Spin paramagnetism in \( d \)-wave superconductors

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The Ginzburg-Landau equations are derived from the microscopic theory for clean layered superconductors with \( d_{x^2-y^2} \) pairing symmetry, including the Pauli paramagnetism effect. The upper critical field \( H_{c2} \) parallel to the \( c \) axis is calculated. A comparison with the experimental data for \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) suggests that, relative to the orbital effect, the Pauli paramagnetism contribution to \( H_{c2} \) is significant. The reversible magnetization \( M \) in high magnetic fields is also calculated, showing strong temperature dependence of the slope \( dM/dH \), as a consequence of the spin paramagnetism. A simple expression for the high-temperature spin susceptibility is derived, in good agreement with the Knight-shift measurements on \( \text{YBa}_2\text{Cu}_3\text{O}_6 \).

I. INTRODUCTION

Among the most important properties of the cuprate superconductors are their magnetic properties, reflecting the highly anisotropic layered structure and unconventional pairing mechanism and symmetry.\(^1\) Nowadays, \( d \)-wave pairing symmetry in hole-doped cuprates,\(^2\) and quasi-two-dimensional (2D) nature of superconductivity,\(^3\) are well established. Although the pairing mechanism is not known, the weak coupling BCS model for 2D \( d \)-wave superconductivity,\(^4\) and the corresponding Ginzburg-Landau (GL) approximation,\(^5\)–\(^8\) are remarkably successful.

The existence of a superconducting phase in the very high magnetic fields and layered structure of high-\( T_c \) superconductors makes the paramagnetic effect in the superconducting state much more important than in conventional superconductors.\(^9\),\(^10\) The purpose of this paper is to derive GL equations for layered \( d \)-wave superconductors including the Pauli paramagnetism effect, giving simple analytical expressions for the upper critical field, magnetization, and spin susceptibility, suitable for comparison with experiments.

The GL equations for conventional (isotropic 3D \( s \)-wave) superconductors were first derived from BCS theory by Gor'kov.\(^11\) For 2D clean superconductors with \( d \)-wave pairing, GL equations are derived by Ren \textit{et al.} \(^5\) and extended by Won and Maki,\(^7\) and Shiraiishi \textit{et al.}\(^9\) to include the higher-order derivative terms. However, in the above references, the magnetic-field influence on the electron spins has been neglected. In their classical papers, Maki and Tsuneto studied the effect of the Pauli paramagnetism in conventional superconductors.\(^12\),\(^13\) Their results mainly refer to the dirty limit.

In many underdoped and overdoped cuprates, where the upper critical fields \( H_{c2} \) and transition temperatures \( T_c \) are relatively low, the upward, positive curvature in the temperature dependence has been observed,\(^14\),\(^15\) unlike the Werthamer-Helfand-Hohenberg theory prediction for the conventional superconductors.\(^16\) A number of theoretical models for this unusual behavior have been proposed, involving, for example, the influence of the scattering by magnetic impurities and of inhomogeneities,\(^17\) the presence of \( d_{xy} \) pairing symmetry,\(^18\) and the higher Landau levels effect.\(^19\)

In the optimally doped cuprates, the temperature dependence of \( H_{c2} \) parallel to the \( c \) axis appears to be qualitatively the same as in the conventional superconductors. However, high values of \( H_{c2} \) and the irreversibility effects pose an obstacle to the study of the field-induced transition to the normal state.\(^20\) The magnetization measurements of \( H_{c2} \) on high-quality single crystals of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) (YBCO) are performed by Welp \textit{et al.}\(^21\) in a temperature interval of about 8 K below \( T_c \). Recently, Nakagawa \textit{et al.}\(^22\),\(^23\) for field parallel and O'Brien \textit{et al.}\(^24\) for field perpendicular to the \( c \) axis, reported data from GHz transport measurements up to 150 T of \( H-T \) phase diagram for YBCO thin films in the whole temperature range.

Yang and Sondhi,\(^9\) and Won \textit{et al.}\(^10\) studied, theoretically, the paramagnetic state of \( d_{x^2-y^2} \) superconductors, neglecting the coupling of the magnetic field to the orbital motion of electrons in the superconducting planes. For perpendicular field, measurements support their theory, strongly suggesting that \( H_{c2} \) is limited by the spin paramagnetism below a certain characteristic temperature \( T^\star \approx 0.85 T_c \).\(^24\) For parallel field the role of the spin paramagnetism should be clarified with regard to the orbital effect.

Magnetization measurements on \( \text{YBa}_2\text{Cu}_4\text{O}_8 \) in intermediate fields of Sok \textit{et al.}\(^25\) are in a good agreement with the GL-like Hao and Clem model.\(^26\) In higher fields, where the magnetic phase diagram contains a vortex fluid,\(^20\) one can presume that the diamagnetism of the vortex fluid resembles closely that of an ideal Abrikosov mixed state at temperatures not too close to \( T_c \).\(^27\) In this regime, the measurements of cuprates should have the same slope in different fields, with strong temperature dependence due to the spin paramagnetism.

Electronic-spin-susceptibility measurements provide evi-
II. GINZBURG-LANDAU EQUATIONS

We have derived the GL equations for clean 2D $d_{x^2-y^2}$ superconductors, extending the procedure used by Ren et al. to include the effect of the spin paramagnetism, analogously to the Maki and Tsuneto approach for $s$-wave superconductors.

For a stack of identical 2D conducting planes in the magnetic field parallel to the $c$ axis, the obtained GL equations are

$$\alpha \psi + \beta |\psi|^2 \psi + \frac{1}{2m} \mathbf{H} \times \psi + \eta H^2 \psi = 0,$$  \hspace{1cm} (2.1)

$$\mathbf{j}_s = -\frac{i e}{m} \left( \psi \frac{\partial \psi^*}{\partial \mathbf{r}} - \psi^* \frac{\partial \psi}{\partial \mathbf{r}} \right) - 4e^2 \frac{a^2}{mc} |\psi|^2 - 2e \mathbf{A} \mathbf{c} \times |\psi|^2,$$  \hspace{1cm} (2.2)

where $\mathbf{A} = -i \hbar \partial / \partial \mathbf{r} - 2e \mathbf{A} \mathbf{c} / c$, and

$$\alpha = \frac{16 \pi^2 k_B^2 T_{c}^2}{\xi^2} \frac{T - T_c}{T_c},$$  \hspace{1cm} (2.3)

$$\beta = \frac{48 \pi^2 k_B^2 T_{c}^2}{\xi^2} \frac{N^*(0)}{m^2 v_F},$$  \hspace{1cm} (2.4)

$$\eta = \frac{4}{mv_F^2 \mu_B^2}.$$  \hspace{1cm} (2.5)

Here, $\mu_B$ is the Bohr magneton, and $N^*(0) = N(0)/\delta$, $\delta$ being the average spacing between the conducting planes.

In this model, the GL equations for a $d$-wave, clean and layered superconductor are of the same form as in the isotropic and clean 3D $s$-wave case. However, there is a factor 2/3 in parameters $\alpha$, $\beta$, $\eta$, and the quantities $m, v_F, N(0) = m/(2\pi h^2)$ refer to the effective mass, Fermi velocity and density of states in 2D conducting $ab$ planes. Therefore, the coherence length is $\xi_{ab} = (h/2m |\alpha|)^{1/2}$, the penetration depth $\lambda_{ab} = (mc^2 \beta/16 \pi e^2 |\alpha|)^{1/2}$, and the GL parameter

$$\kappa = \frac{6c k_B T_c}{e \hbar v_F} \sqrt{\frac{\pi}{7\xi(3)N^*(0)}}.$$  \hspace{1cm} (2.6)

The free-energy density corresponding to Eqs. (2.1) and (2.2) is

$$F = F_n + \alpha |\psi|^2 + \frac{\beta}{2} |\psi|^4 + \frac{1}{2m} \mathbf{H} \cdot \mathbf{H}^* |\psi|^2 + \frac{H^2}{8 \pi} + \eta H^2 |\psi|^2.$$  \hspace{1cm} (2.7)

III. UPPER CRITICAL FIELD

Near the second-order phase transition to the normal phase, from the linearized Eq. (2.1), we obtain for the upper critical magnetic field parallel to the $c$ axis

$$H_{c2} = \frac{e \hbar v_F^2}{8c \mu_B^2} \sqrt{1 + \frac{256 \pi^2 c^2 \mu_B^2 k_B T_{c}^2}{7\xi(3) e^2 H^2 v_F^4 \left( 1 - \frac{T}{T_c} \right)}}.$$  \hspace{1cm} (3.1)

The slope at $T_c$ is

$$\frac{dH_{c2}}{dT} \bigg|_{T_c} = -\frac{16 \pi^2 c k_B T_c}{7\xi(3) e \hbar v_F^2},$$  \hspace{1cm} (3.2)

and the GL expression without the Pauli paramagnetism correction is simply $H_{c2} = |dH_{c2}/dT|_{T_c}(T_c - T)$.

We illustrate our results using the experimental data for YBCO. Samples are in the clean limit, and for fields above 1 T, the spin-orbit scattering can be neglected. Taking the slope $dH_{c2}/dT|_{T_c} = -1.9$ T/K from the magnetization measurements on optimally doped YBCO ($T_c = 92$ K) of Welp et al., we find that at $T = 0.7 T_c$ the spin pair breaking lowers the critical field by 10%, Fig. 1(a). However, it is evident that a larger slope corresponds to the data of Nakagawa et al. We obtain with Eq. (3.1) a good fit of the experimental data for $T \approx 0.5 T_c$, taking the slope $dH_{c2}/dT|_{T_c} = -2.6$ T/K and $T_c = 84.3$ K, Fig. 1(b). In this case the paramagnetic correction is $-15\%$ at $T = 0.7 T_c$.

At low temperatures, where GL theory is not applicable, $H_{c2}(T)$ becomes saturated. From the data of Nakagawa et al., $H_{c2}(0) = 110$ T. The Won and Maki expression $H_{c2}(0) = -0.63 T_c dH_{c2}/dT|_{T_c}^2$ relevant for 2D $d$-wave superconductors, for $T_c = 84.3$ K and the slope $-2.6$ T/K gives $H_{c2}(0) = 138$ T. Therefore, the spin paramagnetism, not taken into account in Ref. 7, but relevant in this case because the Clogston limit is 155 T, should lower $H_{c2}(0)$ by 20%.

Note that the Fermi velocity corresponding to $dH_{c2}/dT|_{T_c} = -2.6$ T/K, $T_c = 84.3$ K is $v_F = 8.3 \times 10^6$ cm/s, in agreement with $v_F = (7.6 \pm 0.9) \times 10^6$ cm/s, obtained from independently measured Fermi energy and the effective mass.

IV. REVERSIBLE MAGNETIZATION

Following the Abrikosov approach, in the vicinity of the upper critical field parallel to the $c$ axis of the layered superconductor, we find that as in the 3D $s$-wave case, the effect of the Pauli paramagnetism on the induction $B$ is included simply by the scaling of $\kappa \to \kappa'$.

$$B = B_{c} - (H_{c2} - H_{c}) \frac{1}{(2\kappa'^2 - 1)} \beta.$$  \hspace{1cm} (4.1)

Here, $H_{c}$ is the external magnetic field, and $\kappa' = \kappa \sqrt{1 + 4 \kappa^2 \gamma}$, with $\gamma = (32 \pi^3 5N^*(0) \mu_B^2 (1 - 1/T_c^2))$. The vortex lattice is again an equilateral triangular lattice with $\beta = 1.16$. The temperature dependence of $\kappa'$ leads to the characteristic variation of the magnetization with temperature. For $\kappa \gtrsim 1$, the slope
is linearly decreasing with temperature, instead of the temperature-independent slope in the GL approach without paramagnetic correction. Due to higher $T_c$ and smaller $v_F$, this effect is not negligible in high-$T_c$ cuprates, in contrast to the conventional superconductors. For example, a strong temperature variation $(dM/dH_c)_{T_c=1} = 2.6(1 - T/T_c)$ corresponds to $dH_{c2}/dT|_{T_c} = -2.6$ T/K.

In high-$T_c$ superconductors, the influence of the vortex lattice melting and the strong fluctuation effects in the vicinity of $T_c$ also affect $dM/dH_c$ temperature dependence. However, because of the strong magnetic effect, we expect that the characteristic increase of $dM/dH_c$ with decreasing temperature, predicted by Eq. (4.2), should persist as a dominant effect for $H \geq 0.5 H_{c2}$, and $0.5 \leq T/T_c \leq 0.9$.

V. SPIN SUSCEPTIBILITY

The current density due to the spin paramagnetism is

$$j_s^{spin}(r) = c \text{ curl}\left[\mathbf{M}_{s}^{spin}(r) - \mathbf{M}_n(r)\right],$$  

where $\mathbf{M}_{s}^{spin}(r)$ and $\mathbf{M}_n(r) = 2N_s(0)\mu_B^2\mathbf{H}$ are the magnetization due to the spin polarization in the superconducting and in the normal state. From Eq. (5.1) and the last term in Eq. (2.2),

$$M_s^{spin} = M_n - \frac{8\mu_B^2}{m^*v_F^2}H|\psi|^2.$$  

In the weak magnetic field $|\psi|^2 = -\alpha/\beta$, and the spin susceptibility $\chi_s = M_s^{spin}/H$ is

$$\chi_s = \chi_n \left[ 1 - \frac{4}{3} \left( 1 - \frac{T}{T_c} \right) \right].$$

This simple GL expression for the spin susceptibility of the layered $d$-wave superconductor is in very good agreement with the microscopic theory result of Won and Maki, in a large temperature range $T \approx 0.6 T_c$. Analogously, the susceptibility in the GL approach for an isotropic 3D $s$-wave, as well as for 2D $s$-wave, superconductor is $\chi_s/\chi_n = 1 - 2(1 - T/T_c)$, in the agreement with the Yosida result from BCS theory. Therefore, $d$-wave and $s$-wave symmetries of the order parameter lead to different slopes of the spin-susceptibility curve in the high-temperature regime.

This is illustrated in Fig. 2, in comparison with the ESR Knight-shift data for YBa$_2$Cu$_4$O$_8$ of Jánossy et al. Small discrepancy of the experimental data and the theoretical predictions at high temperatures could be the consequence of...
the strong coupling effects, or of the presence of a small $s$-wave component of the order parameter, which is possible in Y-based superconductors. 

VI. CONCLUDING REMARKS

The GL equations for layered $d$-wave clean superconductors are derived including the Pauli paramagnetism effect. The parallel upper critical field and the reversible magnetization in the isotropic 3D $s$-wave case, with changed $\kappa$ and $\gamma$. The expression for $H_{c2}$ gives a useful correction to the standard GL result, providing the correct determination of $dH_{c2}/dT$ for high-$T_c$ superconductors from the fit of experimental data in the relatively large range of temperatures below $T_c$. From the comparison with the experimental data for YBa$_2$Cu$_3$O$_{7-\delta}$ thin films, we obtain $dH_{c2}/dT|_{Tc} = -2.6$ T/K, and conclude that the effect of Pauli paramagnetism on $H_{c2}$ parallel to the $c$ axis is significant in comparison to the orbital effect, $-15\%$ at $T=0.7$ $T_c$. At zero temperature, this effect should be greater, about $-20\%$. The strong temperature dependence of the magnetization slope $dM_1/dH_c$ due to the spin paramagnetism influence, should be experimentally detectable in high-field measurements. We have also derived the GL expression for the Knight shift, reflecting the pairing symmetry in accordance with the experimental data.

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