

Theory of magnetic structure in reentrant magnetic superconductors HoMo_6S_8 and ErRh_4B_4

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Magnetic superconductors, in which the first- and the second-order phase transition to the ferromagnetic (FN) state would occur in the absence of superconductivity, are considered. The exchange and the electromagnetic dipolar interactions of localized moments and electrons as well as magnetic anisotropy are taken into account. It is shown that one realizes the transverse domainlike magnetic structure in a superconducting state (the DS phase). Transitions $S \rightarrow \text{DS} \rightarrow \text{FN}$ are considered. The proposed theory (with the second-order transition) explains well the experimental data for HoMo_6S_8 . The experimental data on ErRh_4B_4 may be explained in the framework of the similar microscopic theory with the assumption that it is a first-order magnetic transition and that the critical-temperature (Θ) variations over the sample investigated by Moncton *et al.* [Phys. Rev. Lett. **45**, 2060 (1981)] and by Sinha *et al.* [Phys. Rev. Lett. **48**, 950 (1982)] are due to inhomogeneous stresses.

I. INTRODUCTION

There exists much interest in the problem of the coexistence of ferromagnetism and superconductivity because they are competitive processes.^{1,2} Anderson and Suhl have shown, in a remarkable paper,³ that the competition of these two orderings gives rise to the new superconducting phase with the nonuniform magnetic ordering of localized moments (LM's). They predicted that if the indirect exchange interaction (EX) gives the ferromagnetic order at the temperature Θ (in the absence of superconductivity), then the presence of superconductivity (with the critical temperature $T_{c1} \gg \Theta$) modifies the EX interaction. This effect causes the appearance of the nonuniform magnetic ordering at $T_M \approx \Theta$ by the second-order phase transition. The wave vector of this structure is $Q_M \approx (a^2 \xi_0)^{-1/3}$, where ξ_0 is the superconducting coherence length, and a is the magnetic stiffness—it is of the order of atomic length. The transition temperature T_M practically coincides with Θ , i.e., $T_M - \Theta \approx -\Theta(a/\xi_0)^{2/3}$.

Later Krey,⁴ Blount and Varma,⁵ and Matsumoto *et al.*⁵ came to a similar conclusion assuming that the interaction between LM's and electrons is realized via magnetic field generated by LM's, i.e., via the dipolar electromagnetic interaction (EM). In the framework of their approach the wave vector of magnetic structure (simple spiral) at T_M is $Q_M \approx (a\lambda_L)^{-1/2}$, where λ_L is the London penetra-

tion depth. In both cases the nonuniform magnetic structure is due to the suppression of the zero-wave-vector component of the interaction between LM's, i.e.:

(a) In the case of the EX interaction of LM's and electrons, Cooper pairing suppresses the zero-wave-vector component of electronic paramagnetic susceptibility.

(b) In the case of the EM interaction, the Meissner effect suppresses the uniform magnetic field created by LM's.

Recent experiments on neutron scattering on HoMo_6S_8 , done by Lynn *et al.*,⁶⁻⁸ confirm the prediction of Anderson and Suhl. In these experiments the nonuniform magnetic ordering with the wave vector $Q \approx 0.03 \text{ \AA}^{-1}$ is observed below $T_M \approx 0.7 \text{ K}$ in the superconducting state ($T_{c1} \approx 1.8 \text{ K}$) of HoMo_6S_8 . The nonuniform magnetic ordering was observed also in ErRh_4B_4 by Moncton *et al.*,⁹ below 1.4 K on polycrystalline samples, and by Sinha *et al.*¹⁰ on monocrystal samples ($T_{c1} \approx 8.7 \text{ K}$) below 1.2 K. A first-order transition to the ferromagnetic (FN) state at $T_{c2} \approx 0.75 \text{ K}$ on a monocrystalline sample of ErRh_4B_4 , and at $T_{c2} \approx 0.65 \text{ K}$ on a polycrystalline sample of HoMo_6S_8 , is also observed. These experiments, as well as the antagonistic character of ferromagnetic and superconducting orderings, now provide a challenge for the development of theoretical description.

Recently several models have been proposed that

either neglect the EX interaction^{4,5,11,12} or take into account the EX and the EM interactions but neglect the magnetic anisotropy (MA).^{3,13,14} So first of all we should discuss a very important question: Which type of interactions between LM's and electrons are essential in such real compounds as ErRh₄B₄ and HoMo₆S₈?

Certainly, the dipolar EM interaction contributes significantly to the energy of the magnetically ordered state in ErRh₄B₄ and HoMo₆S₈. The long-range part of this interaction gives the contribution $\frac{1}{3}\Theta_m$ to the energy of the ferromagnetic state (per LM) at $T=0$, where $\Theta_m=2\pi\mu^2n$, with μ the magnetic moment and n the concentration of LM's. Θ_m takes the value 1.8 K in ErRh₄B₄ ($\mu=5.6\mu_B$, $n=10^{22}$ cm⁻³), and 1.3 K in HoMo₆S₈ ($\mu=9\mu_B$, $n\approx 4\times 10^{21}$ cm⁻³). The EX interaction is expected to be small, since the localized f electrons in Er and in Ho responsible for magnetism are "weakly coupled" with the s and d conduction electrons, which are responsible for the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and superconductivity.

However, the question regarding the magnitude and the role of the EX interaction cannot be resolved *a priori*. The answer to this question has to be based in part on the experimental facts, as well as on a correct theoretical treatment of the interplay of the EX and the EM interactions.

First, we remark that it is impossible to explain the transition to the FN state (in real compounds) by taking into account only the EM interaction and neglecting the EX interaction as is done in Refs. 5, 11, and 12. In the case of the EM interaction alone, the suppression of superconductivity is caused by the orbital effect—the influence of the magnetic induction \vec{B} on the orbital motion of electrons. Since both ErRh₄B₄ and HoMo₆S₈ are type-II superconductors, the FN state should be realized whenever $B(T) > H_{c2}^*(T)$, where $H_{c2}^*(T)$ is the upper critical field (including orbital effect only).

Maximal induction $B(0)$, which originates from aligned LM's with magnetic moment μ and concentration n , is $B(0)=4\pi\mu n$, and for ErRh₄B₄ one has $B(0)\approx 6.5$ kOe; for HoMo₆S₈, $B(0)\approx 4.8$ kOe. However, the value $H_{c2}^*(0)$ is 11–14 kOe for ErRh₄B₄, according to the experimental data of Ott *et al.*¹⁵ and Cantor *et al.*¹⁶ On the other hand, $H_{c2}^*(0)\approx 3$ kOe in HoMo₆S₈ according to the experimental data of Ishikawa and Fisher,¹⁷ but what is very important is that one has $B(T_{c2}) < H_{c2}^*(T_{c2})$. So it is impossible to explain the transition, of both compounds, to the FN state by the orbital effect (the EM interaction) only, because $B(T_{c2}) < H_{c2}^*(T_{c2})$. Undoubtedly, this means that the EX interaction between LM's and superconducting electrons must be taken into account, if one wishes the explanation of

the destruction of superconductivity by magnetic ordering at T_{c2} .

The EX energy (per LM) Θ_{EX} of the compound ErRh₄B₄, which contributes to the energy of the FN state at $T=0$, may be estimated from the measurements of $T_{c1}(x)$ in Er_xY_{1-x}Rh₄B₄ and Er_xLu_{1-x}Rh₄B₄ with nonmagnetic atoms Y and Lu.^{1,18}

It is well known from the earlier theory that

$$\frac{dT_{c1}(x)}{dx} \approx \pi^2\Theta_{EX}/2$$

when $x\rightarrow 0$. From the measurements of Okuda *et al.*¹⁸ one has $dT_{c1}(x)/dx \approx 3$ K, which gives $\Theta_{EX}^{Er} \approx 0.6$ K. Therefore, the value of Θ_{EX} is of the order of $\Theta_m/3$ in ErRh₄B₄. A similar estimation for the suppression of T_{c1} in HoMo₆S₈, based on experimental data of Ishikawa and Muller,¹⁹ gives $\Theta_{EX}^{Ho} \approx 0.15$ K. This means that the EX contribution to the energy of the FN state is not very small compared with the EM energy: They are of the same order.

Theoretical treatment of magnetic reentrant superconductors, which includes both the EX and the EM interaction, was given in Ref. 14 (in the framework of the microscopic theory—the isotropic model). It is predicted there that helical (simple spiral) order of LM's in superconducting state could be realized below the temperature T_M of the second-order phase transition. This conclusion is based on the mean-field theory, and it is well known that fluctuations induce the first-order phase transition²⁰—minima of inverse susceptibility on the sphere. But experimentally a magnetic structure on the strictly determined wave vector \vec{Q} , is observed, and probably the fluctuations do not induce the first-order phase transition.

In Ref. 14 it is also shown that the role of the EX and the EM interaction (in the coexisting phase) is rather different. It turns out (see Sec. II D) that the magnitude of the wave vector \vec{Q} of magnetic structure, as well as the depression of superconductivity, are determined by the EX interaction if the condition $\Theta_{EX} \gg \Theta_m(\lambda_L Q)^{-2}$ is fulfilled. In that case, the role of the EM interaction is only to make the structure transverse and also to renormalize the parameters Θ and a . So if $(\lambda_L Q)^2 \gg 1$, which is realized in ErRh₄B₄ and HoMo₆S₈, the EX interaction determines the nonuniform magnetic structure in the superconducting phase below T_M , if Θ_{EX} is not extremely small compared with Θ_m .

However, the theory¹⁴ did not take into account the MA of real compounds (as well as other anisotropies of the system, such as the anisotropy of Fermi surface, etc.), which could modify the helical structure. So we must consider the EX and EM in-

teractions, as well as the magnetic anisotropy and the anisotropy of electronic system. Moreover, the problem must be treated in the framework of the microscopic theory, since we deal with the low-temperature region, i.e., $0 < T < T_M \ll T_{c1}$. Such a theory, based on the BCS model and on the assumption of a second-order phase transition to the FN state (in the absence of superconductivity), is presented in Secs. III and IV.

In what follows we shall show that the conclusions stated in Ref. 14 regarding the role of the EX and the EM interactions are valid for any nonuniform magnetic structure. We shall also show that in the presence of the MA and below T_M magnetic structure should be domainlike and transverse—this phase is called the DS phase. The direction of \vec{Q} is in the plane perpendicular to the easy-magnetization axis, and it is determined by the anisotropy of the Fermi surface, and the magnetic stiffness a . The magnitude of \vec{Q} is of the order $(a\xi_0)^{-1/2}$, and below T_M it slowly decreases with the decrease of temperature. Magnetization of the DS phase differs insignificantly from its value, which would be in the absence of superconductivity.

We shall also show that in the crystals with the MA of an easy-magnetization axis, critical magnetic fluctuations are suppressed by the EM interaction, except in the narrow temperature interval

$$\tau \equiv (T - T_M)/T_M \leq \tau_g \approx (a/\xi_0)^{2/3}.$$

For $\tau \gg \tau_g$, the fluctuations are of the ferromagnetic type.

The theory predicts that if the condition $\Theta_{EX} > \Theta_{EX}^c \approx T_{c1}^3/E_F T_M$ is fulfilled, then the first-order phase transition from the DS to the FN phase takes place, with the small jump of magnetization and with the latent heat of the order of superconducting condensation energy. (E_F is the Fermi energy.)

The results of this theory are in an agreement with the neutron scattering data⁶⁻⁸ for HoMo_6S_8 (the MA is of an easy-magnetization axis)—see Sec. VI. However, some conclusions seem to be in a contradiction with the data for ErRh_4B_4 .^{9,10}

We infer that the assumption of the second-order phase transition, from paramagnetic to ferromagnetic state (in the absence of superconductivity), is valid for the compound HoMo_6S_8 , but evidently it is not valid for ErRh_4B_4 . *Magnetic transition in ErRh_4B_4 may be of first order* in the absence of superconductivity. Magnetic measurements, done by Behroozi *et al.*,²¹ give evidence of this assumption. In Sec. V we shall describe the process of the formation of the DS phase—in the case of the first-order phase transition, of course. It is shown that the experimental data on ErRh_4B_4 may be explained by the DS phase theory, if one assumes that (a) it is a first-order magnetic transition and that (b) critical temperature Θ varies over the sample of the crystal because of inhomogeneous stresses.

II. FREE-ENERGY FUNCTIONAL OF MAGNETIC SUPERCONDUCTORS

A. Hamiltonian of the system

Let us consider the system of electrons with Cooper pairing and the system of LM's \vec{J}_i distributed regularly in the crystal lattice at sites $\{\vec{r}_i\}$. We shall treat the LM's in the mean-field approximation (MFA), i.e., \vec{J}_i are replaced by $\vec{S}_i \equiv \langle \vec{J}_i \rangle$. The MFA neglects exchange scattering of electrons on LM's. This scattering can be neglected since $\Theta_{EX} \ll T_{c1}$, and it is shown (see further text) that it is good approximation for ErRh_4B_4 and HoMo_6S_8 .

The Hamiltonian of the system is given by

$$\begin{aligned} \hat{H} = \int d^3r & \left[\frac{1}{2} \psi^\dagger(\vec{r}) \left[\vec{p} - \frac{e}{c} \vec{A} \right] (\hat{m}^{-1}) \left[\vec{p} - \frac{e}{c} \vec{A} \right] \psi(\vec{r}) + \Delta(\vec{r}) \psi^\dagger(\vec{r}) i \sigma_y \psi(\vec{r}) - \Delta^*(\vec{r}) \psi(\vec{r}) i \sigma_y \psi^\dagger(\vec{r}) \right. \\ & \left. + \sum_i \tilde{J}(\vec{r} - \vec{r}_i) \psi^\dagger(\vec{r}) \vec{\sigma} (g - 1) \hat{J}_i \psi(\vec{r}) + \frac{|\Delta(\vec{r})|^2}{g_{e-ph}} + \frac{\vec{B}^2}{8\pi} \right] \\ & + \sum_i [-B(\vec{r}_i) g \mu_B \hat{J}_i + \hat{H}_0(\hat{J}_i)] + \hat{H}_{sc}, \end{aligned} \quad (1)$$

$$\vec{B}(\vec{r}) = \text{rot} \vec{A}.$$

Here $\Delta(\vec{r})$ means the superconducting order parameter for the singlet pairing of electrons; $\psi(\vec{r})$ is spinor; $\vec{\sigma}$ are Pauli matrices; \vec{A} is the vector potential; $\tilde{J}(\vec{r})$ is the exchange integral; g_{e-ph} is the param-

eter of the electron-phonon coupling. The term $\hat{H}_0(\hat{J}_i)$ describes the effect of the crystal field, and \hat{H}_{sc} describes electron scattering on nonmagnetic impurities. Writing (1), we have supposed the model

of the effective electronic mass, where \hat{m}^{-1} is the tensor of the inverse mass. We have also neglected, in (1), the term which describes the effect of magnetic induction \vec{B} on the spins of conducting electrons, because this effect is small in comparison with the exchange one.

Using the Hamiltonian (1), one can get the free-energy functional $F[\vec{A}, \Delta, \vec{S}_i]$, where \vec{A} , Δ , and $\vec{S}_i \equiv \langle \vec{J}_i \rangle$ are unknown functions (see Refs. 13 and 14). Minimization of F with respect to \vec{A} gives the Maxwell equations for \vec{A} , and after exclusion of \vec{A} from them we get the functional $F[\vec{S}_i, \Delta(\vec{r}); T]$. The functional F can be written in the form

$$F = F_M[\vec{S}_i; T] + F_s[\Delta(\vec{r}); T] \\ + F_{\text{int}}[\vec{S}_i, \Delta(\vec{r}); T],$$

where F_M means the functional for the magnetic subsystem, F_s is for the superconducting subsystem, and F_{int} is the part which describes the interaction between magnetic and superconducting subsystems—it includes the EX and the EM interactions.

B. The magnetic functional

The magnetic free-energy functional can be written in the form

$$F_M[\vec{S}_i; T] = E_M[\vec{S}_i] + \sum E_0[\vec{S}_i; T],$$

where E_M describes the EX and the EM contributions to F_M and E_0 is the single-ion contribution to F_M . For $\hat{H}_0 = 0$, one has

$$E_0[\vec{S}_i] = -T \int_0^s b_s(x) dx,$$

where b_s is the inverse Brillouin function of the moment \vec{S}_i .

Let us consider the term E_M carefully. According to Eq. (1) the exchange field, which acts on superconducting electrons, is proportional to $\vec{S}_i = \sum_q \vec{S}_q e^{i\vec{q} \cdot \vec{r}_i}$. In the sum over \vec{q} there are terms with $|\vec{q}| \ll 2\pi n^{1/3}$, and terms with large wave vectors

$$\vec{q} + \vec{K},$$

where \vec{K} are the wave vectors of the inverse lattice of LM's, i.e., $|\vec{K}| \geq 2\pi n^{1/3}$. The latter terms are the consequence of the discreteness of the lattice, and they describe the short-wave part of the indirect exchange (RKKY) interaction of LM's. The influence of this rapidly oscillating part of the field on superconductivity is negligible, due to the small parameter $Q/K \approx (a/\xi_0)^{1/2}$ (see Refs. 13 and 22). The contribution of this part to E_M , together with the contribution from the direct exchange is described by the term

$$\left[- \sum_{\vec{q}} \Theta'_{\text{EX}} (1 - \vec{q} \cdot \hat{\alpha}_1 \cdot \vec{q}) \vec{S}_q \cdot \vec{S}_{-\vec{q}} \right].$$

Θ'_{EX} and the tensor $\hat{\alpha}_1$ depend on the electron band structure and the type of the lattice.

The long-wavelength component of the exchange field is given by

$$\vec{h}_{-\vec{q}} = (g-1)nJ(\vec{q})\vec{S}_q \equiv h_0\vec{S}_q,$$

where

$$h_0 = (g-1)nJ(0),$$

$$J(\vec{q}) = \int d^3r \vec{J}(\vec{r}) e^{i\vec{q} \cdot \vec{r}}.$$

When $q \ll 2\pi n^{1/3}$ it is correct to put $J(q) \approx J(0)$. The long-wavelength components of the exchange field influence superconductivity, and vice versa. This interaction is described by the term F_{int} (explained later).

The contribution of the long-wavelength part of the EX interaction to E_M is given by

$$- \sum_q \chi_n(\vec{q}) \vec{h}_q \cdot \vec{h}_{-\vec{q}} / 2\mu_B^2 \\ \equiv - \sum_{\vec{q}} \Theta_{\text{EX}} (1 - \vec{q} \cdot \hat{\alpha}_2 \cdot \vec{q}) \vec{S}_q \cdot \vec{S}_{-\vec{q}},$$

where $\chi_n(\vec{q})$ means the electronic paramagnetic susceptibility in the normal state; $\Theta_{\text{EX}} = N(0)h_0^2$, where $N(0)$ is the density of states (per LM) on the Fermi surface. The tensor $\hat{\alpha}_2$ is positive definite, and in the framework of the Hamiltonian (1) it has the form $\hat{\alpha}_2 = [(\hat{m}^{-1})/24]E_F$.

We shall also divide the EM interaction into the long- and short-range parts. The contribution of the EM interaction to E_M can be written in the form

$$E_{M_d} = -\frac{1}{2} \sum_i \vec{B}(\vec{r}_i) \cdot (\mu_B g \vec{S}_i), \quad (2)$$

where the field $\vec{B}(\vec{r}_i)$ (at \vec{r}_i) is created by all localized moments, except the i th dipole. Let us surround the i th dipole by the sphere of radius R so that $n^{-1/3} \ll R \ll 2\pi Q^{-1}$ (Ref. 23) is fulfilled. The interaction of all dipoles inside the sphere with the i th dipole represents the short-range part of the EM interaction, which does not interplay with superconductivity because $R \ll 2\pi Q^{-1} \ll \lambda_L$. The contribution of this part to E_M has the following form

$$- \Theta'_m \sum_q (1 - \vec{q} \cdot \hat{\alpha}_3 \cdot \vec{q}) \vec{S}_q \cdot \vec{S}_{-\vec{q}},$$

where the signs of Θ'_m and $\hat{\alpha}_3$ depend on the type of magnetic lattice. The interaction of the i th dipole with the dipoles outside the sphere may be described by the continuum approximation. By solving the

$$F_M[\vec{S}_i; T] = \sum_q \left[-\Theta_0(1 - \vec{q} \cdot \hat{\alpha} \cdot \vec{q}) \vec{S}_{\vec{q}} \cdot \vec{S}_{-\vec{q}} + \frac{1}{2} \Theta_m \frac{(\vec{q} \cdot \vec{S}_{\vec{q}})(\vec{q} \cdot \vec{S}_{-\vec{q}})}{q^2} \right] + \sum_i E_0[\vec{S}_i; T], \quad (3)$$

$$\Theta_m = 2\pi g^2 n \mu_B^2 S^2(0), \quad \hat{\alpha} = \hat{\alpha}_1 + \hat{\alpha}_2 + \hat{\alpha}_3, \quad \Theta_0 = \Theta'_{EX} + \Theta_{EX} + \Theta'_m + \frac{1}{3} \Theta_m.$$

We suppose, in (3), that $\Theta_0 > 0$, and $\vec{q} \cdot \hat{\alpha} \cdot \vec{q} > 0$, because we study the situation when the ground state of the system is ferromagnetic (in the absence of superconductivity). In Eq. (3) we use the normalized values \vec{S}_i , i.e., we replace \vec{S}_i by $S(0)\vec{S}_i$ with $|\vec{S}_i| \leq 1$. $S(0)$ is the value of the moment in the FN state at $T=0$.

Further, we shall study systems with an easy-magnetization axis anisotropy (in HoMo_6S_8 the crystal structure is of the rhombohedral type with small distortions from cubic symmetry), as well as systems with an easy-magnetization plane anisotropy (in ErRh_4B_4 the crystal structure is tetragonal with an easy-magnetization plane perpendicular to the c axis). Tensors \hat{m}^{-1} and $\hat{\alpha}$ are used in the diagonalized forms, i.e., $(\hat{m})_{ij}^{-1} = m_i^{-1} \delta_{ij}$, and $(\hat{\alpha})_{ij} = a_i^2 \delta_{ij}$.

C. The superconducting and interaction functionals F_s and F_{int}

As we shall see later, the wavelength Q^{-1} (of the nonuniform magnetic structure) is of order $(a\xi_0)^{1/2} \ll \xi_0, \lambda_L$. Therefore, the exchange field and magnetic induction oscillate rapidly, which means that we must consider the problem of superconducting pairing in the presence of the fields with the wavelength $q^{-1} \ll \xi_0, \lambda_L$. We shall study (and solve) this problem in the case of dirty superconductors with electronic mean free path l , which satisfies the following conditions: $\xi_0 \gg l \gg Q^{-1}$; $(lh_q/v_F)^2$, $(elA_q/c)^2 \ll 1$. These conditions ensure an effective averaging of the exchange field and magnetic induction over the distances of order ξ_0 , as well as an isotropization of the electronic motion. It turns out that the effect of the EX and EM fields on superconductivity (in dirty samples) is equivalent to the effect of magnetic impurities, as is shown in the Appendix. Moreover, for the compounds with $T \leq T_M \ll T_{c1}$ it is correct to put $T=0$ when we consider the superconducting part (F_s) of the free-energy functional. It is shown in the Appendix that under all these conditions, the coordinate dependence of $\Delta(r)$ can be neglected. The following func-

Maxwell equations one gets the long-range contribution to E_M (in the normal state). Summing all contributions of the EX and the EM interactions, we obtain the magnetic free-energy functional

tionals F_s and F_{int} are obtained:

$$F_s[\Delta] = -\frac{1}{2} N(0) \Delta^2 \ln \frac{e\Delta_0^2}{\Delta^2}, \quad e = 2.78$$

$$F_{\text{int}}[\vec{S}_{\vec{q}}, \Delta, \vec{B}_{\vec{q}}] = N(0) \left[\frac{\pi\Delta}{2\tau_m} - \frac{1}{3\tau_m^2} \right], \quad \tau_m \Delta \geq 1$$

$$\tau_m^{-1} = v_F^{-1} \sum_q \left[\frac{\pi \vec{h}_{\vec{q}} \cdot \vec{h}_{-\vec{q}}}{q} L_1(ql) + \frac{3\vec{B}_{\vec{q}} \cdot \vec{B}_{-\vec{q}}}{16\lambda_L^2 n N(0) q^3} L_2(ql) \right], \quad (4)$$

$$L_1(y) = \frac{2y \arctan y}{\pi(y - \arctan y)},$$

$$L_2(y) = \frac{2}{\pi} \left[\left[1 + \frac{1}{y^2} \right] \arctan y - \frac{1}{y} \right].$$

Here Δ_0 is the superconducting gap at $T=0$ in the absence of LM's. The induction $\vec{B}_{\vec{q}}$ is determined by the minimization of the Gibbs free energy, i.e., the sum of the long-range part of Eq. (2) and F_{int} . One gets

$$\vec{B}_{\vec{q}} = \frac{4\pi n g \mu_B [q^2 \vec{S}_{\vec{q}} - \vec{q}(\vec{q} \cdot \vec{S}_{\vec{q}})]}{q^2 + 4\pi P_0(q)(1 - 4/3\pi\tau_m\Delta)}, \quad (5)$$

where

$$P_0(q) = 3\pi\Delta L_2(ql) / 16v_F\lambda_L^2 q$$

means the electromagnetic kernel of a superconductor at $T=0$ (in the absence of LM's). In further considerations we assume that $Ql \gg 1$, which gives $L_1, L_2 \approx 1$. The case $Ql \ll 1$ will be discussed in Sec. III E.

D. The domainlike magnetic structure in the superconducting state

It follows from Eqs. (4) and (5) that the EM contribution to τ_m^{-1} is much smaller than that of the EX one, if $3\Theta_m/(\lambda_L Q)^2 \ll 2\Theta_{EX}$.¹⁴ Certainly, this condition is fulfilled in real compounds due to the large value $(\lambda_L Q)^2 \approx (\lambda_L^2/a\xi_0) \gg 1$. So we can reject the second term in the expression for τ_m^{-1} , and only retain the EX term in F_{int} , which determines magnetic structure in the superconducting state (together with terms quadratic in q).

If we omit the term $(3\tau_m)^{-2}$ in the expression for F_{int} , then F_{int} depends on $\vec{S}_{\vec{q}}$ and q in the same way as the magnetic field energy, which stems from a dissipation of magnetic flux lines out of the ferromagnetic plate. The latter may be obtained from the EX contribution (to F_{int}) by changing $\Theta_{EX}\Delta/v_F \approx \pi\Theta_{EX}/\xi_0$ by $\Theta_m/2L$, where L is the thickness of the ferromagnetic plate (see Ref. 24). We know that in such a plate the 180° one-dimensional plane-parallel domain structure is realized with the wave vector $Q \approx (aL)^{-1/2}$. This structure minimizes the free energy of the ferromagnetic plate. In that way, we argue that the same domainlike magnetic structure, but with $Q \approx (a\xi_0)^{-1/2}$, should minimize the total free-energy functional (see Fig. 1). In the next section we shall find all equilibrium parameters of the proposed DS phase minimizing $F[\vec{S}_{\vec{q}}, \Delta; T] = F_M + F_s + F_{int}$, and we shall prove that the 180° one-dimensional domain structure actually minimizes free energy.

III. STRUCTURE OF THE COEXISTING PHASE IN THE CASE OF THE SECOND-ORDER MAGNETIC TRANSITION

A. The Anderson-Suhl result near T_M

Now we consider the system with the second-order magnetic transition (in the absence of superconductivity). This implies the positive sign of the quartic term in the expansion of F_M over \vec{S} . Near the temperature Θ , the functional (3) has the usual Ginzburg-Landau (GL) form with the anisotropy energy

$$\epsilon_a(\vec{S}) = D(S_y^2 + S_z^2), \quad D > 0 \quad (6a)$$

$$\epsilon_a(\vec{S}) = DS_z^2 + \bar{D}S_x^2S_y^2, \quad D > 0 \quad (6b)$$

where (6a) describes an easy-magnetization axis anisotropy and (6b) an easy-magnetization plane anisotropy.

First, we shall study the system with an easy-magnetization axis anisotropy. In the vicinity of the transition temperature T_M ($\sim \Theta$) we can retain only

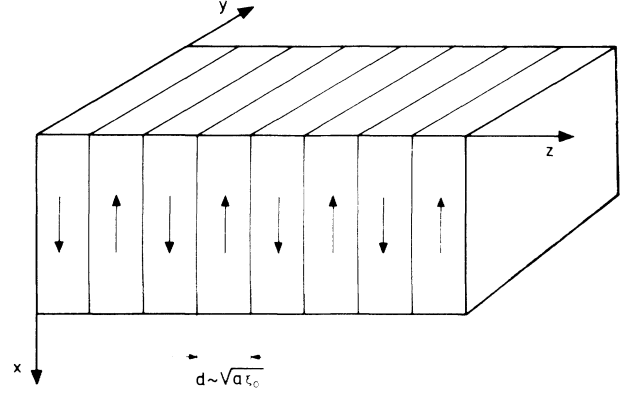


FIG. 1. Domainlike magnetic structure, in the DS phase, with the thickness of domain $d = \pi/Q$. The arrows show the direction of magnetization inside domains.

quadratic terms of S_{xq} in $F_M + F_{int}$. One sees that the transverse structure has the minimal energy, which is due to the EM contribution to F_M —the second term in the large parentheses of Eq. (3).

This means that \vec{Q} should be perpendicular to $\vec{S}(\vec{r})$, i.e., \vec{Q} should lie in the yz plane. In that case (at $T \approx T_M$), the sinusoidal structure

$$S_x(\vec{r}) \sim \sin(\vec{Q}_M \cdot \vec{r}),$$

$$Q_M = (\pi^2 \Theta_{EX} \Delta_0 / 4 \Theta_0 a^2 v_F)^{1/3}, \quad (7)$$

$$\vec{Q}_M = (0, Q_M \sin \phi, Q_M \cos \phi),$$

should appear, which is the consequence of Eqs. (3) and (4). The angle ϕ can be determined minimizing the term $a^2(\phi)/v_F(\phi)$. In Eq. (7), and from now on, it is understood that a and v_F take the value of $a(\phi)$ and $v_F(\phi)$ which minimizes the free energy.

The sinusoidal solution (7) is realized at the point T_M , and it transforms continuously into the domainlike solution—by lowering the temperature. The crossover region is very narrow, and for this reason we do not study a magnetic structure in this region. We remark only that in this region Q decreases from $(a^2 \xi_0)^{-1/3}$ [see Eq. (7)] to the value $\sim (a \xi_0)^{-1/2}$ (in the DS phase).

B. The energy of the domain wall

We now consider the temperature interval where the domain structure is well established, i.e., where the thickness of the domain wall (DW) is much smaller than the thickness ($d = \pi/Q$) of domain. In that case we can neglect the effect of F_{int} on the structure of the DW. Therefore, the DW structure is determined by F_M only. We shall find firstly the

surface energy ζ of the DW, and after that rewrite the functional $F_M[\vec{S}_{\vec{q}}; T]$ in the form

$$[-\Theta_0 S^2 - T\sigma(S) + \zeta(S, \phi; T) \cdot Q/\pi],$$

where S means the normalized value of the moment inside the domain, and therefore it is not coordinate dependent. As a result, F_M will depend on S, Q, ϕ instead of $\{\vec{S}_{\vec{q}}\}$.

To obtain the temperature dependence of the DS structure, we should know ζ for all temperatures $T < T_M$. The latter problem is solved by Ginzburg and one of the authors (L.N.B.) in the framework of the GL functional.²⁵ The GL approach is correct if

$$\tau = (T_M - T)/T_M \ll 1,$$

and we start from this case.

Let us determine the DW structure if Q is along the z axis. If $\tau < 2D/\Theta_0$, one gets the linear-type DW with the following coordinate dependence of magnetization:

$$S_x(z) = S \tanh\left[\frac{z\sqrt{\tau}}{a_z}\right], \quad S_y = S_z = 0, \quad (8)$$

$$\zeta = \frac{2\sqrt{2}}{3} a_z \Theta_0 S^2 \tau^{1/2}.$$

The DW thickness is $a_z/\sqrt{\tau}$, the condition $a_z/\sqrt{\tau} \ll d$ (or $a_z^2 Q^2/\pi^2 \ll \tau < 2D/\Theta_0$) determines the lower boundary on D , and the temperature interval where the DS phase is well established. F_{int} does not influence the DW structure—under the same conditions, of course.

If one has $2D/\Theta_0 < \tau \ll 1$, we get the DW with rotating moments

$$S_x(z) = S \tanh\left[\frac{z}{a_z} \left[\frac{D}{\Theta_0}\right]^{1/2}\right],$$

$$S_y(z) = \left[1 - \frac{2D}{\Theta_0 \tau}\right]^{1/2} S \cosh^{-1}\left[\frac{z}{a_z} \left[\frac{D}{\Theta_0}\right]^{1/2}\right], \quad (9)$$

$$S_z = 0, \quad \zeta = 2a_z S^2 (\Theta_0 D)^{1/2} \left[1 - \frac{2D}{3\Theta_0 \tau}\right],$$

which transforms into the Bloch DW when $2D/\Theta_0 \ll \tau$. For the arbitrary value of ϕ , the plane of rotation of moments is determined by the minimization of F_M including the EM interaction too. In this case we obtain solutions analogous to (8) and (9), but a_z should be replaced by $a(\phi)$ and D by the effective anisotropy parameter $D_e(\phi, \Theta_m, D, \bar{D})$. The surface energy depends, in this case, on S, ϕ , and T . For $D > \Theta_0$, the rotating solution is absent, and if τ is not very small one gets $\zeta = C_0 a(\phi) \Theta_0 S^2$, where C_0 is a numerical factor of order unity.

C. Equilibrium parameters of the DS phase

In the DS phase (for $\tau \gg a^2 Q^2/\pi^2$) $\vec{S}(\vec{r})$ is the periodic function with the period $2d$ and with the property $\vec{S}(\vec{r}) = -\vec{S}(\vec{r} + \vec{d})$. The function $\vec{S}(\vec{r})$ may be approximated by the step function $S_x(\vec{r}) = \pm S$, which is shown in Fig. 1. The Fourier expansion of $S_x(r)$ has the following form:

$$S_x(\vec{r}) = \frac{4S}{\pi} \sum_{n=0}^{\infty} \frac{\sin[(2n+1)\vec{Q} \cdot \vec{r}]}{2n+1},$$

$$S_y = S_z = 0. \quad (10)$$

Setting this function into the expressions for τ_m^{-1} in Eq. (4), one gets the free-energy functional of the DS phase— S is the variational parameter. As is mentioned above, the magnetic structure should be transverse with the wave vector $\vec{Q} = Q\{0, \sin\phi, \cos\phi\}$. So we have the following in the DS phase:

$$F[S, \Delta, Q, \phi; T] = -\Theta_0 S^2 + E_0[S; T] + \zeta(S, \phi, T)Q/\pi - \frac{1}{2}N(0)\Delta^2 \ln \frac{e\Delta_0^2}{\Delta^2} + N(0) \left[\frac{\pi\Delta}{2\tau_m} - \frac{1}{3\tau_m^2} \right], \quad (11)$$

$$\tau_m^{-1} = 8Ch_0^2 S^2 / \pi v_F(\phi)Q, \quad C = 1.05.$$

The minimum of F with respect to ϕ is reached for the minimal value of $\zeta(\phi)/v_F(\phi)$. From Eqs. (8) and (11) it follows that there exists a well-established domain structure when $\tau \gg (a/\xi_0)^{2/3}$. It follows also from Eq. (11) that the equilibrium value of $S(T)$ (in the DS phase) is practically the same as in the absence of superconductivity, but with the small shift of the critical temperature

$$\frac{\delta\Theta}{\Theta} \approx \frac{4C\Theta_{EX}}{\pi\xi_0 Q\Theta_0}. \quad (12)$$

For the equilibrium values $\Delta(T)$, $Q(T)$, and $F(T)$, we get [from Eq. (11)] the following expressions:

$$\Delta(T) = \Delta_0 e^{-\pi x/4}, \quad Q(T) = Q_0(T) e^{-\pi x/8} \left[1 - \frac{4x}{3\pi} \right]^{1/2}, \quad (13a)$$

$$F(T) = -\Theta_0 S^2 + E_0(T, S(T)) - \frac{1}{2} N(0) \Delta_0^2 e^{-\pi x/2} \left[1 - \frac{3\pi x}{2} + 2x^2 \right], \quad (13b)$$

$$Q_0^2(T) = \frac{4\pi C \Delta_0 S^2(T) \Theta_{EX}}{v_F \zeta(T)}, \quad \zeta(T) \equiv \zeta(S(T), T), \quad (13c)$$

where $x = (\Delta \tau_m)^{-1} \leq 1$ is determined by the equation

$$E(x) \equiv x e^{-3\pi x/8} \left[1 - \frac{4x}{3\pi} \right]^{1/2} = \frac{2}{\pi N(0) \Delta_0^2} \left[\frac{C \Delta_0 \zeta(T) S^2(T) \Theta_{EX}}{\pi v_F} \right]^{1/2}. \quad (13d)$$

Equation (13d) has one or two solutions, and the minimal value of F is reached for the minimal value of x . It is seen from Eqs. (13) that for $(a/\xi_0)^{2/3} \ll \tau \ll 1$, one has

$$Q(T) = 2.1 (\Theta_{EX}/a \xi_0 \Theta_0)^{1/2} \tau^{-1/4}.$$

In the temperature region $\tau \leq 1$, $Q(T)$ decreases slowly with the decrease of temperature due to the growth of $S(T)[\zeta(T)]^{1/2}$, on the right-hand side (rhs) of (13d), and corresponding growth of x . The superconducting parameter $\Delta(T)$ decreases by lowering temperature, and its value is of the order of Δ_0 (see Sec. III D); $Q(T)$ is of the order of $(a \xi_0)^{-1/2}$ for practically all temperatures where the DS phase exists. The temperature dependence of Q , Δ , and S in the DS and FN phases is shown in Fig. 2.

D. The first-order transition DS-FN

At the temperature T_{c2} , where the energies of the DS and FN phases are equal, the first-order phase transition takes place. The condition $F_{DS}(T_{c2}) = F_{FN}(T_{c2})$ gives the equation for x , i.e.,

$$2x^2 + 1 - 3x/2 = 0.$$

The solution is $x_{c2} = 0.235$, and

$$\frac{S_{c2}^2}{Q_{c2}} \equiv \frac{S^2(T_{c2})}{Q(T_{c2})} = 0.0766 \frac{\Delta_0 v_F N(0)}{\Theta_{EX}}. \quad (14)$$

We would like to point out that we consider the sample with the thickness $L \gg \xi_0$, which allows us to neglect the energy of magnetic field dissipated outside the sample.

At T_{c2} one has $\Delta_{c2} = 0.831 \Delta_0$, and the domain structure (with the wave vector Q_{c2}) disappears for $T < T_{c2}$ (see the comment on metastable states). The magnitude of S jumps at T_{c2} , from S_{c2} to $S_{c2} + \delta S$, where $\delta S \approx (a/\xi_0)^{1/2}$. The latent heat of this transition is given by

$$\begin{aligned} \tilde{q} &= \delta S \left[\frac{\partial^2 F_M(S, T)}{\partial S \partial T} \right]_{S_{c2}, T_{c2}} \\ &= 0.459 N(0) \Delta_0^2 \left[\frac{d \ln S^2}{d \ln T} \right]_{T_{c2}}. \end{aligned} \quad (15)$$

From Eq. (14) and the condition $S \leq 1$, it follows that the DS phase is stable down to $T = 0$ if

$$\Theta_{EX} < \Theta_{EX}^c \approx 0.025 N(0)^2 \Delta_0^3 v_F / a (D \Theta_0)^{1/2},$$

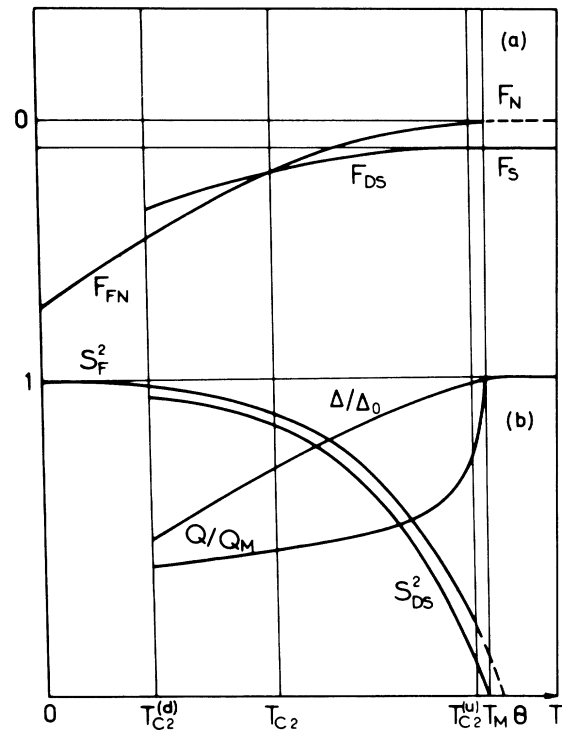


FIG. 2. Case of the second-order magnetic transition. (a) Free energies $F(T)$ of the N , S , DS , and FN phases. (b) Parameters S^2 , Δ , Q in the DS and the FN phase schematically.

where Θ_{EX}^c is of the order of $T_{c1}^3/T_M E_F$. The function $E(x)$, on the left-hand side (lhs) of Eq. (13), has a maximum at $X_{c2}^{(d)} = 0.68$, which means that the DS phase may exist down to $T_{c2}^{(d)} < T_{c2}$, where the supercooling temperature $T_{c2}^{(d)}$ is determined by Eq. (14), replacing T_{c2}, Q_{c2} by $T_{c2}^{(d)}, Q_{c2}^{(d)}$, and numerical factor 0.0766 by 0.146. Since $\xi(T)/S^2(T)$ does not depend on T for $T \leq T_{c2}$, then $S^2(T_{c2}^{(d)}) = 1.59S^2(T_{c2})$, $Q_{c2}^{(d)} = 0.75Q_{c2}$, and $\Delta_{c2}^{(d)} = 0.586\Delta_0$.

The overheating temperature $T_{c2}^{(u)}$ of the FN phase is determined by the condition (see Ref. 26)

$$S^2(T_{c2}^{(u)}) = \frac{\Delta_0}{2h_0} \left[1 + \left[\frac{2\pi\mu n \Delta_0}{h_0 H_{c2}^*(0)} \right]^2 \right]^{-1/2}.$$

(16)

For $h_0 \gg \Delta_0$, $T_{c2}^{(u)}$ practically coincides with T_M . At temperatures far from $T_{c2}^{(u)}$ the energy needed for the creation of critical nucleus of the DS phase (in the FN one) is large, because the domain walls must appear in the region of the size $\xi \approx (\xi_0 l)^{1/2}$. Therefore, the nucleation probability of the DS phase, in the metastable FN phase, is very small far from $T_{c2}^{(u)}$. So, we can go over, by heating, from the FN state directly to the nonmagnetic superconducting state without passing through the DS phase.

In Sec. III C we found that Q should decrease on cooling, i.e., the number of the domain walls should decrease. The wave vector Q may change its direction, with the change of temperature, because at T_M its direction is determined by the minimum of a^2/v_F , and far from T_M by the minimum of ξ/v_F . The change of the number of domain walls and their reorientation occurs by activation processes. This means that if the activation energy is large, the time for the change of the direction of \vec{Q} , may be long enough. For this reason, the change of the direction of \vec{Q} may go very slowly in real experiments. In this case one has to put, in the lhs of Eq. (14), the coefficient K , where $1 \leq K < 2$, and $K = 1$ for the equilibrium value of Q .

E. Conditions for the applicability of the obtained results

We shall give now the conditions on Θ_{EX} and D under which the DS phase is realized. The following conditions for the MA and the EX interaction are used in obtaining the domain magnetic structure: $D \gg \Theta_0 a^2 Q^2 / 2\pi^2$ and $2\Theta_{EX} \gg 3\Theta_m (\lambda_L Q)^{-2}$. We have used the following conditions on the electronic mean free path l : $Q \gg l^{-1}$ and $(h_0 l / v_F)^2 \ll 1$. These conditions permit us to treat the problem quantitatively. All the above conditions can be written in the following form:

$$(\Theta_{EX}/\Theta_0)^2 \gg (D/\Theta_0)^{1/2} a \xi_0 / \lambda_L^2,$$

$$D/\Theta_0 \gg (a \Theta_{EX} / \xi_0 \Theta_0)^{2/3}, \quad (17a)$$

$$\Theta_{EX}/\Theta_0 \ll E_F a^2 / \Theta l^2,$$

$$\Theta_{EX}/\Theta_0 \gg (D/\Theta_0)^{1/2} a \xi_0 / l^2. \quad (17b)$$

These conditions are fulfilled if $D/\Theta_0 \gg a/\xi_0$, $(a/l)^2$, and if

$$a^2 E_F / l^2 \Theta_0 \gg \Theta_{EX} / \Theta_0 \gg (a/\lambda_L), (a^2 \xi_0 / l^3).$$

There is no doubt that in reality, for not very dirty compounds, the EX interaction and the MA are sufficiently large to satisfy these conditions. If the first condition (17a) is reversed, then the magnetic structure (in the superconducting state) is determined by the EM interaction—the possible structures, in that case, are considered in Refs. 4, 5, 11, and 12. If the first condition is fulfilled but the second is reversed, then the helical structure is realized.^{13,14} If only the first condition in (17b) is reversed (very clean superconductors), then the DS phase is realized below T_M , and it is of the plane-parallel type. In that case, the region of its existence is broader than in the case considered here—these results will be published elsewhere. If only the second condition in (17b) is not fulfilled (very dirty superconductors), then the accurate expression for $L_1(ql)$ in Eq. (4) should be used. The region where the DS phase exists narrows when l decreases. If l approaches interatomic distances, the period d is drastically decreased, and the theory breaks down if the condition $d \gg n^{-1/3}$ is violated. We think that for such a small value of l the DS phase cannot be realized, and the first-order transition from nonmagnetic superconducting state to the FN one takes place.

In the study of the DS phase, the spin-orbital scattering has been neglected, which is correct if $Q^2 l_{SO} l \gg \pi^2$, where l_{SO} means the electronic mean free path for the spin-orbital scattering.²⁷ This condition is fulfilled as long as $l \gg a$.

In our consideration, the exchange scattering of electrons on LM's is neglected, i.e., on the spin-wave excitations in the DS phase. To take it into account, we must add the quantity τ_s^{-1} to τ_m^{-1} in Eq. (11), where τ_s means the exchange scattering time (see Appendix). The quantity τ_s does not depend on Q , because $Q \ll k_F$, but it may depend on T . The presence of τ_s modifies the function $E(x)$; the lhs of Eq. (13) should be multiplied by the factor $(1 - x_0 / x e^{\pi x / 4})$, where $x_0 \equiv (\tau_s \Delta_0)^{-1}$. Besides this, we should add the term $\pi x_0 e^{\pi x / 4} (1 - 4x / 3\pi)$ to the factor $(1 + 2x^2 - 3\pi x / 2)$ in the expression for $F(T)$ in Eq. (13b). The approximation $x_0 \approx 0$, which we used in the study of the DS phase, is correct for ErRh_4B_4 , where $x_0 \approx \Theta_{EX} / \Delta_0 \approx 0.04$, as well as for HoMo_6S_8 where $x_0 \approx 0.05$.

IV. CRITICAL MAGNETIC FLUCTUATIONS NEAR T_M

In the magnetic crystals with the critical temperatures $\Theta \gg \Theta_m$ the region of critical magnetic fluctuations is sufficiently large, due to the smallness of the characteristic length of the exchange interaction. For the systems like ErRh_4B_4 and HoMo_6S_8 , the critical temperature T_M is of the same order as Θ_m —which characterizes the long-range dipolar in-

$$F[S_{x,\vec{q}}, \Delta_0, T] = \sum_{|\vec{q}| \gg \xi_0^{-1}} S_{x,\vec{q}} S_{x,-\vec{q}} \left[\Theta_0 \left(a_i^2 q_i^2 + \frac{\pi \Theta_{\text{EX}} \Delta_0}{2 \Theta_0 (v_{F_i}^2 q_i^2)^{1/2}} + \tau \right) + \frac{1}{2} \Theta_m \frac{q_x^2}{q^2} \right]. \quad (18)$$

Fluctuations of the Fourier components of $S_x(r)$ with $q \leq \xi_0^{-1}$ near T_M are suppressed by superconductivity. If τ is much larger than the minimal value of the term

$$(a^2 q^2 + \pi \Theta_{\text{EX}} / 2 \xi_0 \Theta_0 q),$$

i.e., if $\tau \gg a^2 Q_M^2$, then fluctuations are practically the same as in a ferromagnet with an easy-magnetization axis MA and strong magnetic dipolar interaction. We know that in the latter case the fluctuations are four dimensional and logarithmically weak.²⁸ When $\tau \lesssim \tau_g \approx (a/\xi_0)^{2/3}$, fluctuations are strong, and in this region the peak at the wave vector Q_M appears. We think that below T_M the fluctuation region is characterized by the same parameter τ_g . This means that fluctuations are small in the temperature region where the domain structure is well established, and therefore the MFA is correct.

In the case of an easy-magnetization plane MA, the EM interaction does not suppress the transverse fluctuations of the moments in the easy-magnetization plane. However, for $\tau \gg a^2 Q_M^2 \approx (a/\xi_0)^{2/3}$, fluctuations are of the ferromagnetic type again, and the peak at Q_M appears only at $\tau \lesssim (a/\xi_0)^{2/3}$. So, we come to the conclusion that in systems like ErRh_4B_4 and HoMo_6S_8 fluctuations at $T > T_M$ are practically the same as in the absence of superconductivity, i.e., they are of the ferromagnetic type.

V. INHOMOGENEOUS MAGNETIC STRUCTURE IN SYSTEMS WITH THE FIRST-ORDER PHASE TRANSITION

A. The equilibrium state

Let us study which modifications of the theory, presented in Sec. III, are needed if the functional $F_M[S; T]$ gives the first-order transition $N \rightarrow \text{FN}$ at the point Θ . To obtain the equilibrium value $S(T)$

teraction. For this reason, in the crystals with an easy-magnetization axis anisotropy fluctuations are small at all temperatures except the very narrow region near T_M (see Ref. 37). To estimate the region of strong fluctuations, in systems with an easy-magnetization axis MA, we consider the quadratic part of the GL functional near T_M , i.e., for $T > T_M$. In the case where $\lambda_L > \xi_0$, this functional has the form

in superconducting (S) state, it is possible to neglect again the influence of superconductivity on $S(T)$. Therefore, at the temperature $T_M \approx \Theta$ the first-order phase transition takes place, from nonmagnetic S state to the S state with an inhomogeneous magnetic order. At T_M magnetization jumps from zero to S_M .

Parameters of the inhomogeneous magnetic structure are determined by Eqs. (12) and (13) with $S(T)$ and $\zeta(T)$, which correspond to the new functional $F_M[S; T]$. At $T < T_M$ the DW thickness is small compared with the domain thickness d , and here the DS phase is realized too. In this case the DW energy ζ depends weakly on T [the multiplicative factor $\tau^{1/2}$ in Eq. (8) is absent in the case of the first-order transition], and $\zeta(T) \approx a \Theta_0 S^2(T)$. By this reason, the value of Q changes very weakly with temperature, and $Q^{-1} \approx (\Theta_0 a \xi_0 / \Theta_{\text{EX}})^{1/2}$. The temperature dependences of F , S^2 , Δ , and Q in the DS, FN, S and N phases are shown in Fig. 3.

So, as far as we are interested in the equilibrium properties of magnetic structure, modifications of Sec. III for the description of the case when the first-order transition takes place are not essential. The difference is that now the domainlike magnetic structure should appear by a nucleation process in the nonmagnetic S state. In the following section we shall study the process of nucleation of the DS phase.

B. Formation of the DS phase by a nucleation process

Let us study the DS nucleation in the S phase at temperatures $T_{c2}^* < T < T_M$, where $F_{\text{DS}}(T) < F_S(T) < F_{\text{FN}}(T)$, and T_{c2}^* is determined by the condition $F_{\text{FN}}(T_{c2}^*) = F_S(T_{c2}^*)$, i.e., $F_{\text{FN}}(\Theta) - F_{\text{FN}}(T_{c2}^*) = N(0) \Delta_0^2 / 2$. At the beginning, the small region with ferromagnetic order appears. Its magnetization is directed along an easy-magnetization axis, and the

critical nuclei has the form of the rod stretched along the easy-magnetization axis. This is due to the magnetic dipole interaction (see Ref. 29).

Superconductivity has no effect on the creation of the critical nuclei, because its thickness is small in comparison with ξ_0 . The activation energy ϵ_a of the critical nuclei is²⁹

$$\epsilon_a = \frac{1}{2} \pi^2 \kappa^2 \xi^3 n \Theta_m^{1/2} (\delta F)^{-5/2}, \quad (19)$$

$$\kappa = 1 + \Theta_m / D, \quad \delta F = F_{FN}(\Theta) - F_{FN}(T),$$

where $\delta F(T)$ is the gain of the free energy due to the ferromagnetic ordering in normal state; $\delta F(T) \sim \Theta - T$ at temperatures near Θ .

When the critical (ferromagnetic) nucleus appears, it may grow infinitely in the form of a thin fer-

romagnetic plate (the thickness d of the plate being much smaller than ξ_0), with the magnetic moment parallel to the surface of the plate. Such a plate does not destroy overall Cooper pairing, and its free energy is lower than in the nonmagnetic S phase. This is due to the magnetic ordering. So, the initial ferromagnetic rod transforms into the plate, whose volume grows by the infinite increase of the plate area. We determine now (1) the maximum possible value of d , (2) the orientation of the plate, and (3) the temperature at which the area of the plate may grow infinitely. Let us calculate the free energy of the ferromagnetic plate with the thickness $d \ll \xi_0, l$ and with $(h\tau)^2 \ll 1$. In this case the free energy (per unit area) is given by

$$F_{PL}(d, T) = n [-\delta F(T)d + \zeta(T) + F_{int}(d, T)],$$

$$F_{int} = \int d\vec{q} \int d\vec{r}_1 \int d\vec{r}_2 e^{i\vec{q} \cdot (\vec{r}_1 - \vec{r}_2)} S^2(T) \left[\frac{1}{4} \Theta_{EX} [\chi_n(q) - \chi_s(q)] \mu_B^{-2} + \Theta_m \frac{4\pi P_0(q)}{q^2 + 4\pi P_0(q)} \right], \quad (20)$$

$$\frac{1}{2} [\chi_n(q) - \chi_s(q)] \mu_B^{-2} = \int d\omega \frac{2\Delta^2}{v_F q (\omega^2 + \Delta_0^2)} \arctan \frac{qv_F}{2(\omega^2 + \Delta_0^2)^{1/2}},$$

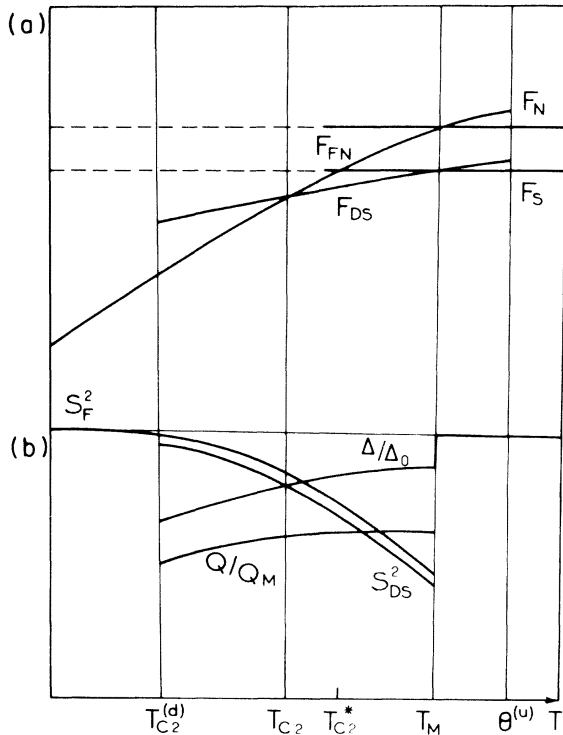


FIG. 3. Case of the first-order magnetic transition: (a) $F(T)$ in the N , S , DS , and FN phases. (b) S^2 , Δ , Q in the DS and the FN phase.

where $S(T)$ is the normalized magnetization inside the plate, which is practically the same as in the FN phase. The term F_{int} describes the increase of magnetic energy due to the change of electronic susceptibility on the presence of superconductivity. The integrations over r_1 and r_2 are over the volume of the plate. The calculation gives

$$F_{PL}(d, T) \cong n \left[-\delta F(T)d + \zeta(T) + \frac{1}{2} \Theta_{EX} S^2(T) \frac{d^2}{\xi_0} \ln \frac{\pi \xi_0}{8de^{1/2}} \right]. \quad (21)$$

The electromagnetic contribution to F_{int} is neglected, because it is smaller than the exchange one, due to the small parameter $\Theta_m d^2 / \Theta_{EX} \lambda_L^2$.

Minimizing F_{PL} with respect to d , one gets the thickness d_m of the ferromagnetic plate in the superconductor, i.e., $d_m \approx \xi_0 \delta F(T) / \Theta_{EX} S^2(T)$. The condition $F_{PL}(d, T) < 0$ determines the temperature at which the plate may appear. For $\delta F(T)$ one gets

$$\delta F(T) \geq (\Theta_{EX} S^2 \xi / \xi_0)^{1/2} \approx \Theta S^2 (a / \xi_0)^{1/2},$$

i.e.,

$$\Theta - T_M \approx \Theta (a / \xi_0)^{1/2},$$

and

$$d_m \gtrsim (\Theta_0 a \xi_0 / \Theta_{EX})^{1/2}.$$

We see that the condition $d \ll \xi_0$ is actually fulfilled near T_M , and $T_M > T_{c2}^*$ if $S(T_M) < S(T_{c2})$.

From Eq. (21) it follows that F_{PL} is minimal if the normal to the surface of the plate is perpendicular to the easy-magnetization axis. This means that the orientation of the plate is obtained from the minimization of the DW energy ξ .

Now, we can determine the activation energy of the critical nucleus at the temperature where the plate may grow infinitely (along an easy-magnetization axis). Here we may rewrite Eq. (19) in the following form:

$$\epsilon_a(T) \approx \frac{1}{2} \pi^3 \kappa^2 n a^3 (\xi_0 / d_m)^{5/2} \Theta, \quad d_m = \frac{\delta F(T)}{S^2 \Theta_{EX}}. \quad (22)$$

Therefore, the plate appears at T_M , where $F_{PL}(d, T) < 0$ and where $\epsilon_a(T)/T$ is not very large. These two conditions are fulfilled if $\Theta - T_M$ is of the order of $\Theta(a/\xi_0)^{1/2}$, and if $d_m \sim (a\xi_0)^{1/2}$.

Superconductivity stops further growth of the thickness of the plate above d_m at $T > T_{c2}^*$, because the destruction of superconductivity cannot be compensated by the gain of magnetic energy. So, further increase of the magnetically ordered region is possible by the creation of a new nucleus only. The appearance of the new nucleus is in its neighborhood, because the plate induces the exchange and magnetic fields at distances of ξ_0 and λ_L , respectively. Far enough from the plate, i.e., on distances $r \gg k_F^{-1}$, directions of induced fields are opposite to that of the moment inside the plate, due to superconducting overscreening. The value of the exchange field is approximately $-(4 \ln 2) \Theta_{EX} S d / \xi_0$ in the region $\xi_0 \gg r \gg k_F^{-1}$. This field causes the decrease of the activation energy of the second nucleus, and it turns out that for the second nucleus δF is increased by a factor of 3. This means that the activation energy of the second critical nucleus ϵ_a (near the initial plate) is only $\frac{1}{15}$ of ϵ'_a (ϵ'_a is the activation energy for the first critical nucleus). Practically, the second nucleus appear immediately after the formation of the first plate; this second nucleus transforms then into the second plate adjoined to the initial one—the moment of the second plate being opposite to the moment of the first one. The repetition of this process gives the transverse one-dimensional domainlike magnetic structure with the wave vector $Q_m = \pi/d_m$ and with $d_m \gtrsim (\Theta_0 a \xi_0 / \Theta_{EX})^{1/2}$. We remark that the parameter Q_m of this structure may be smaller than the Q value of the

equilibrium DS phase. If so, the formation of new domain walls is necessary in order to achieve the equilibrium state. The activation energy of such a process is large enough, and probably it is not possible to achieve the optimal value of Q in real experiments. In a such metastable DS phase only optimal values of $S(T)$ and $\Delta(T)$ are established, and they are determined by the minimization of the functional (11) at the given value $Q_m = \pi/d_m$.

On cooling, the transition DS \rightarrow FN may take place at T_{c2} determined by Eq. (14) with the coefficient K on the rhs. Below T_{c2} the part of domains may survive in the FN phase, due to the pinning of DW's on imperfections of the crystal.

VI. THEORETICAL RESULTS AND COMPARISON WITH EXPERIMENTAL DATA

A. Results in the case of the second-order transition

We summarize here the main theoretical predictions for the case of the second-order phase transition.

(a) The inhomogeneous magnetic structure of transverse domainlike-type should appear on cooling from T_M (at T_M the sinusoidal structure appears). This structure gives peaks at $(2n+1)Q$ in neutron scattering [$Q \approx (a\xi_0)^{-1/2}$], where intensities of peaks are proportional to $(2n+1)^{-2}$ for a monocrystal and to $(2n+1)^{-4}$ for a polycrystal.

(b) The equilibrium value of the normalized magnetization $S(T)$ inside domains is given by the same temperature dependence as in the absence of superconductivity, but with a small shift of the critical temperature—of the order of $(a/\xi_0)^{1/2}$ [see Eq. (12)]. Intensities of peaks at $(2n+1)Q$ are proportional to $S^2(T)$, and near T_M one has $S^2(T) \sim \tau$.

(c) The equilibrium value of Q decreases with the decrease of temperature, i.e., $Q \sim \tau^{-1/4}$ near T_M —see Fig. 2.

(d) Fluctuations are of the ferromagnetic type for $T > T_M$, and they are small in the crystal with an easy-magnetization axis MA. The crossover from the ferromagnetic-type fluctuations to those of the inhomogeneous structure takes place in the very narrow region near T_M —it is of order $\tau_g \approx (a/\xi_0)^{2/3}$.

(e) Bragg scattering, in the DS phase, should give the satellite peaks $[m \pm (2k+1)Q_x a / 2\pi, n \pm (2k+1)Q_y b / 2\pi, p \pm (2k+1)Q_z c / 2\pi]$ around the ferromagnetic peaks (m, n, p) of the FN phase, the ferromagnetic peaks being absent in the DS phase. The net intensity of all satellites is proportional to $S_{DS}^2(T)$; it differs from the ferromagnetic peak intensity $S_F^2(T)$, which would be observed in the absence of superconductivity, by the shift of the critical temperature $\delta\Theta$ —according to Eq. (12).

(f) The transition DS→FN takes place at T_{c2} for the systems with the parameter Θ_{EX} which satisfies $\Theta_{EX} > \Theta_{EX}^c$. The point T_{c2} is determined by Eq. (14). Owing to the pinning of domain walls, the part of domains with “wrong” direction of magnetization (relative to the main coherent structure) may be conserved at $T < T_{c2}$ (in the FN state). These residual domains give rise to the small-angle neutron scattering and to the broad diffuse component around ferromagnetic peaks in Bragg scattering.

(g) The FN phase remains metastable, on warming, up to the temperature $T_{c2}^{(u)}$ which is practically the same as T_M [see Eq. (16)]. The lifetime of the metastable FN phase is sufficiently large, and this phase may survive up to T_M (on warming). In this case the normalized intensity of the neutron scattering on small angles, as well as Bragg scattering, are proportional to $S_F^2(T)$ —the value which would be observed in the absence of superconductivity.

(h) At the transition DS→FN the jump of $S^2(T)$ is of the order of $(a/\xi_0)^{1/2}$, and the latent heat given by Eq. (15) should be observed.

B. Experimental data for HoMo_6S_8 and their interpretation

It is important to mention that up to now the polycrystalline samples of HoMo_6S_8 were studied only. Most of the theoretical predictions, stated above in (a)–(h), are confirmed by the following experimental results:

(a) The peak in neutron intensity (in the small-angle scattering) is observed, at $T_M \approx 0.7$ K on cooling, which comes from the transverse inhomogeneous magnetic structure with the wave vector $Q \approx 0.03$ Å. The peak at $2Q$ is absent with a great accuracy. The peak at $3Q$ is not observed too. This fact is not in a contradiction with the theoretical prediction, because its intensity should be very small in polycrystalline sample: $I_{3Q} < I_Q/81$.

(b) The intensity of the peak at Q is proportional to τ (near T_M), which means that $I_Q \sim S^2(T)$, ($S^2 \sim \tau$). This is typical for the second-order phase transition with very narrow fluctuation region (see Fig. 4).

(c) Q decreases slowly on cooling.⁶

(d) The peak at Q disappears at $T_{c2} \approx 0.65$ K on cooling, and at T_{c2} one has $S_{c2}^2 \approx 0.35$.^{7,8} This value for S_{c2}^2 is obtained from the normalized intensity of small-angle scattering. From the results of Sec. III D, the DS phase could survive down to $T_{c2}^{(d)}$, where $S^2(T_{c2}^{(d)}) \approx 0.56$ and $T_{c2}^{(d)} \approx 0.6$ K. Lynn *et al.*⁸ observed this metastable (DS) phase down to 0.62 K. The residual domains in the FN phase were observed in HoMo_6S_8 (Refs. 6–8) by the small-angle neutron scattering.

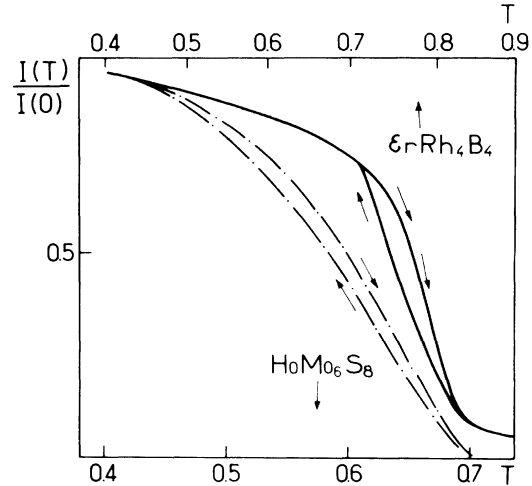


FIG. 4. Dashed lines are the normalized intensities of the Bragg peak (100) in HoMo_6S_8 on cooling and on warming (Ref. 8). The solid lines are the intensities of the (101) peak in ErRh_4B_4 (Ref. 10). Our proposal is as follows: the magnetic transition in HoMo_6S_8 is the second order at 0.7 K, while in ErRh_4B_4 it is the first order. The part of the curve from 0.75 K up to 1.2 K is due to inhomogeneous stresses.

(e) On cooling down to 0.65 K, the intensity of (100) Bragg peak is given by the same curve as on warming (in the FN phase) but with the shift of critical temperature $\delta\Theta/\Theta \approx 0.015$ (Fig. 4). With the use of Eq. (14) and the experimental values $S_{c2}^2 \approx 0.35$, $\Delta_0 \approx 3.5$ K (also using $\Theta_0 \approx \Theta$), we get the reasonable value for the density of electronic states $N(0)^{-1} \approx 3600$ K. Taking $v_F \approx 10^7$ cm/sec, one gets $\Theta_{EX} \approx 0.15$ K.

(g) The FN phase is preserved, on warming up to 0.7 K; this is observed in the small-angle neutron scattering experiments.^{6–8}

We point out here that, on warming from the FN phase, the intensity of the small-angle neutron scattering, as well as that of the Bragg peak, gives the function $S(T)$, which is very close to the Brillouin one. This fact confirms the assumption of the second-order phase transition in HoMo_6S_8 —in the absence of superconductivity. So, the proposed theory presented in Secs. III and IV is applicable to this compound, and it is in an agreement with experimental data.^{6–8} Comparison of experimental data on HoMo_6S_8 in a magnetic field^{7,8} with theoretical predictions in Ref. 30 gives additional confirmation of the theory presented in Secs. III and IV.

C. Experimental data for ErRh_4B_4 and their interpretation

Sinha *et al.*¹⁰ observed in the monocrystal of ErRh_4B_4 at $T < 1.2$ K (on cooling) the growth of

ferromagnetic peaks and of their satellites. At temperatures lower than 0.7 K the ferromagnetic peaks (F peaks) were observed only. The net intensity I_S of satellites is much less than the intensity I_0 of F peaks at all temperatures—it is reported that $I_S \lesssim 0.1I_0$. Besides that, the shape of the temperature dependence of $I_0(T)$ (on cooling) seems to be in contradiction with the assumption of the second-order phase transition, because the intensity of the F peak grows very rapidly—from 0.13 $I_0(0)$ up to 0.7 $I_0(0)$ near $T \approx 0.75$ K [see the data on $I_0(T)$ for ErRh_4B_4 in Fig.4]. On warming, the behavior is similar—a rapid decrease of intensity was observed at 0.8 K, and a width of hysteresis of order 0.06 K is present in the region $0.7 < T < 0.85$ K. Moreover, the broad diffuse component around each F peak at $T < 1.2$ K was observed (B line), caused by the presence of small ferromagnetic regions of sizes ~ 100 Å which are incoherent with the main ferromagnetic region (also in the FN phase). The volume of these incoherent regions is 9% of the ferromagnetic region at 0.75 K, and 5% at 0.54 K. Measurements of resistivity give the superconducting transition to the normal state at 0.73 K on cooling, and from FN to superconducting state at 0.8 K on warming.

The inhomogeneous magnetic ordering is characterized by the wave vector $Q_{\text{expt}} \approx 0.06 \text{ \AA}^{-1}$ —it is practically independent of temperature. The intensity of the $3Q$ peak (if it exists at all) is less than 2% of the intensity of the Q peak.

In polycrystalline samples of ErRh_4B_4 ,⁹ Bragg peaks appear at 1.4 K, and the hysteresis was observed in the interval 0.6–1.2 K. To explain these data we assume the following:

(1) The magnetic transition in ErRh_4B_4 in the absence of superconductivity would be of the first order, or very close to it—the transition temperature being Θ .

(2) The value of Θ varies over the sample investigated in Refs. 9 and 10 due to the inhomogeneous stresses—the distribution function of $\Theta f(\Theta)$ is introduced with the peak near $\Theta \approx 0.75$ K.

The first-order phase transition may be caused by magnetostriction or by the crystalline-field effect (a type of induced ferromagnetism³¹), i.e., the large magnetic contribution of the lowest excited doublet of Er^{3+} ions, the splitting of two lowest doublets being less than 1 K.³² The strong evidence for this assumption is the measurements of magnetization $M(H)$ done by Behroozi *et al.*,²¹ which show that the coefficient of the S^4 term, in the free-energy functional, is very small (near zero) and could be negative—they found the following dependence: $M \sim H^{1/5}$. We remark also that the discontinuous behavior of the value $dI_0(T)/dT$ (at $T \approx 0.75$ K on warming) and large hysteresis of $I_0(T)$ in monocrys-

tal¹⁰ give the evidence in favor of the first-order transition. The second assumption is a natural consequence of the first one, because the strong dependence of Θ on stresses is typical for induced magnetism. It may also explain the observation of Sinha *et al.*,¹⁰ that stresses arising from mounting give a preferential direction of moments along the a axis, but in the crystal without stresses the a and b axis are equivalent.

Independently of the type of transition, the DS phase appears at $T_M = \Theta - \delta\Theta$ [the value of $\delta\Theta$ is of the order $\Theta(a/\xi_0)^{1/2}$], and then the transition DS \rightarrow FN (on cooling) takes place when $S(T, \Theta)$ reaches the value S_{c2} [see Eq. (14) with the coefficient K on the lhs]. In different regions of the sample the transition occurs at different temperatures due to the distribution of Θ . Therefore, at some temperature T the following three phases are present in the sample: (a) the nonmagnetic superconducting S phase in the regions where $\Theta - \delta\Theta < T$, (b) the DS phase in the regions where $0 < S(T, \Theta - \delta\Theta) < S_{c2}$; (it gives satellites), (c) the FN phase with remnant domains in the regions where $S(T, \Theta - \delta\Theta) > S_{c2}$; it gives F peaks and B line.

For the regular domain structure we obtain the normalized intensities $I_S(T), I_0(T)$, and volumes V_S, V_0 of superconducting and normal phases, respectively,

$$I_S(T) = \frac{8}{\pi^2} \int_T^{\Theta_1(T)} f(\Theta) S^2(T, \Theta - \delta\Theta) d\Theta, \quad (23a)$$

$$V_S(T) = \int_T^{\Theta_1(T)} f(\Theta) d\Theta,$$

$$I_0(T) = \int_{\Theta_1(T)}^{\infty} f(\Theta) S^2(T, \Theta) d\Theta, \quad (23b)$$

$$V_0(T) = \int_{\Theta_1(T)}^{\infty} f(\Theta) d\Theta,$$

where Θ_1 is determined by the condition $S(T, \Theta_1 - \delta\Theta_1) = S_{c2}$ on cooling, and by the condition $S_{c2} \geq S(T, \Theta_1 - \delta\Theta_1) \geq S(\Theta^{(u)})$ on warming [in Eq. (23b)], because the FN phase may be overheated up to $\Theta^{(u)}$ (see Fig. 3). The possible increase of Θ_1 , on warming, is the origin of the hysteresis in the neutron scattering experiment and the measurements of the resistance.

We argue that the small relative net intensity I_S/I_0 is caused by the increase of $S(T, \Theta)$ up to S_{c2} in the very small temperature interval (T_M, T_{c2}) —it is much smaller than the interval of the Θ distribution, being narrow due to the first-order transition. In this picture, and in the crude approximation, $I_S(T)$ is proportional to the distribution function $f(T)$, and at $T > 0.75$ K we get

$$I_S(T) \approx \frac{8 \langle S_{DS}^2 \rangle}{\pi^2 \langle S_F^2 \rangle} (T_M - T_{c2}) \frac{dI_0}{dT}, \quad (24)$$

where $\langle S_{DS}^2 \rangle$, and $\langle S_F^2 \rangle$ are the mean values of $S^2(T)$ over the temperature interval where the DS and FN phase exist. The experimental data given in Ref. 10 are in accordance with the relation (24), giving $(T_M - T_{c2})\langle S_{DS}^2 \rangle / \langle S_F^2 \rangle \approx 0.02$ K. We remark that the value of Q does not depend on temperature (in this picture), because the DW energy ξ depends on Θ very weakly, and because the temperature interval (T_M, T_{c2}) , where the DS phase exists, is very small. The absence of the peak at $3Q$ may be explained by irregularities of domain structure, i.e., the irregularities of the domain thickness caused by the crystal imperfections. We remark that intensities of higher peaks decrease rapidly, when irregularities of the domain structure increase. The superconducting resistance transition should have the percolation character, and it occurs when $V_s(T) = 0.25$.³³

Finally, we can verify the condition (14) for ErRh_4B_4 . Taking the parameters presented later in Sec. VII, one gets $S_{c2}^2 \approx 1$ —the experimental value being ~ 0.5 .

So, our assumptions give the possibility for the explanation of the experimental data on ErRh_4B_4 .^{9,10} The second assumption is necessary to explain the simultaneous existence of F — and satellite—peaks, while the first assumption ensures the small intensity of satellites. The full quantitative description is impossible, at this moment, since the functions $f(\Theta)$, and $S(T, \Theta)$ are unknown. Magnetic measurements on monocrystalline sample of ErRh_4B_4 may give an additional information on the magnetic functional $F_M\{S; T\}$ and on $S(T)$. Measurements of magnetization and resistance under axial pressure may check our assumption of the strong dependence of Θ on stresses.

VII. PARAMETERS OF ErRh_4B_4 AND HoMo_6S_8 COMPOUNDS

Relevant energy, and length parameters of ErRh_4B_4 and HoMo_6S_8 compounds are given in Tables I and II.

The parameters l and electron-collision time τ for polycrystal samples of ErRh_4B_4 were obtained from the diffusion constant $D = 4.15$ cm²/sec. The value for v_F is obtained from the data for $N(0)$ and $H_{c1}, H_{c2}^*(0)$, at $T > 1.4$ K, using well-known expressions for H_{c1}, H_{c2}^* , and λ_L . The measurements of H_{c1} done by Schneider *et al.*³⁴ on polycrystals, give $H_{c1} \approx 1.5$ kOe at 2.5 K. Then we get $v_F \approx 2 \times 10^7$

cm/sec. The value for Θ_{EX} is estimated in the Introduction. The parameter a is estimated from the experimental value for d , using the following expressions: $\xi_0 = v_F / \pi \Delta_0$, $\Delta_0 = 1.76 T_{c1}$, and $d \gtrsim (\Theta_0 a \xi_0 / \Theta_{EX})^{1/2}$.

For HoMo_6S_8 , the value for $N(0)$ is taken from the data in Ref. 19; ξ is calculated from the curve $H_{c2}(T)$,¹⁷ and Θ_{EX} is estimated in the Introduction. All other parameters of this compound are unknown.

Finally, we can verify the conditions under which the theory of the DS phase is developed. It is assumed that the parameters $3\Theta_m / 2\Theta_{EX}(\lambda_L Q)^2$, $(h_0 \tau)^2$, $(\tau \Delta)$, $(eA_Q l / c)^2$, and $(Ql)^{-1}$ are small. Indeed, we obtain for the polycrystalline sample of ErRh_4B_4 , 0.006, 0.015, 0.06, 0.002, and 0.3, respectively.

VIII. SUMMARY AND CONCLUSIONS

Let us summarize the main results.

(a) The magnetic structure in the superconducting state is found.

(b) The EX interaction, in the presence of superconductivity, gives rise to very rapid spacial oscillations of magnetic moments, with the wave vector $Q \gg \xi_0^{-1}$.

(c) The EM interaction between superconducting electrons and LM's does not influence the magnetic structure. The direct dipole-dipole interaction between LM's make the structure transverse.

(d) The magnetic anisotropy (MA), with the EX interaction, makes the structure domainlike.

(e) The domain structure is one dimensional, because the superconducting condensation energy depends very weakly on the type of magnetic structure (for given value of the wave vector Q), while the energy of the DW's is minimal for the one-dimensional structure.

(f) If the transition $S \rightarrow DS$ is of the second order, then the DS phase develops continuously starting from the sinusoidal structure with a small amplitude. We argue that this situation is realized in HoMo_6S_8 .

(g) If the transition $S \rightarrow DS$ is of the first order, then the domain structure appears (in real experiments) by the creation of series of nuclei with alternating opposite directions of magnetization, the creation of the first critical nucleus being the bottle neck.

TABLE I. Energy parameters (measured in K).

	$N^{-1}(0)$	$v_F Q_{c2}$	τ^{-1}	h_0	Δ_0	Θ_m	Θ_{EX}
ErRh_4B_4	1850	840	250	30	15	1.3	0.5
HoMo_6S_8	3600			20	3.5	1.3	0.15

TABLE II. Characteristic length parameters (measured in Å).

	λ_L	ξ_0	l	$2d=2\pi/Q$	a	$\xi=(l\xi_0)^{1/2}$
ErRh ₄ B ₄	400	300	60	100	$\lesssim 4$	130
HoMo ₆ S ₈				200		250

(h) For the explanation of experimental data on the monocrystalline sample of ErRh₄B₄, we propose that there exists a first-order magnetic phase transition, in the absence of superconductivity, and (2) there is a strong dependence of the magnetic critical temperature on inhomogeneous stresses in the real sample.

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APPENDIX

We consider Cooper pairing of electrons in the presence of the exchange field $\mathbf{h}(\vec{r})=(0,0,h(\vec{r}))$ the vector potential $\vec{A}(\vec{r})$, and nonmagnetic impurities with scattering potential of the δ -function type.

The Green's function of electrons, averaged over random positions of impurities, is given by the expression

$$\hat{G}(\vec{r},\vec{r}')=\begin{bmatrix} G & -F \\ F^\dagger & \hat{G} \end{bmatrix}=\sum_{\vec{p}}\hat{G}(\vec{p},\vec{r})e^{i\vec{p}\cdot(\vec{r}-\vec{r}')}. \quad (\text{A1})$$

\hat{G} is the solution of Gor'kov equation $\hat{G}^{-1}\hat{G}=1$, where the matrix operator \hat{G}^{-1} is given by

$$\begin{aligned} \hat{G}^{-1}(\vec{p},\vec{r}) &= [-\xi + \vec{v}(i\nabla) + \hat{H}(\vec{r})], \\ \hat{H}(\vec{r}) &= i\left[\omega - \frac{ie}{c}\vec{v}\cdot\vec{A}\right]\hat{\tau}_z + h(\vec{r})\sigma_z\hat{\tau}_z + \Delta^*\sigma_x\hat{\tau}_- \\ &+ \Delta\sigma_x\hat{\tau}_+ - \frac{1}{2\tau}\int\frac{d\Omega}{4\pi}\hat{g}(\vec{v},\vec{r}), \end{aligned} \quad (\text{A2})$$

$$\hat{\tau}_\pm = \hat{\tau}_x \pm i\hat{\tau}_y, \quad \hat{g}(\vec{v},\vec{r}) = \int\frac{d\xi}{2\pi}\hat{G}(\vec{p},\vec{r}).$$

Here, ξ is the electron energy with respect to the Fermi energy; \vec{v} is the velocity on the Fermi surface. The integral over Ω means the integral over the direction of \vec{v} ; $\hat{\tau}$ and $\hat{\sigma}$ are Pauli matrices in "iso-spin" and spin space. The integration of the relation $\hat{G}^{-1}\hat{G} - \hat{G}\hat{G}^{-1} = 0$ over ξ gives the Eilenberger-type equations³⁵

$$-i(\vec{v}\cdot\vec{\nabla})\hat{g} = \hat{H}\hat{g} - \hat{g}\hat{H}. \quad (\text{A3})$$

In our case, we obtain from (A3) two equivalent matrix equations, in the 2×2 space for the spin projections $\pm\frac{1}{2}$. For the spin projection $-\frac{1}{2}$, we get

$$\begin{aligned} \left[\tilde{\omega} + ih(r) + i\frac{e}{c}\vec{v}\vec{A}(\vec{r}) + \frac{1}{2}\vec{v}\cdot\vec{\nabla}\right]f(\vec{v},\vec{r}) \\ = \tilde{\Delta}(\vec{r})g(\vec{v},\vec{r}), \\ \left[\tilde{\omega} + ih(r) + i\frac{e}{c}\vec{v}\vec{A}(\vec{r}) - \frac{1}{2}\vec{v}\cdot\vec{\nabla}\right]f^\dagger(\vec{v},\vec{r}) \\ = \tilde{\Delta}(\vec{r})g(\vec{v},\vec{r}), \end{aligned}$$

$$\begin{aligned} \tilde{\Delta}(r) &= \Delta(r) + \frac{1}{2\tau}\int\frac{d\Omega}{4\pi}f(\vec{v},\vec{r}), \\ \tilde{\Delta}^\dagger(\vec{r}) &= \Delta^*(\vec{r}) + \frac{1}{2\tau}\int\frac{d\Omega}{4\pi}f^\dagger(\vec{v},\vec{r}), \end{aligned} \quad (\text{A4})$$

$$\tilde{\omega}(\vec{r}) = \omega + \frac{1}{2\tau}\int\frac{d\Omega}{4\pi}g(\vec{v},\vec{r}),$$

$$f(\vec{v},\vec{r}) = -\int\frac{d\xi}{2\pi}F_{11}(\xi,\vec{v},\vec{r}),$$

$$f^\dagger(\vec{v},\vec{r}) = \int\frac{d\xi}{2\pi}F_{11}(\xi,\vec{v},\vec{r}),$$

$$g(\vec{v},\vec{r}) = i\int\frac{d\xi}{2\pi}G_{11}(\xi,\vec{v},\vec{r}).$$

Multiplying (A3) by \hat{g} one gets the equation

$$(\vec{v}\cdot\vec{\nabla})[g^2(\vec{v},\vec{r}) + f(\vec{v},\vec{r})f^\dagger(\vec{v},\vec{r})] = 0, \quad (\text{A5})$$

and for $h\rightarrow 0, \vec{A}\rightarrow 0$ we should have $g^2 + ff^\dagger = 1$. So

$$g^2(\vec{v},\vec{r}) + f(\vec{v},\vec{r})f^\dagger(\vec{v},\vec{r}) = 1. \quad (\text{A6})$$

Equations (A4) and (A6) together with the self-consistency equation

$$\Delta(\vec{r}) = \pi g_{e\text{-ph}}N(0)T\sum_{\omega>0}\int\frac{d\Omega}{4\pi}f(\vec{v},\vec{r}) \quad (\text{A7})$$

gives the complete system of equations for the functions $f(\vec{v}, \vec{r})$, $f^\dagger(\vec{v}, \vec{r})$, $\Delta(\vec{r})$, and $g(\vec{v}, \vec{r})$.

We solve these equations for the periodic function $\vec{S}(\vec{r}) = \vec{S}(\vec{r} + 2\vec{d})$, $\vec{S}(\vec{r}) = -\vec{S}(\vec{r} + \vec{d})$, where $2\vec{d}$ is the period of the magnetic structure. In general, the parameter $\Delta(\vec{r})$ is complex and its phase is $\phi(\vec{r})$. We choose the gauge so that $\Delta(\vec{r})$ is a real function. Then in (A4) $\vec{A}(\vec{r})$ should be changed by $\vec{A}_1 = \vec{A} - ic\nabla\phi/2e$. We write $h(r)$ in the form

$$ih(r) = \frac{1}{2} \sum_{m=0} h_{2m+1} (e^{i\vec{Q}\cdot\vec{r}(2m+1)} - e^{-i\vec{Q}\cdot\vec{r}(2m+1)}), \quad (\text{A8})$$

and similarly for $\vec{A}_1(r)$, where \vec{Q} is the inverse lattice vector of magnetic structure. We write the solution also in the form of Fourier expansion, i.e.,

$$f(\vec{v}, \vec{r}) = f_0(\vec{v}) + \sum_{k \neq 0} f_k(\vec{v}) e^{ik\vec{Q}\cdot\vec{r}}, \quad (\text{A9})$$

$$\Delta(\vec{r}) = \Delta + \sum_{k \neq 0} \Delta_k e^{ik\vec{Q}\cdot\vec{r}},$$

and the similar one for $f^\dagger(\vec{v}, \vec{r})$ and $g(\vec{v}, \vec{r})$.

Now we put (A8) and (A9) into (A4) and neglect the terms which are quadratic in g_k and f_k with $k \neq 0$. Later we shall see that these neglected terms are small, due to the small parameters $(h_q\tau)^2$ and $(e\ell A_q/c)^2$. Therefore, we obtain for $k \neq 0$

$$\bar{f}_0^2 + \bar{g}_0^2 = 1, \quad \omega \bar{f}_0 - \Delta \bar{g}_0 = -2\bar{f}_0 \bar{g}_0 \tau_m^{-1}, \quad (\text{A12})$$

$$\tau_m^{-1} = \frac{1}{2} \sum_{k \neq 0} \int \frac{d\Omega}{4\pi} \frac{\tau \left[\tilde{h}_k + \frac{\vec{v}}{c} \vec{A}_{1,+k} \right] \left[\tilde{h}_{-k} + \frac{\vec{v}}{c} \vec{A}_{1,-k} \right]}{1 + k^2 \tau^2 (\vec{v}\vec{Q})^2}.$$

Using (A12) and the self-consistency equation (A7), one gets the equation for Δ at $T=0$ in the form

$$\ln \frac{\Delta_0}{\Delta} - f(x) = 0, \quad x \equiv (\tau_m \Delta)^{-1} \quad (\text{A13a})$$

$$f(x) = \begin{cases} \pi x/4, & x \leq 1 \\ \text{arccosh}(x) + \frac{1}{2} \left[x \text{arcsinh} \left(\frac{1}{x} \right) - (1-x^{-2})^{1/2} \right], & x > 1. \end{cases} \quad (\text{A13b})$$

In obtaining (A13) we assumed that the Debye frequency $\omega_D \gg \Delta_0$. By solving (A13) we can calculate, in the usual way, the free energy $F_s[\Delta, \tau_m]$ of the superconductor for given value of $\tau_m[\vec{S}_q]$, add the functional $F_M[\vec{S}_q, T]$, and then minimize their

$$g_k(\vec{v}) = -\frac{f_0(\vec{v})}{2g_0(\vec{v})} [f_k(\vec{v}) + f_k(-\vec{v})],$$

$$f_k(\vec{v}) = \frac{1}{2} S_k(\vec{v}) \left[1 - \frac{i}{2} \frac{k(\vec{v}\cdot\vec{Q})}{\omega + \frac{1}{2\tau} \bar{g}_0} \right],$$

$$S_k(\vec{v}) = -\frac{2\tau f_0(\vec{v}) g_0(\vec{v}) \left[\tilde{h}_k + \frac{\vec{v}}{c} \vec{A}_{1k} \right]}{1 + k^2 \tau^2 (\vec{v}\vec{Q})^2}, \quad (\text{A10})$$

$$\tilde{h}_k = h_k \left[1 - \frac{\arctan(Qlk)}{Qlk} \right]^{-1},$$

$$\bar{g}_0 = \int \frac{d\Omega}{4\pi} g_0(\vec{v}),$$

$$\bar{f}_0 = \int \frac{d\Omega}{4\pi} f_0(\vec{v}).$$

To obtain (A10) we neglected also terms which contain the small parameter $\Delta\tau$. Using (A10), we get the equations for $g_0(\vec{v})$ and $f_0(\vec{v})$

$$g_0^2(\vec{v}) + f_0^2(\vec{v}) = 1, \quad (\text{A11})$$

$$\begin{aligned} \left[\omega + \frac{1}{2\tau} \bar{g}_0 \right] f_0(\vec{v}) - \left[\Delta + \frac{\bar{f}_0}{2\tau} \right] g_0(\vec{v}) \\ = \frac{1}{2} \sum_{k \neq 0} \left[h_{-k} + \frac{\vec{v}}{c} \vec{A}_{1,-k} \right] S_k(\vec{v}). \end{aligned}$$

From (A11), neglecting terms proportional to small parameters (see above), we get the following equations for \bar{g}_0 and \bar{f}_0 :

sum with respect to \vec{S}_q . In that way one gets the equilibrium magnetic structure. To find F_s , we know that minimization of F_s with respect to Δ should give (A13). We also know that for $T=0$ and $\tau_m^{-1}=0$ the equilibrium value of F_s is $-N(0)\Delta_0^2/2$.

From these conditions it follows unambiguously $F_s[\tau_m, \Delta]$, if we multiply the lhs of (A13a) by $2N(0)\Delta$, and integrate over Δ from $\Delta=0$ to Δ . The final result has very simple form

$$F_s[\tau_m, \Delta] = F_s[\Delta] + F_{\text{int}}[\tau_m, \Delta],$$

$$F_s[\Delta] = -\frac{1}{2}N(0)\Delta^2 \ln \frac{e\Delta_0^2}{\Delta^2}, \quad (\text{A14})$$

$$F_{\text{int}}[\tau_m, \Delta] = N(0) \left[\frac{\pi\Delta}{2\tau_m} - \frac{1}{3\tau_m^2} \right], \quad \tau_m\Delta \geq 1.$$

The expression for F_{int} is written only in the case $\tau_m\Delta \geq 1$, because this case is relevant for real sys-

tems (ErRh₄B₄, HoMo₆S₈). We have been considering from the beginning the case $\vec{h}(r) = (0, 0, h)$. The simple generalization to the case of arbitrary direction of $\vec{h}(r)$ and minimization of (A14) with respect to the phase ϕ of superconducting order parameter give the result (4).

From (A13), and (A14) we see that, the effect of magnetic structure of the DS phase on the superconducting condensation energy is similar to the effect of magnetic impurities with the magnetic scattering time τ_m .³⁶ By this reason, the effect of magnetic scattering, in the DS phase, may be taken into account replacing τ_m^{-1} in (A13) by the quantity $\tau_m^{-1} + \tau_s^{-1}$, where τ_s is the time of the exchange scattering due to the spin excitation in the DS phase.

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