Critical magnetic field in layered superconductors

Yu. N. Ovchinnikov*
Institute for Condensed Matter Theory, University of Karlsruhe, Karlsruhe, 76128 Germany

Vladimir Z. Kresin
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

(Received 27 February 1995; revised manuscript received 15 March 1995)

The nature of the dependence $H_{c2}(T)$ is greatly affected by the presence of magnetic impurities. The scattering leads to change in the sign of the curvature and, consequently, to an increase in the value of H_{c2}. The theory allows one to explain recent experimental data with the overdoped cuprates; the behavior drastically different from the conventional picture has been observed. The calculations are in excellent agreement with the data.

This paper is concerned with the magnetic properties of layered superconductors (e.g., cuprates, organic materials, etc.). More specifically, we focus on the unusual temperature dependence of the critical magnetic field H_{c2} in these materials. As is known, the behavior of conventional superconductors is described by Helfand-Werthamer (HW) theory[1] (see also Refs. 2–6) and is characterized by a saturation of $H_{c2}(T)$ for $T\to 0$. It will be shown below that the behavior of layered superconductors can be entirely different. Contrary to the conventional picture, they can display a positive curvature of $H_{c2}(T)$ over a wide temperature range. Moreover, near $T=0$ K, H_{c2} increases almost linearly toward the finite value $H_{c2}(0)\equiv H_{c2}(T=0\text{ K})$, which greatly exceeds the value $H_{c2}(0)_{\text{conv}}$. The effect is caused by the presence of magnetic impurities and their ordering at low temperatures.

Our study was motivated by the recent observation of an exotic temperature dependence of $H_{c2}(T)$ reported in Refs. 7 and 8. These experiments studied $H_{c2}(T)$ in $Tl_2Ba_2CuO_6$ (Ref. 7) and $Bi_2Sr_2CuO_6$ (Ref. 8) and observed a dependence drastically different from the conventional picture.[1] It turns out that in the cuprates $\partial^2 H_{c2}/\partial T^2>0$; near $T\to 0$ one observed an almost linear temperature dependence. Similar behavior was observed previously for other layered superconductors,[9–11] including other cuprates[9] and organics.[10]

Consider a layered superconductor containing magnetic impurities. Near the critical field H_{c2}, the order parameter $\Delta(r)$ satisfies the linear equation

$$-\partial_-^2\Delta=2\lambda\Delta\ . \tag{1}$$

Here $\partial_-=(\partial/\partial\boldsymbol{\rho}-2ie\,\mathbf{A})$ and $\lambda=eH_{c2}$.[1] Our approach is based on the method of integrated Green's functions developed in Refs. 2 and 3. The critical magnetic field H_{c2} can be determined from the following general equation, which is valid in three-dimensional (3D) as well as in 2D cases:[4,5]

$$\ln(T_c/T)=2\pi T\sum_{n\geq 0}[\omega_n^{-1}-2D_1(\omega_n,H)], \tag{2}$$

where

$$D_1(\omega_n,H)=(\text{sgn}\,\omega_n)J(\omega_n,H)[1-(\tau^{-1}-\tau_s^{-1})J(\omega_n,H)]^{-1},$$

$\omega_n=(2n+1)\pi T$, τ_s is the spin-flip scattering relaxation time, and τ describes the usual elastic scattering. The explicit expression for the kernel $J(\omega,H)$ depends on dimensionality. The expression for the 3D case was derived in Refs. 4,5. We are interested in layered structures. One can show that in this case the function $J(\omega,H)$ has the form

$$J(\omega_n,H)=(2/\pi eHv^2)^{1/2}\times\int_0^\infty dy\,\exp(-y)\arctan[v(2eHy)^{1/2}/\alpha]\ , \tag{3}$$

where $\alpha=2\omega_n+\tau^{-1}+\tau_s^{-1}$ and v is the Fermi velocity. We focus on the case $\mathbf{H}\|c$.

We consider the case when the parameter τ^{-1} is small ($\tau^{-1}\ll\pi T_C$, "clean" case). On the other hand, we allow the spin-flip scattering channel to be important, so that $\Gamma=\tau_s^{-1}\gtrsim\pi T_c$. The presence of a layered structure allows one to combine the condition $\Gamma=\tau_s^{-1}\gtrsim\pi T_c$ with the condition $\tau^{-1}\ll\pi T_c$ (clean case) for the usual elastic scattering. This is due to the possibility of an out-of-plane location of the magnetic moments (see Ref. 12); as a result, the in-plane momentum transfer can be small, despite the large value of the amplitude for the spin-flip process. Based on Eqs. (2) and (3), we obtain

$$\ln(2\gamma\Gamma_{\text{cr}}/\pi T)-\{\psi[0.5+(\Gamma/2\pi T)]-\psi(0.5)\}=f(H,T)\ , \tag{4}$$

$$f(H,T)=(eHv^2/\Gamma)(\Gamma^{-1}\{\psi[0.5+(\Gamma/2\pi T)]-\psi[0.5+(\Gamma/4\pi T)]\}-(4\pi T)^{-1}\times\psi'[0.5+(\Gamma/2\pi T)])\ .$$

Here ψ is the psi function, $\ln\gamma=C=0.58$ is the Euler constant, and Γ_{cr} corresponds to the complete suppression of superconductivity ($T_c=0$). We assume also that $\alpha=eHv^2/\Gamma\ll 1$. If $\alpha>1$, one should use directly Eqs. (2) and (3). One should note that, strictly speaking, Γ depends on temperature (see below), so that $\Gamma=\Gamma(T)$.

Equation (4) is a general equation which allows one to evaluate the temperature dependence of $H=H_{c2}$ in the presence of magnetic impurities. If the condition $\Gamma \gg \pi T_c$ is satisfied, one can reduce Eq. (4) to the form

$$\ln(\Gamma_{cr}/\Gamma) - 0.17(\pi T/\Gamma)^2 + 0.12(\pi T/\Gamma)^4 = (eHv^2/\Gamma^2)[\ln 2 - 0.5 - 0.67(\pi T/\Gamma)^2 + 0.12(\pi T/\Gamma)^4] \ . \quad (5)$$

Let us focus on the low-temperature region $T \ll T_c$. If the parameter Γ is temperature independent (cf. Refs. 13–15), then we obtain a smooth, almost quadratic dependence $H_{c2}(T)$. However, if Γ depends on the temperature (see below), the picture becomes entirely different. It is important to stress that, because of the large value of Γ, a relatively small change in the latter leads to a noticeable change in the value of H_{c2}. Of course, the smallness of τ^{-1} is also an important factor.

The parameter Γ describes the spin-flip process. In almost the entire temperature region (except for a small area near $T=0$; see below), this parameter can be treated as temperature independent: $\Gamma=\Gamma_\alpha=\text{const}$ for $T \gg \theta$, $\theta \ll T_c$ (θ is defined below). The picture is similar to that described in Refs. 13 and 14. Indeed, the leading term in the amplitude of spin-flip scattering by magnetic impurities does not depend on the temperature; the small Kondo term was neglected in Refs. 13 and 14. However, in the low-temperature region, the impurities become ordered (see, e.g., the discussion in Ref. 15). This ordered state arises for $T<\theta$. The value of θ depends on the material; for the cuprates $\theta \simeq 1$ K, see below. In this region the usual spin-flip process, described by the amplitude Γ_α and based on total spin conservation, is forbidden. However, the spin-flip processes can also be governed by the dipole-dipole interaction (see, e.g., Ref. 16): $V=-3\mu_e\mu_i(\sigma_e \cdot \mathbf{r})\,(\sigma_i \cdot \mathbf{r})/r^5$, where r is the distance between the electron and the impurity; this interaction does not conserve total spin. Its amplitude at $T=0$ is related [see Eq. (6)] to the value of $H_{c2}(0)$. Note that $\Gamma_0 < \Gamma_\alpha$. As a result, the amplitude Γ becomes *temperature dependent* in the low-temperature region $T<\theta$ as Γ decreases from Γ_α to Γ_0. The temperature dependence $\Gamma(T)$ can be represented in the form

$$\Gamma = \Gamma_0 f(\tau), \quad f(\tau) = (1+\beta\tau)/(1+\tau) \ . \quad (6)$$

Here $\tau = T\theta^{-1}$ and $\beta = \Gamma_\alpha/\Gamma_0$; in our case, $\tau_c \gg 1$.

Equation (4) can be rewritten in the form

$$\ln(T_c/T) - \{\psi[0.5 + (\Gamma/2\pi T)] - \psi[0.5 + \beta(\Gamma_0/2\pi T_c)]\} = f(H,T) \ . \quad (7)$$

If we start with an ordinary layered superconductor characterized by an intrinsic critical temperature T_{c0} and add to it magnetic impurities; then, Eq. (5) at $T=T_c$ (here $H=H_{c2}=0$) and the relation $\Gamma_{cr}=\pi T_{c0}/2\gamma$ allow us to calculate the value of $\Gamma_\alpha = \beta\Gamma_0$, which is determined by the concentration of impurities (see Ref. 13). As for the parameter Γ_0, its value is directly related to the value of $H_{c2}(0)$. Therefore, the dependence $\Gamma(T)$ and, consequently, the shape of the function $H_{c2}(T)$ are determined by a single parameter θ. This parameter is sample dependent, although, in principle, its value can be derived from microscopic theory. Nevertheless it can be used as a single adjustable parameter, allowing one to describe the dependence $H_{c2}(T)$.

The situation with the cuprates[7,8] is different. The fact of the matter is that even samples with optimum doping (when $T_c = T_{c,\max}$) contain some magnetic impurities (see the review in Ref. 17). For the $Tl_2Ba_2CuO_6$ compound studied in Ref. 7, the presence of impurities was directly demonstrated by heat capacity data in Ref. 18. In this case the value of Γ_{cr} is unknown, and it is more convenient to use Eq. (7). Then we have two adjustable parameters β and θ. It is remarkable, nevertheless, that we can describe the dependence $H_{c2}(T)$ in the *entire* temperature range $T<T_c$. One can see from Figs. 1 and 2 (see below) that our simple model provides a description which is in excellent agreement with experimental data.

Equations (4)–(7) allow one to reconstruct the dependence $H_{c2}(T)$. One can see directly from Eq. (6) that the temperature dependence $\Gamma(T)$ leads to a deviation of H_{c2} from the simple quadratic HW form[1] and to an increase in the value of H_{c2}. It is also perfectly realistic for the curvature of this dependence to change (see below and Fig. 1). As was noted above, the peculiar structure of Eqs. (5) and (6) leads to the situation when even a relatively small change in Γ leads to a very strong effect on H_{c2} (see below). The ordering leads to a weakening of the pair-breaking effect. As a result, the superconducting properties are less depressed and this leads to an increase in H_{c2}.

We believe that the model is directly related to the situation observed in Refs. 7 and 8, and we now turn our attention to these data. Experimental studies, carried out on overdoped samples of $Tl_2Ba_2CuO_6$ (Ref. 7) ($T_c=14$ K) and $Bi_2Sr_2CuO_6$ (Ref. 8) ($T_c=18.5$ K) revealed unusual magnetic properties of the cuprates. Overdoping leads to a drastic decrease in T_c, and this allows one to measure H_{c2} in the entire temperature region. We conclude that the strong depression of T_c [for example, the critical temperature of the sample prior to overdoping was 85 K (Ref. 7)] is due to the presence of magnetic impurities.

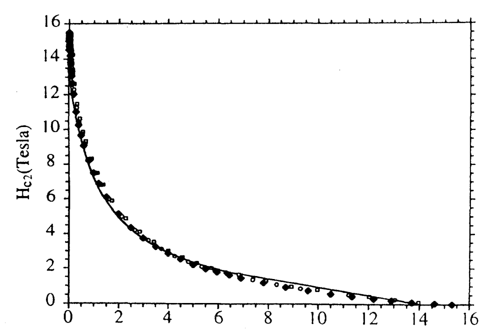

FIG. 1. Dependence $H_{c2}(T)$ for $Tl_2Ba_2CuO_6$; diamonds, open circles, and open squares, experimental data (Ref. 2); solid line, theory.

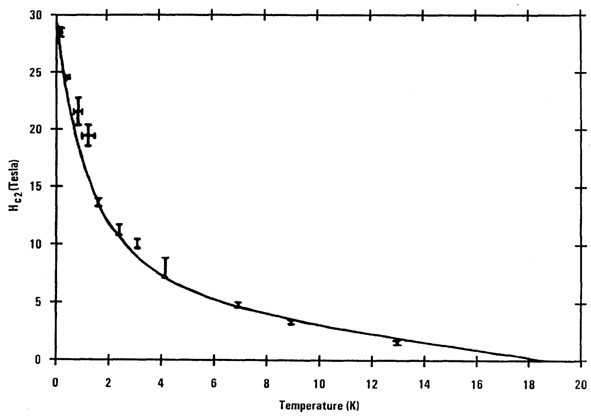

FIG. 2. Dependence $H_{c2}(T)$ for $Bi_2Sr_2CuO_6$; solid squares, experimental data (Ref. 3); solid line, theory.

Indeed, it has been reported in a number of papers (see, e.g., Refs. 19 and 20 and also the review in Ref. 21) that the overdoped cuprates are characterized by gapless behavior. According to Ref. 21, this gaplessness is caused by the presence of localized magnetic moments; as was noted above, their presence has been detected in Ref. 17. Note also that the sample used in Ref. 7 is in a clean state. (According to Refs. 7 and 22, the sample is characterized by a large mean free path $l = 10^3$ Å; this value is determined from normal conductivity data. Probably, this means that the doping is accompanied by the formation of the local magnetic moment on the O-Ba layer.) As a result, the model described above is fully applicable.

Let us consider the $Tl_2Ba_2CuO_6$ compound.[7] For this material $T_c = 14$ K and $H_{c2}(0) = 15.8$ T (Ref. 7) (one can also use $v = 10^7$ cm/sec; see, e.g., Ref. 23). The parameter Γ_0 is calculated to be $\Gamma_0 = 95$ K; see the discussion preceding Eq. (7). Based on Eq. (6), we calculated the dependence $H_{c2}(T)$ for the entire temperature range (the parameters are $\theta = 1.05$ K, $\beta = 1.37$). Figure 1 demonstrates that the theory is in excellent agreement with the experimental data.

One can see that, indeed, a relatively small change in the scattering amplitude ($\simeq 0.35$) leads to a sharp increase in the value of H_{c2}, particularly in the low-temperature region. Such an increase leads to a positive curvature; its sign is opposite to that in conventional HW theory. The sample studied in Ref. 7 has $T_c = 14$ K. The value of T_c can be varied by changing the oxygen content δ_k (see Ref. 18); the index k denotes samples with T_{ck}. According to Eq. (4), $\Gamma_{cr} = 40.6$. Using Eq. (4) for sample k at $T_c = T_{ck}$, one can calculate Γ_0. Then, based on Eqs. (6) and (7), one can evaluate the dependence $H_{c2}(T)$ for sample k using just one adjustable parameter θ_κ. We would like to point out that it would be interesting to measure $H_{c2}(T)$ for samples with various levels of overdoping. The data obtained in Ref. 8 for the $Bi_2Sr_2CuO_6$ compound also can be analyzed in a similar way (see Fig. 2; the parameters are $\theta = 1.67$ K, $\beta = 1.43$). One can see again that there is very good agreement with the data.

In summary, the magnetic properties of layered superconductors, and, in particular, the nature of the dependence $H_{c2}(T)$ are greatly affected by magnetic impurities. Their presence leads to a positive curvature, in complete contrast with the conventional picture[1] and, consequently, to an increase in the value of H_{c2}. Our theoretical calculation allows one to explain recent experimental data[7,8] on overdoped cuprates; Figs. 1 and 2 demonstrate an excellent agreement of the theory with the data. It is interesting to note that this analysis not only allows one to explain experimental data such as the data in Refs. 7 and 8, but indicates that it is possible to modify the behavior of H_{c2} by the additional magnetic impurities.

The authors are grateful to A. Mackenzie, J. Cooper, L. Gor'kov, and S. Wolf for fruitful discussions. We are grateful to J. Tallon and B. Pannetier for providing us with manuscripts prior to publication. One of us (Y.N.O.) wishes to acknowledge the support of the Humboldt Foundation. The research of V.Z.K. is supported by the U.S. Office of Naval Research under Contract No. N00014-94-F0006.

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*Permanent address: L. D. Landau Institute for Theoretical Physics, Russian Academy of Sciences, Kosygin 2, Moscow, 11733V.

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