Effect of Pauli Paramagnetism on Magnetic Properties of High-Field Superconductors*

KAZUMI MAKI†
Department of Physics, University of California, San Diego, La Jolla, California

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We study the magnetic properties of type-II superconductivity having a large Pauli term. Firstly we restrict ourselves to the case where the spin-orbit scattering is absent. In this case the transition changes at low temperatures from second to first order for $\alpha=3\mu/\tau_{tr}v^2e\geq 1$, where μ, τ_{tr}, and v are the Bohr magneton, the transport collision time, and the Fermi velocity, respectively. This involves a revised calculation of the parameter $\kappa_2(t)$, which appears in the expression for the magnetization. The calculation of $\kappa_2(t)$ in an earlier paper on this subject by the author was incorrect, as was recently point out by de Gennes and co-workers. Secondly, we extend the theory to include the effect of spin-orbit scattering, which may be important in the materials usually considered. In particular, we obtain explicitly expressions for the parameters $\kappa_1(t)$ and $\kappa_2(t)$ in the limit of short spin-orbital mean free path, where $\kappa_1(t)=H_{c2}(t)/\sqrt{2}H_c(t)$, and $H_{c2}(t)$, $H_c(t)$ are the upper critical field and the thermodynamical critical field, respectively. It is shown that in this limit the transition is always second order, independent of temperature.

I. INTRODUCTION

IN his classic paper, Abrikosov[1] showed that the magnetic properties of type-II superconductivity are quite different from that of ordinary superconductivity (now called type I). The magnetic field begins to penetrate into the bulk at the field $H=H_{c1}$ (the lower critical field) and a mixed state appears in which the magnetic flux threads the bulk in the form of quantized vortex lines. Only at H_{c2} (the upper critical field) is superconductivity finally suppressed. Abrikosov's work, however, was based on the Ginzburg-Landau theory, which is only valid in a narrow temperature region close to the transition temperature.[2] Recent theoretical studies[3] have mainly focused on a generalization of Abrikosov's theory to all temperatures.

In certain alloys or compound systems it is known that Abrikosov's expression[1] for H_{c2}',

$$H_{c2}'=\sqrt{2}\kappa H_c\,, \tag{1}$$

gives an enormously large value of H_{c2}, since κ is of the order of 50–100. Here κ is the Ginzburg-Landau parameter and H_c is the thermodynamical critical field. In such a situation it was suggested by several people[4] that the Pauli susceptibility energy might play an important role in suppressing the superconducting state. The effect of the Pauli terms at finite temperature was studied in detail by Sarma[5] and by Maki and Tsuneto.[6] The generalized Ginzburg-Landau equations containing the term due to the Pauli susceptibility have been given by the author.[7] Unfortunately, the author's original theory[8] of type-II superconductivity contained some serious algebraic errors, as Caroli, Cyrot, and de Gennes[9] have recently noted. As a result, it is necessary to revise certain calculations given in Ref. 7.

The purpose of the present paper is twofold. Firstly, we present a corrected version of our theory of type-II superconductivity having a large Pauli paramagnetism. Secondly, we extend this theory to include the effect of the spin-orbit scattering, which may be important in some materials. The data[10] on H_{c2} accumulated recently seem to indicate that the effect of the spin-orbit interaction is appreciable.

In the following we shall first discuss the effect of the Pauli term in the absence of spin orbit scattering due to impurities. The two parameters $\kappa_1(t)$ and $\kappa_2(t)$ are defined by

$$H_{c2}(t)=\sqrt{2}\kappa_1(t)H_c(t)\,, \tag{2}$$

$$-4\pi(M_s-M_n)=(H_{c2}-H_0)/(2\kappa_2^2(t)-1)\beta_A \tag{3}$$

where M_s and M_n are the magnetization of the superconducting state and the normal state, respectively ($M_n\neq 0$, due to Pauli paramagnetism). The corrected calculation shows that transition changes from second to first order if $\alpha=H_{c2}'/\sqrt{2}H_P\cong 1$, where H_{c2}' is the upper critical field in the absence of the Pauli term and $H_P=\Delta_{00}/\sqrt{2}\mu=18\,400\ T_c$, the critical field due to the

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† On leave of absence from the Research Institute for Mathematical Sciences, Kyoto University, Kyoto, Japan.

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Pauli term alone. Experimentally, the predicted change of the order of the transition has not been observed[10] even for superconductors having a comparatively large $\alpha \simeq 1 \sim 5$. This may be due to the presence of strong spin-orbit scattering.

The generalization needed to include spin-orbit scattering due to impurity atoms or to dislocations is carried out in Secs. III and IV. When this scattering is strong, we find that the transition is always second order. In addition, the temperature dependence of $\kappa_1(t)$ and $\kappa_2(t)$ are calculated numerically.

II. GENERALIZED GINSBURG-LANDAU EQUATIONS CONTAINING THE PAULI TERM

In this section we would like to briefly recapitulate our previous work on the magnetic properties of type-II superconductors having a large Pauli term. As discussed in the introduction, the Pauli term becomes large generally in type-II superconductors having a large κ parameter. This will serve to give the general background of the theory. The effect of the Pauli paramagnetism as well as the diamagnetic current is described by the following Hamiltonian

$$\mathcal{H} = -\frac{1}{2m}\int (i\nabla + e\mathbf{A})\psi^\dagger (i\nabla - e\mathbf{A})\psi d^3r + \mu\int \psi^\dagger(\boldsymbol{\sigma}\mathbf{H})\psi d^3r, \quad (4)$$

where $\mathbf{A}$, $\mathbf{H}$, μ, and $\boldsymbol{\sigma}$ are the vector potential, the magnetic field, the Bohr magneton and the Pauli spin matrix, respectively.

Since we are interested in the properties of superconductors in fields $\mathbf{H}$ slightly smaller than the upper critical field, we expect that the order parameter $\Delta(\mathbf{r})$ is still small. In the absence of impurity scattering we obtain by expanding Gor'kov's equation in powers of Δ,[7]

$$\Delta^\dagger(\mathbf{r}) = |g| R \sum_n \tfrac{1}{2}\,\mathrm{Tr}\int G_\omega{}^\sigma(\mathbf{r}',\mathbf{r})G_{-\omega}{}^\sigma(\mathbf{r}',\mathbf{r})\Delta^\dagger(\mathbf{r}')\,d^3r' - |g| T\sum_n \tfrac{1}{2}\,\mathrm{Tr}\int\int\int G_\omega{}^\sigma(\mathbf{s},\mathbf{r}) \times G_{-\omega}{}^\sigma(\mathbf{s},\mathbf{l})G_\omega{}^\sigma(\mathbf{m},\mathbf{l})G_{-\omega}{}^\sigma(\mathbf{m},\mathbf{s})\Delta^\dagger(\mathbf{s})\Delta(\mathbf{l})\Delta^\dagger(\mathbf{m})\,d^3s\,d^3l\,d^3m, \quad (5)$$

where $G_\omega{}^\sigma(\mathbf{r},\mathbf{r}')$ is the Green's function of an electron in a normal metal:

$$G_\omega{}^\sigma(\mathbf{r},\mathbf{r}') = \exp\left\{ie\int_{\mathbf{r}}^{\mathbf{r}'} A(\mathbf{l})d\mathbf{l}\right\}\int \frac{d^3p}{(2\pi)^3} e^{\tau\mathbf{p}\cdot(\mathbf{r}-\mathbf{r}')}(i\omega^2 - \mu(\boldsymbol{\sigma}H) - \xi)^{-1}, \quad (6)$$

and $\xi = p^2/2m - \mu$. Here we have taken account of the magnetic field by a change of phase (that is, in the classical approximation).

On the other hand, the current is given by

$$\mathbf{j}_s(\mathbf{r}) = \frac{1}{2mi}(\nabla_r - \nabla_{r'} + 2eiA(\mathbf{r}))T\sum_n \tfrac{1}{2}\,\mathrm{Tr}\left\{\int\int G_\omega{}^\sigma(\mathbf{r},\mathbf{s})G_{-\omega}{}^\sigma(\mathbf{l},\mathbf{s})G_\omega{}^\sigma(\mathbf{l},\mathbf{r}')\Delta(\mathbf{s})\Delta^\dagger(\mathbf{l})d^3s d^3l\right\}\Bigg|_{r=r'} - i\nabla_r \times \mu T\sum_n \tfrac{1}{2}\,\mathrm{Tr}\left\{\int\int \boldsymbol{\sigma} G_\omega{}^\sigma(\mathbf{r},\mathbf{s})G_{-\omega}{}^\sigma(\mathbf{l},\mathbf{s})G_\omega{}^\sigma(\mathbf{l},\mathbf{r}')\Delta(\mathbf{s})\Delta^\dagger(\mathbf{l})d^3s d^3l\right\}, \quad (7)$$

where the first term is due to diamagnetic current and the second term is due to the difference between the paramagnetic current associated with the electron spins in the normal and the superconducting state.

The effect of the impurity scattering can be taken into account by the standard renormalization procedures. In the absence of the spin-orbit scattering, mixing between the spin-up state and spin-down state does not occur, and the necessary renormalization is given by the replacement[8]

$$\omega_n \text{ by } \tilde{\omega}_n = \omega_n\eta_\omega, \quad \text{and} \quad \Delta_q \text{ by } \tilde{\Delta}_q = \eta_{\omega q}\Delta_q,$$

where

$$\eta_\omega = (1 + 1/2\tau|\omega|),$$

and

$$\eta_{\omega q} = \{1 - (1/2\tau|\omega|)(1 - \tfrac{1}{3}\tau\tau_{tr}v^2q^2\}^{-1}. \quad (8)$$

Here τ, τ_{tr}, and v are the collision lifetime, the transport lifetime of electron, and the Fermi velocity, respectively. Thus in the present case, we arrive at

$$\left\{\ln\frac{T}{T_{c0}} + \mathrm{Re}\psi\left(\frac{1}{2} + \frac{Q}{2\pi T}\right) - \psi(\tfrac{1}{2})\right\}\Delta^\dagger(r) + \frac{1}{8(\pi T)^2}\mathrm{Re}\left\{\sum_{n=0}^{\infty}\left[n + \tfrac{1}{2} + \frac{i\mu H}{2\pi T} + \frac{\tau_{tr}v^2}{48\pi T}((\mathbf{p}_1 - \mathbf{p}_3)^2 + (\mathbf{p}_2 - \mathbf{p}_4)^2)\right]\prod_{i=1}^{4} X_{ni}{}^{-1}\right\}\Delta^\dagger(1)\Delta(2)\Delta^\dagger(3)|_{1=2=3=r} = 0, \quad (9)$$

and

$$\mathbf{j}_s(\mathbf{r}) = -\frac{e\tau_{tr}N}{4\pi mT}(i\nabla_1 - i\nabla_2 + 4e\mathbf{A}(\mathbf{r}))\,\mathrm{Re}\{\sum_{n=0}^{\infty}\prod_{i=1}^{2}X_{ni}^{-1}\}\Delta^{\dagger}(1)\Delta(2)|_{1=2=r} - \frac{3N\mu}{2\pi mv^2T}\nabla\times[\mathrm{Im}\{\sum_{n=0}^{\infty}\prod_{i=1}^{2}X_{ni}^{-1}\}|\Delta(r)|^2], \quad (10)$$

where

$$Q_i = \tfrac{1}{6}\tau_{tr}v^2(i\nabla_i - 2e(-1)^i\mathbf{A})^2 + i\mu H(\mathbf{i}),$$
$$\mathbf{p}_i = i\nabla_i - 2e(-1)^iA(\mathbf{i}), \quad X_{ni} = n+1+Q_i/2\pi T. \quad (11)$$

Here Re, Im indicate the real and the imaginary part of the function in the bracket, $\psi(z)$ is the digamma function, and N is the density of electrons.

We note that the term $[(\mathbf{p}_1-\mathbf{p}_3)^2+(\mathbf{p}_2-\mathbf{p}_4)^2]$ in Eq. (9) was missing in the author's original discussion,[8] as was recently pointed out by Caroli, Cryot, and de Gennes.[9] Their calculations indicate that the temperature dependence of the $\kappa_2(t)$ parameter is almost identical to that of $\kappa_1(t)$ in the dirty superconductors without the Pauli term. In our original (incorrect) calculation, the temperature dependence of $\kappa_2(t)$ was quite different from $\kappa_1(t)$. The correction of this error removes one of the major conflicts between experiment and theory.

We should also mention that in the fields with which we are concerned, the paramagnetic current in the normal state is not negligible and the total current is given by

$$\mathbf{j}(\mathbf{r}) = \mathbf{j}_s(\mathbf{r}) - (3N\mu^2/2mv^2)\nabla\times\mathbf{H}(\mathbf{r}). \quad (12)$$

Using Eq. (9) the upper critical field is given as a solution of a transcendental equation,[7]

$$\ln\frac{T}{T_{c0}} + \mathrm{Re}\,\psi\left(\frac{1}{2}+\frac{\tau_{tr}v^2eH_{c2}}{6\pi T}+\frac{\tau\mu H_{c2}}{2\pi T}\right) - \psi(\tfrac{1}{2}) = 0. \quad (13)$$

The asymptotic solutions are

$$H_{c2}(t) = \frac{3\Delta_{00}}{2e\tau_{tr}v^2}(1+\alpha^2)^{-1/2}\left(1-\tfrac{2}{3}\gamma^2t^2\frac{i-\alpha^2}{1+\alpha^2}\right), \quad \text{for } t\ll 1, \quad (14)$$

$$= \frac{12T_{c0}}{\pi e\tau_{tr}v^2}\theta\left(1-\theta\left(\frac{1}{2}-\frac{28\zeta(3)}{\pi^4}(1-\alpha^2)\right)\right), \quad \text{for } t\lesssim 1, \quad (15)$$

where $\theta = 1-t$, $t = T/T_{c0}$ and $\gamma = 1.78$. The parameter α is defined by

$$\alpha = 3\mu/e\tau_{tr}v^2 = \sqrt{2}H_{c2}'(0)/H_{cP},$$

where $H_{c2}'(0)$ is the upper critical field at $t=0$ in the absence of the Pauli term and $H_{cP} = \Delta_{00}/\sqrt{2}\mu$.

It is convenient to introduce in this connection a new parameter $\kappa_1(t)$, which is defined by

$$H_{c2}(t) = \sqrt{2}\kappa_1(t)H_c(t). \quad (16)$$

Using the expressions for $H_{c2}(t)$, we have

$$\kappa_1(t) = \frac{\kappa\pi^2}{2(14\zeta(3))^{1/2}}(1+\alpha^2)^{-1/2}\left(1-\tfrac{1}{3}\gamma^2t^2\frac{1-2\alpha^2}{1+\alpha^2}\right)$$
$$= 1.20\frac{\kappa}{(1+\alpha^2)^{1/2}}\left(1-1.05\frac{1-2\alpha^2}{1+\alpha^2}t^2\right), \quad \text{for } t\ll 1, \quad (17)$$

$$= \kappa\left[1+\left(\frac{28\zeta(3)}{\pi^4}(1-\alpha^2)-\frac{31\zeta(5)}{98\zeta^2(3)}\right)\theta\right]$$
$$= \kappa(1+(0.119-0.346\alpha^2)\theta), \quad \text{for } t\simeq 1. \quad (18)$$

We have made use of the expressions

$$H_c(t) = \Delta_{00}(2mp_0/\pi)^{1/2}(1-\tfrac{1}{3}\gamma^2t^2), \quad \text{for } t\ll 1, \quad (19)$$

$$= H_c(0)\gamma(8/7\zeta(3))^{1/2}\theta \times\left[1-\left(\frac{1}{2}-\frac{31\zeta(5)}{98\zeta^2(3)}\right)\theta\right], \quad \text{for } t\simeq 1, \quad (20)$$

and[2]

$$\kappa = \frac{3m}{2\pi^2e\tau_{tr}}\left(\frac{2\pi m}{p_0^5}7\zeta(3)\right)^{1/2}. \quad (21)$$

Following Abrikosov[1,9] we can calculate the free energy of the mixed state, with the result

$$F_s - F_n = -\frac{1}{8\pi}\left[B^2+\frac{(H_{c2}-B)^2}{(2\kappa_2^2(t)-1)\beta_A+1}\right], \quad (22)$$

where $B = \langle H(\mathbf{r})\rangle_{av}$ is the magnetic induction and $\beta_A = 1.16$. $\kappa_2(t)$ is given by

$$\kappa_2(t) = \left(\frac{3mf_1(\rho)}{8\pi N}\right)^{1/2}(e\tau_{tr}vg(\rho))^{-1}, \quad (23)$$

where

$$g(\rho) = \mathrm{Re}\left\{\sum_{n=0}^{\infty}(1+i\alpha)\frac{1}{(n+\frac{1}{2}+\rho)^2}\right\}, \quad (24)$$

$$f_1(\rho) = \mathrm{Re}\left\{\sum_{n=0}^{\infty}\frac{1}{(n+\frac{1}{2}+\rho)^3}\right\}, \quad (25)$$

and $\rho = (\tau_{tr}v^2/6\pi T)eH_{c2}(1+i\alpha)$.

For some limiting cases we note

$$\kappa_2(t)=\frac{\kappa\pi^2}{2(14\zeta(3))^{1/2}}\frac{(1-\alpha^2)^{1/2}}{1+\alpha^2}\left(1-\tfrac{2}{3}\gamma^2\frac{(1-14\alpha^2+d^4)}{1-\alpha^4}t^2\right)$$

$$=1.20\kappa\frac{(1-\alpha^2)^{1/2}}{1+\alpha^2}\left(1-2.10\frac{(1-14\alpha^2+\alpha^4)}{1-\alpha^4}t^2\right)$$

$$\text{for } t\ll 1, \qquad (26)$$

$$=\kappa\left\{1+\left(\frac{56\zeta(3)}{\pi^4}(1-\alpha^2)-\frac{45\zeta(4)}{7\pi^2\zeta(3)}\right)\theta\right\}$$

$$=\kappa(1+(0.106-0.692\alpha^2)\theta), \quad \text{for } t\simeq 1. \qquad (27)$$

We remark that $\kappa_2(t)>0$ for $\alpha^2\leq 1$ and $\kappa_2(t)$ vanishes at $\alpha=1$ at $T=0$°K, which indicates that the transition is first order for $\alpha\geq 1$ at low temperature. Incidentally, $[\partial H_{c2}(t)/\partial t]|_{t=0}$ becomes negative for $\alpha\leq 1$, implying that $H_{c2}(t)$ is a monotonic function of temperature as long as the transition is of the second order. This contrasts sharply with the previous incorrect results of the author.[7]

No experimental evidence has been found for this change of order of the transition, which probably indicates the importance of spin-orbital scattering neglected so far.

The magnetization as well as the jump of the specific heat at the transition are given by

$$-4\pi M=(H_{c2}-H_0)/1.16(2\kappa_2^2(t)-1), \qquad (28)$$

and

$$\Delta C_s=\frac{1}{4\pi}\left(\frac{\partial H_{c2}}{\partial T}\right)^2\Big/1.16(2\kappa_2^2(t)-1). \qquad (29)$$

Here M is the difference of the magnetization in the superconducting and the normal state.

III. EFFECT OF SPIN-ORBITAL SCATTERING

It is well known that spin-orbital scattering plays a crucial role in the phenomena which involve the Pauli terms such as the Knight shift in superconductivity and the coexistence of ferromagnetism and superconductivity.

In the preceding section we noticed that there are experimental indications that spin-orbital scattering might also be important in the magnetic properties of high-field superconductors. We shall consider here the effect of spin-orbital scattering due to impurity atoms and/or dislocations on the upper critical field as well as the magnetization of type-II superconductors having a large Pauli term. The impurity-averaging technique necessary

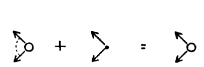

FIG. 1. The self-consistent equations for the renormalized vertex [see Eq. (33)]. Note that the renormalized vertex has two components $\tilde{\Delta}_+$ and $\tilde{\Delta}_-$.

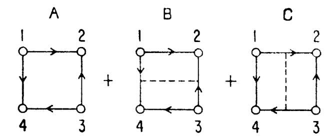

FIG. 2. The function $\mathfrak{F}_\omega(q_i)$ is given by the sum of the contributions from the above diagrams.

for present study has already been discussed in great detail by Abrikosov and Gor'kov.[11] We assume that the interaction between a conduction electron and an impurity atom is given by a potential

$$V(\mathbf{p},\mathbf{p}')=V_1(|\mathbf{p}-\mathbf{p}'|)+iV_{so}(1/p_0^2)[\mathbf{p}\times\mathbf{p}']\cdot\boldsymbol{\sigma}, \qquad (30)$$

where $\mathbf{p}$ and $\mathbf{p}'$ are momenta of incoming and outgoing electrons, respectively. The second term gives rise to spin-orbital scattering.

The effect of impurity scattering on the conduction electrons is taken into account by the following renormalizations.[8] First, the frequency ω in Green's functions should be replaced by $\tilde{\omega}=\omega\eta_\omega$ where η_ω is already given in Eq. (8). Here $1/\tau$ is given by

$$1/\tau=1/\tau_1+1/\tau_{so}, \qquad (31)$$

where

$$\frac{1}{\tau_1}=\frac{nmp_0}{2\pi^2}\int d\Omega|V_1(\theta)|^2,$$

and

$$\frac{1}{\tau_{so}}=\frac{nmp_0}{2\pi^2}\int d\Omega|V_{so}(\theta)|^2\sin^2\theta, \qquad (32)$$

n being the density of impurity atoms. Secondly, we need to consider the vertex renormalization. Solving the integral equation for the superconducting order parameter (which is shown diagrammatically in Fig. 1),

$$\tilde{\Delta}_{\pm\mathbf{q}}=\Delta_{\mathbf{q}}+(2\tau\tilde{\omega}_\pm)^{-1}(1-\tfrac{1}{3}\tau\tau_{tr}v^2q^2)\tilde{\Delta}_{\pm\mathbf{q}}$$
$$+(3\tau_{so}\tilde{\omega}_\pm)^{-1}(1-\tfrac{1}{3}\tau\tau_{tr}v^2q^2)\tilde{\Delta}_{\mp\mathbf{q}}, \qquad (33)$$

where $\tilde{\omega}_\pm=\tilde{\omega}\pm i\mu H$, $|\tilde{\omega}_\pm|=|\tilde{\omega}|\pm i\mu H$, and $\Delta_{\mathbf{q}}$ is the Fourier transform of the order parameter, we have

$$\tilde{\Delta}_{\pm\mathbf{q}}=\eta_{\pm\omega q}\Delta_{\mathbf{q}}, \qquad (34)$$

where

$$\eta_{\pm\omega q}=\frac{(\omega\eta\pm iI)(\omega\mp iI+a+b)}{((\omega+a)^2-b^2+I^2)},$$

$$I=\mu H, \quad a=\frac{1}{3\tau_{so}}+\frac{\tau\tau_{tr}v^2}{6}\mathbf{q}^2 \quad \text{and} \quad b=\frac{1}{3\tau_{so}}. \qquad (35)$$

In the derivation of Eq. (33), we have assumed $\tau\Delta_{00}\ll 1$, (the dirty limit) and[12] $\tau/\tau_{so}\ll 1$ where Δ_{00} is the B.C.S. order parameter at $T=0$°K. Using the above results it is not difficult to write down the generalized Ginzburg-

[11] A. A. Abrikosov and L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. 42, 1088 (1962) [English transl.: Soviet Phys.—JETP 15, 752 (1962)].

[12] This condition is discussed in great detail in a recent report by N. R. Werthamer, E. Helfand and P. C. Hohenberg, where they obtained independently the equation equivalent to Eq. (41); Phys. Rev. 147, 295 (1966).

Landau equations

$$\left\{\ln\frac{T}{T_{c0}}+\left[\frac{1}{2}\left(1+\frac{b}{(b^2-I^2)^{1/2}}\right)\psi(\tfrac{1}{2}+\rho_-)+\frac{1}{2}\left(1-\frac{b}{(b^2-I^2)^{1/2}}\right)\psi(\tfrac{1}{2}+\rho_+)\right]-\psi(\tfrac{1}{2})\right\}\Delta^\dagger(\mathbf{r})$$
$$+\frac{1}{8(\pi T)^2}\sum_{n=0}^{\infty}\mathfrak{F}_{\omega n}(q_i)\Delta^\dagger(1)\Delta(2)\Delta^\dagger(3)|_{1=2=3=r}=0, \quad (36)$$

and

$$\mathbf{j}_s(\mathbf{r})=-\frac{e\tau_{\mathrm{tr}}N}{4\pi mT}(i\nabla_1-i\nabla_2+4e\mathbf{A}(\mathbf{r}))\left[\mathrm{Re}\sum_{n=0}^{\infty}\left(\frac{\omega-iI+a+b}{(\omega+a)^2-b^2+I^2}\right)^2\right]\Delta^\dagger(1)\Delta(2)|_{1=2=r}$$
$$-\frac{3N\mu}{2\pi mv^2T}\nabla\times\left\{\left[\mathrm{Im}\sum_{n=0}^{\infty}\left(\frac{\omega-iI+a+b}{(\omega+a)^2-b^2+I^2}\right)^2\right]|\Delta(r)|^2\right\}, \quad (37)$$

where

$$\rho_\pm=(1/2\pi T)(a\pm(b^2-I^2)^{1/2}). \quad (38)$$

In Eq. (32) we have set[8]

$$a=1/3\tau_{\mathrm{so}}+\tfrac{1}{3}\tau\tau_{\mathrm{tr}}v^2eH_{c2}, \quad (39)$$

since we are interested in the type-II superconductivity in a magnetic field close to H_{c2}.

The function $\mathfrak{F}_\omega(\mathbf{q}_i)$ in Eq. (36) is obtained from the diagrams given in Fig. 2. After a somewhat lengthy calculation we have (see Appendix A)

$$\mathfrak{F}_\omega(\mathbf{q}_i)=(2\pi T)^3\left\{\mathrm{Re}\left(\frac{\omega-iI+a+b}{(\omega+a)^2-b^2+I^2}\right)^3-2I^2b\frac{(\omega+a+b)^2+I^2}{[(\omega+a)^2-b^2+I^2]^4}\right\}. \quad (40)$$

Here a is also given by Eq. (39).

The above set of equations completely determines the magnetic properties of the system provided $\Delta(\mathbf{r})$ is small.

The upper critical field is obtained from

$$\ln\frac{T}{T_{c0}}+\frac{1}{2}\left[\left(1+\frac{b}{(b^2-I^2)^{1/2}}\right)\psi(\tfrac{1}{2}+\rho_-)+\left(1-\frac{b}{(b^2-I^2)^{1/2}}\right)\psi(\tfrac{1}{2}+\rho_+)\right]-\psi(\tfrac{1}{2})=0 \quad (41)$$

where b, I, and $\rho_\pm$ have been already defined, and a is given by Eq. (39).

The free-energy difference is expressed as

$$F_s-F_n=-\frac{1}{8\pi}\left\{B^2+\frac{(H_{c2}-B)^2}{(2\kappa_2{}^2(t)-1)\beta_A+1}\right\}, \quad (42)$$

where

$$\kappa_2(t)=(3mf_1(\rho,b)/8\pi N)^{1/2}(e\tau_{\mathrm{tr}}vg(\rho,b))^{-1}. \quad (43)$$

Here $g(\rho,b)$ and $f_1(\rho,b)$ are given by

$$g(\rho,b)=\mathrm{Re}\left\{\sum_{n=0}^{\infty}(1+i\alpha)\left(\frac{n+\frac{1}{2}-iI'+a'+b'}{(n+\frac{1}{2}+a')^2-b'^2+I'^2}\right)^2\right\}, \quad (44)$$

and

$$f_1(\rho,b)=\mathrm{Re}\left\{\sum_{n=0}^{\infty}\left[\left(\frac{n+\frac{1}{2}-iI'+a'+b'}{(n+\frac{1}{2}+a')^2-b'^2+I'^2}\right)^3-2I'^2b'\frac{(n+\frac{1}{2}+a'+b')^2+I'^2}{[(n+\frac{1}{2}+a')^2-b'^2+I'^2]^4}\right]\right\}, \quad (45)$$

where

$$a'=(1/2\pi T)\{\tfrac{1}{3}\tau_{\mathrm{tr}}v^2eH_{c2}+1/3\tau_{\mathrm{so}}\},$$
$$b'=1/6\pi\tau_{\mathrm{so}}T \quad\text{and}\quad I'=\mu H_{c2}/2\pi T. \quad (46)$$

We note that in the limit $1/\pi\tau_{\mathrm{so}}T\ll1$, the above expressions reduce to those given in Sec. II.

IV. STRONG SPIN-ORBITAL-SCATTERING LIMIT

Using the expressions in the preceding section, we can discuss quite generally the magnetic properties of superconducting alloys having a large Pauli term. However, in the following we shall restrict our consideration to the short spin-orbital mean-free-path limit ($\tau_{\mathrm{so}}\Delta_{00}\ll1$), where the expressions involved simplify remarkably.

The upper critical field is given by (in the above-mentioned limit)

$$\ln t+\psi(\tfrac{1}{2}+\rho)-\psi(\tfrac{1}{2})=0, \quad (47)$$

where $t=T/T_{c0}$ and

$$\rho=(1/2\pi T)(\tfrac{1}{3}\tau_{\mathrm{tr}}v^2eH_{c2}+\tfrac{3}{2}\tau_{\mathrm{so}}(\mu H_{c2})^2). \quad (48)$$

Equation (48) may be solved for $H_{c2}(t)$, with the result

$$H_{c2}(t)=(3\Delta_{00}/\tau_{\mathrm{tr}}v^2e)\beta^{-2}\phi(t), \quad (49)$$

where

$$\phi(t)=4\gamma\beta^2t\rho/[1+(1+4\gamma\beta^2t\rho)^{1/2}], \quad (50)$$

$$\beta=(3\mu/\tau_{\mathrm{tr}}v^2e)(3\tau_{\mathrm{so}}\Delta_{00})^{1/2}, \quad\text{and}\quad \gamma=1.78. \quad (51)$$

Alternatively, we can express $H_{c2}(t)$ in terms of H_{c2}' (the upper critical field in the absence of the Pauli term) by

$$H_{c2}(t)=\frac{2H_{c2}'(t)}{1+[1+(12\tau_{\mathrm{so}}\mu^2/\tau_{\mathrm{tr}}v^2e)H_{c2}'(t)]^{1/2}}. \quad (52)$$

The asymptotic forms of $\kappa_1(t)=H_{c2}(t)/\sqrt{2}H_c(t)$ in the present case are given by

$$\kappa_1(t)=\frac{\kappa\pi^2}{2(14\zeta(3))^{1/2}}\frac{2}{1+(1+\beta^2)^{1/2}}\left(1-\frac{\gamma^2}{3}\frac{t^2}{(1+\beta^2)^{1/2}}\right)$$

$$=1.20\kappa\frac{2}{1+(1+\beta^2)^{1/2}}\left(1-1.05\frac{t^2}{(1+\beta^2)^{1/2}}\right),\quad \text{for } t\ll 1, \tag{53}$$

$$=\kappa\left[1-\left(\frac{28\zeta(3)}{\pi^4}-\frac{31\zeta(5)}{98\zeta^2(3)}-\frac{2\gamma}{\pi^2}\beta^2\right)\theta\right]$$

$$=\kappa(1+(0.119-0.361\beta^2)\theta),\quad \text{for } t\simeq 1. \tag{54}$$

The expressions for $g(\rho,b)$ and $f_1(\rho,b)$ simplify to

$$g(\rho,b)=\left(1+\frac{9\tau_{\text{so}}\mu^2}{\tau_{\text{tr}}v^2e}H_{c2}(t)\right)\sum_{n=0}^{\infty}\frac{1}{(n+\frac{1}{2}+\rho)^2}$$

$$=(1+\phi(t))\sum_{n=0}^{\infty}\frac{1}{(n+\frac{1}{2}+\rho)^2}, \tag{55}$$

and

$$f_1(\rho,b)=\sum_{n=0}^{\infty}\left\{\frac{1}{(n+\frac{1}{2}+\rho)^3}-\frac{3\tau_{\text{so}}(\mu H_{c2})^2}{4\pi T}\frac{1}{(n+\frac{1}{2}+\rho)^4}\right\}$$

$$=\sum_{n=0}^{\infty}\left\{\frac{1}{(n+\frac{1}{2}+\rho)^3}-\frac{\phi^2(t)}{4\gamma\beta^2 t}\frac{1}{(n+\frac{1}{2}+\rho)^4}\right\}, \tag{56}$$

respectively.

Substituting the above expressions in Eq. (43) we can calculate $\kappa_2(t)$. The asymptotic forms of $\kappa_2(t)$ are given by

$$\kappa_2(t)=\frac{\kappa\pi^2}{2(14\zeta(3))^{1/2}}\frac{\{2+2(1+\beta^2)^{1/2}+\frac{1}{3}\beta^2-4\gamma^2t^2[2+2(1+\beta^2)^{1/2}-\frac{1}{3}\beta^2-\frac{1}{9}(\beta^2/(1+\beta^2)^{1/2})]\}^{1/2}}{(1+\beta^2)^{1/2}(1+(1+\beta^2)^{1/2})(1-\gamma^2\beta^2t^2/3(1+\beta^2))}$$

$$\cong 1.20\kappa\frac{(2+2(1+\beta^2)^{1/2}+\frac{1}{3}\beta^2)^{1/2}}{(1+\beta^2)^{1/2}(1+(1+\beta^2)^{1/2})}(1-0t^2),\quad \text{for } t\ll 1, \tag{57}$$

$$=\kappa\left[1+\left(\frac{56}{\pi^4}\zeta(3)-\frac{45\zeta(4)}{7\pi^2\zeta(3)}-\frac{4\gamma}{\pi^2}\beta^2\right)\theta\right]$$

$$=\kappa(1+(0.106-0.722\beta^2)\theta),\quad \text{for } t\simeq 1. \tag{58}$$

The temperature dependence of $\kappa_1(t)$ and $\kappa_2(t)$ has been numerically calculated and the results are presented in Figs. 3 and 4. We see from the above expressions that $\kappa_2(t)$ never vanishes and conclude that in the present limit ($\tau_{\text{so}}\Delta_{00}\ll 1$) the transition is always of second order, since in the material of interest, κ is always larger than 1.

We further note that the relation

$$1\geq\kappa_2(0)/\kappa_1(0)=(2+2(1+\beta^2)^{1/2}+\tfrac{1}{3}\beta^2)^{1/2}/2(1+\beta^2)^{1/2}\geq 1/2\sqrt{3} \tag{59}$$

holds. $\kappa_2(0)$ is generally smaller than $\kappa_1(0)$ if there is an appreciable effect of the Pauli term independently of the value of $\tau_{\text{so}}\Delta_{00}$. For example, in the other limit $\tau_{\text{so}}\Delta_{00}\gg 1$, we have

$$\kappa_2(0)/\kappa_1(0)=((1-\alpha^2)/(1+\alpha^2))^{1/2}. \tag{60}$$

We note also that in the present limit the tunneling density of state is given by[13]

$$N(\mathbf{r},\omega)=N(0)(1-\tfrac{1}{2}|\Delta(\mathbf{r})|^2(\Gamma_s^2-\omega^2)/(\omega^2+\Gamma_s^2)^2), \tag{61}$$

where $\Gamma_s=2\pi T\rho(t)$ and $\rho(t)$ is given by Eq. (42). The normalization of $|\Delta(\mathbf{r})|^2$ is given by[8]

$$\langle|\Delta(\mathbf{r})|^2\rangle_{\text{av}}=\frac{eT}{\sigma}\frac{H_{c2}-H_0}{(2\kappa_2^2(t)-1)\beta_A}(g(\rho,b))^{-1}, \tag{62}$$

where σ is the conductivity in the normal state.

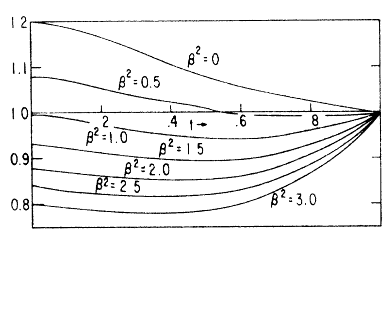

FIG. 3. Numerical results for the temperature dependence of κ_1/κ for $\beta^2=0$, 0.5, 1, 1.5, 2, 2.5, 3. See Eq. (51) for the definition of β^2.

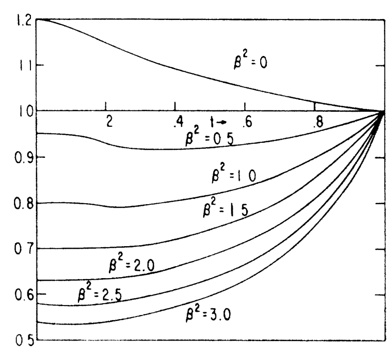

FIG. 4. Numerical results for the temperature dependence of κ_2/κ for $\beta^2=0$, 0.5, 1, 1.5, 2, 2.5, 3.

[13] P. G. de Gennes, Physik Kondensierten Materie 3, 79 (1964).

V. CONCLUDING REMARKS

We have discussed how spin-orbital scattering strongly modifies the effect of the Pauli term on the magnetic properties of high field superconductors. We have also established in these general situations that we can describe the magnetic properties of the system in terms of Abrikosov's theory, except that we have to introduce two parameters: $\kappa_1(t)$ and $\kappa_2(t)$. We have shown that in the situation where the Pauli term is important, the relation $\kappa_2(t)\leq\kappa_1(t)$ generally holds.

It is not difficult to calculate various transport coefficients in the gapless region (or $\Delta(\mathbf{r})\ll\pi T_{c0}$). Especially in the limit $l_{so}/\xi_0<1$ (where ξ_0 is the BCS coherence length), expressions for the transport coefficients such as electromagnetic conductivity[14] and thermal conductivity[15] are similar to those for type-I superconductors with paramagnetic impurities. (Only the normalization of $\langle|\Delta|^2\rangle$ is different in the present case.)[16]

We might point out that the spin contribution to the magnetization can be measured separately from the Knight shift in these specimens. This is discussed in Appendix B.

ACKNOWLEDGMENTS

I would like to thank Professor P. G. de Gennes and Dr. C. Caroli for giving me the opportunity of seeing their work before publication.

APPENDIX A: THE CALCULATION OF $\mathfrak{F}_\omega(q_i)$

The contribution from the diagram A of Fig. 2 (we denote it as A) is given by

$$A=\sum_{\pm}\int\frac{d^3p}{(2\pi)^3}\left(\frac{1}{i\tilde{\omega}_\pm-\xi}\eta_{\pm q_1}\frac{1}{i\tilde{\omega}_\pm+\xi+\mathbf{v}\cdot\mathbf{q}_1}\eta_{\pm q_2}\frac{1}{i\tilde{\omega}_\pm-\xi-\mathbf{v}\cdot(\mathbf{q}_1+\mathbf{q}_2)}\eta_{\pm q_3}\frac{1}{i\tilde{\omega}_\pm-\xi-\mathbf{v}\cdot\mathbf{q}_4}\eta_{\pm q_4}\right)$$

$$=-\frac{mp_0}{2(2\pi^2)}\sum_{\pm}i\int d\Omega\Big(\prod_{i=1}^{4}\eta_{\pm q_i}\Big)\left(\sum_{\text{cyclic}}\frac{1}{2i\tilde{\omega}_\pm+\mathbf{v}\cdot\mathbf{q}_1}\frac{1}{2i\tilde{\omega}_\pm-\mathbf{v}\cdot\mathbf{q}_2}\frac{1}{2i\tilde{\omega}_\pm+\mathbf{v}\cdot\mathbf{q}_3}\right)$$

$$=\frac{mp_0}{2\pi^2}\sum_{\pm}\frac{2}{(2\tilde{\omega}_\pm)^3}\Big(\prod_{i=1}^{4}\eta_{\pm q_i}\Big)\left\{1-\tau\left(\frac{\tau v^2}{3}\right)(\mathbf{q}_1^2+\mathbf{q}_2^2+\mathbf{q}_3^2+\mathbf{q}_4^2+\mathbf{q}_1\cdot\mathbf{q}_3+\mathbf{q}_2\cdot\mathbf{q}_4)\right\}, \qquad \text{(A1a)}$$

while the contributions from B and C are given as

$$B=\frac{1}{2i}\left(\frac{2\pi^2}{mp_0}\right)\sum_{\pm}\Big(\prod_{i=1}^{4}\eta_{\pm q_i}\Big)\int\frac{d^3p}{(2\pi)^3}\left(\frac{1}{i\tilde{\omega}_\pm-\xi}\frac{1}{i\tilde{\omega}_\pm+\xi+\mathbf{v}\cdot\mathbf{q}_1}\frac{1}{i\tilde{\omega}_\pm-\xi-\mathbf{v}\cdot(\mathbf{q}_1+\mathbf{q}_2)}\right)$$
$$\times\int\frac{d^3p'}{(2\pi)^3}\left(\frac{1}{i\tilde{\omega}_\pm-\xi'}\frac{1}{i\tilde{\omega}_\pm+\xi'-\mathbf{v}\cdot\mathbf{q}_4}\frac{1}{i\tilde{\omega}_\pm-\xi-\mathbf{v}\cdot(\mathbf{q}_1+\mathbf{q}_2)}\right)$$
$$+\frac{1}{3\tau_{so}}\left(\frac{2\pi^2}{mp_0}\right)\sum_{\pm}(\eta_{\pm q_1}\eta_{\pm q_2}\eta_{\mp q_3}\eta_{\mp q_4})\int\frac{d^3p}{(2\pi)^3}\left(\frac{1}{i\tilde{\omega}_\pm-\xi}\frac{1}{i\tilde{\omega}_\pm+\xi+\mathbf{v}\cdot\mathbf{q}_1}\frac{1}{i\tilde{\omega}_\pm-\xi-\mathbf{v}\cdot(\mathbf{q}_1+\mathbf{q}_2)}\right)$$
$$\times\int\frac{d^3p'}{(2\pi)^3}\left(\frac{1}{i\tilde{\omega}_\mp-\xi'}\frac{1}{i\tilde{\omega}_\mp+\xi'-\mathbf{v}\cdot\mathbf{q}_4}\frac{1}{i\tilde{\omega}_\mp-\xi'-\mathbf{v}\cdot(\mathbf{q}_1+\mathbf{q}_2)}\right) \qquad \text{(A1b)}$$

$$=-\frac{mp_0}{2\pi^2}\sum_{\pm}\left\{\frac{1}{2\tau}\Big(\prod_{i=1}^{4}\eta_{\pm q_i}\Big)\frac{1}{(2\tilde{\omega}_\pm)^4}\left[1-\tau(\tfrac{1}{3}\tau v^2)(\mathbf{q}_1^2+\mathbf{q}_2^2+\mathbf{q}_3^2+\mathbf{q}_4^2-\mathbf{q}_1\cdot\mathbf{q}_2-\mathbf{q}_3\cdot\mathbf{q}_4)\right]\right.$$
$$\left.+\frac{1}{3\tau_{so}}(\eta_{\pm q_1}\eta_{\pm q_2}\eta_{\mp q_3}\eta_{\mp q_4})\frac{1}{(2\tilde{\omega}_+)^2(2\tilde{\omega}_-)^2}\left[1-\tau(\tfrac{1}{3}\tau v^2)(\mathbf{q}_1^2+\mathbf{q}_2^2+\mathbf{q}_3^2+\mathbf{q}_4^2-\mathbf{q}_1\cdot\mathbf{q}_2-\mathbf{q}_3\cdot\mathbf{q}_4)\right]\right\}, \qquad \text{(A2)}$$

$$C=-\frac{mp_0}{2\pi^2}\sum_{\pm}\left[\frac{1}{2\tau}\Big(\prod_{i=1}^{4}\eta_{\pm q_i}\Big)\frac{1}{(2\tilde{\omega}_\pm)^4}(1-\tau(\tfrac{1}{3}\tau v^2)(\mathbf{q}_1^2+\mathbf{q}_2^2+\mathbf{q}_3^2+\mathbf{q}_4^2-\mathbf{q}_1\cdot\mathbf{q}_4-\mathbf{q}_2\cdot\mathbf{q}_3))\right.$$
$$\left.+\frac{1}{3\tau_{so}}(\eta_{\pm q_1}\cdot\eta_{\mp q_2}\cdot\eta_{\mp q_3}\cdot\eta_{\pm q_4})\frac{1}{(2\tilde{\omega}_+)^2(2\tilde{\omega}_-)^2}(1-\tau(\tfrac{1}{3}\tau v^2)(\mathbf{q}_1^2+\mathbf{q}_2^2+\mathbf{q}_3^2+\mathbf{q}_4^2-\mathbf{q}_1\cdot\mathbf{q}_4-\mathbf{q}_2\cdot\mathbf{q}_3))\right]. \qquad \text{(A3)}$$

[14] K. Maki, Phys. Rev. **141**, 331 (1966).
[15] C. Caroli and M. Cyrot, Physik Kondensierten Materie **4**, 285 (1965).
[16] The general proof proceeds in a similar fashion as given in K. Maki and P. Fulde, Phys. Rev. **140**, A1586 (1965).

Summing up the above contributions we finally have

$$A+B+C=\frac{2mp_0}{2\pi^2}\sum_{\pm}\left\{(\prod_{i=1}^{4}\eta_{\pm\mathbf{q}_i})\frac{1}{(2\tilde{\omega}_\pm)^4}\left[\omega_\pm+\frac{\tau v^2}{24}((\mathbf{q}_1-\mathbf{q}_3)^2+(\mathbf{q}_2-\mathbf{q}_4)^2)\right]\right.$$
$$\left.-\frac{2}{3\tau_{so}}(\eta_{\pm q_1}\eta_{\pm q_2}\eta_{\mp q_3}\eta_{\mp q_4}+\eta_{\pm q_1}\eta_{\mp q_2}\eta_{\mp q_3}\eta_{\pm q_4})\frac{1}{(2\tilde{\omega}_+)^2(2\tilde{\omega}_-)^2}\right\}, \quad (A4)$$

and the $\mathfrak{F}_\omega(q_i)$ are given as

$$\mathfrak{F}_\omega(\mathbf{q}_i)=(\pi^2/mp_0)(\pi T)^3[A+B+C]. \quad (A5)$$

APPENDIX B: THE KNIGHT SHIFT IN TYPE-II SUPERCONDUCTORS

The current due to the spin paramagnetism of electrons in the superconducting state [the second term in Eq. (37)] can be rewritten as

$$\mathbf{j}_s(\mathbf{r})|_{\text{spin}}=\nabla\times(\boldsymbol{M}_s(\mathbf{r})-\boldsymbol{M}_n(\mathbf{r}))_{\text{spin}} \quad (B1)$$

where $M_s|_{\text{spin}}(\mathbf{r})$ and $M_n(\mathbf{r})(=N(0)\mu^2H)$ are the magnetization due to spin polarization of the electrons in the superconducting and normal state, respectively.

Making use of Eq. (37) we have

$$M_s(\mathbf{r})|_{\text{spin}}=M_n\left\{1-\frac{\pi T}{I}|\Delta(\mathbf{r})|^2\left[\text{Im}\sum_n^{\infty}\left(\frac{\omega+iI+a+b}{(\omega+a)^2-b^2+I^2}\right)^2\right]\right\}$$
$$=M_n\left\{1-\frac{\pi T|\Delta(\mathbf{r})|^2}{4(b^2-I^2)^{1/2}}\left[\frac{1}{4\pi T}\left[\left(1+\frac{b}{(b^2-I^2)^{1/2}}\right)\psi'(\tfrac{1}{2}+\rho_-)\right.\right.\right.$$
$$\left.\left.\left.-\left(1-\frac{b}{(b^2-I^2)^{1/2}}\right)\psi'(\tfrac{1}{2}+\rho_+)\right]+\frac{b}{b^2-I^2}[\psi(\tfrac{1}{2}+\rho_-)-\psi(\tfrac{1}{2}+\rho_+)]\right]\right\}, \quad (B2)$$

where

$$\rho_\pm=\frac{1}{2\pi T}[a\pm(b^2-I^2)^{1/2}],$$
$$a=\tau_{tr}v^2eH_{c2}/6+1/3\tau_{so},\quad b=1/3\tau_{so},\quad \text{and}\quad I=\mu H. \quad (B3)$$

Here $\psi(z)$ and $\psi'(z)$ are the usual digamma and trigamma functions.

We note that the shift in the nuclear-magnetic-resonance frequency (the Knight shift) is proportional to

$$\delta\omega=4\pi\mu M_s|_{\text{spin}}(\mathbf{r}). \quad (B4)$$

In principle, by measurement of the Knight shift in type-II superconductors, we can separate the total magnetization into the term due to the diamagnetic current and that due to the spin susceptibility.

In the limit $I\rightarrow 0$, we have from Eq. (B2)

$$\frac{\chi_s(\mathbf{r})}{\chi_n}=1-\pi T|\Delta(\mathbf{r})|^2\sum_n^{\infty}\frac{1}{(\omega_n+a+b)(\omega_n+a-b)^2}$$
$$=1-\frac{2|\Delta(\mathbf{r})|^2}{(\pi T)^2\rho_c}\left\{\tfrac{1}{4}\psi'(\tfrac{1}{2}+\rho)+\frac{1}{2\rho_c}[\psi(\tfrac{1}{2}+\rho)-\psi(\tfrac{1}{2}+\rho+\tfrac{1}{2}\rho_c)]\right\}, \quad (B5)$$

where $\rho_c=2/(3\pi\tau_{so}T)$. The above expression is equivalent to the spin susceptibility in superconducting thin films in a parallel magnetic field.[17]

[17] P. Fulde and K. Maki, Phys. Rev. **139**, A788 (1965); A. I. Larkin, Zh. Eksperim. i Teor. Fiz. **48**, 232 (1965) [English transl.: Soviet Phys.—JETP **21**, 153 (1965)].