Magnetic properties of triplet superconductors in the nonunitary state

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(Submitted 21 October 1986)
Zh. Eksp. Teor. Fiz. **92**, 1110-1117 (March 1987)

The structure of the mixed state of a triplet superconductor with a nonunitary order parameter, a large value of $\varkappa$, and weak spin-orbit interaction is considered. It is shown that a magnetic field (greater than H_{c1}) penetrates such a superconductor without the formation of singularities (Abrikosov vortices), and a distinctive spin texture is formed. One of the possible textures is investigated, and expressions for H_{c1} and the magnetization curve near H_{c1} are obtained. The transition from the superconducting to the mixed state can be first-order or second-order, depending on the mutual orientation of the external magnetic field and the axes of the spin-orbit anisotropy.

1. INTRODUCTION

The superconducting properties of organic compounds of the type M_2X (Bechgaard salts), where M is the molecule TMTSF or TMTTF and X are different monovalent anions (e.g., PF_6, $C1O_4$, Br), are ususual in many respects (see, e.g., the reviews in Refs. 1 and 2). Superconductivity in Bechgaard salts is strongly suppressed by lattice defects,[3] and as a result it has already been postulated that the superconductivity has a triplet character ($S = 1$) in these substances.[4] The values of the upper critical field H_{c2} along one of the axes of the crystal are anomalously large[5] and are two-three times greater than the Clogston paramagnetic limit. As already discussed in Ref. 6, this fact also argues in favor of p-pairing.

The aim of the present work is to investigate, in the approximation of the Ginzburg-Landau functional (i.e., near T_c), the structure of the triplet state in a magnetic field in the case when the superconducting order parameter is nonunitary (the spin of the electron pair has a definite direction) and the spin-orbit interaction is weak. We note here that the electronic specific heats of the Bechgaard salts apparently have an exponential dependence on the temperature below T_c (Ref. 7), while for a nonunitary state we should expect a linear dependence (with a density of states equal to half that in the normal phase), because the energy gap vanishes exactly for half the elementary excitations (see, e.g., Ref. 8). However, even if the superconducting order parameter in Bechgaard salts is unitary, i.e., the average pair-spin projection along any axis is equal to zero, the study of the nonunitary superconducting phase is of definite interest because of the unusual magnetic properties that it possesses. The extra degree of freedom associated with rotation of the spins of the superconducting pairs in the case when the spin-orbit interaction is small leads to the result that for a sufficiently strong magnetic field a distinctive spin texture, analogous to the nonsingular vortex in the A-phase of superfluid ^{3}He (see, e.g., Ref. 9), is formed. In the given case the superconducting current cannot be represented in the form of the gradient of the phase. This leads to the result that a magnetic field (greater than H_{c1}) penetrates the crystal without the formation of singularities (Abrikosov vortices). Expressions are obtained for the lower critical field H_{c1} and the magnetization curve near H_{c1}. The large values of the Ginzburg-Landau parameter $\varkappa \cong 20$–100 considerably simplify the theoretical analysis, because of the local character of the electrodynamics. It is shown that the phase transition from the superconducting to the mixed state can be first-order or second-order, depending on the mutual orientation of the external field and the axis of the spin-orbit anisotropy.

We note that the results discussed in this paper are not applicable to superconductors with so-called heavy fermions (e.g., UBe_{13}, $CeCu_2Si_2$, etc.), in which the pairing is also apparently p-type[10] but the spin-orbit interaction is large and the spins of the superconducting pairs are frozen into the crystal lattice and cannot form an independent spin texture. In the organic compounds discussed in the present paper the spin-orbit interaction is relatively weak.

2. THE FREE ENERGY AND GINZBURG-LANDAU EQUATIONS

The order parameter of the superconductor in the case of p-pairing can be written in the form (see, e.g., Ref. 11)

$$\Delta(\mathbf{p}) = (\hat{\boldsymbol{\sigma}}\mathbf{d})\, i\hat{\sigma}_y \psi(\mathbf{p}), \tag{1}$$

where $\psi(\mathbf{p})$ is the orbital part of the pair wavefunction and $(\hat{\boldsymbol{\sigma}}\mathbf{d}) i\hat{\sigma}_y$ is a symmetric spinor with $S = 1$; $(\hat{\boldsymbol{\sigma}}\mathbf{d})$ is the scalar product of $\boldsymbol{\sigma}$ and $\mathbf{d}$.

We write the Ginzburg-Landau free energy in the form

$$F = \int \Big\{ \alpha \mathbf{d}^*\mathbf{d} + \gamma_{ik}\Big(\nabla_i + \frac{2ie}{c}A_i\Big) d_j^* \Big(\nabla_k - \frac{2ie}{c}A_k\Big) d_j + \beta_1 (\mathbf{d}^*\mathbf{d})^2 + \beta_2 (\mathbf{d}^*\mathbf{d}^*)(\mathbf{d}\mathbf{d}) + f_{ij} d_i^* d_j + \frac{\mathbf{h}^2}{8\pi}\Big\} d^3r, \tag{2}$$

where $\alpha \sim T - T_c$, and the spin-orbit interaction is described by the term $f_{ij} d_i^* d_j$. It can be seen from (2) that for $\beta_2 < 0$ a real vector $\mathbf{d}$ is energetically favored, i.e., the structure of the order parameter (1) is such that the spin of the electron pair has any orientation in the plane perpendicular to $\mathbf{d}$ with equal probability (analogously to the A-phase of ^{3}He). For $\beta_2 > 0$ a complex $\mathbf{d}$, with the condition $\mathbf{dd} = 0$, is favored. This order parameter corresponds to a definite pair spin $\mathbf{S} = i[\mathbf{dd}^*]/dd^*$, where $[dd^*]$ is the vector product of $\mathbf{d}$ and $\mathbf{d}^*$. From the derivation of (2) in the weak-coupling approximation it follows that $\beta_1 > 0$ and $\beta_2 < 0$. Assuming, however, that the order parameter (1) is nonunitary (i.e., that $\mathbf{d}$ is complex), we shall not specify the values of the coefficients β_1 and β_2 in what follows.

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Near T_c the principal contribution to the gradient part of the free energy (2) is made by soft modes associated with rotation of the vector **d** and, correspondingly, of the pair spin **S**. When the vector **d** is normalized by the condition $\mathbf{dd}^* = 2$ we have

$$S = {}^1/_2 i[\mathbf{dd}^*]. \qquad (3)$$

We now derive the Ginzburg-Landau equations describing the nonunitary superconducting phase. Varying (2) with respect to **A**, we have, as usual,

$$j_i = \frac{ien_s}{2} m_{ik}^{-1}\left\{ (\mathbf{d}\nabla_k \mathbf{d}^* - \mathbf{d}^*\nabla_k \mathbf{d}) + \frac{4ie}{c} A_k(\mathbf{dd}^*)\right\}, \qquad (4)$$

where we have taken $\gamma_{ik} = n_s m_{ik}^{-1}/4$. Taking the curl of both sides of (4), after certain transformations we find

$$\operatorname{curl}(\hat{m}\mathbf{j}) = -\frac{4e^2 n_s}{c}\mathbf{h} + (en_s) e_{\alpha pq} S_\alpha [\nabla S_p, \nabla S_q], \qquad (5)$$

which is analogous to the Mermin-Ho equation[12] for $\operatorname{curl}\mathbf{v}_s$ in the A-phase of ^{3}He.

Varying (2) with respect to **d** and **d*** with the condition $\mathbf{d}(\delta\mathbf{d}^*) + \mathbf{d}^*(\delta\mathbf{d}) = 0$, we obtain

$$\frac{1}{4e}(\mathbf{j}\nabla) S_\alpha - \frac{n_s}{4} m_{ik}^{-1}\left[\mathbf{S}, \frac{\partial^2 \mathbf{S}}{\partial x_i \partial x_k}\right]_\alpha - f_{ij} e_{\alpha\beta i} S_j S_\beta = 0. \qquad (6)$$

Together with the Maxwell equation

$$\operatorname{curl}\mathbf{h} = \frac{4\pi}{c}\mathbf{j} \qquad (7)$$

Eqs. (5) and (6) form the complete system of Ginzburg-Landau equations.

We note, finally, that the gradient part of the free energy (2) can be brought, after certain transformations, to the form

$$F = \int\left\{\frac{1}{8e^2 n_s}(\hat{m}\mathbf{j})^2 + \frac{n_s}{4} m_{ik}^{-1}\nabla_i S_j \nabla_k S_j + f_{ii}\mathbf{S}^2 - f_{ij} S_i S_j + \frac{\mathbf{h}^2}{8\pi}\right\} d^3 r \qquad (8)$$

(we have used the conditions $\mathbf{dd} = 0$ and $f_{ij} = f_{ji}$).

The crystal lattice of the Bechgaard salts is almost orthorhombic. We take the crystal axes as our coordinate axes, and let the xy plane coincide with the surface of the sample; the external field **H** is perpendicular to the surface (see Fig. 1). In this case, obviously, **S** and **h** do not depend on the coordinate z. We shall assume that in the spin-orbit interaction tensor f_{ij} only one component $f_{zz} = f$ is nonzero. Then the contribution of the spin-orbit interaction to the free energy F is equal to

$$f_{ij} d_i^* d_j = f d_z^* d_z = f(\mathbf{S}^2 - S_z^2),$$

which for $f > 0$ corresponds to an easy axis (z), and for $f < 0$ corresponds to an easy plane (xy).

We write the electron-pair spin S in polar coordinates:

$$S_x = \sin\theta\cos\varphi, \quad S_y = \sin\theta\sin\varphi, \quad S_z = \cos\theta,$$

and, with the aid of (7), rewrite (5) in the form

$$\lambda_y^2 \frac{\partial^2 h}{\partial x^2} + \lambda_x^2 \frac{\partial^2 h}{\partial y^2} + h = \frac{c}{2e}\left(\frac{\partial\theta}{\partial x}\frac{\partial\varphi}{\partial y} - \frac{\partial\theta}{\partial y}\frac{\partial\varphi}{\partial x}\right)\sin\theta, \qquad (9)$$

where $h = h_z$ (since $h_x = h_y = 0$) and $\lambda_i^2 = m_i c^2/16\pi e^2 n_s$.

We now transform (6). We note that when (6) is multiplied by S_α the result is identically zero, i.e., only two components of (6) are independent, as should be the case. By taking the sum of the projections of (6) along the x and y axies, and also the projection along z, we obtain, respectively,

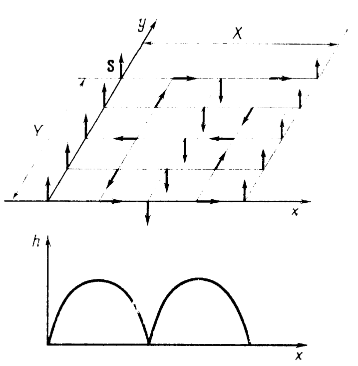

FIG. 1. Unit cell of the spin texture. The cell (nonsingular vortex) is rectangular and carries four quanta of magnetic flux. The coordinate axes are chosen so that $m_y < m_x$. The polar angle θ of the spin vector **S** depends only on the coordinate x and the azimuthal angle $\varphi = ky$. On the dashed line the direction of rotation of the spin is reversed (i.e., $k \to (-k)$). The magnetic field is $h = h(x)$.

$$\sin\theta\left(j_x\frac{\partial}{\partial x} + j_y\frac{\partial}{\partial y}\right)\varphi - en_s\left(\frac{1}{m_x}\frac{\partial^2}{\partial x^2} + \frac{1}{m_y}\frac{\partial^2}{\partial y^2}\right)\theta$$

$$+ en_s\sin\theta\cos\theta\left[\frac{1}{m_x}\left(\frac{\partial\varphi}{\partial x}\right)^2 + \frac{1}{m_y}\left(\frac{\partial\varphi}{\partial y}\right)^2\right]$$

$$+ 4ef\sin\theta\cos\theta = 0, \qquad (10)$$

$$\left(j_x\frac{\partial}{\partial x} + j_y\frac{\partial}{\partial y}\right)\theta + 2en_s\cos\theta\left(\frac{1}{m_x}\frac{\partial\theta}{\partial x}\frac{\partial\varphi}{\partial x} + \frac{1}{m_y}\frac{\partial\theta}{\partial y}\frac{\partial\varphi}{\partial y}\right)$$

$$+ en_s\sin\theta\left(\frac{1}{m_x}\frac{\partial^2}{\partial x^2} + \frac{1}{m_y}\frac{\partial^2}{\partial y^2}\right)\varphi = 0. \qquad (11)$$

3. THE SPIN TEXTURE, LOWER CRITICAL FIELD H_{c1}, AND MAGNETIZATION CURVES

One of the possible solutions of the system of Ginzburg-Landau equations (7), (9), (11) will be sought in the form

$$\varphi = ky, \quad j = j_y(x) \qquad (12)$$

(for (12) to be realized the condition $m_y < m_x$ should be fulfilled, and everywhere below we shall assume this to be so). Such one-dimensional behavior of the superconducting current **j** can be expected in view of the strong anisotropy of the mass tensor of the Bechgaad salts.

By rewriting the Ginzburg-Landau equations (9), (10), and (11) with the condition (12), and eliminating j_y from them by means of (7), we can obtain

$$-\lambda_y^2\frac{\partial^2 h}{\partial x^2} + h - \frac{ck}{2e}\frac{\partial\theta}{\partial x}\sin\theta = 0, \qquad (9')$$

$$\frac{4e}{c}\lambda_y^2 k\frac{\partial h}{\partial x}\sin\theta + \frac{m_y}{m_x}\frac{\partial^2\theta}{\partial x^2} - (k^2 + f)\sin\theta\cos\theta = 0, \qquad (10')$$

$$\frac{cm_y}{8\pi en_s}\frac{\partial h}{\partial x}\frac{\partial\theta}{\partial y} - k\frac{\partial\theta}{\partial y}\cos\theta = 0, \qquad (11')$$

where $\tilde{f}=4m_y f/n_s$. From (11′) it follows that $\partial\theta/\partial y=0$, i.e., $\theta=\theta(x)$ (otherwise, if $\partial\theta/\partial y\neq 0$, from (9′) and (11′) we immediately obtain $h=0$). The equations (9′) and (10′) have the first integral

$$\frac{m_y}{m_x}\left(\frac{\partial\theta}{\partial x}\right)^2+\frac{8e^2}{c^2}\lambda_y^2h^2+\frac{1}{2}(k^2+\tilde{f})\cos 2\theta-8\frac{e^2\lambda_y^4}{c^2}\left(\frac{\partial h}{\partial x}\right)^2=A. \quad (13)$$

For $H-H_{c1}\ll H_{c1}$ the characteristic lengths of the spin texture are much greater than λ_y (as will be confirmed below); therefore, we neglect the last term in (13) and the first term in (9′) and rewrite (13) in the form

$$\left(\frac{dx}{d\theta}\right)^2=\frac{m_y}{m_x}\frac{1+2\lambda_x^2k^2\sin^2\theta}{A-{}^1/_2(k^2+\tilde{f})\cos 2\theta}. \quad (13')$$

From (9′) we then have

$$h=\frac{ck}{2e}\frac{d\theta}{dx}\sin\theta. \quad (14)$$

The unit cell of the spin texture is depicted in Fig. 1. Its periods are

$$X=2\int_0^\pi\frac{dx}{d\theta}d\theta=2\left(\frac{m_y}{m_x}\right)^{1/2}\int_0^\pi\left[\frac{1+2\lambda_x^2k^2\sin^2\theta}{A-{}^1/_2(k^2+\tilde{f})\cos 2\theta}\right]^{1/2}d\theta,$$

$$Y=2\pi/k.$$

Below it will be established that $X\gg Y$ near H_{c1}, and since, in addition, $\lambda_x>\lambda_y$, we can neglect terms of the form $\lambda_y^2(\partial^2/\partial x^2)$, as was done above, while retaining terms containing $\lambda_x^2k^2$.

Using (14) we find the flux through a unit cell:

$$\Phi=Y\int_0^X h\,dx=\frac{2\pi}{k}\cdot 2\int_0^\pi h\frac{dx}{d\theta}d\theta=4\frac{\pi c}{e}, \quad (15)$$

i.e., four quanta of flux. This result can be obtained immediately from (9) by integrating both sides of (9) over the area of the cell and making use of the fact that the integral of the right-hand side is equal to $4\pi N$, where N is the degree of the mapping, implemented by the vector $\mathbf{S}(x,y)$, of the unit cell onto the unit sphere. Our cell is chosen in such a way that $N=2$, since the polar angle θ runs twice over all values from 0 to π (see Fig. 1).

To determine the texture parameters X and Y, the lower critical field H_{c1}, and the magnetization curve, we write out the Gibbs free energy $G=F-\mathbf{BH}/4\pi$ per unit volume (B is the magnetic induction averaged over the texture). Using (8) and neglecting the first term in the integrand, we can obtain

$$G=\frac{1}{XY}\int\left\{\frac{n_s}{4}\left[\frac{1}{m_x}\left(\frac{\partial\theta}{\partial x}\right)^2+\frac{\sin^2\theta}{m_y}k^2\right]+f\sin^2\theta+\frac{h^2}{8\pi}\right\}dx\,dy-\frac{BH}{4\pi}, \quad (16)$$

where the integration is performed over the area of a unit cell of the texture. Taking as the independent parameters

$$B=4\pi c/eXY,\qquad p=[(k^2+\tilde{f})/(A+{}^1/_2(k^2+\tilde{f}))]^{1/2}$$

in place of k and A, after certain calculations we rewrite (16) in the form

$$G=\alpha(p)B^2+\frac{n_s}{8m_yp^2}\left[2\frac{E(p)}{K(p)}-(1-p^2)\right]\times\left\{\left[\tilde{f}^2+\frac{m_y}{m_x}\left(\frac{4epK(p)}{c}B\right)^2\right]^{1/2}+\tilde{f}\right\}-\frac{BH}{4\pi}, \quad (17)$$

where

$$\alpha(p)=\frac{1}{24\pi p^2}[3E^2(p)+(4-2p^2)E(p)K(p)-(1-p^2)K^2(p)],$$

and $K(p)$ and $E(p)$ are complete elliptic integrals of the first and second kind, respectively. We give asymptotic expressions for them for $p\to 1$, which we shall need below:

$$K(p)\approx\ln\frac{4}{\omega}+\frac{1}{4}\left(\ln\frac{4}{\omega}-1\right)\omega^2$$

$$E(p)\approx 1+\frac{1}{2}\left(\ln\frac{4}{\omega}-\frac{1}{2}\right)\omega^2,$$

where $\omega=(1-p^2)^{1/2}\ll 1$. By finding the minimum of (17) as a function of p and B, we can determine H_{c1}, the periods X and Y, and the magnetization curve near H_{c1}.

We consider first the case $f=0$, i.e., spin-orbit interaction is absent. The free energy (17) then has the form

$$G=\alpha(p)B^2+B\left\{\frac{en_s}{2cp(m_xm_y)^{1/2}}[2E(p)-(1-p^2)K(p)]-\frac{H}{4\pi}\right\}, \quad (18)$$

whence we obtain

$$H_{c1}{}^0=4\pi en_s/c(m_xm_y)^{1/2}=c/4e\lambda_x\lambda_y, \quad (19)$$

which is reached $p=1$. We note that (19) is smaller by a factor of $\ln\varkappa$ than the expression for H_{c1} for an ordinary (with singular vortices) type-II superconductor. In the Bechgaard salts the parameter $\varkappa$, as already stated, is large. Estimating it from the ratio of the critical fields, $\varkappa\cong(H_{c2}/H_{c1})^{1/2}$, we obtain for $(TMTSF)_2ClO_4$ at low temperatures the value $\varkappa\cong 100$ (Refs. 3, 13).

Using the asymptotic expressions for $K(p)$ and $E(p)$ for $p\to 1$, it is not difficult to find from (18) the magnetization curve for $H-H_{c1}^0\ll H_{c1}^0$:

$$H-H_{c1}{}^0={}^1/_3B\ln(H_{c1}{}^0/B), \quad (20)$$

whence it follows that $(dB/dH)_{H_{c1}^0}=0$ (see Fig. 2). The periods of the spin texture are given by

$$X^2=\left(\frac{m_y}{m_x}\right)^{1/2}\frac{4c}{Be}\ln\frac{H_{c1}{}^0}{B},\qquad\frac{X}{Y}=\left(\frac{m_y}{m_x}\right)^{1/2}\frac{1}{\pi}\ln\frac{H_{c1}{}^0}{B}, \quad (21)$$

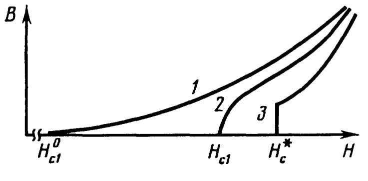

FIG. 2. Form of the magnetization curves. 1) $f=0$, no spin-orbit interaction; 2) $f>0$, easy z axis; 3) $f<0$, easy xy plane.

i.e., near H_{c1}^0 we obtain $X \gg Y$.

We now consider the case $f > 0$ (easy z axis). For $B = 0$ the minimum of the free energy (16) is zero, which is reached at $\theta = 0$, i.e., when all the spins are aligned along the easy z axis. Substituting into (17) the value $p = 1$, we make the replacement $BK(p) = R$ and assume that

$$f^2 \ll (4eR/c)^2 m_y/m_x. \tag{22}$$

Then for H_{c1} we obtain the equation

$$\frac{1}{12\pi} R^2 + \frac{1}{4\pi} R(H_{c1}^0 - H) + f = 0,$$

where H_{c1}^0 is determined by the expression (19). From this is is not difficult to obtain

$$H_{c1} = H_{c1}^0 + (16\pi f/3)^{1/2}. \tag{23}$$

We note that for $B \to 0$ (i.e., $H \to H_{c1}$) R remains finite, i.e., the condition (22) that f be small is fulfilled. It can be rewritten in the form

$$f \ll \frac{1}{m_x m_y} \left(\frac{n_s e}{c} \right)^2. \tag{24}$$

By substituting into (17) the asymptotic expressions for the elliptic integrals, we can convince ourselves that H_{c1} is indeed reached at $p = 1$ (we shall not give the corresponding cumbersome calculations). The form of the magnetization curve, however, differs from that considered in the preceding case in that $(dB/dH)_{H_{c1}} = \infty$ for arbitrarily small f.

The case with $f < 0$, which corresponds to an easy xy plane, is the most interesting. For $B = 0$ the minimum of the free energy (16) is equal to $-|f|$ and is reached at $\theta - \pi/2$, $\varphi = \text{const}$ (i.e., all the spins are oriented along some direction in the easy xy plane). Minimizing (16) for $B \neq 0$, we can convince ourselves that the resulting texture has $k^2 > |f|$ and for the free energy we can use the expression (17). Between these two states a first-order transition occurs at

$$H_c^* = H_{c1}^0 + \left(\frac{8\pi |f|}{3} \ln \frac{c}{e\lambda_x \lambda_y |f|^{1/2}} \right)^{1/2} \tag{25}$$

(we recall that $|f|$ is small and satisfies the condition (24)). In contrast to the preceding case, H_c^* is reached at $p \neq 1$. The form of the magnetization curve is shown in Fig. 2. The discontinuity at $H = H_c^*$ is equal to

$$B(H = H_c^*) = \left[\frac{24\pi |f|}{\ln(c/e\lambda_x\lambda_y |f|^{1/2})} \right]^{1/2}. \tag{26}$$

We note that in this case, as in the preceding case, spin-orbit interaction makes the formation of a spin texture difficult by raising the value of H_{c1}.

4. CONCLUSIONS

We have ascertained that the model that we have considered for a triplet superconductor with a nonunitary order parameter has unusual magnetic properties. A magnetic field can penetrate into it in the form of nonsingular vortices, with the formation of a distinctive spin texture. We recall that here we are discussing superconductors in which the spin-orbit interaction is small and the electron-pair spin is not frozen by the crystal lattice.

One of the possible spin textures was described above. Its unit cell (nonsingular vortex) has a rectangular shape and carries four quanta of magnetic flux. The spin-orbit interaction, by pinning the spin vector, hinders the formation of a spin texture. It raises H_{c1} and influences the type of the phase transition in a magnetic field.

The authors express their thanks to L. P. Gor'kov for useful discussions throughout the course of the work.

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Translated by P. J. Shepherd