Meissner phases in spin-triplet ferromagnetic superconductors

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We present new results for the properties of phases and phase transitions in spin-triplet ferromagnetic superconductors. The superconductivity of the mixed phase of coexistence of ferromagnetism and unconventional superconductivity is triggered by the presence of spontaneous magnetization. The mixed phase is stable but the other superconducting phases that usually exist in unconventional superconductors are either unstable or for particular values of the parameters of the theory some of them are metastable at relatively low temperatures in a quite narrow domain of the phase diagram. Phase transitions from the normal phase to the phase of coexistence is of first order while the phase transition from the ferromagnetic phase to the coexistence phase can be either of first or second order depending on the concrete substance. Cooper pair and crystal anisotropies determine a more precise outline of the phase diagram shape and reduce the degeneration of ground states of the system but they do not change drastically phase stability domains and thermodynamic properties of the respective phases. The results are discussed in view of application to metallic ferromagnets as UGe$_2$, ZrZn$_2$, URhGe.

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I. INTRODUCTION

In 2000, experiments [1] at low temperatures ($T \sim 1$ K) and high pressure ($P \sim 1$ GPa) demonstrated the existence of spin triplet superconducting states in the metallic compound UGe$_2$. The superconductivity is triggered by the spontaneous magnetization of the ferromagnetic phase that occurs at much higher temperatures. It coexists with the superconducting phase in the whole domain of its existence below $T \sim 1$ K; see also experiments from Refs. [2, 3], and the discussion in Ref. [4]. The same phenomenon of existence of superconductivity at low temperatures and high pressure in the domain of the $(T, P)$ phase diagram where the ferromagnetic order is present was observed in other ferromagnetic metallic compounds (ZrZn$_2$ [5] and URhGe [6]) soon after the discovery [1] of superconductivity in UGe$_2$.

In contrast to other superconducting materials, as ternary and Chevrel compounds, where the influence of magnetic order on superconductivity is also substantial (see, e.g., [7, 8, 9, 10]), in these ferromagnetic substances the phase transition temperature ($T_F$) to the ferromagnetic state is much higher than the phase transition temperature ($T_{FS}$) from ferromagnetic to a mixed state of coexistence of ferromagnetism and superconductivity. For example, in UGe$_2$, $T_{FS} = 0.8$ K while the critical temperature of the phase transition from paramagnetic to ferromagnetic state in the same material is $T_f = 35$ K [1, 2].

It can be assumed that in these substances the material parameter $T_s$ defined as the usual critical temperature of the second order phase transition from normal to uniform (Meissner) superconducting state in a zero external magnetic field is much lower than the phase transition temperature $T_{FS}$. The above mentioned experiments on the compounds UGe$_2$, URhGe, and ZrZn$_2$ do not give any evidence for the existence of a standard normal-to-superconducting phase transition in a zero external magnetic field.

It seems that the superconductivity in the metallic compounds mentioned above always coexists with the ferromagnetic order and is enhanced by it. In these systems, as claimed in Ref. [1], the superconductivity probably arises from the same electrons that create the band magnetism and can be most naturally understood rather as a triplet than spin-singlet pairing phenomenon. Metallic compounds UGe$_2$, URhGe, and ZrZn$_2$, are itinerant ferromagnets. An unconventional superconductivity is also suggested [11] as a possible outcome of recent experiments in Fe [12], in which a superconducting phase has been discovered at temperatures below 2 K and pressures between 15 and 30 GPa. There both vortex and Meissner superconducting phases [12] are found in the high-pressure crystal modification of Fe with a hexagonal close-packed lattice for which the strong ferromagnetism of the usual bcc iron crystal probably disappears [11]. It can be hardly claimed that in hexagonal Fe the ferromagnetism and superconductivity coexist but the clear
evidence for a superconductivity is also a remarkable achievement.

The reasonable question whether these examples of superconductivity and coexistence of superconductivity and ferromagnetism are bulk or surface effects can be stated. The earlier experiments performed before 2004 do not answer this question. Recent experiments show that surface superconductivity appears in ZrZn$_2$ and its presence depends essentially on the way of preparation of the sample. But in our study it is important that bulk superconductivity can be considered well established in this substance.

A phenomenological theory that explains the coexistence of ferromagnetism and unconventional spin-triplet superconductivity of Landau-Ginzburg type has been developed recently in Refs. 14, 15 where possible low-order couplings between the superconducting and ferromagnetic order parameters are derived with the help of general symmetry group arguments. On this basis several important features of the superconducting vortex state of unconventional ferromagnetic superconductors were established 14, 15.

In our paper we shall follow the approach from Refs. 14, 15 to investigate the conditions for the occurrence of the Meissner phase and to demonstrate that the presence of ferromagnetic order enhances the p-wave superconductivity. Also we extend the phase diagram of ferromagnetic superconductors in a zero external magnetic field and show that the phase transition to the superconducting state can be either of first or second order depending on the particular substance. We confirm the predictions made in Refs. 14, 15 about the symmetry of the ordered phases.

Our investigation is based on the mean-field approximation 16 as well as on known results about the possible phases in nonmagnetic superconductors with triplet (p-wave) pairing 17, 18, 19, 20. We extend our preceding results 21, 22, 23 and show that taking into account the anisotropy of the spin-triplet Cooper pairs modifies but does not drastically change the thermodynamic properties of the coexistence phase, especially in the temperature domain above the superconducting critical temperature $T_c$. The effect of crystal anisotropy is similar but we shall not make an overall thermodynamic analysis of this problem because we have to consider concrete systems and crystal structures 17, 20 for which there is not enough information from experiment to make conclusions about the parameters of the theory. Our results confirm the general concept that the anisotropy reduces the degree of ground state degeneration, and depending on the symmetry of the crystal, picks up a crystal direction for the ordering.

There exists a formal similarity between the phase diagram we obtain and the phase diagram of certain improper ferroelectrics 24, 25, 26, 27, 28, 29. We shall make use of the concept in the theory of improper ferroelectrics, where the trigger of the primary order parameter by a secondary order parameter (the electric polarization) has been initially introduced and exploited; see Refs. 24, 25, 26, 27, 28, 29. The mechanism of the M-triggered superconductivity in itinerant ferromagnets is formally identical to the mechanism of appearance of structural order triggered by the electric polarization in improper ferroelectrics.

Our aim is to establish the uniform phases which are described by the GL free energy presented in Sec. II. We investigate a quite general GL model in a situation of a lack of concrete information about the values of the parameters of this model for concrete compounds (UGe$_2$, URhGe, ZrZn$_2$) where the ferromagnetic superconductivity has been discovered. On the one hand the lack of information makes impossible a detailed comparison of the theory to available experimental data but on the other hand our results are not bound to one or more concrete substances and can be applied to any unconventional ferromagnetic superconductor. In Sec. III the M-trigger effect will be described when only a linear coupling of the magnetization $M$ to the superconducting order parameter $\psi$ is considered in a model of ferromagnetic superconductors where the spatial dependence of order parameters and all anisotropy effects are ignored. In Sec. IV we analyze the influence of quadratic coupling of magnetization to the superconducting order parameter on the thermodynamics of the ferromagnetic superconductors. The application of our results to experimental $(T, P)$ phase diagrams is discussed in Sec. IV.C. In Sec. V the anisotropy effects are outlined. In Sec. VI we summarize and discuss our findings.

## II. GINZBURG-LANDAU FREE ENERGY

The general GL free energy functional, we shall use in our analysis, is

$$F[\psi, M] = \int d^3x f(\psi, M),$$

(1)

where the free energy density $f(\psi, M)$ (hereafter called “free energy”) of a spin-triplet ferromagnetic superconductor is a sum of five terms 14, 15, 17, namely,

$$f(\psi, M) = f_S(\psi) + f_V(M) + f_{\psi}(\psi, M) + \frac{B^2}{8\pi}B.M.$$  

(2)

In Eq. (2) $\psi = \{\psi_j; j = 1, 2, 3\}$ is a three-dimensional complex vector describing the superconducting order and $B = (H + 4\pi M) = \nabla \times A$ is the magnetic induction; $H$ is the external magnetic field, $A = \{A_j; j = 1, 2, 3\}$ is the magnetic vector potential. The last two terms on r.h.s. of Eq. (2) are related with the magnetic energy which includes both diamagnetic and paramagnetic effects in the superconductor; see, e.g., 5, 30.

The term $f_S(\psi)$ in Eq. (2) describes the superconductivity for $H = M = 0$. It can be written in the form
\[ f_\psi(\psi) = f_{\text{grad}}(\psi) + a_s|\psi|^2 + b_s|\psi|^4 + u_s|\psi|^2 |\psi_j|^2 + v_s \sum_{j=1}^{3} |\psi_j|^4. \]  
\[ (3) \]

where a summation over the indices \( i,j = 1,2,3 \) is assumed and the symbol

\[ D_j = -i\hbar \frac{\partial}{\partial x_i} + \frac{2|\psi|}{c} A_j \]  
\[ (5) \]
of covariant differentiation is introduced. In Eq. \( (3) \), \( b_s > 0 \) and \( a_s = a_s(T - T_s) \), where \( a_s \) is a positive material parameter and \( T_s \) is the critical temperature of the standard second order phase transition which may occur at \( H = |\mathbf{M}| = 0 \); \( H = |\mathbf{H}| \), and \( \mathbf{M} = |\mathbf{M}| \). The parameters \( u_s \) and \( v_s \) describe the anisotropy of the spin-triplet Cooper pair and the crystal anisotropy, respectively. \[ (6) \]

Parameters \( K_j, (j = 1,2,3) \) in Eq. \( (4) \) are related with the effective mass tensor of anisotropic Cooper pairs. \[ (7) \]

The superconducting part \( (3) \) of the free energy \( f(\psi, \mathbf{M}) \) is derived from symmetry group arguments and is independent of particular microscopic models; see, e.g., Refs. \[ 17, 20 \]. According to classifications \[ 17, 20 \], the \( p \)-wave superconductivity in the cubic point group \( O_h \) can be realized through one-, two-, and three-dimensional representations of the order parameter. The expressions \( (3) \) and \( (5) \) incorporate all three possible cases. The coefficients \( b_s, u_s, \) and \( v_s \) in Eq. \( (3) \) are different for weak and strong spin-orbit couplings but in our investigation they are considered as undetermined material parameters which depend on the particular substance.

The free energy of a standard isotropic ferromagnet is given by the term \( f_0(\mathbf{M}) \) in Eq. \( (2) \),

\[ f_0(\mathbf{M}) = c_f \sum_{j=1}^{3} \left| \nabla_j \mathbf{M} \right|^2 + a_f(T_f') M^2 + \frac{b_f}{2} M^4, \]  
\[ (6) \]

where \( \nabla_j = \partial/\partial x_j \) and \( b_f > 0 \). The quantity \( a_f(T_f') = a_f(T - T_s) \) is expressed by the material parameter \( a_f > 0 \) and the temperature \( T_f' \) which is different from the critical temperature \( T_f \) of the ferromagnet and this point will be discussed below. We have already added a negative term \( -2\pi M^2 \) to the total free energy \( f(\psi, \mathbf{M}) \) and that is obvious by setting \( H = 0 \) in Eq. \( (2) \). The negative energy \( -2\pi M^2 \) should be added to \( f_0(\mathbf{M}) \). In this way one obtains the total free energy \( f(\psi, \mathbf{M}) \) of the ferromagnet in a zero external magnetic field that is given by a modification of Eq. \( (6) \) according to the rule

\[ f(\psi, \mathbf{M}) = f_0(\mathbf{M}) + \frac{\gamma_0}{\alpha} \mathbf{M} \cdot (\psi \times \psi^*) + \delta M^2 |\psi|^2. \]  
\[ (9) \]
transition line $T_{c2}(H)$ (see also, Ref. [33]) and applied it with respect to the magnetization $\mathcal{M}$ when $H = 0$ for small values of $|\psi|$ near the phase transition line $T_{c2}(\mathcal{M})$. We are interested in the uniform phases when the order parameters $\psi$ and $\mathbf{M}$ do not depend on the spatial vector $\mathbf{x} \in V$ ($V$ is the volume of the superconductor). Therefore, we present a detailed investigation of the coexistence of Meissner superconductivity and ferromagnetic order and, in particular, we show that the main properties of the uniform phases can be described when the crystal anisotropy is ignored. We claim that some of the main features of the uniform phases in unconventional ferromagnetic superconductors can be reliably outlined even when the Cooper pair anisotropy is neglected.

The magnetization $\mathbf{M}$ can be always assumed uniform outside a quite close vicinity of the magnetic phase transition when the superconducting order parameter $\psi$ is also uniform, i.e., vortex phases are not present in the respective temperature domain. These conditions are directly satisfied in type I superconductors but in type II superconductors the temperature should be sufficiently low and the external magnetic field should be zero. Nevertheless, in type II superconductors these requirements for the appearance of uniform superconducting states may turn insufficient in materials having very high values of the spontaneous magnetization. In this case the uniform (Meissner) superconductivity and, hence, its coexistence with uniform ferromagnetic order may not occur even at zero temperature. Up to now type I unconventional ferromagnetic superconductors are not found experimentally. The predominant amount of experimental data for UGe$_2$, URhGe, and ZrZn$_2$ do not give the possibility to conclude definitely either about the absence or the presence of uniform superconducting states at low and ultra-low temperatures but recently, an experimental evidence of uniform coexistence of superconductivity and ferromagnetism in UGe$_2$ has been reported [34].

If real materials can be modelled by the general GL free energy (1) - (9), their ground state properties will be described by uniform states. The problem about the availability of such states in real materials at finite temperatures is quite subtle at the present stage of experimental research. We shall assume that uniform phases can exist in some unconventional ferromagnetic superconductors, moreover these phases are solutions of the GL equations corresponding to the free energy (1) - (9). These arguments completely justify our study.

In case of a strong easy axis type of magnetic anisotropy, as is in UGe$_2$ [1], the overall complexity of mean-field analysis of the free energy $f(\psi, \mathbf{M})$ can be avoided by doing an Ising-like description: $\mathbf{M} = (0, 0, \mathcal{M})$, where $\mathcal{M} = \pm |\mathbf{M}|$ is the magnetization along the z-axis. Because of the thermodynamic equivalence of up and down physical states ($\pm \mathbf{M}$) the analysis can be done only for $\mathcal{M} \geq 0$. But this approach can be also supported without attracting crystal anisotropy arguments. When the symmetry of magnetic order is continuous, the symmetry of the total free energy $f(\psi, \mathbf{M})$ with respect to $\mathbf{M}$ comes into play and we can avoid the consideration of equivalent thermodynamic states that occur as a result of the respective symmetry breaking at the phase transition point but have no effect on thermodynamics of the system. In the isotropic system one may again choose the magnetization vector to point in the same direction as z-axis ($|\mathbf{M}| = \mathcal{M}$) and this will not influence the generality of thermodynamic analysis. Here we prefer an alternative description for which the ferromagnetic state can occur as two thermodynamically equivalent up and down domains with magnetizations $\mathcal{M}$ and $(-\mathcal{M})$, respectively.

We shall make the mean-field analysis of the uniform phases and the possible phase transitions between such phases in a zero external magnetic field ($\mathbf{H} = 0$) when the crystal anisotropy is neglected ($v_s \equiv 0$). The calculations will be more easy to understand if we use notations that reduce the number of parameters in $f(\psi, \mathbf{M})$ by introducing

$$b = (b_s + u_s + v_s). \quad (10)$$

Then we redefine the order parameters and all other parameters in the following way:

$$\varphi_j = b^{1/4}\psi_j = \phi_j e^{i\psi_j}, \quad M = b_f^{1/4} \mathcal{M}, \quad (11)$$

$$r = \frac{a_s}{\sqrt{b}}, \quad t = \frac{a_f}{\sqrt{b_f}}, \quad w = \frac{u_s}{b}, \quad v = \frac{v_s}{b}, \quad \gamma = \frac{\gamma_0}{b_{1/2} b_f^{1/4}}, \quad \gamma_1 = \frac{\delta}{(bb_f)^{1/2}}. \quad$$

With the help of Eqs. (10) - (11) and using the uniformity of $\psi$ and $\mathbf{M}$ we write the free energy density $f(\psi, \mathbf{M}) = F(\psi, \mathbf{M})/V$, in the form

$$f(\psi, \mathbf{M}) = r \phi^2 + \frac{1}{2} \phi^4 + 2\gamma_1 \phi_2 M \sin(\theta_2 - \theta_1) + \gamma_1 \phi^2 M^2 + t M^2 + \frac{1}{2} M^4 - 2w \left[ \phi_1^2 \phi_2^2 \sin^2(\theta_2 - \theta_1) + \phi_1^2 \phi_3^2 \sin^2(\theta_1 - \theta_3) + \phi_2^2 \phi_3^2 \sin^2(\theta_2 - \theta_3) \right] - v [\phi_1^2 \phi_2^2 + \phi_1^2 \phi_3^2 + \phi_2^2 \phi_3^2]. \quad (12)$$

In the above expression the order parameters $\psi$ and $\mathbf{M}$ are defined per unit volume.
The equilibrium phases are obtained from the equations of state
\[ \frac{\partial f(\mu_0)}{\partial \mu_\alpha} = 0, \tag{13} \]
where \( \mu = \{\mu_\alpha\} = (M, \phi_1, \ldots, \phi_3, \theta_1, \ldots, \theta_3) \) and \( \mu_0 \) denotes an equilibrium phase. The stability matrix \( \bar{F} \) of the phases \( \mu_0 \) is given by
\[ \bar{F}(\mu_0) = \{F_{\alpha\beta}(\mu_0)\} = \frac{\partial^2 f(\mu_0)}{\partial \mu_\alpha \partial \mu_\beta}. \tag{14} \]

An alternative treatment can be done in terms of real \((\psi'_j)\) and imaginary \((\psi''_j)\) parts of the complex numbers \( \psi_j = \psi'_j + i\psi''_j \). The calculation with moduli \( \phi_j \) and phase angles \( \theta_j \) of \( \psi_j \) is more simple but in cases of strongly degenerate phases some of the angles \( \theta_j \) remain unspecified. Then an alternative analysis with the help of the components \( \psi'_j \) and \( \psi''_j \) should be done.

The thermodynamic stability of the phases that are solutions of Eqs. (13) is checked with the help of the matrix (14). An additional stability analysis is done by the comparison of free energies of phases that satisfy (13) and render the stability matrix (14) positive in one and the same domain of parameters \( \{r, t, \gamma, \gamma_1, w, v\} \). This step is important because the complicated form of the free energy generates a great number of solutions of Eqs. (13) and we have to sift out the stable from metastable phases that correspond either to global or local minima of the free energy, respectively.

Some solutions of Eqs. (13) have a marginal stability, i.e., their stability matrix (14) is neither positively nor negatively definite. This is often a result of the degeneration of phases with broken continuous symmetry. If the reason for the lack of a clear positive definiteness of the stability matrix is precisely the mentioned degeneration of the ground state, one may reliably conclude that the respective phase is stable. If there is another reason, the analysis of the matrix (14) will be insufficient to determine the respective stability property. These cases are quite rare and occur for particular values of the parameters \( \{r, t, \gamma, \ldots\} \).

III. SIMPLE CASE OF M-TRIGGERED SUPERCONDUCTIVITY

We shall consider the Walker-Samokhin model \(^{15}\) when only the \( M\phi_1\phi_2 \)–coupling between the order parameters \( \psi \) and \( M \) is taken into account (\( \gamma > 0, \gamma_1 = 0 \)) and the anisotropies \( (w = v = 0) \) are ignored. The uniform phases and the phase diagram in this case were investigated in Refs. \(^{21, 22, 23}\). Here we summarize the main results in order to make a clear comparison with the new results presented in Sections IV and V. Our main aim is the description of a “trigger effect” which consists of the appearance of a “compelled superconductivity” caused by the presence of ferromagnetic order (here, this is a standard uniform ferromagnetic order); see also Refs. \(^{21, 22, 23}\) where this effect has already established and briefly discussed. As mentioned in the Introduction, a similar trigger effect is known in the physics of improper ferroelectrics. We shall set \( \theta_3 = 0 \) and use the notation \( \theta = \Delta \theta = (\theta_2 - \theta_1) \).

A. Phases

The possible (stable, metastable and unstable) phases are given in Table 1 together with the respective existence and stability conditions. The normal or disordered phase, denoted in Table 1 by \( N \), always exists (for all temperatures \( T \geq 0 \)) and is stable for \( t > 0, r > 0 \). The superconducting phase denoted in Table 1 by \( SC1 \) is unstable. The same is valid for the phase of coexistence of ferromagnetism and superconductivity denoted in Table 1 by \( CO2 \). The \( N \)–phase, the ferromagnetic phase (FM), the superconducting phases (SC1–3) and two of the phases of coexistence (CO1–3) are generic phases because they appear also in the decoupled case (\( \gamma = 0 \)). When the \( M\phi_1\phi_2 \)–coupling is not present, the phases SC1–3 are identical and represented by the order parameter \( \phi \) with components \( \phi_j \) that participate on equal footing. The asterisk attached to the stability condition of the second superconductivity phase (SC2) indicates that our analysis is insufficient to determine whether this phase corresponds to a minimum of the free energy. It will be shown that the phase SC2, two other purely superconducting phases and the coexistence phase CO1, have no chance to become stable for \( \gamma \neq 0 \). This is so, because the phase of coexistence of superconductivity and ferromagnetism (FS in Table 1), that does not occur for \( \gamma = 0 \), is stable and has a lower free energy in their domain of stability.

A second domain (\( M < 0 \)) of the FS phase exists and is denoted in Table 1 by \( FS^* \). Here we shall describe only the first domain FS. The domain \( FS^* \) is considered in the same way.

The cubic equation for magnetization of FS-phase (see Table 1) is shown in Fig. 1 for \( \gamma = 1.2 \) and \( t = -0.2 \). For any \( \gamma > 0 \) and \( t \), the stable FS thermodynamic states are given by \( r(M) < r_m = r(M_m) \) for \( M > M_m \), where \( M_m \) corresponds to the maximum of the function \( r(M) \). The dependence of \( M_m(t) \) and \( M_0(t) = (t + \gamma^2/2)^{1/2} = \sqrt{3}M_m(t) \) on \( t \) is drawn in Fig. 2 for \( \gamma = 1.2 \). Functions \( r_m(t) = M_m(t)/\gamma \) for \( t < \gamma^2/2 \) (depicted by the line of circles in Fig. 3) and
\[ r_e(t) = |t|^{1/2}, \tag{15} \]
for \( t < 0 \) define the borderlines of stability and existence of FS.

B. Phase diagram

We have outlined the domain in the \( (t, r) \) plane where the FS phase exists and is a minimum of the free en-
FIG. 1: $h = \gamma r/2$ as a function of $M$ for $\gamma = 1.2$, and $t = -0.2$. The parameters $r$, $t$, and $\gamma$ are given by Eq. (11).
FIG. 2: The magnetization $M$ versus $t$ for $\gamma = 1.2$: the dashed line represents $M_0$, the solid line represents $M_{eq}$, and the dotted line corresponds to $M_m$. 
FIG. 3: The phase diagram in the plane $(t, r)$ with two tricritical points (A and B) and a triple point C; $\gamma = 1.2$. The parameters $r \sim [T - T_s(P)]$ and $t \sim [T - T_f(P)]$ are defined by Eq. (11). The domains of existence and stability of the phases N, FM and FS are shown. The line of circles represents the function $r_m(t)$ given by Eq. (17). The dotted line represents the function $r_e(t)$ given by Eq. (15). On the left of point B, the same dotted curve corresponds to a FM-FS phase transition of second order. The equilibrium lines of N-FS and FM-FS phase transitions of first order are given by the solid lines AC and CB, respectively.
energy. For $r < 0$ the cubic equation for $M$ (see Table 1) and the existence and stability conditions are satisfied for any $M \geq 0$ provided $t \geq \gamma^2$. For $t < \gamma^2$ the condition $M \geq M_0$ have to be fulfilled, here the value $M_0 = (-t + \gamma^2/2)^{1/2}$ of $M$ is obtained from $r(M_0) = 0$. Thus for $r = 0$ the N-phase is stable for $t \geq \gamma^2/2$, and FS is stable for $t \leq \gamma^2/2$. For $r > 0$, the requirement for the stability of FS leads to the inequalities

$$\max \left( \frac{r}{\gamma}, M_m \right) < M < M_0,$$

where $M_m = (M_0/\sqrt{3})$ and $M_0$ should be the positive solution of the cubic equation of state from Table 1; $M_m > 0$ gives a maximum of the function $r(M)$; see also Figs. 1 and 2.

The further analysis defines the existence and stability domain of FS below the line AB denoted by circles (see Fig. 3). In Fig. 3 the curve of circles starts from the point A with coordinates $(\gamma^2/2, 0)$ and touches two other (solid and dotted) curves at the point B with coordinates $(t_B = -\gamma^2/4, r_B = \gamma^2/2)$. Line of circles represents the function $r(M_m) \equiv r_m(t)$ where

$$r_m(t) = \frac{4}{3\sqrt{3}\gamma} \left( \frac{\gamma^2}{2} - t \right)^{3/2}. \quad (17)$$

Dotted line represents $r_e(t)$, defined by Eq. (15). The inequality $r < r_m(t)$ is a condition for the stability of FS, whereas the inequality $r \leq r_e(t)$ for $(-t) \geq \gamma^2/4$ is a condition for the existence of FS as a solution of the respective equation of state. This existence condition for FS is obtained from $\gamma M > r$ (see Table 1).

In the region on the left of the point B in Fig. 3, the FS phase satisfies the existence condition $\gamma M > r$ only below the dotted line. In the domain confined between the lines of circles and the dotted line on the left of the point B the stability condition for FS is satisfied but the existence condition is broken. The inequality $r \geq r_e(t)$ is the stability condition of FM for $0 \leq (-t) \leq \gamma^2/4$. For $(-t) > \gamma^2/4$ the FM phase is stable for all $r \geq r_e(t)$.

In the region confined by the line of circles AB, the dotted line for $0 < (-t) < \gamma^2/4$, and the $t$–axis, the phases N, FS and FM have an overlap of stability domains. The same is valid for FS, the SC phases and CO1 in the third quadrant of the plane $(t, r)$. The comparison of the respective free energies for $r < 0$ shows that the stable phase is FS whereas the other phases are metastable within their domains of stability.

The part of the $t$-axis given by $r = 0$ and $t > \gamma^2/2$ is a phase transition line of second order which describes the N-FS transition. The same transition for $0 < t < \gamma^2/2$ is represented by the solid line AC which is the equilibrium transition line of a first order phase transition. The equilibrium transition curve is given by the function

$$r_{eq}(t) = \frac{1}{4} \left( 3\gamma - (\gamma^2 + 16t)^{1/2} \right) M_{eq}(t). \quad (18)$$

Here

$$M_{eq}(t) = \frac{1}{2\sqrt{2}} \left[ \gamma^2 - 8t + \gamma (\gamma^2 + 16t)^{1/2} \right]^{1/2}$$

is the equilibrium jump of the magnetization. The order of the N-FS transition changes at the tricritical point A.

The domain above the solid line AC and below the line of circles for $t > 0$ is the region of a possible overheating of FS. The domain of overcooling of the N-phase is confined by the solid line AC and the axes $(t > 0, r > 0)$. At the triple point C with coordinates $(0, r_{eq}(0) = \gamma^2/4)$ the phases N, FM, and FS coexist. For $t < 0$ the straight line

$$r_{eq}^*(t) = \frac{\gamma^2}{4} + |t|, \quad t_B < t < 0,$$

describes the extension of the equilibrium phase transition line of the N-FS first order transition to negative values of $t$. For $t < t_B$ the equilibrium phase transition FM-FS is of second order and is given by the dotted line on the left of the point B which is the second tricritical point in this phase diagram. Along the first order transition line $r_{eq}^*(t)$ given by Eq. (20), the equilibrium value of $M$ is $M_{eq} = \gamma/2$, which implies an equilibrium order parameter jump at the FM-FS transition equal to $(\gamma/2 - \sqrt{|t|})$. On the dotted line of the second order FM-FS transition the equilibrium value of $M$ is equal to that

| Phase | order parameter | existence conditions | stability domain |
|-------|-----------------|----------------------|-----------------|
| N     | $\phi_0 = M = 0$ | always               | $t > 0, r > 0$  |
| FM    | $\phi_0 = 0, M^* = -t$ | $t < 0$              | $r > 0, r > r_e(t)$ |
| SC1   | $\phi_1 = M = 0, \phi^* = -r$ | $r < 0$              | unstable       |
| SC2   | $\phi^* = -r, \theta = \pi k, M = 0$ | $r < 0$              | $(t > 0)^*$   |
| SC3   | $\phi_0 = \phi_2 = M = 0, \phi^* = -r$ | $r < 0$              | $r < 0, t > 0$  |
| CO1   | $\phi_1 = \phi_2 = 0, \phi^* = -r, M^* = -t$ | $r < 0, t > 0$      | unstable       |
| CO2   | $\phi_0 = 0, \phi^* = -r, \theta = \theta_2 = \pi k, M^* = -t$ | $r < 0, t < 0$      | unstable       |
| FS    | $2\phi^* = 2\phi_2^* = \phi^2 = -r + \gamma M, \phi_0 = 0, \phi_1 = 0, \theta = 2\pi (k - 1/4)$, $\gamma r = (\gamma^2 - 2t)M - 2M^3$ | $\gamma M > r$     | $3M^2 > (-t + \gamma^2/2)$ |
| FS*   | $2\phi^* = 2\phi_2^* = \phi^2 = -(r + \gamma M), \phi_0 = 0, \phi_1 = 0, \theta = 2\pi (k + 1/4)$, $\gamma r = (2t - \gamma^2)M + 2M^3$ | $\gamma M > r$     | $3M^2 > (-t + \gamma^2/2)$ |

**Table 1:** Phases and their existence and stability properties $[\theta = (\theta_2 - \theta_1), k = 0, \pm 1, \ldots]$.
of the FM phase ($M_{eq} = \sqrt{|t|}$). The FM phase does not exist below \( T_s \) and this is a shortcoming of the model \(^{12}\) with \( \gamma_1 = 0 \).

The equilibrium FM-FS and N-FS phase transition lines in Fig. 3 can be expressed by the respective equilibrium phase transition temperatures \( T_{eq} \) defined by the equations \( r_c = r(T_{eq}), \) \( r_g = r(T_{eq}) \), \( r_{eq}^* = r(T_{eq}) \), and with the help of the relation \( M_{eq} = M(T_{eq}) \). This limits the possible variations of parameters of the theory. For example, the critical temperature \( (T_{eq} \equiv T_c) \) of the FM-FS second order transition \( (\gamma^2/4 < -t) \) is obtained in the form \( T_c = (T_s + 4\pi JM/\alpha_3) \), or, using \( M = (-a_f/b_f)^{1/2} \),

\[
T_c = T_s - \frac{T^*}{2} + \left[ \left( \frac{T^*}{2} \right)^2 + T^*(T_f - T_s) \right]^{1/2}.
\]

Here \( T_f > T_s \), and \( T^* = (4\pi J)^2 a_f/\alpha_3^2 b_f \) is a characteristic temperature of the model \(^{12}\) with \( \gamma_1 = w = v = 0 \). A discussion of Eq. (21) is given in Sec. IV.C.

The investigation of the conditions for the validity of Eq. (21) leads to the conclusion that the FM-FS continuous phase transition \( (\gamma^2 < -t) \) is possible only if the following condition is satisfied:

\[
T_f - T_s > (\varsigma + \sqrt{\varsigma})T^*.
\]

where \( \varsigma = b_f r_0^2/4b_0\gamma r_0^2 \). Therefore, the second order FM-FS transition should disappear for a sufficiently large \( \gamma \)-coupling. Such a condition does not exist for the first order transitions FM-FS and N-FS.

The inclusion of the gradient term \( (4) \) in the free energy \(^{24}\) should lead to a depression of the equilibrium transition temperature. As the magnetization increases with the decrease of the temperature, the vortex state should occur at temperatures which are lower than the equilibrium temperature \( T_{eq} \) of the Meissner state. For example, the critical temperature \( (T_c) \) corresponding to the vortex phase of FS-type has been evaluated \(^{14}\) to be lower than the critical temperature \( (T_c) \) of the Meissner state.

For \( r > 0 \), namely, for temperatures \( T > T_s \) the superconductivity is triggered by the magnetic order through the \( \gamma \)-coupling. The superconducting phase for \( T > T_s \) is entirely in the \( (t, r) \) domain of the ferromagnetic phase. Therefore, the uniform superconducting phase can occur for \( T > T_s \) only through a coexistence with the ferromagnetic order.

In the next Sections we shall focus on the temperature range \( T > T_s \) which seems to be of main practical interest. We shall not dwell on the superconductivity in the fourth quadrant \( (t > 0, r < 0) \) of the \( (t, r) \) diagram where pure superconducting phases can occur for systems with \( T_s > T_f \), but this is not the case for UGe\(_2\), URhGe and ZrZn\(_2\). Also we shall not discuss the possible metastable phases in the third quadrant \( (t < 0, r < 0) \) of the \( (t, r) \) diagram.

\section{C. Magnetic susceptibility}

We consider the longitudinal magnetic susceptibility \( \chi_1 = (\chi_V/V) \) per unit volume \(^{23}\). The external magnetic field \( H = (0, 0, H) \) with \( H = (\partial f/\partial M) \) has the same direction as the magnetization \( M \). We shall calculate the quantity \( \chi = \sqrt{b_f}\chi_1 \) for the equilibrium thermodynamic states \( \mu_0 \) given by Eq. (13). Having in mind the relations (11) between \( M \) and \( \mathcal{M} \), and between \( \psi \) and \( \varphi \) we can write

\[
\chi^{-1} = \frac{d}{dM_0} \left[ \left( \frac{\partial f}{\partial M} \right)_{T,\varphi_f} \right]_{\mu_0},
\]

where the equilibrium magnetization \( M_0 \) and equilibrium superconducting order parameter components \( \varphi_{0j} \) should be taken for the respective equilibrium phase. See Table 1, where the suffix “0” of \( \phi \), \( \theta \), and \( M \) is omitted; hereafter this suffix will be often omitted. The value of the equilibrium magnetization \( M \) in FS is the maximal nonnegative root of the cubic equation in \( M \) given in Table 1.

From Eq. (23) we obtain the susceptibility \( \chi \) of FS phase in the form

\[
\chi^{-1} = -\gamma^2 + 2t + 6M^2.
\]

The susceptibility of the other phases has the usual expression

\[
\chi^{-1} = 2t + 6M^2.
\]

Eq. (25) yields as results the paramagnetic susceptibility \( (\chi_p = 1/2t; \ t > 0) \) of the normal phase and the ferromagnetic susceptibility \( (\chi_F = 1/4|t|; \ t < 0) \) of FM. These susceptibilities can be compared with the susceptibility \( \chi \) of FS which cannot be calculated analytically in the whole domain of the superconducting phase. Therefore, we shall consider the close vicinity of the N-FS and FM-FS phase transition lines.

Near the second order phase transition line on the left of the point \( B \) \((t < t_B)\), the magnetization has a smooth behavior and the magnetic susceptibility does not exhibit any singularities like jump or divergence. For \( t > \gamma^2/2 \), the magnetization is given by \( M = (s_- + s_+) \), where

\[
s_\pm = \left\{ \frac{-\gamma \sqrt{r}}{4} \pm \left( \frac{(t - \gamma^2/2)^3}{27} + \left( \frac{\gamma \sqrt{r}}{4} \right)^2 \right)^{1/3} \right\}^{1/3}.
\]

When \( r = 0 \), it is obvious that also \( M = 0 \). For \( |\gamma|s \ll (t - \gamma^2/2) \) we have \( M \approx \gamma \sqrt{r}/(2t - \gamma^2) \ll 2t \). Therefore, in a close vicinity \((r < 0)\) of \( r = 0 \) along the second order phase transition line \((r = 0, t > \gamma^2/2) \) the magnetic susceptibility is well described by the paramagnetic law \( \chi_p = (1/2t) \). For \( r < 0 \) and \( t \to \gamma^2/2 \), we obtain \( M = -(\gamma \sqrt{r}/2)^{1/3} \) which gives

\[
\chi^{-1} = 6 \left( \frac{\gamma |r|}{2} \right)^{2/3}.
\]
On the phase transition line $AC$

$$M_{eq}(t) = \frac{1}{2\sqrt{2}} \left[ \gamma^2 - 8t + \gamma \left( \gamma^2 + 16t \right)^{1/2} \right]^{1/2}$$

(28)

and, hence,

$$\chi^{-1} = -4t - \frac{\gamma^2}{4} \left[ 1 - 3 \left( 1 + \frac{16t}{\gamma^2} \right)^{1/2} \right].$$

(29)

At the tricritical point $A$ this result gives $\chi^{-1}(A) = 0$, and at the triple point $C$ with coordinates $(0, \gamma^2/4)$ we have $\chi(C) = (2/\gamma^2)$. On the line $BC$ we obtain $M = \gamma/2$ so

$$\chi^{-1} = 2t + \frac{\gamma^2}{2}.$$ 

(30)

At the tricritical point $B$ with coordinates $(-\gamma^2/4, \gamma^2/2)$ the result is $\chi^{-1}(B) = 0$.

To investigate the magnetic susceptibility tensor we shall consider arbitrary orientations of the vectors $H$ and $M$. We denote the spatial directions $(\mathbf{x}, \mathbf{y}, \mathbf{z})$ by $(1, 2, 3)$.

The components of the inverse magnetic susceptibility tensor

$$\chi^{-1}_{ij} = \chi^{-1}_{ij} \sqrt{b_j} = \{\chi^{-1}_{ij}\} \sqrt{b_j}$$

(31)

can be represented in the form

$$\chi^{-1}_{ij} = 2(t + M^2)\delta_{ij} + 4M_i M_j + i\gamma \frac{\partial}{\partial M_j} (\varphi \times \varphi^*)_i,$$ 

(32)

where $M$ and $\varphi_j$ are taken at their equilibrium values: $M_0, \varphi_{0j}, \theta_{0j}$. The last term in r.h.s. of Eq. (29) is equal to zero for all phases in Table 1 except for FS and FS'.

When the second term in Eq. (30) is equal to zero we obtain the known result of the susceptibility tensor for second order phase transitions; see, e.g., [16].

In FS phase $\varphi_j$ depend on $M_j$ and we can choose again $M = (0, 0, M)$ and use the results from Table 1 for the equilibrium values of $\varphi_j, \theta$ and $M$. Then the components $\chi^{-1}_{ij}$ corresponding to FS are

$$\chi^{-1}_{ij} = 2(t + M^2)\delta_{ij} + 4M_i M_j - \gamma^2 \delta_{13}.$$ 

(33)

Thus we have $\chi^{-1}_{i \neq j} = 0$,

$$\chi^{-1}_{11} = \chi^{-1}_{22} = 2(t + M^2),$$ 

(34)

and $\chi^{-1}_{33}$ coincides with the inverse longitudinal susceptibility $\chi^{-1}$ as given by Eq. (24).

D. Entropy and specific heat

The entropy $S(T) = (\tilde{S}/V) = -V \partial(f/\partial T)$ and the specific heat $C(T) = (\tilde{C}/V) = T(\partial S/\partial T)$ per unit volume $V$ are calculated in a standard way [16]. We are interested in the jumps of these quantities on the N-FM, FM-FS, and N-FS transition lines. The behavior of $S(T)$ and $C(T)$ near the N-FM phase transition and near the FM-FS phase transition line of second order on the left of the point $B$ (Fig. 3) is known from the standard theory of critical phenomena and for this reason we focus our attention on the first order phase transitions FS-FM and FS-N for $t > -\gamma^2/4$, i.e., on the right of the point $B$ in Fig. 3.

We make use of the equations for the order parameters $\psi$ and $M$ from Table 1 and apply the standard procedure for the calculation of $S$:

$$S(T) = -\frac{\alpha_s^2}{\sqrt{b_s}} \frac{\gamma^2}{\sqrt{b_f}} M^2.$$ 

(35)

The next step is to calculate the entropies $S_{FS}(T)$ and $S_{FM}$ of the ordered phases FS and FM. We shall stick to the usual convention $F_N = V f_N = 0$ for the free energy of the N-phase, so we must set $S(N) = 0$.

Near the second order phase transition line ($r = 0$, $t > \gamma^2/2$), $S_{FS}(T)$ is a smooth function of $T$ and has no jump but the specific heat $C_{FS}$ has a jump at $T = T_s$, i.e., for $r = 0$. This jump is given by

$$\Delta C_{FS}(T_s) = \frac{\alpha_s^2 T_s}{b_s} \left[ 1 - \frac{1}{1 - 2t(T_s)/\gamma^2} \right].$$ 

(36)

The jump $\Delta C_{FS}(T_s)$ is higher than the usual jump $\Delta C(T_c) = T_c \alpha_s^2 / b_s$ known from the Landau theory of standard second order phase transitions [16].

The entropy jump $\Delta S_{AC}(T) \equiv S_{FS}(T)$ on the line $AC$ is

$$\Delta S_{AC}(T) =$$

$$-M_{eq} \left\{ \frac{\alpha_s \gamma}{4\sqrt{b_s}} \left[ 1 + \left(1 + \frac{M_{eq}}{\gamma} \right)^{1/2} \right] - \frac{\alpha_f}{\sqrt{b_f}} M_{eq} \right\},$$

(37)

where $M_{eq}$ is given by Eq. (19). From Eqs. (19) and (37), we have $\Delta S(t = \gamma^2/2) = 0$, i.e., $\Delta S(T)$ becomes equal to zero at the tricritical point $A$. We find also from Eqs. (19) and (37) that at the triple point $C$ the entropy jump is

$$\Delta S(t = 0) = -\frac{\gamma^2}{4} \left( \frac{\alpha_s}{\sqrt{b_s}} + \frac{\alpha_f}{\sqrt{b_f}} \right).$$ 

(38)

On the line $BC$ the entropy jump is defined by $\Delta S_{BC}(T) = [S_{FS}(T) - S_{FM}(T)]$. We obtain

$$\Delta S_{BC}(T) = \left( |t| - \frac{\gamma^2}{4} \right) \left( \frac{\alpha_s}{\sqrt{b_s}} + \frac{\alpha_f}{\sqrt{b_f}} \right).$$ 

(39)

At the tricritical point $B$ this jump is equal to zero as should be. The calculation of the specific heat jump on the first order phase transition lines $AC$ and $BC$ is redundant for two reasons. Firstly, the jump of the specific heat at a first order phase transition differs from the entropy by a factor of order of unity. Secondly, in calorific experiments where the relevant quantity is the latent heat $Q = T \Delta S(T)$, the specific heat jump can hardly be distinguished.
**E. Note about a simplified theory**

The analysis in this Section can be done following an approximate scheme known from the theory of improper ferroelectrics; see, e.g., Ref. [24]. In this approximation the order parameter $M$ is considered small enough which makes possible to ignore $M^2$-term in the free energy. Then one easily obtains from the data for FS presented in Table 1 or by a direct calculation of the respective reduced free energy that the order parameters $\phi$ and $M$ of FS-phase are described by the simple equalities $r = (\gamma M - \phi^2)$ and $M = (\gamma/2\hbar)\phi^2$. For ferroelectrics working with oversimplified free energy gives a substantial departure of theory from experiment [29]. For ferromagnetic superconductors the domain of reliability of this approximation could be the close vicinity of a small part of phase diagram should be calculated numerically.

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**IV. EFFECT OF SYMMETRY CONSERVING COUPLING**

Here we shall include in our consideration both linear and quadratic couplings of magnetization to the superconducting order parameter which means that both parameters $\gamma$ and $\gamma_1$ in free energy (12) are different from zero. In this way we shall investigate the effect of the symmetry conserving $\gamma_1$-term in the free energy on the thermodynamics of the system. When $\gamma$ is equal to zero but $\gamma_1 \neq 0$ the analysis is easy and the results are known from the theory of birectical and tetracritical points [16, 24, 51, 38]. For the problem of coexistence of conventional superconductivity and ferromagnetic order the analysis ($\gamma = 0, \gamma_1 \neq 0$) was made in Ref. [3].

At this stage we shall not take into account any anisotropy effects because we do not want to obscure the influence of quadratic interaction by considering too many parameters. For $\gamma, \gamma_1 \neq 0$ and $w = 0, v = 0$ the results again can be presented in an analytical form, only a small part of phase diagram should be calculated numerically.

### A. Phases

The calculations show that for temperatures $T > T_s$, i.e., for $r > 0$, we have again three stable phases. Two of them are quite simple: the normal $(N)$- phase with existence and stability domains shown in Table 1, and the FM phase with the existence condition $t < 0$ as shown in Table 1, and a stability domain defined by the inequality $r_e^{(1)} \leq r$. Here

$$r_e^{(1)} = \gamma_1 t + \gamma \sqrt{-t},$$

and one can compare it with the respective expression (15) for $\gamma_1 = 0$. In this paragraph we shall retain the same notations as in Sec. III, but with a superscript (1) in order to distinguish them from the case $\gamma_1 = 0$. The third stable phase for $r > 0$ is a more complex variant of the mixed phase FS and its domain FS*, discussed in Sec. III. The symmetry of the FS phase coincides with that found in [15].

We have to mention that for $r < 0$ there are five pure superconducting $(M = 0, \phi > 0)$ phases. Two of them, $(\phi_1 > 0, \phi_2 = \phi_3 = 0)$ and $(\phi_1 = 0, \phi_2 > 0, \phi_3 > 0)$ are unstable. Two other phases, $(\phi_1 > 0, \phi_2 > 0, \phi_3 = 0, \theta_2 = \theta_1 + \pi k)$ and $(\phi_1 > 0, \phi_2 > 0, \phi_3 > 0, \theta_2 = \theta_1 + \pi k, \theta_3 = \text{arbitrary}; k = 0, \pm 1, \ldots)$ show a marginal stability for $r > \gamma_1 r$.

Only one of the five pure superconducting phases, the phase SC3, given in Table 1, is stable. In case of $\gamma_1 
= 0$ the values of $\phi_j$ and the existence domain of SC3 are the same as shown in Table 1 for $\gamma_1 = 0$ but the stability domain is different and is given by $t > \gamma_1 r$. When the anisotropy effects are taken into account the phases exhibiting marginal stability within the present approximation may become stable. Besides, three other mixed phases $(M \neq 0, \phi > 0)$ exist for $r < 0$ but one of them is metastable $(\gamma_1^2 > 1, t < \gamma_1 r$, and $r < \gamma_1 t)$ and the other two are absolutely unstable. Here the thermodynamic behavior for $r < 0$ is much more abundant in phases than for improper ferroelectrics with two component primary order parameter [27]. However, at this stage of experimental needs about the properties of unconventional ferromagnetic superconductors the investigation of the phases for temperatures $T < T_s$ is not of primary interest and for this reason we shall focus our attention on the temperature domain $r > 0$.

The FS phase for $\gamma_1 \neq 0$ is described by the following equations:

$$\phi_1 = \phi_2 = \frac{\phi}{\sqrt{2}}, \phi_3 = 0,$$  \hspace{1cm} (41)

$$\phi^2 = (\pm \gamma M - r - \gamma_1 M^2),$$  \hspace{1cm} (42)

$$(1 - \gamma_1^2)M^3 \pm \frac{3}{2} \gamma_1 M^2 + \left(\frac{r^2 - \gamma_1 r}{2}\right) M \pm \frac{\gamma r}{2} = 0,$$  \hspace{1cm} (43)

and

$$(\theta_2 - \theta_1) = \frac{\pi}{2} + 2\pi k,$$  \hspace{1cm} (44)
the existence condition \(\phi\) of stability of FS phase is to express with the help of the picture shown in Fig. 4.

Equation (43) becomes

\[ M_{eq} = \frac{\gamma}{2 \gamma_1} \]

(49)

and to substitute the above expression in the existence and stability conditions of FS-phase. It is obvious that there is a special value of \(M\)

\[ M_{S1} = \frac{\gamma}{2 \gamma_1} \]

(49)

for which this procedure cannot be applied and should be considered separately. Note, that \(M_{S1}\) is given by the respective horizontal dashed line in Fig. 4. The analysis shows that in the interval \(t_B^{(1)} < t < \gamma^2/2\) the phase transition is again of first order; here

\[ t_B^{(1)} = -\frac{\gamma^2}{4(1 + \gamma_1)^2}. \]

(51)

To find the equilibrium magnetization of first order phase transition, depicted by the thick line \(ACB\) in Fig. 4 we need the expression for equilibrium free energy of FS-phase. It is obtained from Eq. (12) by setting \((w = 0, v = 0)\) and substituting \(r, \phi_i\) as given by Eqs. (41), (42) and (48). The result is

\[ r_{eq}^{(1)}(t) = \frac{M_{eq}}{(\gamma M_{eq} - \gamma/2)} \left[ (1 - \gamma_1^2)M_{eq}^2 + \frac{3}{2} \gamma_1 M_{eq} + (t - \gamma^2/2) \right], \]

(48)

by the formula

\[ M_{eq}^{(1)} = \frac{\gamma}{2(1 + \gamma_1)}, \]

(53)

and is drawn by thick line \(CB\) in Fig. 4.

The existence and stability analysis shows that for \(r > 0\) the equilibrium magnetization of the first order phase transition should satisfy the condition \(M_{eq}^{(1)} < M_{eq}^{(1)} < M_0^{(1)}\).

By \(M_0^{(1)}\) we denote the positive solution of \(r^{(1)}(M_{eq}) = 0\) and its \(t\)-dependence is drawn in Fig. 4 by the curve.
with circles. \( M_m^{(1)} \) is the smaller positive root of stability condition (47) and also gives the maximum of function \( r_{eq}^{(1)} (M) \); see Eq. (48). The function \( M_m^{(1)} \) is depicted by the dotted curve AB in Fig. 4. When \( t_{S1} < t < M_0^{(1)} \), the existence and stability conditions are fulfilled if \( \sqrt{-t} < M < M_{S1} \), where \( \sqrt{-t} \) is the magnetization of ferromagnetic phase and is drawn by a thin black line on the left of point B in Fig. (4). Here we have two possibilities: \( r > 0 \) for \( \sqrt{-t} < M < M_0^{(1)} \) and \( r < 0 \) for \( M_0^{(1)} < M < M_{S1} \). To the left of \( t_{S1} \) and \( t > t_{S2} \), where

\[
t_{S2} = - \left( \frac{\gamma}{\gamma_1} \right)^2. \tag{54}
\]

the FS phase is stable and exists for \( M_{S1} < M < \sqrt{-t} \). Here \( r \) will be positive when \( M_0^{(1)} < M < \sqrt{-t} \) and \( r < 0 \) for \( M_0^{(1)} > M > M_{S1} \). When \( t < t_{S2} \), \( M < \sqrt{-t} \) and \( r \) is always negative.

On the basis of the existence and stability analysis we draw in Fig. 5 the \((t, r)\)-phase diagram for concrete values of \( \gamma \) and \( \gamma_1 \). As we have mentioned above the order of phase transitions is the same as for \( \gamma_1 = 0 \), see Fig. 3, Sec. III. The phase transition between the normal and FS phases is of first order and goes along the equilibrium line \( AC \) in the interval \( (t_A = \gamma^2/2 \) and \( t_C = 0) \). The function \( r_{eq}^{(1)} (t) \) is given by Eq. (48) with \( M_{eq}^{(1)} \) from Fig. 4.

N, FM, and FS phases coexist at the triple point \( C \) with coordinates \( t = 0 \), and \( r_{eq}^{(1)} = \gamma^2/4(\gamma_1 + 1) \). On the left of \( C \) for \( \gamma_1 < t < 0 \) the phase transition line of first order \( r_{eq}^{(1)} (t) \) is found by substituting in Eq. (48) the respective equilibrium magnetization, given by Eq. (53). In result we obtain

\[
r_{eq}^{(1)} (t) = 4(1 + \gamma_1)^{-1} - t. \tag{55}
\]

This function is illustrated by the line \( BC \) in Fig. 5 that terminates at the tricritical point \( B \) with coordinates \( t_B^{(1)} \) from Eq. (51), and

\[
r_B^{(1)} = \frac{\gamma^2(2 + \gamma_1)}{4(1 + \gamma_1)^2}. \tag{56}
\]

To the left of the tricritical point \( B \) the second order phase transition curve is given by the relation (40). Here the magnetization is \( M = \sqrt{-t} \) and the superconducting order parameter is equal to zero (\( \phi = 0 \)). This line intersects t-axis at \( t_{S2} \) and is well defined also for \( r < 0 \). The function \( r_{eq}^{(1)} (t) \) has a maximum at the point \( (t_{S1}, \gamma^2/4\gamma_1) \); here \( M = M_{S1} \). When this point is approached the second derivative of the free energy with respect to \( M \) tends to infinity. The result for the curves \( r_{eq}^{(1)} (t) \) of equilibrium phase transitions (N-FS and FM-FS) can be used to define the respective equilibrium phase transition temperatures \( T_{FS} \).

We shall not discuss the region, \( t > 0, r < 0 \), because we have supposed from the very beginning that the transition temperature for the ferromagnetic ordering \( T_f \) is higher then the superconducting transition temperature \( T_s \), as is for the known unconventional ferromagnetic superconductors. But this case may become of substantial interest when, as one may expect, materials with \( T_f < T_s \) may be discovered experimentally.

C. Discussion

The shape of the equilibrium phase transition lines corresponding to the phase transitions N-SC, N-FS, and FM-FS is similar to that of the more simple case \( \gamma_1 = 0 \) and we shall not dwell on the variation of the size of the phase domains with the variations of the parameter \( \gamma_1 \) from zero to values constrained by the condition \( \gamma_1^2 < 1 \). Our treatment from Sec. III of the magnetic susceptibility tensor and the thermal quantities can be generalized in order to demonstrate the dependence of these quantities on \( \gamma_1 \). We shall not consider such problems. But an important qualitative difference between the equilibrium phase transition lines shown in Figs. 3 and 5 cannot be omitted. The second order phase transition line \( r_{eq}^{(1)} (t) \) shown by the dotted line on the left of point \( B \) in Fig. 3, tends to large positive values of \( r \) for large negative values of \( t \) and remains in the second quadrant \( (t < 0, r > 0) \) of the plane \((t, r)\) while the respective second order phase transition line \( r_{eq}^{(1)} (t) \) in Fig. 5 crosses the t-axis at the point \( t_{S2} \) and is located in the third quadrant \((t > 0, r < 0)\) for all possible values \( t < t_{S2} \). This means that the ground state (at 0 K) of systems with \( \gamma_1 = 0 \) will be always the FS phase whereas two types of ground states, FM and FS, can exist for systems with \( 0 < \gamma_1^2 < 1 \). The latter seems more realistic when we compare theory and experiment, especially, in ferromagnetic compounds like UGe$_2$, URhGe, and ZrZn$_2$. Neglecting the \( \gamma_1 \)-term does not allow to describe the experimentally observed presence of FM phase at very low temperatures and relatively low pressure \( P \).

The final aim of the phase diagram investigation is the outline of the \((T, P)\) diagram. Important conclusions about the shape of the \((T, P)\) diagram can be made from the form of the \((t, r)\) diagram without an additional information about the values of the relevant material parameters \((a_s, a_f, ...)\) and their dependence on the pressure \( P \). One should know also the characteristic temperature \( T_s \), which has a lower value than the experimentally observed phase transition temperature \( T_{FS} \sim 1K \) to the coexistence FS-phase. A supposition about the dependence of the parameters \( a_s, a_f, ... \) on the pressure \( P \) was made in Ref. 15. Our results for \( T_f \gg T_s \) show that the phase transition temperature \( T_{FS} \) varies with the variation of the system parameters \((a_s, a_f, ...)\) from values which are higher than the characteristic temperature \( T_s \) down to zero temperature. This is seen from Fig. 5.
FIG. 4: The dependence $M(t)$ as an illustration of stability analysis for $\gamma = 1.2$, $\gamma_1 = 0.8$ and $w = 0$. The parameters of the theory $(r, t, \gamma, \gamma_1, w, \ldots)$ are defined by Eq. (11). The horizontal dashed lines represent the quantities $M_{S1}$ given by Eq. (49) and $M_{S2} = 2M_{S1}$. The line of circles $AS_1S_2$ describes the positive solution of Eq. (48). The thick line $AC$ gives the equilibrium magnetization for $t > 0$. The thick line $BC$ represents the equilibrium magnetization for $t < 0$ as given by Eq. (53). The dotted curve is the smaller positive solution of the stability condition (47). The thin solid line $BS_1S_2$ is the magnetization $M = \sqrt{-t}$. The arrow indicates the triple point $C$. $A$ and $B$ are tricritical points of phase transition. The point $S_1$ corresponds to the maximum of the curve (40) for $t < 0$, and the point $S_2$ corresponds to $r_e^{(1)}(t) = 0$ in Eq. (40).
FIG. 5: The phase diagram in the \((t, \gamma)\) plane for \(\gamma = 1.2, \gamma_1 = 0.8\) and \(w = 0\). The parameters of the theory \((r, t, \gamma, \gamma_1, w, \ldots)\) are defined by Eq. (11). The domains of stability of the phases N, FM and FS are indicated. A and B are tricritical points of phase transitions separating the dashed lines (on the left of point B and on the right of point A) of second order phase transitions from the solid line ABC of first order phase transitions. The FS phase is stable in the whole domain of the \((t, \gamma)\) plane below the solid and dashed lines. The vertical dashed line coinciding with the \(\gamma\)-axis above the triple point C indicates the N-FM phase transition of second order.
In systems where a pure superconducting phase is not observed for temperatures $T \sim T_f$ or $T \sim T_{FS}$, we can set $T_c \sim 0$ in Eq. (21). Neglecting $T_c$ in Eq. (21) and assuming that $(T^*/T_f) \ll 1$ we obtain that $T_c \equiv T_{FS} \sim (T^*/T_f)^{1/2}$. Note that the first $(T^*/T_f)^{1/2}$-correction to this result has a negative sign which means that a suitable dependence of the characteristic temperature $T^*$ on the pressure $P$ may be used in attempts to describe the experimental shape of the FM-FS phase transition line in the $(T, P)$ diagrams of UGe$_2$ and ZrZn$_2$; see, for example, Fig. 2 in Ref. 3, Fig. 3 in Ref. 4, Fig. 4 in Ref. 5. The experimental phase diagrams indicate that $T_f(P)$ is a smooth monotonically decreasing function of the pressure $P$ and $T_f(P)$ tends to zero when the pressure $P$ exceeds some critical value $P_c \sim 1$ GPa. Postulating the respective experimental shape of the function $T_f(P)$ one may try to give a theoretical prediction for the shape of the curve $T_{FS}$ describing the FM-FS phase transition line. The lack of experimental data about important parameters of the theory forces us to make some suppositions about the behavior of the function $T^*(P)$. The phase transition temperature $T_f$ will qualitatively follow the shape of $T_f(P)$ provided the dependence $T^*(P)$ is very smooth. This is in accord with the experimental shapes of these curves near the critical pressure $P_c$ where both $T_f$ and $T_{FS}$ are very small.

The substantial difference between $T_f$ and $T_{FS}$ at lower pressure ($P < P_c$) can be explained with the negative sign of the correction term to the leading dependence $T_{FS}(P) \sim [T^*(P)T_f(P)]^{1/2}$ mentioned above and a convenient supposition for the form of the function $T^*(P)$.

Eq. (21) presents a rather simplified theoretical result for $T_C \equiv T_{FS}$ because the effect of $M^2|\psi|^2$ coupling is not taken into account. But following the same ideas, used in our discussion of Eq. (21), a more reliable theoretical prediction of the shape of FM-FS phase transition line can be given on the basis of Eq. (40). Using the knowledge about the experimentally found shape of $T_f(P)$ and the definition of the parameters $r$ and $t$ by Eq. (11) we substitute $T = T_{FS}(P)$ in Eq. (40). In doing this we have applied the following approximations, namely, that $T_s \sim 0$ for any pressure $P$, $T_{FS}(P_s) \sim T_f(P_s) \sim 0$ and for substantially lower pressure ($P < P_c$), $T_f(P) \gg T_{FS}(P)$.

Then near the critical pressure $P_c$, we easily obtain the transition temperature $T_{FS} \sim 0$, as should be. For substantially lower values of the pressure there exists an experimental requirement $(T_{FS} - T_s) \ll (T_f - T_{FS})$. Using the latter we establish the approximate formula $(T_f - T_{FS}) = \gamma^2 b_{ij}/\gamma^2_0 \alpha_f$. The same formula for $(T_f - T_{FS})$ can be obtained from the parameter $t_{FS}(T_{FS})$ given by Eq. (54). The pressure dependence of the parameters included in this formula defines two qualitatively different types of behavior of $T_{FS}(P)$ at relatively low pressures ($P \ll P_c$): (a) $T_{FS}(P) \sim 0$ below some (second) critical value of the pressure ($P_r^2 < P_c$), and (b) finite $T_{FS}(P)$ up to $P \sim 0$. Therefore, we can estimate the value of the pressure $P_c < P_r$ in UGe$_2$, where $T_{FS}(P_c) \sim 0$. It can be obtained from the equation $T_f(P_r') = (\gamma^2 b_{ij}/\gamma^2_0 \alpha_f)$ provided the pressure dependence of the respective material parameters is known.

So, the above consideration is consistent with the theoretical prediction that the dashed line in Fig. 5 crosses the axis $r = 0$ and for this reason we have the opportunity to describe two ordered phases at low temperatures and broad variations of the pressure. Our theory allows also a description of the shape of the transition line $T_{FS}(P)$ in ZrZn$_2$ and URhGe, where the transition temperature $T_{FS}$ is finite at ambient pressure. To avoid a misunderstanding, let us note that the diagram in Fig. 5 is quite general and the domain containing the point $r = 0$ of the phase transition line for negative $t$ may not be permitted in some ferromagnetic compounds.

Up to now we have discussed experimental curves of second order phase transitions. Our analysis gives the opportunity to describe also first order phase transition lines. Our investigation of the free energy (12) leads to the prediction of triple $(C)$ and tricritical points $(A$ and $B)$; see Figs. 3 and 5. We shall not dwell on the possible application of these results to the phase diagrams of real substances, where first order phase transitions and multicritical phenomena occur; see, e.g., Refs. 34, 35, where first order phase transitions and tricritical points have been observed. The consideration of such problems, in particular, the explanation of the phase transition lines in Refs. 34, 35 requires further theoretical studies, that can be done on the basis of a convenient extension of the free energy (12). For example, the investigation of vortex phases in Ref. 34 needs taking into account the gradient terms (4). Another generalization should be done in order to explain the observation of two FM phases in Refs. 36, 37. Note, that the experimentalists are not completely certain whether the FS phase is a uniform or a vortex phase, and this is a crucial point for the orientation of the further investigations. But we find quite encouraging that our studies naturally lead to the prediction of the same variety of phase transition lines and multicritical points that has been observed in recent experiments 36, 37.

V. ANISOTROPY EFFECTS

Our analysis demonstrates that when the anisotropy of Cooper pairs is taken in consideration, there will be no drastic changes in the shape the phase diagram for $r > 0$ and the order of the respective phase transitions. Of course, there will be some changes in the size of the phase domains and the formulae for the thermodynamic quantities. It is readily seen from Figs. 6 and 7 that the temperature domain of first order phase transitions and the temperature domain of stability of FS above $T_s$ essentially vary with the variations of the anisotropy parameter $w$. The parameter $w$ will also insert changes in the values of the thermodynamic quantities like the magnetic susceptibility and the entropy and specific heat jumps at the phase transition points.

Besides, and this seems to be the main anisotropy ef-
fect, the $w$- and $v$-terms in the free energy lead to a stabilization of the order along the main crystal directions which, in other words, means that the degeneration of the possible ground states (FM, SC, and FS) is considerably reduced. This means also a smaller number of marginally stable states.

The dimensionless anisotropy parameter $w = u_s/(b_s + u_s)$ can be either positive or negative depending on the sign of $u_s$. Obviously when $u_s > 0$, the parameter $w$ will be positive too and will be in the interval $0 < w < 1$ to ensure the positiveness of parameter $b$ from Eq. (10). When $w < 0$, the latter condition is obeyed if the original parameters of free energy (3) satisfy the inequality $-b_s < u_s < 0$.

We should mention here that a new phase of coexistence of superconductivity and ferromagnetism occurs as a solution of Eqs. (13). It is defined in the following way:

$$\phi_1^2 + \phi_2^2 = \frac{1}{1 - \gamma_1^2} \left[ \gamma_1 (t + \gamma_2^2/2w) - r \right], \quad (57)$$

and

$$M^2 = \frac{1}{1 - \gamma_1^2} \left[ \gamma_1 r - (t + \gamma_2^2/2w) \right],$$

and

$$2w \sin(\theta_2 - \theta_1) = \gamma M, \quad \cos(\theta_2 - \theta_1) \neq 0. \quad (58)$$

In the present approximation the phase (57)-(58) is unstable, but this may be changed when crystal anisotropy is taken into account.

We shall write the equations for order parameters $M$ and $\phi_1$ of the FS phase in order to illustrate the changes when $w \neq 0$

$$\phi_1^2 = \frac{\pm \gamma M - r - \gamma_1 M^2}{(1 - w)} \geq 0, \quad (59)$$

and

$$(1 - w - \gamma_1^2) M^3 
\pm \frac{3}{2} \gamma_1 M^2 + \left[ t(1 - w) - \frac{\gamma^2}{2} - \gamma_1 r \right] M \pm \frac{\gamma r}{2} = 0, \quad (60)$$

where the meaning of the upper and lower sign is the same as explained just below Eq. (44). The difference in the stability conditions is more pronounced and gives new effects that will be explained further,

$$\frac{(2 - w) \gamma M - r - \gamma_1 M^2}{1 - w} \geq 0, \quad (61)$$

and

$$\gamma M - w r - w \gamma_1 M^2 \geq 0, \quad (62)$$

and

$$\frac{3(1 - w - \gamma_1^2) M^2 + 3\gamma_1 M + t(1 - w) - \gamma_2^2/2 - \gamma_1 r}{1 - w} \geq 0. \quad (63)$$

The calculations of the phase diagram in $(t, r)$ parameter space are done in the same way as in case of $w = 0$ and show that for $w > 0$ there is no qualitative change of the phase diagram. Quantitatively, the region of first order phase transition widens both with respect to $t$ and $r$ as illustrated in Fig. 6. On the contrary, when $w < 0$ the first order phase transition region becomes more narrow but the condition (62) limits the stability of FS for $r < 0$. This is seen from Fig. 7 where FS is stable above the straight dotted line for $r < 0$ and $t < 0$. So, purely superconducting (Meissner) phases occur also as ground states together with FS and FM phases.

VI. CONCLUSION

We investigated the M-trigger effect in unconventional ferromagnetic superconductors. This effect arises from the $M\psi_1\psi_2$-coupling term in the GL free energy and brings into existence a superconductivity in a domain of the system’s phase diagram that is entirely occupied by the ferromagnetic phase. The coexistence of unconventional superconductivity and ferromagnetic order is possible for temperatures above and below the critical temperature $T_s$, that corresponds to the standard second-order phase transition from normal to Meissner phase – usual uniform superconductivity in a zero external magnetic field which occurs outside the domain of existence of ferromagnetic order. Our investigation has been mainly intended to clarify the thermodynamic behavior at temperatures $T_s < T < T_f$ where the superconductivity cannot appear without the mechanism of M-triggering. We have described the possible ordered phases (FM and FS) in this most interesting temperature interval.

The Cooper pair and crystal anisotropies have also been investigated and their main effects on the thermodynamics of the triggered phase of coexistence is established. In discussions of concrete real material one should consider the respective crystal symmetry. But when the low symmetry and low order (in both $M$ and $\omega$) $\gamma$-term is present in the free energy, the dependence of essential thermodynamic properties on the type of crystal symmetry is not substantial.

Below the superconducting critical temperature $T_s$ a variety of pure superconducting and mixed phases of coexistence of superconductivity and ferromagnetism exists and the thermodynamic behavior at these relatively low temperatures is more complex than in known cases of improper ferroelectrics. The case $T_f < T_s$ also needs a special investigation.

Our results are referred to the possible uniform superconducting and ferromagnetic states. Vortex and other nonuniform phases need a separate study.

The relation of the present investigation to properties of real ferromagnetic compounds, such as UGe$_2$, URhGe, and ZrZn$_2$, has been discussed throughout the text. In these compounds the ferromagnetic critical temperature is much larger than the superconducting
FIG. 6: Phase diagram in the \((t, r)\) plane for \(\gamma = 1.2, \gamma_1 = 0.8, \) and \(w = 0.4\). The meaning of lines and points is the same as given in Fig. 5.
FIG. 7: Phase diagram in the \((t, r)\) plane for \(\gamma = 1.2, \gamma_1 = 0.8,\) and \(w = -2.\) The straight dotted line for \(r < 0\) indicates an instability of the FS phase. The meaning of other lines and notations is the same as given in Fig. 5.
critical temperature \( (T_f \gg T_s) \) and that is why the M-triggering of the spin-triplet superconductivity is very strong. Moreover, the \( \gamma_1 \)-term is important to stabilize the FM order up to the absolute zero (0 K), as is in the known spin-triplet ferromagnetic superconductors. Ignoring the symmetry conserving \( \gamma_1 \)-term does not allow a proper description of the known real substances of this type. More experimental information about the values of the material parameters \( (a_s, a_f, \ldots) \) included in the free energy (12) is required in order to outline the thermodynamic behavior and the phase diagram in terms of thermodynamic parameters \( T \) and \( P \). In particular, a reliable knowledge about the dependence of the parameters \( a_s \) and \( a_f \) on the pressure \( P \), the value of the characteristic temperature \( T_s \) and the ratio \( a_s/a_f \) at zero temperature are of primary interest.

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