Recovery of superconductivity and the critical field in layered superconductors

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

V. Z. Kresin
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720
(Received 22 December 1995)

The presence of magnetic impurities greatly affects the temperature dependence of the critical field H_{c2}. We focus, mainly, on layered superconductors. As a result of correlations among magnetic impurities, the scattering amplitude Γ_s decreased with $T\rightarrow 0$ K. This temperature dependence leads to a weakening of the pair-breaking effect and consequently to a positive curvature of $H_{c2}(T)$ and a drastic increase in $H_{c2}(0)$. The calculations are in excellent agreement with recent experimental data. It is also shown that the presence of inhomogeneities of a certain size also modifies the temperature dependence of H_{c2}. [S0163-1829(96)05126-0]

I. INTRODUCTION

This paper is concerned with the magnetic properties of layered superconductors and, foremost among them, the cuprates. We will focus on the critical field and its temperature dependence. One can observe a behavior drastically different from that of ordinary superconductors.[1,2] Two basic scenarios can greatly modify the dependence of H_{c2} on T. The first involves scattering by magnetic impurities, while the second is based on the influence of inhomogeneities. The former method was briefly discussed by us in Ref. 3. The present paper contains a detailed description as well as a number of new results.

An unusual behavior of the critical field has been reported in several papers.[4–13] First of all, we would like to mention the study of overdoped 2201 Tl-based[4] and Bi-based[5] compounds. Overdoping leads to a drastic decrease in T_c (e.g., for $Tl_2Ba_2CuO_6$ the value of T_c decreases from $T_{c\max}=90$ K to $T_c=14$ K). This decrease in T_c is accompanied by a decrease in the value of H_{c2}, allowing direct measurements of the dependence $H_{c2}(T)$. The latter appears to be entirely different from the conventional picture.[1,2] According to this picture (and as has, indeed, been observed in many conventional superconductors), $H_{c2}(T)$ is characterized by a linear increase near T_c. In addition, $H_{c2}(T)$ saturates as $T\rightarrow 0$ K. Contrary to this picture, the critical field in overdoped cuprates displays[4,5] a positive curvature in the entire temperature range accompanied by a sharp increase in the low-temperature region (see below, Figs. 2, 3), without any sign of saturation. The measurements in Ref. 4 were performed up to $T=10$ mK. It is remarkable that the data[4] show a sharp, almost linear increase in $H_{c2}(T)$ towards $T=0$ K, so that the value of $H_{c2}(0)$ appears to be almost an *order of magnitude* larger than one could expect from conventional theory. A similar effect of positive curvature with an accompanying large increase in H_{c2} has been also observed for $Sm_{1.85}Ce_{0.15}CuO_{4-y}$ (Ref. 6) (see below, Fig. 4) and for the overdoped La-Sr-Cu-O compound,[7] although measurements have not been performed below 1 K. The most recent observation was made with an underdoped Y-Ba-Cu-O compound with partial Zn substitution.[8] Behavior similar to that for the Tl- and Bi-based compounds[4,5] has been observed (see below, Fig. 4).

It will be shown in this paper that the observed unusual behavior of the critical field can be described with high accuracy by the scenarios mentioned above, that is, by magnetic scattering or by the effects of spatial inhomogeneity. One should note also that positive curvature has also been observed in three-dimensional (3D) systems.[9–13] This case will be also discussed below, although we are going to focus, primarily, on layered systems.

The structure of the paper is as follows. A detailed description of the theoretical models is presented in Secs. II and III. Section II is concerned with the effect of magnetic impurities on $H_{c2}(T)$. The influence of inhomogeneities is described in Sec. III. An analysis of various experimental data is given in Sec. IV.

II. THEORY: MAGNETIC IMPURITIES AND H_{c2}

A. Qualitative picture

Consider a layered superconductor containing magnetic impurities with concentration n_M; the impurities cause a spin-flip scattering of the carriers. Such scattering leads to the pair-breaking effect.[14–19] As a result, at some critical value of the impurity concentration $n_{M,\mathrm{cr}}$ superconductivity becomes totally supressed ($T_c=0$). For $n_M<n_{\mathrm{cr}}$, the superconducting state is depressed. This means that the values of T_c, the energy gap, and the critical field are lower than their intrinsic values (that is, values in the absence of magnetic impurities). Moreover, a gapless state[14–19] arises prior to the total suppression of superconductivity. All of the above phenomena are caused by the pair-breaking effect of magnetic impurities. At high temperatures ($T\gtrsim T_c$), impurities can be treated as independent scattering centers[14–17] (see also Ref. 19). The electron pair-breaking effect is accompanied by the impurity spin-flip process, ensuring the conservation of total spin. However, as the temperature decreases, the interaction between impurities (ordering trend) become important. Sometimes ordering can be observed at a finite temperature (see below), whereas other cases correspond to formation of a spin-glass state. Such correlation processes frustrate spin-

flip scattering and, thereby, the pair-breaking effect, because it becomes impossible to satisfy the spin conservation law. As a result, the superconducting state becomes less depressed, which leads to an increase (''recovery'') of various quantities, first of all, the value of the critical field. Such a recovery of H_{c2} is a major focus of the present paper. Correlation leads also to a decrease of the ''normal'' component, accompanied by changes in the penetration depth, heat capacity, etc.

One should note that even complete ordering does not imply the total disappearance of the pair-breaking effect. This effect can still be provided by the dipole-dipole interaction [see below, Eq. (5)]: This interaction does not conserve total spin (it translates into orbital motion).

Strictly speaking, the impurity ordering trend makes the pair-breaking amplitude *temperature dependent*. This temperature dependence is a key factor in our approach. One can show (see below) that a relatively small change in the amplitude, namely, its small decrease caused by the correlations, can lead to noticeable changes in H_{c2} and in its temperature dependence.

B. Theory: Magnetic impurities

Let us focus on the evaluation of the critical field H_{c2} for a layered superconductor in the presence of magnetic impurities. Near the critical field H_{c2}, the order parameter $\Delta(\mathbf{r})$ satisfies the linear equation

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

Here $\partial_- = (\partial/\partial\rho - 2ie\mathbf{A})$, $\lambda = eH_{c2}$,[2] and the order parameter is an eigenfunction of the operator ∂_-^2. The critical field can be determined from the following equation (see Refs. 20, 21):

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

where

$$D_1(\omega_n,H) = (\mathrm{sgn}\,\omega_n)\,\frac{J(\omega_n,H)}{1-(\tau^{-1}-\tau_s^{-1})J(\omega_n,H)}, \tag{2'}$$

$\omega_n = (2n+1)\pi T$, and τ_s is the spin-flip scattering relaxation time (this channel is caused by the presence of magnetic impurities). The term τ^{-1} describes the usual elastic scattering; $T_c^0 = T_c$ $(\tau_s = \infty)$.

The evaluation of the kernel $J(\omega_n,H)$ is based on the method of integrated Green's functions.[22,23] For the usual 3D case, such a calculation has been performed in Ref. 20 and by one of the authors in Ref. 21. Here we are dealing with a layered structure, and this requires a special treatment. One can show (see the Appendix) that for a layered superconductor $J(\omega_n,H)$ has the form

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

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

The set of equations (2) and (3) determines the critical field H_{c2} and its temperature dependence. One can see directly from these equations that the nature of this dependence is directly affected by behavior of the amplitude $\Gamma_s = \tau_s^{-1}$ for spin-flip scattering.

The total Hamiltonian in the presence of magnetic impurities can be written in the form

$$H = H_e + H_{\mathrm{imp}} + H_{\mathrm{int}}, \tag{4}$$

where H_e and H_{imp} describe the carrier and impurity subsystems, respectively; H_{int} describes the carrier-impurity interaction and contains the term $H^{s\text{-}s}$ (see, e.g., Refs. 24, 25):

$$H^{s\text{-}s} = \mu_i\mu_e\sum\int_0^\infty d\mathbf{r}_e d\mathbf{R}_i\{\Psi_e^\dagger\Psi_i^\dagger S_i S_e\Psi_e\Psi_i|\mathbf{r}_e-\mathbf{R}_i|^{-3} - (8\pi/3)\,\delta(\mathbf{r}_e-\mathbf{R}_i)\Psi_e^\dagger\Psi_i^\dagger S_i S_e\Psi_e\Psi_i - 3\Psi_e^\dagger\Psi_i^\dagger[S_i(\mathbf{r}_e-\mathbf{R}_i)(S_e(\mathbf{r}_e-\mathbf{R}_i)]\Psi_e\Psi_i|\mathbf{r}_e-\mathbf{R}_i|^{-5}\}. \tag{5}$$

Here Ψ_e and Ψ_i describe carrier and impurity states, while S_e and S_i are the spin operators. In many cases one can neglect the interaction between impurity atoms and treat them as totally independent. This corresponds to the model studied in Refs. 14, 17; the authors of Ref. 14 considered the second term in the brackets (this term is called the ''contact interaction'': see Ref. 24; the first and third terms represent the dipole-dipole interaction). The averaging performed in Ref. 14 assumes that the impurity spins form a chaotic noninteracting system. If it is necessary to take into account correlations between the impurity spins as $T\to 0$ K, the interaction becomes essential, and then the averaging should be performed with considerable care. In addition, the last term in (5) becomes important. Indeed, the first two terms in brackets conserve total spin. As a result, pair-breaking effects described by these two terms require a change in the impurity spin (spin-flip impurity scattering) in order to satisfy spin conservation. However, the third term (studied in detail by one of the authors in Ref. 25) does not conserve the total spin (it transforms into orbital motion). If the usual spin-flip scattering is frustrated by impurity spin correlations or even by their ordering (see below), then the contribution of this process becomes important.

Equations (2) and (3) contain the spin-flip relaxation time τ_s and the relaxation time τ describing the usual nonmagnetic scattering. In ordinary 3D systems magnetic impurities provide the pair-breaking effect as well as the elastic scattering. If $l < \xi_0$ $(l = l_{\mathrm{elas}})$, we are dealing with the ''dirty'' case. The situation with layered superconductors is quite different. Because of the layering, we must distinguish two different cases. One of them (in-plane location of the magnetic impurities) corresponds to the dirty case (strong pair breaking), as in the 3D case. However, another interesting situation provides a ''clean'' case. For example, in a layered structure it is possible to combine a strong spin-flip channel $(\Gamma_s = \tau_s^{-1} > \pi T_c)$ with the condition $\tau^{-1} \ll \pi T_c$ (clean case) for elastic scattering. Generally speaking, it is realistic that $\Gamma_s \gg \tau^{-1}$. This is possible if the magnetic moments are located out of the plane, in which case, in-plane momentum transfer can remain small, despite the large amplitude of the

spin-flip process. Indeed, as is known, the main contribution to the cross section for scattering by charged impurities comes from small angles and strongly depends on the screening length λ_{scr} (see, e.g., Ref. 26). Placing the impurities out of the plane leads to an effective increase of the transport collision time τ_{tr}, which is larger than the increase in the usual carrier-impurity collision time. It is important to recognize that both the clean and dirty cases, as well as the intermediate case, are realized in layered cuprates (see below, Sec. IV).

C. Clean case

The basic equations (2) and (3) allow one to evaluate the dependence $H_{c2}(T)$ for the general case of both spin-flip and nonmagnetic channels, making comparable contributions to the scattering. Indeed, if the relaxation times τ^s and τ have the same order of magnitude, one has to use these equations directly; we have performed such calculations for some cases (see below, Sec. IV B). However, in some limiting cases one can simplify Eqs. (2) and (3). Consider, first, the clean case for nonmagnetic scattering, as defined above. Then one can neglect the term τ^{-1}.

As is known,[14] the value of $\Gamma_s=\tau_s^{-1}$ depends on the concentration of magnetic impurities n_M, so that $\Gamma_s \propto n_M$. At some value of the concentration $n_{M,\mathrm{cr}}$ (corresponds $\Gamma_s=\Gamma_{s,\mathrm{cr}}$), the superconducting state becomes totally suppressed.

Assume that $\zeta=eHv^2/\Gamma_s \ll 1$; then, one can simplify Eqs. (2) and (3). Using the relation $\zeta \ll 1$ and introducing the parameter $\Gamma_{s,\mathrm{cr}}$, we arrive, after some manipulations, at the equation

$$\ln(2\gamma\Gamma_{s,\mathrm{cr}}/\pi T)-\{\psi[0.5+(\Gamma_s/2\pi T)]-\psi(0.5)\}=f(H,T), \tag{6}$$

where

$$\begin{aligned} f(H,T)=(eHv^2/\Gamma_s)(\Gamma_s^{-1}\{&\psi[0.5+(\Gamma_s/2\pi T)] \\ &-\psi[0.5+(\Gamma_s/4\pi T)]\} \\ &-(4\pi T)^{-1}\psi'[0.5+(\Gamma_s/2\pi T)]). \end{aligned} \tag{7}$$

Here ψ is the psi function and $\ln\gamma=C=0.58$ is the Euler constant. If, in addition, the condition $\Gamma_s \gg \pi T_c$ is satisfied, Eq. (6) reduces to the form

$$\begin{aligned} \ln(\Gamma_{s,\mathrm{cr}}/\Gamma_s)-0.17(\pi T/\Gamma_s)^2+0.12(\pi T/\Gamma_s)^4 \\ =(eHv^2/\Gamma_s^2)[\ln 2-0.5-0.67(\pi T/\Gamma_s)^2 \\ +0.12(\pi T/\Gamma_s)^4]. \end{aligned} \tag{8}$$

A more exact solution (for any value of ζ) can be obtained from Eqs. (1)–(3); see Sec. IV. It is very important that the amplitude Γ_s, generally speaking, depend on temperature (see below), so that $\Gamma_s=\Gamma_s(T)$.

At $T=T_c$, H_{c2} vanishes, and we obtain, from Eq. (6),

$$\ln(2\gamma\Gamma_{s,\mathrm{cr}}/\pi T_c)-\{\psi(0.5+[\Gamma_s(T_c)/2\pi T_c)]-\psi(0.5)\}=0. \tag{9}$$

The value of T_c depends on the concentration of magnetic impurities in the way described by Eq. (9). Putting $n_M=0$, one arrives at the relation[14] $\Gamma_{s,\mathrm{cr}}=\pi T_c^0/2\gamma$, where T_c^0 is the value of the critical temperature in the absence of magnetic impurities (''intrinsic'' T_c). Based on Eqs. (6) and (9), we arrive at the following equation for the critical field H_{c2}:

$$\begin{aligned} \ln(T_c/T)-\{&\psi[0.5+\Gamma_s(T)/2\pi T)] \\ &-\psi[0.5+(\Gamma_s(T_c)/2\pi T_c)]\}=f(H,T). \end{aligned} \tag{10}$$

Here $f(H,T)$ is defined by Eq. (7).

D. Spin-flip scattering

Equations (2), (3), (7), and (10), as well as Eq. (19) (see below), form the basis of our theory. They allow one to evaluate the dependence $H_{c2}(T)$ for a layered superconductor in the presence of magnetic impurities. One can see directly from Eqs. (7) and (10) that the functional form of H_{c2} is greatly affected by the behavior of the spin-flip scattering amplitude Γ_s. If the parameter Γ_s is temperature independent, we obtain a smooth, almost quadratic dependence of H_{c2} on (T). However, if Γ_s depends on the temperature, the picture becomes entirely different.

The amplitude Γ_s describes the spin-flip scattering of the carriers by localized magnetic moments. As stated above, this leads to the pair-breaking effect and to the depression of T_c, H_{c2}, etc. At almost all temperatures this parameter can be treated as temperature independent: $\Gamma_s=\Gamma_\alpha=\mathrm{const}$. This picture is similar to that described in Refs. 14–19, where the impurities were treated as noninteracting and scattering was described by the ''contact'' interaction [see Eq. (5)]. Averaging over impurity spins[14] does not include any correlation between them. Then $\Gamma_s=\tau_s^{-1}=JS(S+1)$. This leading term in the amplitude of spin-flip scattering by magnetic impurities, indeed, does not depend on the temperature (the small ''Kondo'' term was neglected in Ref. 14).

However, at low temperatures the picture becomes different. As was noted above [see the discussion after Eq. (5)], the decrease in thermal motion for $T \to 0$ K leads to the correlations between impurity spins becoming important. These correlations affect the second term in (4). The manifestation of such a correlation is different in various systems, and we shall consider different examples in Sec. IV. In some cases, this effect may become so strong that ordering arises at a finite temperature $T_{\mathrm{ord}}>0$; in addition, the width of the phase transition region δT is such that $T_{\mathrm{ord}}-\delta T>0$. Such a case is realized, for example, in the Sm-Ce-Cu-O system[6] and in the Ni/V compound;[12] see Sec. IV E. Frequently, one observes a trend towards total ordering, but even if the transition occurs, $T_{\mathrm{ord}}<\delta T$. In this case one sees an increase in the magnetic succeptibility near $T=0$ K (such an increase was observed in Ref. 27). A perfectly realistic is a formation of a ''spin glass'' (see, e.g., Ref. 28). In addition to the susceptibility, this ordering trend is manifested in the appearance of Shottky anomalies in the heat capacity. These anomalies have been observed in many cases (see, e.g., Refs. 29–31). Note also that the impurities may be distributed in such a way that two impurity atoms end up much closer to each other than the average distance between impurities. The spin interaction for such pairs can be particularly strong.

As a result of spin-spin correlations of the impurity atoms, spin-flip scattering becomes frustrated. The relative contribu-

tion of the dipole-dipole interaction (which does not conserve spin) increases, and averaging no longer leads to a simple expression.[14] Note that the increase of the spin-flip scattering time for a spin glass has been observed experimentally in Ref. 32. In other words, correlations make the amplitude $\Gamma_s=\tau_s^{-1}$ temperature dependent; this dependence is particularly important in the low-temperature region.

An exact evaluation of this temperature dependence is an interesting problem of microscopic theory, and we will address some aspects of this problem elsewhere. However, the main subject of this paper is the behavior of the critical field $H_{c2}(T)$, and we restrict ourselves to a semiphenomenological approach.

It is essential that as a result of the correlation among magnetic moments the amplitude Γ_s become *temperature dependent*. Based on the described physical picture, we now study the impact of this temperature dependence on the behavior of the critical field.

Let us discuss this temperature dependence. We introduce a characteristic temperature θ, which describes the ordering trend. In the region $T\gg\theta$ the magnetic impurities can be treated as independent, and the amplitude $\Gamma_s=\Gamma_\alpha=$const. Let us denote the value of the amplitude at $T=0$ K by Γ_0. The temperature dependence $\Gamma_s(T)$ in the entire range can be represented in the form (this is similar to the Pado approximation)

$$\Gamma_s=\Gamma_0 f(\tau), \tag{11}$$

$$f(\tau)=(1+\beta\tau)/(1+\tau). \tag{12}$$

Here $\tau=T\theta^{-1}$, $\beta=\Gamma_\alpha/\Gamma_0$; we assume that $\tau_c=T_c\theta^{-1}\gg1$, $\beta>1$. Indeed, one can see that at $T=0$ the amplitude Γ_s is equal to Γ_0, whereas in the region $T\gg\theta$ ($\tau\gg1$) we obtain $\Gamma_s=\Gamma_0\beta=\Gamma_\alpha$. Equation (11) can be rewritten in the dimensionless form

$$\gamma_s=\gamma_0 f(\tau), \tag{13}$$

where $\gamma_s=\gamma_s(T)=\Gamma_s/T_c$, $\gamma_0=\Gamma_0/T_c$, and $f(\tau)$ is described by Eq. (12). Equation (10) can be rewritten in the form

$$\ln(T_c/T)-\{\psi[0.5+\gamma_s(T)(T_c/2\pi T)]-\psi[0.5+(\beta\gamma_0/2\pi)]\} =f(H,T), \tag{14}$$

where

$$f(H,T)=(eHv^2/\gamma_s T_c)((\gamma_s T_c)^{-1}\{\psi[0.5+(\gamma_s T_c/2\pi T)] -\psi[0.5+\gamma_s T_c/4\pi T)]\} -(4\pi T)^{-1}\psi'(0.5+\gamma_s T_c/2\pi T)). \tag{15}$$

Equations (14) and (15), along with (12) and (13), allow us to evaluate the functional form of $H_{c2}(T)$.

Let us discuss the dependence (13) in more detail. Generally speaking, Eqs. (12) and (13) contain three parameters: the characteristic temperature θ, the value of the amplitude at $T=0$ (γ_0), and the ratio of the amplitudes $\beta=\gamma_\alpha/\gamma_0$. As was noted above, these parameters can be evaluated from microscopic theory, and we will address this question elsewhere. For our present purposes, we will use values extracted from experimental measurements (as described below) or use them as adjustable parameters.

If we are interested in the shape of the dependence $H_{c2}(T)$, we can employ a measured value of $H_{c2}(0)$ in order to determine γ_0. Furthermore, if we consider a layered superconductor which does not contain any localized magnetic moments (in which case its T_c coincides with the intrinsic critical temperature T_c^0) and add to it magnetic impurities, then Eq. (9) together with the relation $\Gamma_{cr}=\pi T_c^0/2\gamma$ allows us to calculate the value of $\Gamma_\alpha=\Gamma_s(T_c)$, which is determined by the concentration of impurities. As was noted above, the value of the parameter Γ_0 is directly related to the value of $H_{c2}(0)$. Therefore, the values of the parameters β and Γ_0 can be expressed in terms of experimentally measured values of T_c^0 and $H_{c2}(0)$. As a result, in this case 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 is different if the superconductor contains some magnetic impurities even in the original state. This is actually an important case (see below, Sec. IV). In this case the value of Γ_{cr} is unknown and it is more convenient to use Eqs. (12)–(14) directly. Then we have two adjustable parameters β and θ. Note that the value of θ can also be determined from magnetic susceptibility or heat capacity (Shottky anomaly) measurements. In the following, we will use β and θ as adjustable parameters. It is, nevertheless, remarkable that in this way we can describe the dependence $H_{c2}(T)$ in the *entire* temperature range $T<T_c$. It will be shown (see Sec. IV) that our simple model provides a description which is in excellent agreement with the experimental data.

One should note that if the superconductor contains magnetic impurities from the beginning, then its $T_c=T_{c,\mathrm{init}}$ (which is the experimentally observed quantity) is actually depressed by the pair-breaking effect. The intrinsic critical temperature $T_{c,\mathrm{intr}}=T_c^0$, which is defined as the value of T_c in the absence of magnetic impurities, always exceeds $T_{c,\mathrm{init}}$. It turns out that precisely such a situation occurs in the cuprates (see below, Sec. IV A).

Let us make several more comments. First of all, the value of the amplitude γ_s appears to be large ($\gamma_s\gg1$), especially in the gapless region (see below). As a result (and this a remarkable and important feature of the model), even a relatively small change in γ_s makes a very strong impact on H_{c2} (see below, Sec. IV A).

Furthermore, let us consider the special case, mentioned above, when the impurity spin system undergoes a phase transition at some temperature T_m and the width of the critical region δT is such that $T_m-\delta T>0$. Then the amplitude Γ_s is temperature independent near $T=0$ K, although it differs from the value $\Gamma_\alpha=\Gamma(T_c)$. In this special case one can expect saturation near $T=0$ K and the amplitude can be described by the expression

$$\gamma_s=\gamma_0 f(\tau),\quad f(\tau)=(1+\beta\tau)/(1+\tau)\quad (T>T_1)$$

$$f(\tau)=1\quad (T<T_1). \tag{16}$$

Here $\tau=(T-T_1)/\theta$, $T_1=T_m-\alpha\theta$, $\alpha\simeq1$; $\alpha\theta=\delta T$, and other parameters are defined by Eqs. (12) and (13). Note that the

Sm-Ce-Cu-O and Nb/V systems correspond to precisely this situation (see Secs. IV B, IV E).

We focus on the case $T_c \gg \theta$ [($\tau \gg 1$; see Eq. (12)], that is, where the value of T_c greatly exceeds the value of the characteristic temperature θ. This case corresponds to the experiments described in Refs. 4, 5 (see below, Sec. IV). However, as the concentration of magnetic impurities continues to increase, T_c keeps on decreasing until at some value of n_M there is a crossover and T_c becomes smaller than θ. As a result, the dependence $T_c(n_M)$ begins to deviate from that obtained in Ref. 14, since the spin-flip scattering amplitude decreases with the outset of ordering. The decrease in T_c slows down, resulting in the appearance of a tail. We will discuss this interesting case elsewhere.

Note, also, that the ordering trend can be important in the 3D case as well; the relevant data will be discussed in Sec. IV. However, as was pointed out above, the presence of layering is essential, because it provides for the out-of-plane location of the magnetic moments, thus combining a clean nonmagnetic scattering with a strong pair-breaking effect. The absence of the term τ^{-1} [see Eqs. (2) and (3)] makes the spin-flip scattering channel dominant, leading to a stronger influence on the dependence $H_{c2}(T)$.

E. Dirty case: Superconductor–normal-metal sandwich

Above, we described the clean case [Eqs. (11)–(15)] when the spin-flip interaction is the dominant scattering channel. This case arises for an out-of-plane location of magnetic impurities. Now let us consider the dirty limit of the usual elastic scattering with the free path $l_{\rm tr} \ll \xi_0$. There are now two possibilities. One of them corresponds to the in-plane location of the magnetic impurities. These impurities provide both a strong pair-breaking effect and the usual elastic scattering. Another possibility is when spin-flip scattering is provided by out-of-plane magnetic moments, whereas nonmagnetic scattering is caused by other nonmagnetic in-plane impurities and defects.

We will consider a more general case, namely, the proximity superconductor–normal-metal (S-N) system which consists of two dirty films with thicknesses d_1 and d_2 and densities of states ν_1 and ν_2. The equations describing pairing in these films can be written in the form

$$0.5D_1\partial_-^2\beta_1+\Delta=(\omega+\Gamma_1)\beta_1, \tag{17a}$$

$$0.5D_2\partial_-^2\beta_2-\beta_2(\omega+\Gamma_2)=0. \tag{17b}$$

Here β_1 and β_2 are the Green's functions describing pairing in the S and N films, respectively, D_i are the diffusion coefficients, and Δ is the order parameter which is finite only in the S film; $\partial_-^2=(\partial^2/\partial z^2+\partial_-^2)$, where ∂_- is defined by Eq. (1). The pairing states are connected by the boundary condition

$$(\nu D\partial\beta/\partial n)_+=(\nu D\partial\beta/\partial n)_-; \quad \beta_+=\beta_-. \tag{18}$$

Based on Eqs. (17) and the condition (18), one arrives at the following equation (see the Appendix) which permits an evaluation of the dependence $H_{c2}(T)$:

$$\ln(T_c^0/T)=\Psi\{0.5+[\Gamma(T)/2\pi T]+eHD/2\pi T\}-\Psi(0.5), \tag{19}$$

where $D=(D_1+kD_2)/(1+k)$, $T_c^0=T_c^0(\pi T_c^0/2\gamma\Omega)^k$, and $\Omega=\Omega_D$; T_c^0 is the transition temperature of the superconducting layer in the absence of magnetic impurities;

$$\Gamma=(\Gamma_1+k\Gamma_2)/(1+k); \quad k=\nu_1 d_1/\nu_2 d_2 \tag{20}$$

(cf. Ref. 33). If $k=0$ (no proximity effect), one can use Eq. (19) with $D=D_1$. For layered superconductors one has to use the expression $D=(v_F l_{\rm tr})/2$ for the diffusion coefficient. The temperature dependence $\Gamma(T)$ of the electronic spin-flip amplitude (pair breaking) is described by Eqs. (11) and (12).

Equation (19) can be written in the form

$$\ln(T_c^0/T)=\Psi[0.5+(\gamma_s T_c/2\pi T)+eHD/2\pi T]-\Psi(0.5), \tag{19'}$$

where γ_s is defined by Eq. (13).

Equations (11), (12), (19), and (20) allow one to reconstruct the dependence $H_{c2}(T)$. The ordering of magnetic impurities and, consequently, the appearance of a temperature dependence of $\gamma_s(T)=\Gamma_s(T)/T_c$ lead to a deviation of H_{c2} from the Helfand-Werthamer (HW) dependence[2] and to an increase in the value of H_{c2}.

III. THEORY: CRITICAL FIELD AND INHOMOGENITIES

In this section we discuss another aspect related to the magnetic properties of layered superconductors. In fact, a noticeable deviation from the conventional HW behavior can be caused not only by magnetic impurities, but by a different mechanism as well. Namely, positive curvature and a corresponding increase in H_{c2} relative to the HW value[2] can be caused by the presence of 2D in-plane inhomogeneities. Indeed, consider a dielectric island as an example of such an inhomogeneity. The function $H_{c2}(T)$ satisfies Eq. (1) even in the presence of inhomogeneities, but their presence leads to an additional boundary condition

$$\mathbf{n}(\partial/\partial\rho-2ie\mathbf{A})\Delta_{\rm isl}=0, \tag{21}$$

where $\mathbf{n}$ is perpendicular to the boundary and ρ is a 2D vector. The parameter λ is equal to eH_{c2}^0 (see Ref. 2), H_{c2}^0 is the value of H_{c2} in the absence of the inhomogeneity, and the dependence $H_{c2}^0(T)$ is described by the HW theory.

Now let us focus on the effect of the inhomogeneities. Assume that there are no magnetic impurities, that is, $\tau_s^{-1}=0$. The function $H_{c2}(T)$ enters the left-hand side of Eq. (1), and to determine this function, one has to apply the boundary condition. This condition results in a deviation from the HW solution.

For concreteness, consider a dielectric island in the shape of a disk of radius ρ_0. The order parameter $\Delta(\rho)$ can be written in the form $\Delta(\rho)=\exp(in\phi)f(\rho)$, $n=0,1,2,\ldots$. Based on Eqs. (1) and (21), one can show that the radial function has the form $f(\rho)=y^{-1/2}W_{(n+\lambda/eH)/2;n/2}(y)$ ($y=eH\rho^2$ and W is the Whitteker function) with the boundary condition $(\partial f/\partial y)_{y_0}=0$; $y_0=eH\rho_0^2$. As a result, we arrive at the following equation allowing one to evaluate $H_{c2}(T)$:

$$(n/y_0)=1+2g_1/g_2. \tag{22}$$

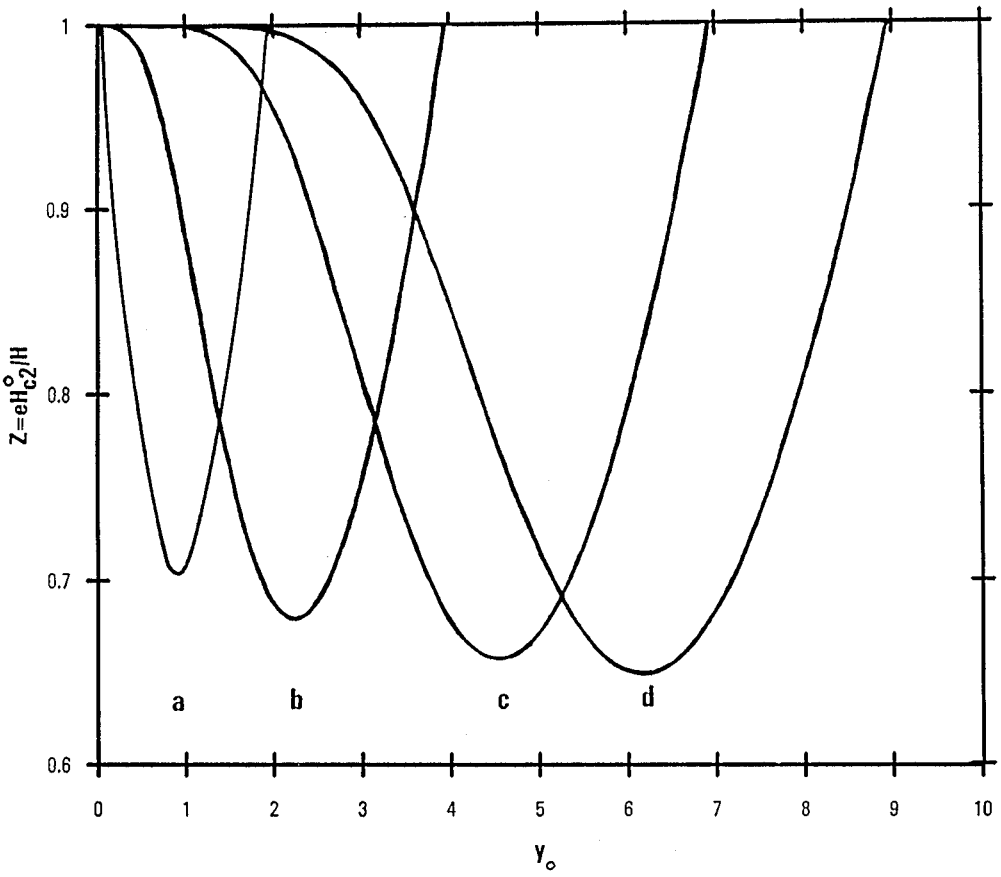

FIG. 1. Dependence $Z_n=H^0_{c2}/H_{c2}$ on $y_0=eH\rho_0^2$ for different n: (a) $n=2$, (b) $n=4$, (c) $n=7$, (d) $n=9$.

Here $g_i=g_i(y,n,Z)=\int_0^\infty dt\ \varphi_i$, $i=\{1,2\}$, $\varphi_1=e^{-yt}t^{1/2-(z/2)}(1+t)^{n-1/2+(z/2)}$, $\varphi_2=\varphi_1/t$, and $Z=H^0_{c2}/H_{c2}$.

Equation (22) determines $Z_n=H^0_{c2}/H_{c2}$ as a function of y_0 for a given n; see Fig. 1. Note that the function $Z_n=H^0_{c2}/H_{c2}$ was studied in Ref. 34 for the 3D case.

Let us describe the main features of the solution for layered superconductors. First of all, one can see directly from Fig. 1 that $Z_n<1$. This means that the presence of inhomogeneities leads to an increase in the value of H_{c2} relative to the HW value H^0_{c2}. Note also that an increase in ρ_0 provides for a larger effect. The minimum value of Z_n is equal to $Z_{n,\min}=0.6$, corresponding to $H_{c2,\max}=1.7H^0_{c2}$. This value is similar to the ratio H_{c3}/H_{c2} in conventional superconductors [see, e.g., Ref. 16(a)]. However, in the present case we are dealing with the critical field for bulk superconductivity. In addition, the ratio H_{c2}/H^0_{c2} can be modified by varying ρ_0. It is important to note that for inhomogeneity dimensions corresponding to $Z_n^{-1}>1.4$, positive curvature appears.

We wish also to emphasize that the effect of inhomogeneities becomes noticeable for $y=1$, that is, if the size ρ_0 is of the order of the coherence length ξ. As a result, the dependence $H_{c2}(T)$ for temperatures near T_c is not affected by such inhomogeneities. However, in the low-temperature region where H_{c2} is large and the parameter y_0 can become of the order of unity (for $\rho_0=\xi$), there arises a noticeable increase in H_{c2} relative to the HW value (by up to a factor of 1.7). In addition, the curvature becomes positive in this region. However, we should remark that the mechanism due to magnetic impurities, discussed above, leads to a larger effect (see Figs. 2 and 3).

The size of inhomogeneities can be estimated from the broadening of the transition in the region near $T=0$ K. For example, for the compound studied in Ref. 4, $\rho_0=10^{-7}$ cm, that is, $\rho_0\ll\xi_0$ ($\xi_0>50$ Å for this overdoped material; see Ref. 4). The effect observed in Ref. 4 is due to magnetic impurities (see below); in addition, the scale of the increase observed in Ref. 4 greatly exceeds that which could be caused by inhomogeneities. Nevertheless, the appearance of a positive curvature due to inhomogeneities may be relevant to other systems, such as those encountered in Refs. 9, 10, 13; we shall discuss this problem in Sec. IV G; see below. It is important to note that this strong modification of the magnetic properties can be induced by making layers with inhomogeneities of a well-defined size. It would be interesting to carry out experiments of this type to confirm our conclusion.

IV. EXPERIMENTAL DATA: DISCUSSION

A temperature dependence of the critical field drastically different from that of the ordinary picture[1,2] has been observed in a number of layered superconductors.[4–8] In this section we will present a detailed analysis for several such systems.

An analysis of experimental data has to be carried out with considerable care because in many cases one observes a broadening of the transition in a magnetic field, caused by

inhomogeneities (see Sec. III). This broadening frustrates a determination of the value of H_{c2}. However, in a number of cases (see, e.g., Refs. 4, 5), the applied field leads to a shift of the transition, which allows one to determine the dependence $H_{c2}(T)$ with high accuracy. Indeed (see, e.g., the discussion in Ref. 4), one can employ different ways of determining H_{c2}. Namely, H_{c2} could be defined as the field leading to some fixed loss in the resistance. It turns out that the shape of the dependence $H_{c2}(T)$ for systems analyzed below is universal and insensitive to the specific definition.

A. Overdoped cuprates

Experimental studies carried out on overdoped samples $Tl_2Ba_2CuO_6$ (Ref. 4) ($T_c=14$ K) and $Bi_2Sr_2CuO_6$ (Ref. 5) ($T_c=18.5$ K) revealed unusual magnetic properties of the cuprates. Overdoping leads to a drastic decrease in T_c, and, correspondingly, in H_{c2}, thus making it possible 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 Tl-based sample prior to overdoping was 90 K (Ref. 4); thus, the decrease was truly drastic: $90\rightarrow 14$ K] is due to the presence of magnetic impurities. In fact, their presence has been established by several independent experiments. For example, heat capacity measurements[29–31] have revealed Schottky anomalies; these anomalies are a direct manifestation of the presence of magnetic impurities. As is known (see Refs. 14–17), the presence of magnetic impurities leads to the pair-breaking effect. And, indeed, muon-spin-resonance (μSR) data[35,36] show that the normal component decreases upon overdoping, corresponding to pair breaking. These data also demonstrated the appearance of a gapless state. The gaplessness is caused by the presence of localized magnetic moments; as was noted above, the presence of the latter was detected in Refs. 29, 30. In addition, magnetic scattering also leads to a decrease in T_c and, eventually, to suppression of superconductivity. Such a decrease in T_c was indeed observed in Refs. 4, 5.

Note that at the same time the 2201 Tl-based sample used in Ref. 4 is in the clean state (the sample is characterized by a large mean free path $l\simeq 10^3$ Å, as determined from normal conductivity data). This means that doping results in the formation of out-of-plane local magnetic moments (see the discussion, Sec. II B). As a result, the model described in Sec. II C above is fully applicable.

Let us now consider the $Tl_2Ba_2CuO_6$ compound.[4] For this material $T_c=14$ K and $H_{c2}(0)=15.8$ T (Ref. 4) (we use $v_F=1.3\times 10^7$ cm/sec, see, e.g., Ref. 37). Using this value of $H_{c2}(0)$, one can calculate the parameter γ_0 [see the discussion following Eq. (15)], which appears to be equal to $\gamma_0=6.8$. Based on Eqs. (12)–(15), we have calculated the dependence $H_{c2}(T)$ for the entire temperature range (the parameters are $\theta=1$ K, $\beta=1.26$). Figure 2 demonstrates that the present theory is in excellent agreement with the experimental data.

One can see that, indeed, a relatively small change in the scattering amplitude ($\Delta\Gamma/\Gamma_0=0.37$) leads to a sharp increase in the value of H_{c2}, particularly at low temperatures. Such an increase results in positive curvature; that is, its sign is opposite to that in the conventional HW theory.

The data obtained in Ref. 5 for the $Bi_2Sr_2CuO_6$ compound can be analyzed in a similar way. For this material $T_c=18.5$ K, $H_{c2}(0)=28$ T. Similar to the case with the Tl-based cuprates[4] (see above), we can describe the dependence $H_{c2}(T)$ in the entire temperature range with the use of only two adjustable parameters. Their values are equal to $\theta=1.6$ K, $\beta=1.38$ (with the same value of $v_F=1.3\times 10^7$ cm/sec). One can see again that there is very good agreement with the data (see Fig. 3).

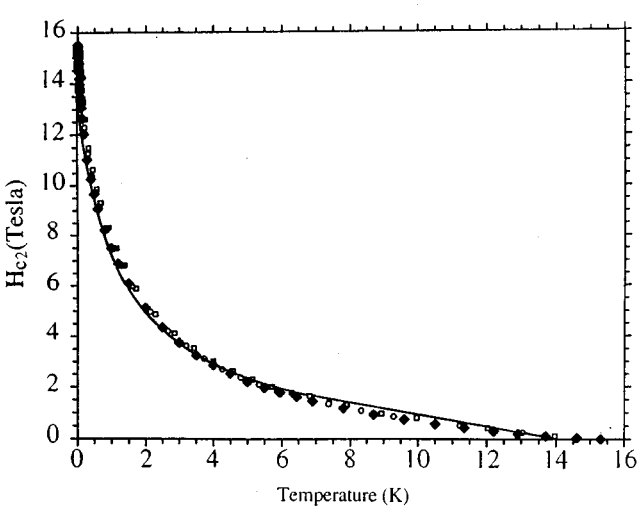

FIG. 2. Dependence $H_{c2}(T)$ for $Tl_2Ba_2CuO_6$; diamonds, open circles, and open squares, experimental data; Ref. 4; solid line, theory.

One can evaluate the value Γ_{cr} which corresponds to $T_c=0$. Based on this value, one can then estimate the value $T_c^0=2\gamma\Gamma_{cr}/\pi$ (this intrinsic value corresponds to absence of magnetic impurities). It turns out that T_c^0 is greater than the experimental values guided in Ref. 4. For example, we obtain $T_c^0=155$ K for the 2201 Tl-based sample;[4] the experimentally measured value at optimum doping is $T_{c,m}=90$ K. This means that the materials in Ref. 4 contained magnetic impurities prior to overdoping and their T_c was depressed. As a result, the value of the intrinsic critical temperature $T_{c,\mathrm{intr}}=T_c^0$ (this quantity was introduced in Sec. III D above) for this material greatly exceeds the currently observed maximum value. Therefore, the T_c of these materials can be raised further.

B. Sm-Ce-Cu-O system

A very interesting experimental study of the $Sm_{1.85}Ce_{0.15}CuO_{4-y}$ compound has been described in Ref. 6. The relatively low value of $T_c=9.5$ K (the transition is relatively broad, leading to some uncertainty in determining T_c) permits measurements of the dependence of $H_{c2}(T)$. Such measurements[6] show that the dependence H_{c2} is characterized by positive curvature, similar to that observed in the overdoped cuprates (see Sec. IV A). The measurements were performed for $T>1$ K. The absence of experimental data for $T<1$ K does not allow one to carry out a numerical analysis with the same accuracy as that for the $Tl_2Ba_2CuO_6$ compound (see above). In addition, the noticeable broadening of the transition (3 K) means that this system is not homogeneous and therefore requires special treatment. Nevertheless, the physical picture appears to be similar, which is supported by the magnetic susceptibility measurements[6] on the same

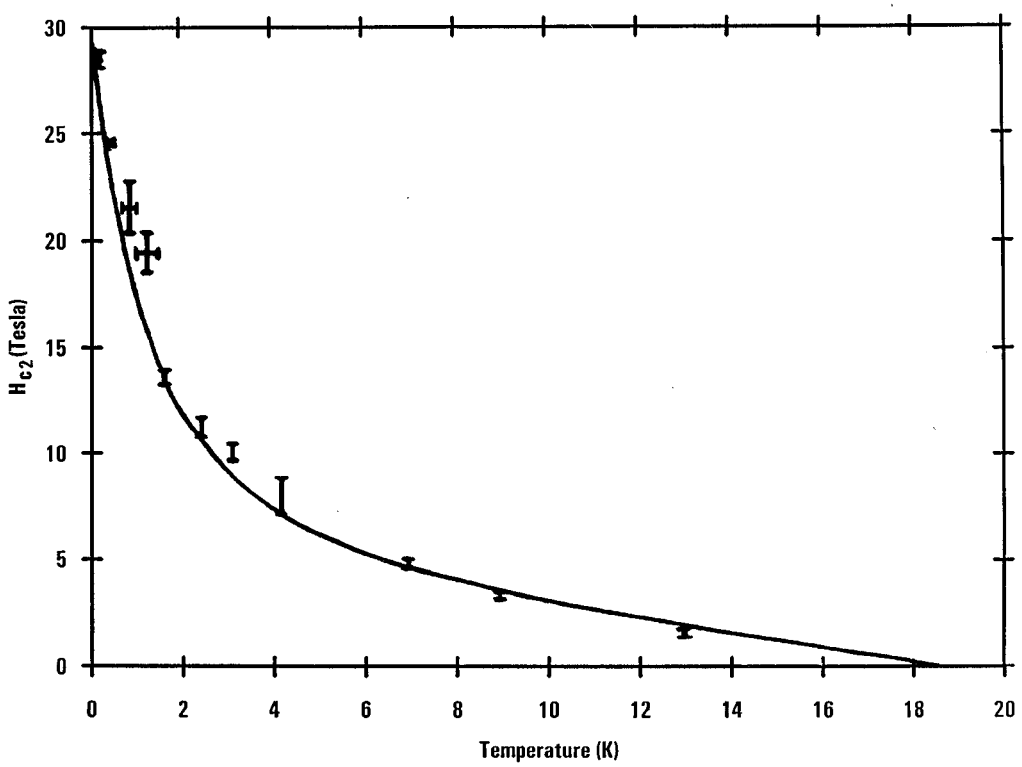

FIG. 3. Dependence $H_{c2}(T)$ for $Bi_2Sr_2CuO_6$: solid squares, experimental data, Ref. 5; solid line, theory.

sample. These measurements have established the existence of antiferromagnetic ordering of Sm^+ ions. It is remarkable that, according to Ref. 6, the departure from the HW curve correlates with a drop in the susceptibility caused by the ordering.

Therefore, the model, described by Eqs. (2), (3), (11), and (12), appears to be fully applicable to the $Sm_{1.85}Ce_{0.15}CuO_{4-y}$ compound. In addition, the data[6] show that increasing the magnetic field leads, mainly, to a parallel shift, allowing us to carry out an analysis similar to that for the overdoped cuprates. The value of the mean free path can be estimated from the broadening, yielding $l=30$ Å. The nonmagnetic relaxation time is $\tau=3\times10^{-14}$ sec (with $v_F=2\times10^7$ cm sec^{-1}). Since the situation does not correspond to any limiting case, we used the exact equations (2) and (3). This provided a description of the dependence $H_{c2}(T)$ for $Sm_{1.85}Ce_{0.15}CuO_{4-y}$ for the entire temperature range (the values of the parameters were $\theta=2$ K, $\beta=1.15$).

As shown in Fig. 4, there is good agreement with the data.[6] In addition, one expects a saturation near $T=0$ K, which makes the $Sm_{1.85}Ce_{0.15}CuO_{4-y}$ system somewhat different from the overdoped Tl- and Bi-based cuprates. This saturation is due to the ordering at a finite temperature $T_1=4.3$ K with $T_1-\delta T=1$ K (this ordering is manifested in the observed behavior of the susceptibility).[6] It would be interesting to measure the dependence $H_{c2}(T)$ in the region $T<1$ K to verify the above prediction.

C. Underdoped cuprates

A very interesting study of the Y-Ba-Cu-O system was published recently in Ref. 8. The authors used Zn substitution with subsequent oxygen depletion and obtained a sample with $T_c=10$ K. As a result of such a procedure, a relatively sharp resistive transition with a parallel shift in an applied magnetic field has been observed. Therefore, it was possible to determine the dependence $H_{c2}(T)$, which turned out to display an upward curvature similar to that observed for the overdoped cuprates (see above, Sec. IV A and Figs. 2 and 3). Recent NMR measurements[38] show that magnetic pair breaking can fully account for the observed decrease in T_c; this observation supports our model. One should note, however, that according to Ref. 8, the low-temperature resistivity of the underdoped $YBa(Cu_{0.97}Zn_{0.03})_3O_{7-\delta}$ sample is almost two orders of magnitude greater than that of the $Tl_2Ba_2CuO_6$ compound.[4] This means that the data and the dependence $H_{c2}(T)$ can be analyzed, but the situation corresponds to the dirty case (see Sec. III E).

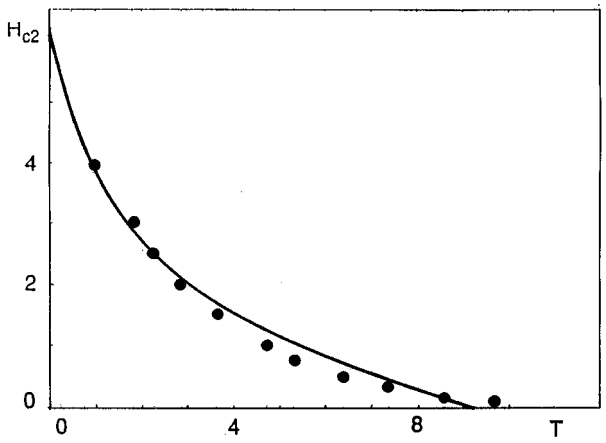

FIG. 4. Dependence $H_{c2}(T)$ for $Sm_{1.85}Ce_{0.15}CuO_{4-y}$: circles, experimental data, Ref. 6; solid line, theory.

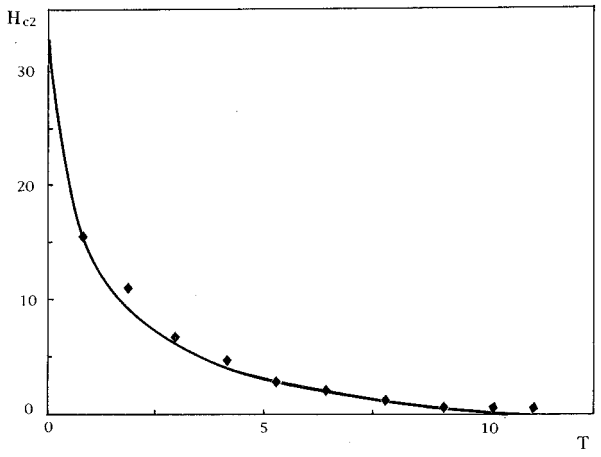

FIG. 5. Dependence $H_{c2}(T)$ for YBa_2 $(CuO_{0.97}Zn_{0.03})_3CuO_{7-\delta}$: diamonds, experimental data, Ref. 8; solid line, theory.

The analysis is based on Eqs. (13), (19), and (20). The value of $H_{c2}(0)=35$ T implies $\gamma_0=0.6$. The values of the adjustable parameters are $\beta=1.28$ (note that this value is very close to that for the overdoped cuprates), $\theta=0.7$ K, and the diffusion coefficient $D=0.8$ cm^2/sec. Using these values, we get excellent agreement with the data;[8] see Fig. 5. In addition, the diffusion coefficient D allows one to evaluate the mean free path which appears to be equal to 12 Å; this value is in remarkable agreement with the value $l_{tr}=10$ Å measured in Ref. 8.

Based on Eq. (20), one can also determine the intrinsic value of the critical temperature [cf. Eqs. (9) and discussions in Secs. III D and IV A]. As a result, we obtain $T_c^0=170$ K. This is a very interesting result, because it shows, in analogy with overdoped cuprates (see Sec. IV A), that the intrinsic T_c exceeds the value of critical temperature at optimum doping $T_{c,\mathrm{opt}}$, implying that the latter is actually depressed by the pair-breaking effect. In other words, even at optimum doping (in the absence of Zn atoms) the samples contain magnetic impurities which depress T_c relative to its intrinsic value. It is interesting that these intrinsic critical temperatures appear to be close (160–170 K) for all cuprates studied (see above), including Tl- and Bi-based samples, as well as the Y-Ba-Cu-O compound. All of them contain the Cu-O plane as a basic unit, and probably the value T_c^0 is an important intrinsic parameter of this structural unit. If this is true, we can conclude that the Hg-based cuprate ($T_c\simeq 150$ K) is not far from this limit.

One can raise the very interesting question of the location of the magnetic moments responsible for pair breaking. An unconventional temperature dependence of H_{c2} can be observed for different cases (see above), since any set of magnetic moments located in the Cu-O plane or near it will provide a pair-breaking mechanism depressing T_c, H_{c2}, etc. Transport data showing a large mean free path[4] lead one to the conclusion that in Tl-based cuprates the magnetic moments are located on the apical oxygen layer (see the discussion in Sec. IV A above). We can assume that the same is true for the Y-Ba-Cu-O compound. In connection with this, it is interesting to note that the calculation carried out in Ref. 39 has shown that the doping ($YBa_2Cu_3O_6\rightarrow YBa_2Cu_3O_7$) is accompanied by change of the charge Z^* on the O(4) side, so that $Z^*=-2.07\rightarrow 0.67$. This calculation explicitly shows that the doping leads to formation of local magnetic moments on the Ba-O layer. The calculation[39] provides a good description of the nuclear-quadrupole-resonance (NQR) data and supports our approach. However, one should note that for the Y-Ba-Cu-O system the intrinsic value of T_c can be also depressed by other pair breakers. The fact of the matter is that the mean free path for the Y-Ba-Cu-O compound is not so large, indicating the possible presence of in-plane pair breakers. In connection with this fact, it is interesting to note that a number of recent studies (see, e.g., Refs. 40–42) have indicated that the cuprates (including the Y-Ba-Cu-O compound) are intrinsically inhomogeneous even at optimum doping. Phase separation and formation of small spin-polarized clusters can also lead to the pair-breaking effect. Our analysis of the dependence $H_{c2}(T)$ leads to the conclusion that pair breakers are present in Y-Ba-Cu-O even at optimum doping, but the question of the nature of these magnetic moments (i.e., whether we are dealing with the magnetic in-plane inclusions or whether there is also a contribution from the Ba-O layer) deserves special additional study.

In this paper we have focused on the case of a single layer in the unit cell. This is appropriate, because the experiments[4,5] were performed on 2201 samples with relatively low critical temperatures. Nevertheless, there exists an interesting question of the dependence of $H_{c2}(T)$ on the number of layers in the unit cell. This problem has been studied in detail in Refs. 43, 44 for layered systems without magnetic impurities. As regards magnetic scattering, which is the subject of the present paper, one should note that a study of overdoped 2212 compounds would be of definite interest. Such a study would allow one to distinguish between two types of magnetic scatterers (see above) or, if both types are present, to estimate their relative contributions. If the magnetic moments are located on the apical oxygen site, then one should expect a relative decrease in the curvature of $H_{c2}(T)$; in other words, the ratio $H_{c2}(0)/H_{c2}(0)_{\mathrm{nm}}$ will decrease [$H_{c2}(0)_{\mathrm{nm}}$ corresponds to the absence of magnetic impurities; see Refs. 2, 43, 44]. Indeed, the Cu-O plane in the 2201 compound is affected by two O^- apical ions. At the same time, the effect of magnetic scattering is weaker for the 2223 compound, because the apical oxygens are located outside of the set of three planes. As for the in-plane magnetic ''clusters,'' their contribution does not depend on the number of the layers. Note that the value of T_c is also affected by the total number of the layers. The observed increase in T_c with increasing number of layers (e.g., $T_c=90$ K for the 2201 Tl-based cuprate and $T_c=125$ K for the 2223 compound) provides support for our concept of the magnetic moments being located on the apical oxygen site.

The experiments[4,5] have generated a lot of interest. A number of models leading to upward curvature in $H_{c2}(T)$ have been developed (see, e.g., Ref. 45). However, if we want to relate the theoretical model explicitly to the phenomenon observed in Refs. 4, 5, one should not only have a theoretically rigorous model, but one should carry out a detailed comparative analysis of the data.[4,5] From this point of view, for example, the bypolaronic model[45(a)] leads to a divergence at $T\rightarrow 0$, whereas the data[4] strongly indicate an

almost linear dependence in this region without any sign of divergence.

We think that our approach, based on the presence of magnetic impurities and inhomogeneities, is directly related to the data[4–13] as well as to other similar observations; our theory does not contain any additional assumptions, since the presence of magnetic impurities, as well as inhomogeneities, is directly determined by number of other techniques. $H_{c2}(T)$ saturates as $T\rightarrow 0$ K for samples with $T_c^{\rm opt}=90$ K;[46] this agrees with our theory since $T_c^{\rm opt}$ corresponds to a smaller n_M.

D. Other systems: Superlattices, YNi_2B_2C compound

In addition to the systems discussed in detail above, a peculiar temperature dependence of the critical field, drastically different from the conventional picture,[1,2] has been observed for other systems as well.

As was noted above (Sec. II B), a change in the functional form of $H_{c2}(T)$, due to the pair-breaking effect of magnetic impurities and their ordering trend at low temperatures, can be observed in 3D systems as well. In connection with this, one should mention a very interesting study of Ni/V superlattices.[12] A decrease in the thickness of the Ni films was seen to result in a change in the curvature of $H_{c2}(T)$. We think that this effect can be explained by the picture described here. Indeed, according to Ref. 12, a decrease in Ni thickness below 10 Å also leads to a decrease in the Curie temperature $T_{\rm Cu}$. Eventually, $T_{\rm Cu}$ drops below the superconducting critical temperature: $T_{\rm Cu}<T_c$. Then, in the region $T_c>T>T_{\rm Cu}$, one deals with the usual spin-flip scattering and pair-breaking effects, whereas the ordering occurring at $T_{\rm Cu}$ frustrates such scattering and gives rise to a ''recovery'' of H_{c2}, that is, to the type of behavior described in this paper. At larger thicknesses $T_c<T_{\rm Cu}$ and below T_c, the magnetic moments are always ordered; as a result, the scattering amplitude (here the dipole-dipole interaction plays an important role; see Sec. II B) only weakly depends on the temperature. The scenario, described here, accounts for the situation, observed in Ref. 12, whereby the $T_c-T_{\rm Cu}$ crossover drastically affects the dependence $H_{c2}(T)$: The functional form of the temperature dependence changes dramatically in the region $T<T_{\rm Cu}<T_c$.

The YNi_2B_2C compound, studied recently in Ref. 11, has a layered structure, but, nevertheless, displays isotropic superconducting properties. Increasing magnetic field leads, primarily, to a parallel shift in the I-V curve, permitting reliable measurements of the dependence $H_{c2}(T)$. The measurements in Ref. 11 were performed in the region $T>4$–5 K ($T_c=15$–16 K), but the data, nevertheless, clearly show a strong deviation from the HW picture with the appearance of a positive curvature similar to that observed in Refs. 4–6. We think that this effect is caused by spin-flip scattering of Ni ions and their ordering. It would be interesting to measure $H_{c2}(T)$ near $T=0$ K in order to observe a large increase in H_{c2}.

E. Other properties

Ordering of magnetic impurities leads to a frustration of pair breaking, and consequently to a decrease in the normal component. This decrease is manifested not only in the behavior of the critical field, but in a number of other properties as well. For example, according to Ref. 14, the gaplessness gives rise to a linear temperature dependence of the electronic heat capacity; the value of the Sommerfeld constant is directly related to the amount of the normal component. The presence of magnetic impurities and their ordering trend near $T=0$ K decreases the pair breaking and, consequently, further lowers the normal component. As a result, the temperature dependence of $C_{\rm el}$ becomes nonlinear. Such an effect has, indeed, been observed experimentally.[30] Note also that since the Sommerfeld constant is directly related to the normal density of states, which for a layered structure is proportional to the effective mass,[37] one might conclude that the ordering of magnetic impurities effectively decreases m^*. What is happening in reality is a decrease in the normal component. The latter is also manifested also in the penetration depth, electronic thermal conductivity, etc.

A study of the $SmRh_4B_4$ system has shown that the superconducting order parameter measured by Josephson tunneling increases below the antiferromagnetic ordering temperature.[46] This observation is also directly related to the picture we have discussed. Indeed, ordering leads to a less depressed superconducting state, which is not only manifested in an increase in H_{c2}, but is also accompanied by an increase in the superconducting order parameter. As a result, the amplitude of the Josephson current increases.

To summarize the previous discussion, our approach is based on a specific physical model. Namely, the temperature dependence $H_{c2}(T)$, which is our major focus, is greatly affected by the correlation of magnetic impurities at low temperatures. The trend weakens the pair-breaking effect, enhances superconductivity, and increases H_{c2}.

F. Critical field and inhomogeneities

In Secs. II and III we discussed two different scenarios leading to a positive curvature for a dependence $H_{c2}(T)$. Such a curvature can appear as a result of spin-flip scattering by magnetic impurities, based on the temperature dependence of the scattering amplitude. The other possibility is related to presence of inhomogeneites. As described above, in both cases one can observe a positive curvature. However, the second scenario provides for a smaller increase of $H_{c2}(0)$ relative to its conventional HW value.

The presence of inhomogeneites is responsible for the upward curvature of $H_{c2}(T)$ in the $Ba_{1-x}K_xBiO_3$ compound. This effect has been observed in Ref. 9. The compound has a cubic structure and does not contain localized magnetic moments. This is the reason why the spin-flip scattering mechanism is irrelevant in this case. However, it is known that samples of this material are of multiphase composition. Indeed, the superconducting state of this material occurs only for the cubic phase with $x>0.37$ (see, e.g., Ref. 48). At the same time, a very close value of the concentration ($x=0.35$) corresponds to a coexistence with the semiconducting phase. Indeed, the upper critical field H_{c2} was measured in Ref. 9 for samples with x in the range $x<0.4$. Under such conditions superconducting and semiconducting phases coexist; that is, the sample is inhomogeneous. The scale of this inhomogeneity is on the order of 100 Å.[9,48] The coherence length ξ_0 is of order of 50 Å. In this case the model described in

Sec. III is applicable. The presence of inhomogeneities leads to the dependence $H_{c2}^{*}(T)=\beta(T)H_{c2}^{\mathrm{HW}}(T)$ [here H_{c2}^{*} is the nucleation field, and $H_{c2}^{\mathrm{HW}}(T)$ is the conventional HW dependence[2]]. The function $\beta(T)$ changes from $\beta=1$ at $T=T_c$ to $\beta=1.7$ at $T=0$, which is, indeed, the scale of the increase observed in Ref. 9.

Note also that the semiconducting phase has an energy gap. The size of this gap is smaller that the value of T_c; the appearance of this gap is probably related to the charge-density-wave (CDW) transition. In this case one has to take into account the proximity effect between superconducting and semiconducting regions (particularly near T_c). The proximity effect leads to an effective decrease in T_c and, correspondingly, to a broadening of the transition. Because the value of H_{c2} was determined from resistivity measurements, this broadening leads to the appearance of a tail in the dependence $H_{c2}(T)$; such a tail has been, indeed, observed in Ref. 9. One should add also that the presence of the CDW gap makes the number of carriers in the semiconducting phase decrease as $T\rightarrow 0$ K. This greatly diminishes the proximity effect and depression of the superconducting state (including T_c). Such a decrease in the proximity effect acts similar to magnetic impurity ordering and can also increase $H_{c2}(0)$. We will describe this effect in detail elsewhere.

The presence of semiconducting phase islands leads to pinning in such materials and, consequently, to a finite value of the critical current (see Ref. 48).

The observed dependence $H_{c2}(T)$ in $Ba_{1-x}K_xBiO_3$ can be described with good accuracy by the model described in Sec. III, that is, by the presence of inhomogeneities.

A positive curvature caused by the presence of inhomogeneites has been observed also in Ref. 13. The authors prepared holes in the Al films and observed changes in the dependence $H_{c2}(T)$ caused by the presence of such holes.

A positive curvature has been also observed in the organic layered superconductor k(BEDT-TTF)$_2$ Cu(NCS)$_2$ compound,[10] $T_c=10.5$ K. Although the curvature is positive, in contrast to the conventional HW picture,[2] no sharp increase, similar to that observed in the cuprates, is seen at low temperatures. The effect, observed in Ref. 10, is caused by inhomogeneities. This conclusion is supported by the large value of the pinning force reported in Ref. 49, which greatly exceeds the value in the cuprates.

V. CONCLUSION

The main results of this paper can be summarized as follows.

(1) The magnetic properties of layered superconductors and, in particular, the nature of the temperature dependence of H_{c2} are greatly affected by magnetic impurities. The temperature dependence of the scattering amplitude leads to an increase in the value of H_{c2} and results in a positive curvature, in complete contrast with the conventional picture.[1,2]

(2) Our theoretical calculation explains recent experimental data[4,5] on the critical field behavior in overdoped cuprates; Figs. 2 and 3 demonstrate an excellent agreement of the theory with the data.

The analysis is also in a very good agreement with the data on the Sm-Ce-Cu-O system[6] and the underdoped Y-Ba-Zn-Cu-O compound.[8]

(3) The material contains magnetic impurities even at optimum doping, which implies that the intrinsic T_c exceed $T_{c,\mathrm{opt}}^{\mathrm{dop}}$.

(4) The dependence $H_{c2}(T)$ can be also modified by 2D in-plane inhomogeneities.

The treatment developed above not only allows one to explain the experimental data such as Refs. 4–13, but indicates that it is possible to modify the behavior of H_{c2} by means of adding magnetic impurities or selected inhomogeneities.

ACKNOWLEDGMENTS

The authors are grateful to S. Wolf, A. Mackenzie, L. Gor'kov, and I. Schuller for fruitful discussions. One of us (Y.N.O.) wishes to acknowledge support of the Humboldt Foundation. The research of V.Z.K. is supported by the U.S. Office of Naval Research under Contract No. N00014-95-F0006.

APPENDIX

(I) The critical field is determined by Eqs. (2) and (3). In order to derive expression (3) for the kernel $J(\omega_n,H)$ in the 2D case, consider the relation

$$\Delta=\lambda\nu 2\pi T\sum_{n\geqslant 0}\int_0^{\infty}(d\varphi/2\pi)\beta_p. \tag{A1}$$

Here Δ is the order parameter and β_p is the Green's function. As was shown by one of the authors,[21] β_p is satisfied by the following linear equation (at $H=H_{c2}$):

$$\left\{\mathrm{sgn}\ \omega_n v\partial_- +2\omega+\tau^{-1}+\tau_s^{-1}-nv\int_0^{\infty}d\Omega_{p_1}\sigma_{pp_1}+\tau_s^{-1}\int_0^{\infty}(d\varphi/2\pi)\right\}\beta_p=2\Delta. \tag{A2}$$

The operator ∂_- is defined by Eq. (1); the vector potential $\mathbf{A}=H(0,x,0)$. One can seek the solution in the form $\Delta=\Delta_0\exp(-eHx^2)$. The solution of Eq. (A2) can be written in the form

$$\beta_p=\mathrm{sgn}\ \omega_n\left[1+\mathrm{sgn}\ \omega_n(\tau^{-1}-\tau_s^{-1})D_1(\omega_n)\times\int_0^{\infty}dx_1K_p(x,x_1)\exp(-eHx_1^2),\right. \tag{A3}$$

where

$$K_p(x,x_1)=\theta(x-x_1)Rs^{-1},\ \cos\varphi>0,$$

$$\theta(x_1-x)Rs^{-1},\ \cos\varphi<0,$$

$$R=\exp[-\alpha(x-x_1)s^{-1}+ieH(x^2-x_1^2)\tan\varphi];\ \ s=v\cos\varphi. \tag{A4}$$

α is defined by Eq. (3).

One can write also the following equation for $D_1(\omega)$:

$$D_1(\omega_n)=[\mathrm{sgn}\omega_n+(\tau^{-1}-\tau_s^{-1})]D_1(\omega_n)$$
$$\times\int_0^\infty (d\varphi/2\pi)dx_1K_p(x,x_1)\exp[-eH(x_1^2-x_2^2)]. \tag{A5}$$

Based on Eqs. (A4) and (A5), we arrive at Eq. (2′) with

$$J(\omega_n,H)=\int_0^\infty dx_1[2\pi eHv^2(x^2-x_1^2)]^{-1/2}$$
$$\times\exp[-\alpha^2(2eHv^2)^{-1}(x-x_1)(x+x_1)^{-1}]. \tag{A6}$$

Equation (A6) can be reduced to Eq. (3).

(H) Consider the S-N system. The order parameter is not equal to zero only in the first (S) layer, and we have

$$\Delta=\lambda\nu_1 2\pi T\sum_{n\geqslant 0}\beta_1 \tag{A7}$$

(λ and ν_1 are the coupling constant and density of states for the first layer; for the dirty case, the Green's function does not have, in a first approximation, the angular dependence). One can write expressions for the Green's functions β_i ($i=1,2$) in the form $\beta_i=C_i(z,\omega)\exp(-eHx^2)$.[50] Assume also that the thicknesses $d_i\ll\xi_i$.

The functions C_i can be written in the form

$$C_1(z,\omega)=C_{11}(\omega)+C_{12}(\omega)(d_1-z)^2,\quad 0<z<d_1,$$
$$C_2(z,\omega)=C_{22}(\omega)+C_{21}(\omega)(d_2+z)^2,\quad -d_2<z<0. \tag{A8}$$

With use of the boundary conditions, we obtain $C_{11}=C_{22}$, $C_{12}=-C_{21}$ (κ_2/κ_1), and $\kappa_\iota=\nu_\iota D_i d_i$. Based on Eqs. (17a), (17b), and (A8) and the expression $\Delta=\Delta_0\exp(-eHx^2)$, one can obtain

$$C_{11}=\Delta_0[\omega(1+k)+(\Gamma_1+k\Gamma_2)+eH(D_1+kD_2)]^{-1}. \tag{A9}$$

k is defined by Eq. (20). Inserting this expression in (A7) and introducing the notation $\widetilde{T}_c^0=T_c^0\ (\pi T_c^0/2\gamma\omega_D)^k$, $\Gamma=(\Gamma_1+k\Gamma_2)/(1+k)$, and $D=(D_1+kD_2)/(1+k)$, we arrive, after some manipulations, at Eq. (19).

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

[1] L. Gor'kov, Sov. Phys. JETP **10**, 593 (1960).

[2] E. Helfand and N. R. Werthamer, Phys. Rev. Lett. **13**, 686 (1964); Phys. Rev. **147**, 288 (1966).

[3] Yu. N. Ovchinnikov and V. Z. Kresin, Phys. Rev. B **52**, 3075 (1995).

[4] A. P. Mackenzie *et al.*, Phys. Rev. Lett. **71**, 1938 (1993); J. Supercond. **7**, 27 (1994); Physica C **235-240**, 233 (1994); A. Carrington *et al.*, Phys. Rev. B **49**, 13 243 (1994).

[5] M. Osofski *et al.*, Phys. Rev. Lett. **71**, 2315 (1993); J. Supercond. **7**, 279 (1994).

[6] Y. Dalichaouch *et al.*, Phys. Rev. Lett. **64**, 599 (1990).

[7] M. Suzuki and M. Hikita, Phys. Rev. B **44**, 249 (1991).

[8] D. Warner *et al.*, Phys. Rev. B **51**, 9375 (1995).

[9] C. Escribe-Filippini *et al.*, Physica C **210**, 133 (1993).

[10] K. Oshima *et al.*, Physica C **153**, 1148 (1988).

[11] M. Xu *et al.*, Physica C **227**, 321 (1994); **235–240**, 2533 (1994).

[12] H. Homma *et al.*, Phys. Rev. B **33**, 3562 (1986).

[13] 11. A. Bezryadin and B. Pannetier, J. Low Temp. Phys. **98**, 251 (1995).

[14] A. Abrikosov and L. Gor'kov, Sov. Phys. JETP **12**, 1243 (1961).

[15] P. de Gennes, Phys. Condens. Matter **3**, 79 (1964); *Superconductivity in Metals and Alloys* (Benjamin, New York, 1966).

[16] (a) D. Saint-James, G. Sarma, and E. Thomas, *Type II Superconductors* (Pergamon, Oxford, 1969); (b) K. Maki, in *Superconductivity*, edited by R. Parks (Dekker, New York, 1969), p. 1035.

[17] S. Skalski, O. Betbeder, and P. Weiss, Phys. Rev. **136**, A1500 (1964).

[18] V. Kresin and S. Wolf, Phys. Rev. B **51**, 1229 (1995).

[19] A. Abrikosov, *Fundamentals of the Theory of Metals* (North-Holland, Amsterdam, 1988).

[20] O. Fischer, Helv. Phys. Acta **45**, 332 (1972).

[21] Yu. Ovchinnikov, Sov. Phys. JETP **39**, 538 (1974).

[22] G. Eilenberger, Z. Phys. **214**, 195 (1968).

[23] A. Larkin and Yu. Ovchinnikov, Sov. Phys. JETP **28**, 1200 (1969).

[24] J. Winter, *Magnetic Resonance in Metals* (Oxford University Press, New York, 1971), Chap. 2.

[25] Yu. Ovchinnikov, I. Vagner, and A. Dyagaev, JETP Lett. **59**, 569 (1994).

[26] V. Gantmakher and Y. Levinson, *Carrier Scattering in Metals and Semiconductors* (North-Holland, Amsterdam, 1987).

[27] Y. Shimakawa, Physica C **204**, 247 (1993).

[28] K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, Amsterdam, 1984); K. Fisher and J. Hertz, *Spin Glasses* (Cambridge University Press, Cambridge, England, 1991).

[29] N. Phillips, R. Fisher and J. Gordon, in *Progress in Low-Temperature Physics* **13**, edited by D. Brewer (North-Holland, Amsterdam, 1992), p. 267.

[30] J. Wade *et al.*, J. Supercond. **7**, 261 (1994).

[31] A. Judod, in *Studies of High Temperature Superconductors*, edited by A. Narlikar (Nova Science, Commack, NY, in press); we are grateful to the author for sending the manuscript prior to publication.

[32] I. Schuller, R. Orbach, and P. Chaikin, Phys. Rev. Lett. **41**, 1413 (1978).

[33] K. Biagi and V. Kogan, J. Clem, Phys. Rev. B **32**, 7165 (1985).

[34] Yu. N. Ovchinnikov, Sov. Phys. JETP **52**, 755 (1980).

[35] C. Niedermayer *et al.*, Phys. Rev. Lett. **71**, 1764 (1993); J. Supercond. **7**, 165 (1994).

[36] J. Tallon *et al.*, Phys. Rev. Lett. **74**, 1008 (1995).

[37] V. Kresin and S. Wolf, Phys. Rev. B **41**, 4278 (1990).

[38] G. Williams, J. Tallon, and R. Meinhold, Phys. Rev. B **52**, 7034 (1995).

[39] V. Gusakov, Physica C **235–240**, 813 (1994).

[40] *Phase Separation in Cuprate Superconductors*, edited by E. Sigmund and K. A. Mueller (Springer-Verlag, Berlin, 1994).

[41] A. Furrer *et al.*, Ref. 39, p. 101; J. Supercond. (to be published).

[42] M. Osofsky *et al.*, Appl. Phys. Lett. **68**, 2729 (1996).

[43] A. Rajagopal and S. Jha, Physica C **174**, 161 (1991); **185-189**, 1543 (1991).
[44] S. Jha and A. Rajagopal, Pramana J. Phys. **39**, 615 (1992).
[45] (a) A. S. Alexandrov, Phys. Rev. B **44**, 10 571 (1993); (b) J. Cooper *et al.*, *ibid*. **51**, 6179 (1995).
[46] J. Smith *et al.*, J. Supercond. **7**, 269 (1994); A. Bykov *et al.*, Physica B **211**, 248 (1995).
[47] R. Vaglio *et al.*, Phys. Rev. Lett. **53**, 1489 (1984).
[48] S. Pei *et al.*, Phys. Rev. B **41**, 4126 (1990).
[49] A. Swanson *et al.*, Solid State Commun. **73**, 353 (1990).
[50] A. Larkin and Yu. Ovchinnikov, in *Nonequilibrium Superconductivity*, edited by D. Landerberg and A. Larkin (Elsevier, Amsterdam, 1986), p. 493.