

Magnetic structures in the superconductivity–weak-ferromagnetism coexistence phase

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Magnetic superconductors whose symmetry admits of the existence of weak ferromagnetism in the normal state are considered. It is shown that the superconducting state below the T_N of such superconductors can exhibit either a phase with an inhomogeneous antiferromagnetic and a weak ferromagnetic moment of the domain structure type or a Meissner phase with a weak homogeneous ferromagnetic moment. The type of ordering is determined by the magnitude of the Dzyaloshinskii–Moriya interaction constant β , the homogeneous phase occurring at small values of β . On cooling these phases may be replaced by a self-induced vortex structure. A phase diagram of the states in the (T, β) plane is constructed. The possibility of explaining the modulation, revealed by neutron diffraction, of the antiferromagnetic moment in NdRh_4B_4 in the basis of the weak-ferromagnetism concept is discussed.

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1. INTRODUCTION

Since the discovery in 1976 of the coexistence of superconductivity and magnetic order in the stoichiometric compounds REMo_6S_8 , where $\text{RE} = \text{DY, Gd, and Tb}$ (Ref. 1), substantial progress has been made in both the synthesis of new magnetic superconductors and the construction of the theory of this phenomenon. We now have about one and a half dozen magnetic superconductors, and their number is still increasing rapidly. With the exception of the two compounds ErRh_4B_4 and HoMo_6S_8 , the magnetic superconductors exhibit an antiferromagnetic ordering of localized magnetic moments that has only a slight effect on the superconductivity.

In ErRh_4B_4 and HoMo_6S_8 in the superconducting phase there occurs at a temperature $T = T_m < T_{c1}$ an inhomogeneous magnetic ordering with characteristic wave vector $Q \sim 0.06 \text{ \AA}^{-1}$ for ErRh_4B_4 and 0.03 \AA^{-1} for HoMo_6S_8 , followed at $T = T_{c2}$, when the temperature is lowered further, by a first-order transition into the ferromagnetic (FN) state. In this connection it has been suggested that these compounds would, in the absence of superconductivity, exhibit a normal ferromagnetic transition, and that the superconductivity is the cause of the inhomogeneous magnetic ordering in the temperature region $T_{c2} < T < T_m$.

Such behavior is due to the mutual antagonism between the superconducting order and the ferromagnetic order. The antagonism between these two types of order was first noted by Ginzburg,² who postulated that the electromagnetic interaction between the localized moments is responsible for the magnetic ordering. Later, Anderson and Suhl³ considered the problem of the coexistence of ferromagnetism and superconductivity in the case when the conduction-electron-mediated indirect exchange interaction between the localized moments (the RKKY interaction) leads to the ferromagnetic ordering of the moments in the absence of superconductivity. They showed that the Cooper pairing of the electrons transforms the interaction between the localized moments in such a way that there appears at the mag-

netic transition point an inhomogeneous magnetic order with wave vector $Q \sim (a^2 \xi_0)^{-1/3}$, where ξ_0 is the superconducting correlation length and a is the magnetic hardness, which is of the order of the interatomic distance. Blount and Varma,⁴ as well as Matsumoto, Umezawa, and Tachiki⁵ arrived at a similar conclusion in investigations of the electromagnetic (EM) (magnetic dipole) interaction between the localized moments in the presence of superconductivity, but they found that the inhomogeneous magnetic structure at the magnetic-transition point has in this case a wave vector $Q \sim (a \lambda_L)^{-1/2}$ where λ_L is the London penetration depth.

In Ref. 6 it is shown that the magnetic anisotropy plays an important role in the formation of the inhomogeneous magnetic structure in the superconducting phase below the magnetic-transition point. In this temperature region a domain-type structure with $Q \sim (a \xi_0)^{-1/2}$ is realized, and the effect of the electromagnetic interaction, for all really conceivable relations between the exchange and the magnetic-dipole interactions, manifests itself only in the fact that the structure is of a transverse nature: in other respects the type of inhomogeneous magnetic order and its effect on the superconducting properties are determined by the exchange interaction between the localized moments and the conduction electrons.

Within the framework of the purely electromagnetic interaction, the domain-type magnetic structure was first proposed as a candidate for the superconductivity-magnetism coexistence phase by Krey,⁷ and later Greenside *et al.*⁸ carried out a detailed investigation of an equivalent structure, which they called the linealry polarized solution. Kuper *et al.*,⁹ as well as Tachiki *et al.*,¹⁰ have proposed another coexistence phase—a structure with spontaneous vortices—within the framework of the electromagnetic interaction. The transition into this phase becomes possible when the magnetic induction $4\pi M$ (where M is the magnetization) attains a value of the order of the lower critical magnetic field H_{c1} of type-II superconductors. The transition from the superconducting phase with spontaneous vortices into the normal ferromagnetic phase then occurs when the induction $4\pi M$ is

higher than the value of the upper critical field.

Superconducting antiferromagnets, in which, as has already been noted, the effect of the magnetism on the superconductivity is not so strong, have attracted much less attention. The latter is quite understandable, since the average values (over the coherence length ξ_0) of the moment and the exchange field in antiferromagnets are equal to zero, and only the occurrence of a gap of the order of the value of the exchange field h in a narrow region of the order of $h/v_F Q$ (where $Q \sim 1/a$ is the antiferromagnetism vector) on the Fermi surface affects the superconductivity. As a result, the superconducting order parameter^{11,12} changes by an amount of the order of $T_c h / \epsilon_F \approx T_N T_c / h$, where T_N is the Néel temperature, and, since $T_N \ll h$, this effect is unimportant. There is another mechanism underlying the suppression of superconductivity in antiferromagnets: the exchange scattering by the magnetic-moment fluctuations above T_N and the spin waves below T_N . Its effect on the superconductivity is also slight—to the extent of the smallness of the parameter T_N/T_c . In the light of the foregoing, the existence of compounds with $T_N > T_c$ is quite unlikely, although it should be noted that a compound with $T_N > T_c$ has been found.¹³

In a sense, the intermediate position between antiferromagnets and ferromagnets is occupied by weak ferromagnets—compounds in which the establishment of the antiferromagnetic order is accompanied by the appearance of a small ferromagnetic moment of the order of 10^{-3} – 10^{-1} of the nominal moment (see, for example, Refs. 14 and 15). In the present paper we theoretically consider the question of the coexistence of superconductivity and weak ferromagnetism. We shall show that a domain structure (a DS phase) can arise at a temperature below the magnetic transition temperature T_N , similarly to what happens in superconducting ferromagnets of the HoMo_6S_8 type. This state is characterized by the fact that each domain possesses a small ferromagnetic moment that changes sign on going from a given domain to a neighboring one (see Fig. 1). As in the case of superconducting “ferromagnets” of the HoMo_6S_8 type, the domain structure is transverse (the wave vector Q is perpendicular to the direction of the small ferromagnetic moment) and one-dimensional. Furthermore, in the case of very weak ferromagnetism there can appear at the point T_N a homogeneous ferromagnetic Meissner state (FS phase), which, typically, does not give rise to a nonzero magnetic induction inside the sample (see Fig. 2).

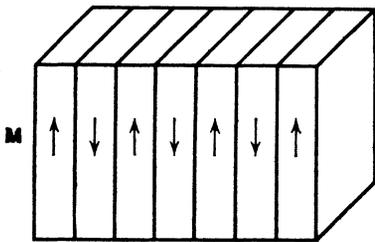


FIG. 1. Domain structure of weak ferromagnets in the superconducting state (the DS phase). The directions of the antiferromagnetism vector L and the ferromagnetism vector M in neighboring domains are opposite. The wave vector of the structure is perpendicular to M . The superconducting order parameter is almost homogeneous over the sample.

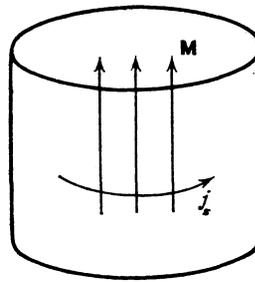


FIG. 2. A weak ferromagnetic Meissner superconducting state (FS). The superconducting current flowing along the sample boundary in a layer of thickness λ_L screens off the surface current due to the jump that occurs in the ferromagnetic moment at the boundary. Inside the sample the magnetic moment is constant, the magnetic induction is equal to zero, and the superconducting order parameter is homogeneous.

It is significant here that, whereas in ferromagnets the major role in the determination of the characteristics of the DS phase is played by the exchange interaction, in weak ferromagnets for which the ratio of the ferromagnetic moment to the antiferromagnetic moment is sufficiently small the dominant role is played by the electromagnetic interaction. Therefore, the domain structure may disappear when the temperature is lowered, but, in contrast to the ferromagnetic case, its disappearance does not lead to the destruction of the superconductivity, and there arises in the system a structure with self-induced vortices (VS phase), as shown in Fig. 3. The Meissner phase may also be transformed into this structure as the temperature decreases, and the small ferromagnetic moment increases correspondingly.

At present we cannot unequivocally point to a superconducting weak ferromagnet. The fact that weak ferromagnetism is widespread among ordinary antiferromagnets^{14,15} allows us to be hopeful of its discovery among superconductors also. The indicated type of magnetic ordering is perhaps realized in the compound NdRh_4B_4 : neutron diffraction data¹⁶ indicate that this antiferromagnet exhibits an antiferromagnetic-moment modulation with wave vector $Q \approx 0.13 \text{ \AA}^{-1}$. The presence of weak ferromagnetism could explain this modulation.

Preliminary symmetry analysis, based on room-temperature x-ray diffraction data, of compounds of the type RERh_4B_4 and REMo_6S_8 indicates that they do not possess weak ferromagnetism. Perhaps, a structural transition that changes the symmetry occurs when the temperature is

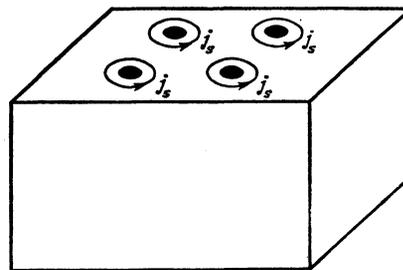


FIG. 3. The superconducting phase with spontaneous vortices (VS) induced by the weak ferromagnetism of the sample. In this phase the ferromagnetic moment is almost homogeneous, the induction and the superconducting currents are highly inhomogeneous, and superconductivity has been destroyed inside the vortex cores.

lowered. Furthermore, the fact that NdRh₄B₄ crystals exist only together with an impurity phase¹⁶ indicates the presence of considerably internal strains that distort the structure in polycrystalline NdRh₄B₄ samples. Owing to piezomagnetism, these distortions can give rise to induced ferromagnetism. It is also as yet not clear what type of magnetic order occurs in the compounds Tb(Ir_xRh_{1-x})₄B₄, Ho(Ir_xRh_{1-x})₄B₄ (Ref. 17) and Y₉Co₇ (Ref. 18). The foregoing and also the progress being made in the synthesis of new magnetic superconductors allow us to hope that superconducting weak ferromagnets exist or will be produced in the near future.

2. TYPE OF MAGNETIC STRUCTURE IN THE SUPERCONDUCTING STATE BELOW THE NÉEL POINT

We now consider compounds that would, in the absence of superconductive pairing below the Néel point T_N exhibit a weak ferromagnetic order. Below we shall investigate compounds with $T_N \ll T_c$, where T_c is the superconducting transition temperature (at the qualitative level the results of the paper will be valid for compounds with $T_N \lesssim T_c$ as well). Furthermore, we shall limit ourselves to the investigation of type-II superconductors, since all the magnetic superconductors known at present are of this type. In the presence of superconductivity, the primary antiferromagnetic structure changes insignificantly, but the superconductivity can radically change the character of the weak "ferromagnetic" order because of the superconductive screening of the long-wave components of the magnetic and exchange fields that occurs in a system with the RKKY interaction.

To investigate the type of magnetic order that exists below the Néel point T_N , we should consider those terms in the system's free energy functional which are quadratic in the magnetic moment. In this case to the ordinary magnetic functional $\mathcal{F}_m\{\mathbf{L}(\mathbf{r}), \mathbf{M}(\mathbf{r}), T\}$ describing the magnetic order in the normal state must be added the functional $\mathcal{F}_s(\Delta, T)$ (where Δ is the superconducting parameter of the system) of the superconducting system for electrons and the functional $\mathcal{F}_{int}\{\mathbf{M}(\mathbf{r}), \Delta, T\}$ describing the interaction between the superconducting and magnetic subsystems. After deriving the total functional, we can determine the type of magnetic order by varying the functional, we can determine the type of magnetic order by varying the functional in \mathbf{L} , \mathbf{M} , and Δ .

Let us, to begin with, consider the magnetic part of the functional. We cannot at this time identify unequivocally the symmetry of the compounds in which superconductivity and weak ferromagnetism can coexist. Therefore, we shall, for definiteness, take the magnetic functional that describes the weak ferromagnetism in magnetic materials with the MnCO₃-type rhombohedral structure.¹⁴ Notice that the specific form of the functional is, naturally, connected with the symmetry of the compound, but the qualitative results of our analysis do not depend on this. Thus, the free-energy density functional of the magnetic subsystem has the form

$$\mathcal{F}_m = \frac{n\Theta_m}{2\pi} \left\{ \frac{A}{2} l^2 + \frac{b}{2} m^2 + \frac{D}{2} l_z^2 + \frac{e}{2} m_z^2 + \beta (l_x m_y - l_y m_x) + \frac{C}{4} l^4 + a^2 \left(\frac{\partial l_i}{\partial x_j} \right)^2 \right\} + \varepsilon_a(1). \quad (1)$$

Here $\mathbf{l} = \mathbf{m}_1 - \mathbf{m}_2$, $\mathbf{m} = \mathbf{m}_1 + \mathbf{m}_2$ are the magnetization vectors of the sublattices $\mathbf{M}_{1,2} = \mu n \mathbf{m}_{1,2}$ where μ is the magnitude of the localized magnetic moment, n is the concentration of the magnetic atoms, and the energy parameter $\Theta_m = 2\pi\mu^2 n$ coincides in order of magnitude with T_N . Weak ferromagnetism arises in the case when the vector \mathbf{l} is perpendicular to the z axis, which is realized when $D > -\beta^2/b$. Below we assume this condition to be fulfilled. The anisotropy energy \mathcal{E}_a determines the direction of \mathbf{l} in the (x, y) plane; the explicit form of \mathcal{E}_a is unimportant, and we shall, for definiteness, assume that the vector \mathbf{l} is oriented along the y axis. In the absence of superconductivity, the functional (1) leads to the appearance of ferromagnetic moment below the Néel temperature T_N and in the case when $l_x = 0, l_y \neq 0$. This moment is oriented along the x axis and its magnitude $m_x = (\beta/b)l_y$, i.e., the ferromagnetic moment is much smaller than the antiferromagnetic moment, since the factor $\gamma = \beta/b \ll 1$.

The characteristic values of the magnetic-transition temperatures of the superconducting magnetic materials range from 1 to 5 K, which corresponds to a maximum induction $B_0 = 4\pi\mu n$ of the order of several kilo-oersted, and the characteristic values of the exchange fields h_0 lie in the range from 20 to 100 K. Notice that in the group of compounds under consideration the contribution of the electromagnetic interaction to the energy of the antiferromagnetic state is, in order of magnitude, equal to $\Theta_m = 2\pi n \mu^2$ (i.e., of the order of 1 K), and it is comparable to the exchange contribution $\Theta_{ex} = h_0^2 N(0)$, where $N(0)$ is the density of electron states. At the same time, the superconducting transition temperatures T_c of these materials lie in the range from 2 to 10 K, and the upper critical fields H_{c2} are of the order of several kilo-oersted. Since in weak ferromagnets the corresponding values of the ferromagnetic moments effectively interacting with the superconductivity are $\gamma \sim 10^2 - 10^3$ times smaller than the nominal values, we see that the ferromagnetic component of the exchange field is small, i.e., $h_f \approx \gamma h_0 \ll T_c$, as is the component of the magnetic induction: $B_f \approx \gamma B_0 \ll H_{c2}$. Thus, weak ferromagnetism cannot destroy the superconductivity, and, furthermore, the interaction between the exchange field of the weak ferromagnet and the superconductivity can always be described within the framework of perturbation theory, since we require for this purpose the fulfillment of the condition $h_f \ll T_c$. The magnetic field that occurs under the conditions of weak ferromagnetism can, generally speaking, be higher than H_{c1} . Then its effect on the superconductivity is not slight: such a field cannot be taken into account by perturbation theory, and it is precisely in this situation that the vortex structure is possible. But near T_N the magnetic field is always weak, and it, like the exchange field, can be taken into account by perturbation theory.

The superconductivity screens off the long-wave parts of the exchange and electromagnetic interactions and at the same time has virtually no effect on the short-wave parts with wave vectors of the order of the reciprocal lattice vector, which is by far greater than the reciprocal superconducting correlation length ξ_0^{-1} . In view of this, we have included in the functional \mathcal{F}_{int} the free-energy difference corre-

sponding to the difference between the long-range interactions in the superconducting and normal phases. The functional \mathcal{F}_{int} has the form

$$\mathcal{F}_{int} = \int \left(\frac{B^2}{8\pi} - \mathbf{B}\mathbf{M} + 2\pi\mathbf{M}^2 \right) d\mathbf{r} + \frac{1}{2} \int \left\{ Q_s(\mathbf{r}-\mathbf{r}') \mathbf{A}(\mathbf{r}) \mathbf{A}(\mathbf{r}') - \alpha_{ex} \frac{\chi_s(\mathbf{r}-\mathbf{r}') - \chi_n(\mathbf{r}-\mathbf{r}')}{\chi_n^0} \mathbf{M}(\mathbf{r}) \mathbf{M}(\mathbf{r}') \right\} d\mathbf{r} d\mathbf{r}'$$

$$\mathbf{B} = \text{rot } \mathbf{A}, \quad \alpha_{ex} \approx \hbar^2 N(0) / (2\pi n \mu^2), \quad \mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2, \quad (2)$$

where $\alpha_{ex} \sim 1$ is a coefficient characterizing the relative contribution of the exchange interaction to the magnetic energy as compared to the electromagnetic contribution, $\chi_n(r)$ and $\chi_s(r)$ are the spin susceptibilities in the normal and superconducting states, $\chi_n^0 = \mu_B^2 N(0)$, $Q_s(r)$ is the electromagnetic superconductive kernel. In the momentum representation for a pure superconductor with mean free path $l \gg \xi_0$ we have

$$Q_s(q) = \frac{1}{4\pi\lambda_L^2}, \quad \frac{\chi_s(q)}{\chi_n^0} = \frac{q^2 v^2}{30\Delta^2} = \frac{\pi^2 q^2 \xi_0^2}{30}, \quad q \ll \xi_0^{-1}; \quad (3a)$$

$$Q_s(q) = \frac{3}{4\xi_0 q \lambda_L^2}, \quad \frac{\chi_s(q)}{\chi_n^0} = 1 - \frac{\pi}{2\xi_0 q}, \quad q \gg \xi_0^{-1}. \quad (3b)$$

We do not need the explicit form of the functional \mathcal{F}_s , since the superconducting condensation energy does not change in second order perturbation theory in terms of \mathbf{M} .

The minimization of the total functional with respect to the Fourier components \mathbf{B}_q and \mathbf{M}_q yields the equations

$$[\mathbf{q} \times \mathbf{B}_q] = \frac{4\pi}{c} \mathbf{j}_q + 4\pi[\mathbf{q} \times \mathbf{M}_q], \quad \mathbf{j}_q = -cQ_s(q) \mathbf{A}_q, \quad \mathbf{L}_q = n\mu \mathbf{l}_q, \\ (bM_{q,x} - \beta L_{q,y}) - (B_{q,x} - 4\pi M_{q,x}) + \alpha_{ex} \frac{\chi_s(q) - \chi_n(q)}{\chi_n^0} M_{q,x} = 0, \\ M_{q,y} = M_{q,z} = 0. \quad (4)$$

To find the free energy as a functional of \mathbf{l} , we must solve the equations (4) and substitute the \mathbf{M} and \mathbf{B} values thus found into the sum of the functionals (1) and (2). As a result, we find that the energy minimum is attained for the transverse structure, i.e., in the equilibrium state the vector \mathbf{q} is perpendicular to the axis. The free energy functional expressed in terms of the antiferromagnetic moment $l_q \equiv l_{q,y}$ had the form

$$\mathcal{F}(l_q) = \sum_q \frac{n\theta_m}{2\pi} \left[\frac{A}{2} l_{q,-q} - \frac{\beta^2}{2B} l_{q,-q} + a^2 q^2 l_{q,-q} + \frac{\beta^2 F(q)}{2b[b+F(q)]} l_{q,-q} \right], \\ F(q) = \alpha_{ex} [1 - \chi_s(q)/\chi_n^0] + \frac{4\pi Q_s(q)}{q^2 + 4\pi Q_s(q)}. \quad (5)$$

As can be seen from (5), the onset of the magnetic order is accompanied by the appearance of a component l_q with $q \neq 0$, and, because of the anisotropy, the solution $l_q(\mathbf{r}) \sim \sin(\mathbf{Q} \cdot \mathbf{r})$, where the direction of the vector \mathbf{Q} in the (y, z) plane is determined by the magnetic-hardness anisotropy and the electron fermi velocity, is realized. Minimizing the energy with respect of the magnitude q of the wave vector, we find for the superstructure wave vector in the case when

$\beta \gg (a/\xi_0)$ the expression

$$Q = \frac{\beta^{3/2}}{2} (a^2 \xi_0)^{-1/2} \left(\frac{\pi \alpha_{ex}}{b^2} \right)^{1/2}. \quad (6)$$

In this region of the parameter β , the wave vector Q is determined largely by the exchange interaction.

The situation $\beta \ll a/\xi_0$ is also entirely possible in weak ferromagnets. In this case the major role in the determination of the magnitude of the wave vector is played by the electromagnetic interaction, and here when $\beta \gg a/\lambda_L$

$$Q = [\beta/2^{1/2}(b + \alpha_{ex})]^{1/2} (a\lambda_L)^{-1/2}. \quad (7)$$

But if β is so small that $\beta < \beta_{cN} = 2^{1/2} a(b + \alpha_{ex} + 1)\lambda_L$, then the appearance of the inhomogeneous state turns out to be disadvantageous, and there is realized in the system a homogeneous magnetization \mathbf{M} with magnetic induction $\mathbf{B} = 0$, since the superconducting currents flowing along the sample surface completely screens off the magnetic field $4\pi\mathbf{M}$ produced by the localized moments (see Figs. 2). Thus, in this case we have the Meissner ferromagnetic (FS) state to deal with. Notice that in this case $F(0) = 4\pi + \alpha_{ex}$, whereas in the absence of superconductivity $F = 0$. As a result the ratio $M/L = \beta/(b + F)$ differs from the corresponding quantity in the normal magnetic material (in which $M/L = \beta/b$); in the case when $T_N > T_c$ this should manifest itself in a decrease in the ratio M/L in the region below T_c .

Summing up, we can conclude that, for sufficiently large values of $\beta > \beta_{cN} \approx a/\lambda_L$, the system exhibits a transverse magnetic superstructure. Typically, the coordinate dependences of the antiferromagnetic and ferromagnetic vectors are sine functions, and the superstructure wave vector Q can be quite large because of the small magnitude of the magnetic hardness a . For small values of the parameter ν (i.e., for $\beta \lesssim \beta_{cN}$) a homogeneous weak ferromagnetism with a magnetic induction that is nonzero only on the sample surface in a layer of thickness of the order of the London penetration depth (i.e., a Meissner state) is realized.

The analysis carried out above is valid only in a small neighborhood of T_N , where the terms of higher order in \mathbf{l} in the free-energy functional can be neglected. Below T_N the variation in magnitude of \mathbf{l} is disadvantageous, and there is realized in the anisotropic system a domain structure such that the ferromagnetic vector \mathbf{M} and the antiferromagnetic vector \mathbf{L} reverse their directions when we go from a given domain to a neighboring one. The nature of the domain wall in weak ferromagnets is investigated in Ref. 19. According to the results obtained there, the domain structure will be well defined, i.e., the wall dimension will be much smaller than the period of the structure, in the temperature region where $[(T_N - T)/T_N]^{1/2} \gg aQ$, i.e., virtually everywhere except in a small neighborhood of T_N .

As the temperature is lowered, and the magnetization intensity increases, it becomes possible for the DS phase to go over into the state with a self-induced vortex structure (the VS phase). The Meissner phase can go over into this same state when the quantity $4\pi M$ exceeds H_{c1} .

Below we shall determine the principal characteristics of each of the indicated states, find their existence domains

and their free energies, and construct the phase diagram in the (T, β) plane.

3. THE DOMAIN STRUCTURE

Let us, to begin with, consider the DS phase. In this case the magnetization M can be approximated by a step function (Fig. 1):

$$M_x(r) = \frac{4M}{\pi} \sum_{n=0}^{\infty} \frac{\sin(2n+1)qr}{2n+1}. \quad (8)$$

In the free energy \mathcal{F}_m of the magnetic subsystem we should replace the $(\partial l / \partial y)^2$ and anisotropy terms by the domain-wall energy $n\Theta_m(Qa)l^2\eta/2\pi$, where $n\eta\Theta a$ is the surface energy of the walls and the quantity η is of the order of unity. We can, in analyzing the domain structure, use perturbation theory in terms of not only the exchange, but also the magnetic field. Indeed, the spatially varying magnetization having a Fourier transform of M_q gives rise to a magnetic field described by the vector potential (in the London gauge)

$$A_q = \frac{4\pi q}{q^2 + 4\pi Q_s(q)} M_q.$$

The condition of applicability of the perturbation theory has the form $2eA_y/c \ll \xi_0^{-1}$, and it is fulfilled when

$$4\pi M \ll \Phi_0(q^2 + 4\pi Q_s)/q\xi_0.$$

After determining the equilibrium value of q , we can easily verify that this condition is fulfilled in the entire existence domain of the DS phase because of the small values of β and a/ξ_0 . Thus, as $\mathcal{F}_{\text{int}} + \mathcal{F}_s$, we can again take the expression (2). The minimization of the functional with respect to q and l gives the equilibrium values of these parameters and the free energy of the system.

When $\beta \gg (a/\xi_0)^{1/2}$, the major role is played by the exchange interaction, and here the period of the DS phase turns out to be small compared to ξ_0 . Using for χ and Q_s the expression in the $q \gg \xi_0^{-1}$ case, and minimizing them with respect to the magnitudes of the wave vector q and the antiferromagnetic moment l , we find the equilibrium value of q and the free energy \mathcal{F}_{DS} for the DS phase:

$$Q = (a\xi_0)^{-1/2} \frac{\beta}{b} \left(\frac{7\xi(3)}{4\eta} \right)^{1/2}, \quad \beta \gg (a/\xi_0)^{1/2},$$

$$\mathcal{F}_{\text{DS}} = \frac{n\Theta}{2\pi} \left[-\frac{\beta^2}{2b} + \frac{\beta}{b} \left(\frac{7\xi(3)a}{\xi_0\eta} \right)^{1/2} \right] l_0^2, \quad (9)$$

where l_0 is the equilibrium value of the antiferromagnetism parameter for $\beta = 0$ and \mathcal{F}_{DS} is the correction that has to be made in the free energy of the antiferromagnet as a result of the presence of the weak antiferromagnetism.

When $\beta \sim (a/\xi_0)^{1/2}$, the electromagnetic and exchange interactions play equally important roles in the formation of the structure, and it is difficult in this case to obtain analytic expressions for Q and \mathcal{F} .

If, on the other hand, $\beta \ll (a/\xi_0)^{1/2}$, then the role of the electromagnetic interaction is dominant, and the free energy functional $\mathcal{F}_{\text{DS}}(Q)$ is, after being minimized with respect to l , equal to

$$\mathcal{F}_{\text{DS}}(Q) = \frac{\Theta}{2\pi} l_0^2 \left\{ \frac{\beta^2}{2(b+\alpha_{\text{ex}})(b+1+\alpha_{\text{ex}})} \times \left[1 - \frac{2Q\lambda_L}{\pi} \left(\frac{\alpha_{\text{ex}}+b}{\alpha_{\text{ex}}+b+1} \right)^{1/2} \right] \times \text{th} \left(\left(\frac{\alpha_{\text{ex}}+b+1}{\alpha_{\text{ex}}+b} \right)^{1/2} \frac{\pi}{2Q\lambda_L} \right) \right\} + \eta Q a. \quad (10)$$

As follows from a direct analysis of the Q dependence of the free energy (10), the minimum of the functional is attained at $Q \neq 0$ only when

$$\beta > \beta_c = (\eta\pi a/\lambda_L)^{1/2} (\alpha_{\text{ex}}+b)^{1/2} (\alpha_{\text{ex}}+1+b)^{1/4}.$$

This value β_c of β is found from the expansion of (10) in powers of Q for $Q \rightarrow 0$ from the requirement that the term linear in Q should vanish. In the case when $\beta < \beta_c$, the energy minimum corresponds to the value $Q = 0$, i.e., the phase with homogeneous magnetization [the Meissner FS phase (see Figs. 2)], which can go over into the VS phase when the temperature is lowered further.

As $\beta \rightarrow \beta_c$, the wave vector Q of the DS phase goes smoothly to zero, and the line $\beta = \beta_c$ is the FS-DS phase boundary.

In the case when we can, by expanding (10) in powers of the quantity $1/Q\lambda_L$, easily obtain analytic expressions for

$$(a/\lambda_L)^{1/2} \ll \beta \ll (a/\xi_0)^{1/2},$$

the equilibrium Q vector and the free energy of the DS phase:

$$Q = (a\lambda_L^2)^{-1/2} \left(\frac{\beta}{b+\alpha_{\text{ex}}} \right)^{1/2} \left(\frac{15\xi(4)}{2\pi^2\eta} \right)^{1/2},$$

$$\mathcal{F}_{\text{DS}} = \frac{\Theta_m}{2\pi} l_0^2 \left[-\frac{\beta^2}{2(b+\alpha_{\text{ex}})} + 3 \left(\frac{15\beta^2\xi(4)\eta^2 a^2}{(b+\alpha_{\text{ex}})^2 16\pi^2\lambda_L^2} \right)^{1/2} \right],$$

$$(a/\lambda_L)^{1/2} \ll \beta \ll (a/\xi_0)^{1/2}. \quad (11)$$

In the region $\beta \gtrsim (a/\lambda_L)^{1/2}$ the wave vector Q is of the order of λ_L^{-1} , and, to find its magnitude as well as the free energy of the DS phase, we must numerically minimize the expression (10) with respect to Q .

Notice that the DS phase exists only when $\beta > \beta_c \sim (a/\lambda_L)^{1/2}$, whereas the inhomogeneous state is formed at the point T_N when $\beta \gtrsim a/\lambda_L$. Thus, there is realized in the region of β values

$$(a/\lambda_L)^{1/2} > \beta \gtrsim (a/\lambda_L)$$

a situation in which the inhomogeneous structure exists only in a very narrow neighborhood of T_N , giving way to the FS phase when the temperature is lowered further.

4. THE VORTEX STRUCTURE

We should, in determining the character of the vortex structure in weak ferromagnets, consider the complete system of equations for \mathbf{L} , \mathbf{M} , and \mathbf{B} . Since the characteristic scales of the vortex structure are large compared to ξ_0 and a , we can neglect the gradient terms in the magnetic functional, and, as for the superconducting part, we can set $\chi_s = 0$. As a result, we arrive at equations determining the magnetization and the magnetic induction in the phase with vortices:

$$(b + \alpha_{ex})M - \beta L - B + 4\pi M = 0,$$

$$\text{rot } \mathbf{B} = \frac{4\pi}{c} \mathbf{j}_s + 4\pi \text{rot } \mathbf{M}, \quad (12)$$

$$\mathbf{j}_s = cQ_s \mathbf{A}.$$

$$\mathbf{M} = \frac{\beta \mathbf{L}_0 + \mathbf{B}}{b + \alpha_{ex} + 4\pi}, \quad L_0^2 = -\frac{A}{C}. \quad (13)$$

In this case the equation for the field distribution will have the form

$$\text{rot rot } \mathbf{B} = 4\pi (1 + 4\pi / (b + \alpha_{ex})) Q_s \mathbf{B}. \quad (14)$$

Equation (14) is similar to the corresponding equation for an ordinary vortex structure²⁰ when the following substitution is made:

$$\lambda_L^2 \rightarrow \lambda_L^2 / [1 + 4\pi / (b + \alpha_{ex})] = \tilde{\lambda}^2. \quad (15)$$

The field-dependent part of the free-energy functional of the weak ferromagnet then has the form

$$\mathcal{F}_{vs}(\mathbf{B}) = \frac{1}{1 + 4\pi / (b + \alpha_{ex})} \left[-\frac{\beta \mathbf{L} \mathbf{B}}{b + \alpha_{ex}} + \frac{B^2}{8\pi} + \frac{A^2}{\tilde{\lambda}^2} \right]. \quad (16)$$

As usual, the second and third terms in the square brackets are the field and superconducting-current energies respectively, while the first term describes the effect of the external homogeneous magnetic field \mathbf{H} , the role of which is played in our case by the quantity $L4\pi\beta/b'$, where $b' = \alpha_{ex} + b$.

When the density of the vortices is low, we can neglect the interaction between them, and write $\mathcal{F}_{vs}(\mathbf{B})$ in the form (see Ref. 20)

$$\left[1 + \frac{4\pi}{b + \alpha_{ex}} \right] \mathcal{F}_{vs}(\mathbf{B}) = n_L \bar{\sigma} - \frac{\beta L}{b'} \mathbf{B}, \quad \bar{\sigma} = \left(\frac{\Phi_0}{4\pi \tilde{\lambda}} \right)^2 \ln \frac{\tilde{\lambda}}{\xi_0}, \quad (17)$$

where n_L is the number of vortex filaments per unit area, $\bar{\sigma}$ is the linear density of the energy of a vortex filament, and Φ_0 is the flux quantum.

Furthermore, since each filament carries one flux quantum Φ_0 , the induction B can be written in the form $B = n_L \Phi_0$. As a result,

$$\mathcal{F}(B) \sim n_L (\bar{\sigma} - \beta L \Phi_0 / b')$$

and the formation of the vortices becomes energetically advantageous when

$$\beta L / b' > \bar{\sigma} / \Phi_0 = \tilde{H}_{c1} / 4\pi,$$

where

$$\tilde{H}_{c1} = \frac{\Phi_0}{4\pi \tilde{\lambda}^2} \ln \frac{\tilde{\lambda}}{\xi_0}.$$

Thus, the condition for the transition from the Meissner FS state into the vortex VS phase to occur has the form

$$\beta L / b' = \tilde{H}_{c1} / 4\pi. \quad (18)$$

The properties of the vortex state in a weak ferromagnet are then entirely similar to those of the ordinary vortex state²⁰ when the substitutions $H/4\pi \rightarrow \beta L/b'$ and $\lambda \rightarrow \tilde{\lambda}$ are made.

In order to analyze the transition from the DS phase into the VS state, we must know the energy of the VS phase

for the intermediate values of the vortex density n_L , i.e., for $\tilde{\lambda}^{-2} \ll n_L \ll \xi_0^{-2}$. Using the corresponding expression for the triangular vortex lattice,²⁰ we can write

$$\mathcal{F}_{vs} = -\frac{\beta^2}{2(b' + 4\pi)} L_0^2 - \frac{b + \alpha_{ex}}{8\pi(b' + 4\pi)} \times \left(\frac{4\pi\beta L_0}{b'} - H_c \frac{\ln(\tilde{\lambda}/d)\pi}{24\tilde{\lambda}} \right)^2, \quad (19)$$

where H_c is the thermodynamic critical field

$$H_c^2 / 8\pi = N(0) \Delta_0^2 / 2, \quad \tilde{\kappa} = \tilde{\lambda} / \xi_0$$

and d is the vortex-lattice constant, which can be found from the condition²⁰

$$\frac{2}{\sqrt{3}} \frac{\Phi_0}{d^2} + \tilde{H}_{c1} \frac{\ln(0.18d/\xi_0)}{\ln \tilde{\kappa}} = \frac{4\pi\beta L}{b'}. \quad (20)$$

The VS phase can compete in terms of energy with the DS phase only when $\beta \lesssim (a/\xi_0)^{1/2}$, since the energy of the DS phase is always lower than that of the VS phase when $\beta \gg (a/\xi_0)^{1/2}$. In the region $\beta \ll (a/\xi_0)^{1/2}$ the free energy is given by the expression (10). In this case the transition into the VS phase with intermediate vortex density will occur only when $\beta \gg (a/\lambda_L)^{1/2}$; when $\beta \gtrsim (a/\lambda_L)^{1/2}$, the vortex density $n_L \gtrsim \lambda_L^{-2}$, and an analytical description of the transition is not possible in this case.

Using for \mathcal{F}_{DS} the expression (11) in the limit $\beta \gg (a/\lambda_L)^{1/2}$, and equating the energies of the VS and DS phases, we find the condition for the transition to occur:

$$L = \frac{1}{3b'} \left[\frac{8\pi^2 \beta \tilde{\lambda}^2 b'^2}{15\zeta(4)\eta^2 a^2} \right]^{1/2} H_c', \quad (21)$$

where

$$H_c' = H_c \ln(\tilde{\lambda}/d) \pi / 24\tilde{\kappa} = \nu H_c \tilde{\kappa},$$

ν is of the order of unity, and ζ is the function $\zeta(4) = \pi^4/90$. As is easy to verify, the condition $4\pi\beta L \gg \tilde{H}_{c1}$ is indeed fulfilled in this case, and the vortex density is not low. The β - or temperature-governed transition between the DS and VS phases should be of first order: a dense ($n_L \gg L^{-2}$) vortex lattice should appear all at once.

As a result, we arrive at the phase diagram, schematically shown in Fig. 4, for the case when $\tilde{H}_{c1} b' / 4\pi\beta_c L_0 < 1$. On the line AB , as β increases, the DS phase develops continuously, i.e., domains appear, and their thickness decreases with increasing β . On the line BC vortices appear in the Meissner state, and the density of the vortex lattice increases with increasing β . The line BD separates the existence regions of the dense vortex lattice and the domain structure.

When $\tilde{H}_{c1} b' / 4\pi\beta_c > 1$, the VS phase does not occur, and the line $\beta = \beta_c$ separates the FS and DS phases right down to absolute zero.

We considered the superconductor to be pure when computing the DS phase. Actually, the asymptotic form (3a) used by us is valid so long as $Ql \ll 1$, while the asymptotic form (3b) is valid so long as $Ql \gg 1$. Therefore, the phase diagram shown in Fig. 4 reproduces qualitatively accurately the behavior of dirty type-II superconductors as well.

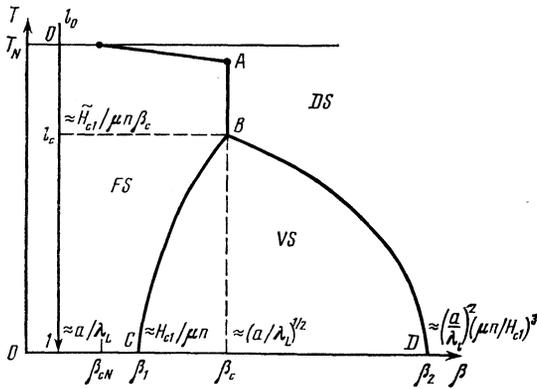


FIG. 4. Phase diagram for type-II superconductors with weak ferromagnetism under conditions when $\tilde{H}_{c1}/\mu_n\beta_c < 1$. The phase FS is the ferromagnetic Meissner superconducting phase; DS, the superconducting phase with a domain structure for weak ferromagnetism; and VS, the superconducting phase with spontaneous vortices. The temperature and the magnitude of the antiferromagnetic vector l have been plotted along the axis of ordinates.

5. PRESSURE-INDUCED WEAK FERROMAGNETISM

Let us also note the interesting possibility of observing the above-described magnetic structures in superconducting antiferromagnets through the application of the phenomenon of piezomagnetism. The essence of this phenomenon consists in the fact that, in certain antiferromagnets, the response to an external pressure is the appearance of a spontaneous ferromagnetic moment (see, for example, Ref. 14). Thus, in the presence of superconductivity, the situation is quite similar to the one considered above. Let us illustrate this for the particular case of magnetic materials with the rhombohedral structure. In this case the crystal symmetry leads to a free-energy expansion (with allowance for the terms linear in the stresses σ_{ij} and the magnetic moment) of the form

$$\mathcal{F}_m = \frac{1}{2}AL^2 + \frac{1}{2}a(L_x^2 + L_y^2) + \frac{1}{2}bM^2 + \frac{1}{2}b_1M_z^2 + \beta(L_xM_y - L_yM_x) + \lambda_1(M_x\sigma_{yz} - M_y\sigma_{zx})L_z + \eta_1(L_y\sigma_{yz} + L_x\sigma_{zx})L_z. \quad (22)$$

In the absence of stresses, weak ferromagnetism can exist only when $a < 0$. Weak ferromagnetism does not occur when $a > 0$ and $L_x = L_y = 0$. Under the action of, for example, the stresses $\sigma_{xz} = 0$, $\sigma_{yz} \neq 0$, there appears in the direction of the x axis a ferromagnetic moment given by the expression

$$M_x = - \left[\frac{\beta\eta_1 + \lambda_1 a}{ba - \beta^2} \right] L_z \sigma_{yz}. \quad (23)$$

As is easy to see, the deviation from weak ferromagnetism lies only in the fact that the role of the parameter β/b is played by the quantity $[(\beta\eta_1 + \lambda_1 a)/(ba - \beta^2)]\sigma_{yz}$, which is proportional to the stress.

Thus, we can vary the ratio in piezomagnetic materials by varying the external pressure, which, in principle, makes it possible for us to obtain the entire phase diagram shown in Fig. 4.

6. CONCLUSIONS

Let us emphasize again that the electromagnetic interaction may turn out to be the controlling factor in the forma-

tion of the magnetic structure in weak superconducting ferromagnets, when $\beta < (a/\xi_0)^{1/2}$. At the same time, in superconducting "ferromagnets" with a realistic relation between \mathcal{O}_{ex} and \mathcal{O}_{em} , the major role is played by the exchange interaction,⁶ and the theory developed in Refs. 7–10 is hardly applicable in this case. A domain structure with $Q \sim (a\xi_0)^{-1/2}$ should appear in them,⁶ and the transition of this structure into the normal ferromagnetic phase on being cooled is also governed by the exchange interaction. Thus, only weak ferromagnets offer us an opportunity of observing those coexistent structures predicted in Refs. 7–10. Comparing our phase diagram (Fig. 4) with the one found in Refs. 7–10, we can conclude that, basically, the phase diagram of weak ferromagnets with $\beta < (a/\xi_0)^{1/2}$ is similar to the phase diagram of superconducting ferromagnets when allowance is made for only the electromagnetic interaction. But there is one significant difference between them: the state with homogeneous magnetization (the FS phase) is possible in weak ferromagnets. For $\beta > (a\xi_0)^{1/2}$ the behavior of weak ferromagnets is similar to the behavior of superconducting ferromagnets, but the phase transition into the FN phase that occurs on cooling can, in the case of weak ferromagnetism, be observed only in exceptional cases (when the value of β is sufficiently large).

The experimental neutron-diffraction data on the antiferromagnetic superconductor NdRh_4B_4 indicate the presence of antiferromagnetism-vector modulation with $Q \sim 0.13 \text{ \AA}^{-1}$ (Ref. 16).¹¹ The fact that this compound exists only in the presence of the stabilizing impurity phase NdRh_6B_6 indicates the presence of appreciable internal strains. In that case, it is possible that there exists a piezomagnetic ferromagnetic moment in NdRh_4B_4 . Then the superconductivity could, in accordance with the results of the present paper, lead to the appearance of a domain structure, and the L and M vectors in neighboring domains would have opposite directions. Thus, this could account for the appearance of the antiferromagnetism-vector modulations.¹⁶

According to the data presented in Ref. 16, the ferromagnetic moment in NdRh_4B_4 does not exceed 10% of the antiferromagnetic moment. In that case, as follows from the expression (9), the wave vector Q of the DS phase would be significantly smaller than the observed value. The cause of this discrepancy in the Q values, which does not support the interpretation of the data of Ref. 16 within the framework of the idea that superconductivity has an effect on weak ferromagnetism, is not clear. Therefore, it would be of interest to verify experimentally whether the presence of the superstructure in NdRh_4B_4 is connected with the superconductivity. This can be done by suppressing the superconductivity with microwave or laser radiation. Notice also, that owing to the presence of the mixed invariant ML in the free-energy functional, an external magnetic field applied at a temperature above the Néel point causes antiferromagnetic ordering, besides magnetization, in weak ferromagnets and piezomagnets.¹⁴ As a result, the $T > T_N$ nonzero-magnetic-field neutron-diffraction data on NdRh_4B_4 would have been expected to exhibit antiferromagnetic peaks if weak ferromagnetism or piezomagnetism existed in this compound.²⁾

¹¹A similar antiferromagnetism-vector modulation with $Q \sim 0.24 \text{ \AA}^{-1}$ was recently observed in TmRh_4B_4 by Majkrzak *et al.*²¹.

²¹Note added in proof (29 June 1983). The fabrication of a new body-centered phase of ErRh_4B_4 , which is an antiferromagnetic superconductor, has recently been reported (H. Iwasaki, M. Isino, K. Tsunokumi, and Y. Muto, *J. Magn. Magnetic Mater.* **31-34**, 521 (1983)). The structure of this phase, as symmetry analysis shows, admits of the existence of weak ferromagnetism.

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