same is true of the direction of **l**). With further cooling, it may turn out that the domain structure is unfavorable from the energy standpoint because of the large energy of the domain walls. In this event we will observe a transition to a structure with spontaneous vortices, shown in Fig. 10b.

As β decreases, Q becomes so small that the exchange interaction gives way to an electromagnetic interaction as the dominant interaction in the formation of the magnetic structure of the coexistence phase. The magnetic structure is then again a domain structure with a wave vector $Q \approx (a\lambda_L)^{-1/2} \beta^{1/2}$. During cooling, the structure may again go into a vortex phase.

Finally, at a very small value of β, we run into a situation with $ab/\beta \gtrsim \lambda_L$. The magnetically modulated phase is then unfavorable, and below T_N there is the Meissner phase shown in Fig. 10b. This phase could not occur in ferromagnetic superconductors, with $\lambda_L, \xi_0 \gg a$. During cooling, the Meissner phase converts into a vortex phase if the magnetic induction $B = 4\pi M$ exceeds H_{c1}. The complete phase diagram of weak ferromagnetic superconductors is shown schematically in Fig. 11.

In order to draw conclusions regarding the presence of weak ferromagnetism in the bct phase of $ErRh_4B_4$ on the basis of symmetry considerations, we need information on its magnetic structure. So far, no neutron-scattering experiments have been carried out on this compound, and we have no direct information on its magnetic structure. The only measurements which have been carried out have been mea-

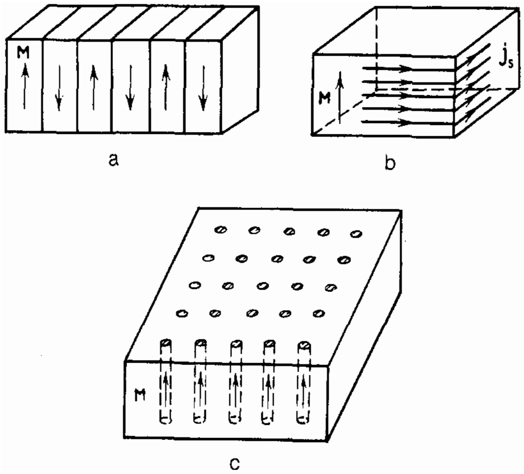

FIG. 10. a: Domain structure of weak ferromagnetic materials in the superconducting state (DS phase); the directions of the antiferromagnetism vector **L** and of the ferromagnetic vector **M** in adjacent domains are opposite; the wave vector of the structure is perpendicular to **M**; the superconducting order parameter is essentially uniform over the sample. b: Weak ferromagnetic Meissner superconducting state (FS); the superconducting current, flowing along the boundary of the sample in a layer of the order of λ_L in thickness, screens the surface current associated with the jump in the ferromagnetic moment at the boundary; the magnetic induction vanishes inside the sample. c: Superconducting phase with spontaneous vortices (VS) caused by the presence of weak ferromagnetism; in this phase, the ferromagnetic moment is nearly uniform, the induction and the superconducting currents are very nonuniform; and the superconductivity is destroyed inside the vortex core.

surements of the magnetization along various crystallographic directions in single crystals of the bct phase of $BrRh_4B_4$ below the Néel point.[66] These results are evidence of a complex antiferromagnetic structure in this crystal. As noted earlier, as the temperature was lowered below the Néel point in the bct phase of $ErRh_4B_4$, a sharp increase was observed in the trapped flux. This effect may have been due to the appearance of a self-induced vortex phase in this compound as a result of weak ferromagnetism.

To conclude this section we wish to point out an interesting possibility for observing the magnetic structures described above in superconducting antiferromagnets whose symmetry allows piezomagnetism. The application of pressure to such a material causes a spontaneous ferromagnetic moment (Ref. 45, for example), since the free energy has mixed invariants $l_i m_k$ with coefficients proportional to the pressure. In the presence of superconductivity, the situation becomes completely analogous to that described above, with the coefficient β proportional to the pressure. By varying the pressure in the superconducting piezomagnet one could in principle generate the entire phase diagram in Fig. 11.

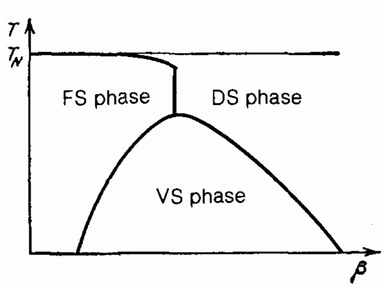

FIG. 11. Schematic phase diagram of a type II superconductor with weak ferromagnetism.

7. SPIN WAVES IN ANTIFERROMAGNETIC SUPERCONDUCTORS

As mentioned earlier, superconductivity has no appreciable effect on the magnetic structure of antiferromagnetic substances, and we would thus be led to expect that the magnetic properties of antiferromagnetic superconductors would differ only slightly from the properties of normal antiferromagnetic materials. Indeed,this turns out to be the case, with one exception: spin waves. In a spin wave, when there is a deviation of the localized moments from their equilibrium positions, a ferromagnetic moment arises; in the case of a long-wave excitation, this moment interacts strongly with superconductivity. As a result, the spin-wave spectrum in an antiferromagnetic superconductor should be of a fundamentally different nature than in an ordinary antiferromagnetic material.[51]

In examining the spectrum of spin waves, we will for clarity restrict the discussion to the case of a two-sublattice antiferromagnetic material with sublattice magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$. Furthermore, since the specific behavior of the spin-wave spectrum is related to the long-wave region ($q \ll a^{-1}$), the magnetic subsystem can be described by the functional (20) with $\beta = 0$, and (21) can be used as $F_{\rm int}$.

Transforming to the complete functional, which contains only the quantities **m** and $\delta\mathbf{l}$ (the deviations of the vectors **M** and **L** from their equilibrium values 0 and $\mathbf{L}_0$, respectively), we find, in the Fourier representation,

$$F = \theta_{\rm ex} \sum_{\mathbf{q}} \Big\{ \Big(\frac{1}{2} D + a^2 q^2 \Big) |\delta \mathbf{l}_{\mathbf{q}}|^2 + \Big[\frac{1}{2} b + 1 - \frac{\chi_s(q)}{\chi_n(0)} + \tilde{\beta} \cdot 4\pi Q_s(q) \times (q^2 + 4\pi Q_s(q))^{-1} \Big] |\mathbf{m}_{\mathbf{q}}|^2 \Big\}, \qquad (23)$$

where $\tilde{\beta} = \theta_{\rm em} / \theta_{\rm ex}$ and where we are assuming the anisotropy to be uniaxial (all the antiferromagnetic superconductors which have been studied by neutron scattering have a uniaxial anisotropy). The parameter D here is a measure of the anisotropy ($D > 0$) of the easy-axis type. Functional (23) deviates from the ordinary functional in that the coefficient of $|\mathbf{m}_{\mathbf{q}}|^2$ is a strong function (at the scales of λ_L^{-1} and ξ_0^{-1}) of the wave vector. Writing the equations of motion for the vectors **m** and **l**, and solving them, we find the spin-wave spectrum:

$$\omega^2 = \gamma^2 (D + a^2 q^2) \Big[b + 2 \Big(1 - \frac{\chi_s(q)}{\chi_n(0)} \Big) + \frac{8\pi\tilde{\beta} Q_s(q)}{q^2 + 4\pi Q_s(q)} \Big], \qquad (24)$$

where $\gamma = g\mu_B L_0/2$. The gap in the spin-wave spectrum at $q = 0$ is $\omega_s(0) = [\gamma D(2 + b + 2\tilde{\beta})]^{1/2}$ and differs from the corresponding value in the absence of superconductivity by a factor $[1 + 2b^{-1}(1 + \tilde{\beta})]^{1/2}$, where the second term in brackets is of order unity. Superconductivity thus increases the size of the gap in the spin-wave spectrum (the antiferromagnetic resonance frequency) by an amount of the order of its value in the normal phase. The expression here for the

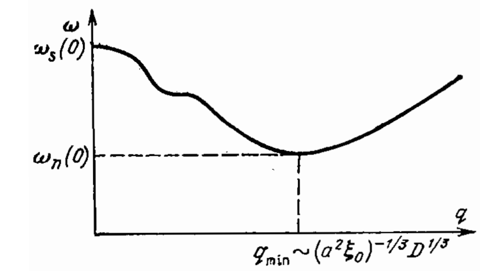

FIG. 12. Typical shape of the spin-wave spectrum in an antiferromagnetic superconductor.

spin-wave spectrum, (24), differs from the ordinary expression in the strong q dependence of ω at the scale values $q\sim\xi_0^{-1}$ and λ_L^{-1} and the presence of a minimum (or possibly two minima) on the $\omega(q)$ curve (Fig. 12). At the minimum, the value of the frequency is essentially equal to the antiferromagnetic resonance frequency in the normal phase, $\omega_{min}\approx\omega_n(0)$, and the minimum is reached at the value $q_{min}\approx(a^2\xi_0)^{-1/3}D^{1/3}$ of the wave vector. A study of the spin-wave spectrum in antiferromagnetic superconductors therefore might yield a rich store of information on both the anisotropy of the magnetic system and the superconducting and magnetic characteristics of the system. Inelastic neutron scattering yields information on primarily the nature of the spectrum $\omega(q)$ at $q\gg q_{min}$, on which superconductivity has essentially no effect. On the other hand, rf methods should "see" a gap $\omega_s(0)=\omega_n(0)[1+2b^{-1}(1+\tilde{\beta})]^{1/2}$, since the typical wave vectors here are much smaller than q_{min}. An increase in the external field from H_{c1} to H_{c2}, i.e., a destruction of the superconductivity should be quite evident in this case from a change in the shape of the long-wave region in the dispersion $\omega(q)$. So far, unfortunately, there has been no experimental study of the spin-wave spectra of superconducting antiferromagnetic materials.

We conclude this section of the paper by noting that there is also the possibility of an inverse effect of spin waves in antiferromagnetic materials on superconductivity: The spin waves weaken the Cooper pairing in the singlet state and may intensify triplet pairing.[52] So far, there are no indications of any sort for a significant interaction of electrons through spin waves.

8. SUPERCONDUCTIVITY AND COLLECTIVIZED MAGNETISM

The discussion above has dealt with antiferromagnetic superconductors with localized magnetic moments (magnetic atoms). In contrast, compounds have now been discovered in which a band (collectivized) magnetism coexists with superconductivity.

These compounds are Y_9Co_7, in which the appearance of a band magnetism ($T_m\approx 6$ K), according to measurements of the magnetic susceptibility and the magnetization, precedes a superconducting transition[53] ($T_c\approx 3$ K); the compounds $LaRh_2Si_2$ and YRh_2Si_2, with $T_N\approx 7$ K and with a transition to the superconducting state at[54] $T_c\approx$3–4 K; and the $Cr_{1-x}Re_x$ alloys.[55] In the latter case, at $x>x_c=0.16$, susceptibility and NMR measurements[55] indicate the appearance of a band antiferromagnetism at $T_N\approx 160$ K, well above the temperature of the superconducting transition, $T_c\approx 3$ K. Interestingly, as x is raised from 0 to x_c, a sharp decrease in T_N (from 500–600 K to 160 K) is observed near x_c, and only at $x>x_c$ does superconductivity appear.

Chaussi *et al.*[56] believe that their measurements of the magnetization in the bct phase of $ErRh_4B_4$ indicate the appearance of band ferromagnetism at a temperature of about 20 K, i.e., above $T_c=7.7$ K, and a Néel temperature $T_N\approx 0.7$ K, at which a magnetic ordering of erbium ions occurs.

Furthermore, the coexistence of superconductivity and a magnetic order of the spin-density-wave type might in principle be observed in organic compounds also.[22,62]

At present there is too little information on superconducting band magnetic materials to determine the reasons for the occurrence of band magnetism and the mechanisms for its interaction with superconductivity. It is, on the other hand, clear that there are important differences between systems with superconductivity and with band magnetism, on the one hand, and superconducting compounds with localized moments, on the other. In the first place, the same band appears to be responsible for both the superconducting and magnetic properties in the former systems, while the bands which determine superconductivity and magnetism in the latter are quite different: The Cooper pairing which occurs in ternary compounds involves the s- and d-bands, while magnetism involves the f-band. Furthermore, the energies of superconductivity and of the band magnetism are determined in order of magnitude by similar expressions, $T_c^2N(0)$ and $T_N^2N(0)$, since both effects involve only a small part of the band near the Fermi surface, of the order of $T_c/\mathscr{E}_F$ and $T_N/\mathscr{E}_F$, respectively. This is the situation, for example, in systems with a magnetic order of the spin-density-wave type, which arises from the circumstance that different parts of the Fermi surface can be brought into coincidence through translation by a vector Q, which determines the wave vector of the magnetization wave.[22,62] The relation between the energies in this case depends on the relation between T_c and T_N. In systems with localized moments, the magnetic energy, of the order of T_N, is generally much larger than the superconducting condensation energy, even if $T_N<T_c$. The interaction mechanism here allows the coexistence of superconductivity and antiferromagnetism under the condition $T_N<T_c$ or $T_N\approx T_c$. However, the competition between band magnetism and superconductivity is not determined exclusively by their relative energies. In principle, there could be a situation in which the two occur on different parts of the Fermi surface, in which case the relation between T_c and T_N could be arbitrary. This circumstance and also the absence of detailed information on the geometry of the Fermi surface in systems with band magnetism hinder the derivation of a systematic theory for the coexistence of superconductivity and band magnetism.

A theoretical approach to the description of band superconducting antiferromagnetic substances which has been proposed in several studies[57–59] is based on Bilbro and McMillan's theory,[60] in which the interaction between two types of transitions results exclusively from the competition for the Fermi surface. This theory[60] was derived for, and used successfully for, a description of the coexistence of superconductivity and a charge density wave. In the case of band antiferromagnetism, on the other hand, an exchange mechanism for an interaction between superconductivity and magnetism and also a scattering of electrons by spin excitations might play an important role.

9. CONCLUSION

The mutual effects of magnetism and superconductivity in antiferromagnetic superconductors are not as obvious as in ferromagnetic substances. Nevertheless, superconductivity in these compounds has many distinctive features: the appearance of a gapless situation, a strong effect of ordinary impurities on the superconducting characteristics below the Néel point, an unusual behavior of the upper critical field near the point of antiferromagnetic transition, and a change in the nature of the transition to the superconducting state, from second-order to first-order. In turn, superconductivity should lead to an unusual spectrum of spin waves in antiferromagnetic superconductors. Finally, very interesting and varied phases of coexistence of superconductivity and magnetism might be studied in weak ferromagnetic materials if the corresponding compounds were produced. There is accordingly the hope that research on antiferromagnetic superconductors will lead to a better understanding of both the behavior of superconductors in strong exchange fields and the effect of superconductivity on magnetism.

We wish to thank S. S. Krotov, M. L. Kulich, and S. V. Panyukov for a useful discussion of these questions.

[1)]An exceptional case is $Tm_2Fe_3Si_5$, in which antiferromagnetic ordering below $T_N \approx 1.1$ K destroys superconductivity.[74] On the other hand, superconductivity exists in this compound only at pressures above 3 kbar; the critical temperature reaches a maximum $T_c \approx 3$ K at $P \approx 9$ kbar and then falls off rapidly with increasing pressure.[74] This anomalously strong pressure dependence of the critical temperature suggests that superconductivity is destroyed in $Tm_2Fe_3Si_5$ not by the antiferromagnetic transition itself but by the striction effects which accompany it.

[2)]A spatial variation of the order parameter has been taken into account in several studies.[15,21] The corresponding Fourier components of this parameter, Δ_{2nQ} ($n = 1, 2, \ldots$), however, are small, of the order of $h_Q/v_F Q$ with respect to Δ. The incorporation of these components leads to corrections of order $(h_Q/\mathscr{E}_F)^2$ in the superconducting characteristics. It is this circumstance which makes the approach of Refs. 14 and 20, where only uniform pairing was considered, essentially equivalent to the approach of Refs. 15 and 21, where nonuniform components of the superconducting order parameter were also taken into account.

[3)]If an exchange field with a wave vector **Q** gives rise to a gap over large regions of the Fermi surface, which can be brought into coincidence by translation by a vector **Q**, the effect of the antiferromagnetism on the superconductivity may be much stronger.[18] In the limiting case of a system with a one-dimensional spectrum, the compound becomes an insulator below the point of the magnetic transition, and there is no superconducting transition. It is this situation which frequently prevails in the family of Bechgord quasi-one-dimensional organic salts.[22,62]

[4)]Spin waves also contribute to the heat capacity, but the contribution is usually exponentially small because of the gap in the spin-wave spectrum (Section 7 below). The size of this gap in ternary compounds can be estimated to be about 1 K.

[5)]In the coexistence phase of ferromagnetic superconductors, this situation does not occur, since a normal ferromagnetic phase is preferred from the energy standpoint under the condition $\tau_s \Delta < 1$.

[6)]Interestingly, while the vanishing of the resistance is determined by percolation along superconducting regions, the behavior of the thermal conductivity in a field should be characterized by percolation along normal regions (since at $T \ll T_c$ a superconductor is essentially a thermal insulator, and the normal regions are primarily responsible for the thermal conductivity). The behavior of the thermal conductivity and that of the resistance in a field in a polycrystalline magnetic superconductor should therefore be intimately related. This question has yet to be studied experimentally.

[7)]In ternary superconducting compounds, the magnitudes of the exchange integrals are exceptionally small (the typical value of the exchange field in these compounds is $\sim$10–50 K, while in ordinary magnetic materials it is of the order of 500–1000 K). The reason is a pronounced spatial separation of conduction electrons, which move primarily along Rh_4B_4 or Mo_6S_8 clusters, and magnetic rare earth ions, which lie between clusters. A cluster structure of this sort also makes possible a coexistence of superconductivity and magnetism.[4] In disordered samples, the magnetic ions evidently approach clusters.

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