Positive curvature of H_{c2} in layered superconductors

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We present a phenomenological calculation of the equilibrium upper critical field H_{c2} in layered superconductors. We find that $H_{c2}(T)$ has positive curvature near T_c, but becomes linear further away, as a result of a proximity effect between neighboring superconducting and nonsuperconducting layers, for low-T_c and high -T_c superconductors alike.

I. INTRODUCTION

The upper critical field H_{c2} of the high-T_c layered superconductors has attracted a lot of attention recently.[1] Early resistance measurements showed that $H_{c2}(T)$ had positive curvature near T_c for all field orientations.[2] These measurements proved unreliable,[3] though, because they were masked by irreversible effects, such as flux creep, and the values of H_{c2} extracted from them should rather be interpreted as a determination of an irreversibility line. Since thermally activated flux creep affects the resistive measurements enormously, reversible measurements would be needed for determining the true thermodynamic H_{c2}. Such measurements were recently undertaken. dc-magnetization measurements[4] on single crystals of $YBa_2Cu_3O_{7-\delta}$ revealed a well-defined onset of diamagnetism, allowing a unique magnetic determination of the true upper critical field. At the magnetically determined nucleation temperatures no distinct features were found in resistive transition curves. $H_{c2}(T)$ had, once again, positive curvature near T_c, but at lower temperatures it became a straight line that intercepted the T axis at around 91.5 K, about 1 K below T_c. The angular dependence of the upper critical field was also measured with dc magnetization,[5] the results being describable with a three-dimensional anisotropic Ginzburg-Landau (GL) theory in the region of linear H_{c2}.

It seems certain then that the H_{c2} of high-T_c superconductors has positive curvature near T_c, for *any* field orientation. Such curvatures have also been observed in low-T_c layered superconductors,[6] such as intercalated TAS_2 and MoS_2, where the flux creep effects are negligible.[3] For the field direction parallel to the layers, the positive curvature is attributed to a kind of dimensional crossover predicted by the Lawrence Doniach model of identical Josephson coupled superconducting layers[7] (for various extensions of the model, and for reviews, see Ref. 8). In fact, $H_{c2}^{\parallel}$ may diverge at low temperatures in this model, if the cores of the vortices fit between the layers. The generic curvature for the field direction parallel to the c axis is still not well understood, however. The GL theory predicts a linear temperature dependence of $H_{c2}^{\perp}$ near T_c, but positive curvature is universal in the naturally occurring superconducting layered compounds.[6] For a review of the various theoretical efforts and experimental results, see Ref. 9.

In this paper we extend the simple physical picture proposed earlier[10] in order to account for the positive curvature of H_{c2} in layered superconductors, low and high T_c alike. We make use of the fact that most of the naturally occurring layered compounds contain not only superconducting (SC) layers, but also nonsuperconducting (NSC) ones. Normally, the order parameter is zero on the NSC layers. The Josephson coupling between neighboring SC and NSC layers, though, makes the order parameter nonzero on the NSC layers as well, through a proximity effect. Thus we have differing order parameters on different layers. The spatial variation of the order parameter from layer to layer in materials where NSC layers are in the proximity of SC layers gives rise to the observed positive curvature.

In particular, we predict that the upper critical field $H_{c2}^{\perp}(T)$ in the direction of the c axis has positive curvature near T_c, but becomes a straight line further away, that intercepts the T axis below T_c. The linear region of $H_{c2}(T)$ is precisely the region where the order parameter on the NSC layers has become negligible, since the proximity effect does not exist there any more. We further predict that the intercept of this straight line with the T axis is independent of the field direction, and that the angle dependence of H_{c2} in the linear region is precisely that of the anisotropic GL theory.

The inclusion of the NSC layers is crucial for this proximity effect. The reason Klemm *et al.*[7] get a linear $H_{c2}^{\perp}(T)$ near T_c is that they use only identical SC layers. It is the presence of inequivalent layers that creates the curvature. Experimental indications for the presence of this proximity effect in the Bi 2:2:1:2 high-T_c superconductors have been reported recently by Briceno and Zettl.[11]

A macroscopic version of this effect exists in superconducting multilayers.[9,12] Theoretical considerations[13] have shown that the temperature dependence of the field

$H_{c2}^{\perp}$ perpendicular to the layers could exhibit anomalous upward curvature under appropriate conditions. When the coherence length is long, near T_c, the order parameter is continuously coupled through the multilayer, but when it becomes small enough compared with the layer thickness, the order parameter decreases exponentially in the NSC layers, producing thus a decoupling of the SC layers. Thus the observed $H_{c2}^{\perp}$ at lower temperatures is that of the SC layers, and consequently very high.

In Sec. II we present the free energy adopted. In Secs. III and IV, we calculate H_{c2} for $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Ba_xCuO_{4-y}$, respectively. Our conclusions are presented in Sec. V.

II. FREE ENERGY

We shall consider a series of superconducting and nonsuperconducting layers. Let ψ_n and $\mathbf{A}_n$ be the order parameter and vector potential on the nth layer. We shall define the discretized magnetic field $\mathbf{H}_n$ on each layer as

$$\mathbf{H}_n = \hat{\mathbf{z}}(\partial A_{yn}/\partial x - \partial A_{xn}/\partial y) + \hat{\mathbf{x}}(\partial A_{zn}/\partial y - DA_{yn}) + \hat{y}(DA_{xn} - \partial A_{zn}/\partial z) \ . \quad (1)$$

Here $\hat{\mathbf{z}}$ is the direction of the c axis and Df_n is the discretized derivative across the layers, $Df_n = (f_{n+1} - f_n)/d_{n,n+1}$, where $d_{n,n+1}$ is the distance between the nth and $(n+1)$th layers. $\mathbf{H}_n$ is invariant under the discrete gauge transformation $\mathbf{A}_n \to \mathbf{A}_n + \nabla_{\parallel}\chi_n + \hat{\mathbf{z}}D\chi_n$, where $\chi_n(x,y)$ is an arbitrary function of x and y, and $\nabla_{\parallel}$ is the gradient along the layers. We also have $\psi_n \to \psi_n \exp(-2ie\chi_n/\hbar c)$ under the gauge transformation. Near H_{c2}, we may neglect the influence of ψ_n on $\mathbf{A}_n$, since ψ_n is very small, and we may also neglect quartic terms and pure gauge terms. Then the gauge invariant Gibbs free energy describing layered superconductors, where the Cooper pairs consist of holes, is

$$\int dx\, dy \sum_n [a_n(T)|\psi_n|^2 + |-i\hbar\nabla_{\parallel}\psi_n + 2e\,\mathbf{A}_n^{\parallel}\psi_n/c|^2/2m_n^* + \eta_{n,n+1}|\psi_{n+1} - \psi_n \exp(-2ied_{n,n+1}A_{zn}/\hbar c)|^2] \ . \quad (2)$$

This generalizes the free energy[7] used to describe a stack of equidistant superconducting layers by introducing inequivalent layers. The fact that we may be integrating over unequal distances is incorporated into the constants $a_n, m_n^*, \eta_{n,n+1}$, the use of which makes manifest the introduction of inequivalent layers. The concept of differing order parameters on different interacting subunits was introduced first in Ref. 14, where it was realized in the context of a hypothetical layered structure with alternating sheets having different types of conductivity. Attention was drawn to the fact that the properties of low-T_c superconductors differ radically from those of purely two-dimensional systems, due to the role of the interacting subunits, of which these superconductors consist.[14] Recently this idea has been explored systematically in the context of high-T_c superconductivity,[10,15] where free energies of the form of Eq. (2) have been used.

We shall assume that $\mathbf{H}_n = H(\cos\theta\hat{\mathbf{z}} + \sin\theta\hat{\mathbf{y}})$, i.e., $\mathbf{A}_n = xH(\cos\theta\hat{\mathbf{y}} - \sin\theta\hat{\mathbf{z}})$. Then the free energy is minimized if ψ_n is a function of x only, in which case it takes the form

$$\int dx \sum_n [a_n(T)|\psi_n|^2 + \hbar^2(|\partial\psi_n/\partial x|^2 + y^2\cos^2\theta|\psi_n|^2)/2m_n^* + \eta_{n,n+1}|\psi_{n+1} - \psi_n \exp(id_{n,n+1}y\sin\theta)|^2] \ , \quad (3)$$

with $y = 2exH/\hbar c$.

Let us deal explicitly with two examples: $La_{2-x}Ba_xCuO_{4-y}$ and $YBA_2Cu_3O_{7-\delta}$, two high-T_c superconductors. $La_{2-x}Ba_xCuO_{4-y}$ consists of superconducting CuO_2 and nonsuperconducting La(Ba)O layers, in the order CuO_2-LaO-LaO-CuO_2-LaO-LaO-. . ., etc. The distance d_2 between neighboring LaO layers is 2.3629 Å, while the distance d_1 between neighboring LaO and CuO_2 layers is 2.1307 Å.[16] $YBa_2Cu_3O_{7-\delta}$, on the other hand, consists of Y-Cu(2)-BaO-Cu(1)-BaO-Cu(2)-Y-. . . layers, where only the Cu(2) layers are superconducting.[17] The distance d_1 between the Cu(1) "chains" and the Cu(2) planes is 4.1252 Å, while the distance d_2 between neighboring Cu(2) planes is 3.3830 Å.[18] We shall neglect in this paper the role of the BaO and Y layers in the $YBa_2Cu_3O_{7-\delta}$ superconductors, for simplicity, assuming that their effect may be approximately incorporated into the Josephson couplings.

Note that in both of these materials we have a single layer of one kind A, followed by a pair of layers of another kind B. In $La_{2-x}Ba_xCuO_{4-y}$, A is CuO_2 and B is LaO. In $YBa_2Cu_3O_{7-\delta}$, A is Cu(1) and B is Cu(2). Let the order parameter of the nth layer of kind A be ϕ_n. Let the order parameters of kind B that surround it on either side be ψ_n^L and ψ_n^R. If we denote the constants associated with kinds A and B by the subscripts 1 and 2, respectively, we get the free energy

$$\int dx \sum_n [a_1(T)|\phi_n|^2 + a_2(T)(|\psi_n^L|^2 + |\psi_n^R|^2) + \hbar^2(|\partial\phi_n/\partial x|^2 + y^2\cos^2\theta|\phi_n|^2)/2m_1^* + \hbar^2(|\partial\psi_n^L/\partial x|^2 + |\partial\psi_n^R/\partial x|^2 + y^2\cos^2\theta|\psi_n^L|^2 + y^2\cos^2\theta|\psi_n^R|^2)/2m_2^* + \eta_1|\psi_n^R - \phi_n e^{id_1 y\sin\theta}|^2 + \eta_1|\phi_n - \psi_n^L e^{id_1 y\sin\theta}|^2 + \eta_2|\psi_{n+1}^L - \psi_n^R e^{id_2 y\sin\theta}|^2] \ . \quad (4)$$

This complicated free energy describes both $La_{2-x}Ba_xCuO_{4-y}$ and $YBa_2Cu_3O_{7-\delta}$ near T_c. Here η_1 and d_1 are the Josephson coupling and the distance between neighboring layers of type A and B, while η_2 and d_2 are the same quantities between neighboring type B layers.

We sum the equations that minimize this free energy over all n, defining $\phi=\sum_n \phi_n/\sqrt{N}$, $\psi_L=\sum_n \psi_n^L/\sqrt{N}$, and $\psi_R=\sum_n \psi_n^R/\sqrt{N}$, where N is the number of layers considered. The symmetries of these equations allow ϕ to be real and $\psi_L=\psi_R^*$. We obtain thus

$$\hbar^2(\partial^2\phi/\partial x^2)/2m_1^* = [a_1(T)+\hbar^2 y^2\cos^2\theta/2m_1^* + 2\eta_1]\phi - \eta_1(\psi_L e^{id_1 y\sin\theta}+\text{c.c.}) , \tag{5}$$

$$\hbar^2(\partial^2\psi_L/\partial x^2)/2m_2^* = [a_2(T)+\hbar^2 y^2\cos^2\theta/2m_2^* + \eta_1+\eta_2]\psi_L - \eta_1\phi e^{-id_1 y\sin\theta} - \eta_2\psi_L^* e^{id_2 y\sin\theta} . \tag{6}$$

These equations are also the equations that minimize the effective functional

$$\int dx[a_1(T)\phi^2+2a_2(T)|\psi|^2+\hbar^2y^2\cos^2\theta\phi^2/2m_1^*+\hbar^2y^2\cos^2\theta|\psi|^2/m_2^*$$
$$+2\eta_1|\phi-\psi\exp(id_1y\sin\theta)|^2+\eta_2|\psi-\psi^*\exp(id_2y\sin\theta)|^2+\hbar^2(\partial\phi/\partial x)^2/2m_1^*+\hbar^2|\partial\psi/\partial x|^2/m_2^*] , \tag{7}$$

where for convenience we have dropped the subscript L of ψ. This functional contains all of the physical information, and it will be used extensively in what follows.

III. $YBa_2CU_3O_{7-\delta}$

Let us begin the calculation of H_{c2} by examining $YBa_2Cu_3O_{7-\delta}$. Here the superconducting Cu(2) layers are the ones associated with the subscript 2, the nonsuperconducting ones are associated with the subscript 1. We assume $a_2(T)=a_2(T-T_0)/T_c$ and $a_1(T)=a_1$, where T_0 is some phenomenological temperature, and a_1,a_2 are positive constants. Thus temperature dependence appears explicitly only in the coefficients associated with the Cu(2) layers. We can rewrite the effective functional of Eq. (7) in terms of $\lambda_1=2\eta_1^2/a_2(a_1+2\eta_1)$, $\lambda_2=\eta_2/a_2$, as well as $l_1^2=\lambda_1\hbar^2/m_1^*(a_1+2\eta_1)$, $l_2^2=\hbar^2/2m_2^*a_2$ and T_c, by using the rescaled order parameters $\phi'=\phi\eta_1/\lambda_1$ and $\psi'=a_2\psi$, and noting that $T_0/T_c=1+(\eta_1/a_2)-\lambda_1$. Then our effective functional becomes

$$G_{\text{eff}}=\int dx[2(T/T_c-1)|\psi|^2+2\lambda_1|\phi-\psi\exp(id_1y\sin\theta)|^2+\lambda_2|\psi-\psi^*\exp(id_2y\sin\theta)|^2$$
$$+l_1^2(y^2\cos^2\theta\phi^2+(\partial\phi/\partial x)^2)+2l_2^2(y^2\cos^2\theta|\psi|^2+|\partial\psi/\partial x|^2)] , \tag{8}$$

where we dropped the primes, for brevity. This is our basic expression for the free energy of $YBa_2Cu_3O_{7-\delta}$, and all our subsequent work will be based on this. Note that there appear only four parameters here: λ_1 [the Josephson coupling between Cu(1) and Cu(2) layers], λ_2 [the Josephson coupling between neighboring Cu(2) layers], and l_1 and l_2 [the phenomenological lengths associated with the Cu(1) and Cu(2) layers]. These are parameters that will be determined by fitting our H_{c2} predictions to the experimental data. The old parameters that had been used before, such as m_n^*, are not physical observables, in the sense that only their combinations that make up these four new parameters can be determined from experiments conducted in the Ginzburg-Landau regime. In the remainder of this section we shall use the functional of Eq. (8) in order to determine the upper critical field of $YBa_2Cu_3O_{7-\delta}$, using the techniques of Ref. 10.

Let us begin with finding the upper critical field $H_{c2}^{\perp}$ parallel to the c axis ($\theta=0$). In that case both ψ and ϕ may be taken to be real. So we get the equations of motion

$$l_2^2\partial^2\psi/\partial x^2=\lambda_1(\psi-\phi)+l_2^2y^2\psi+(T/T_c-1)\psi , \tag{9}$$

$$l_1^2\partial^2\phi/\partial x^2=2\lambda_1(\phi-\psi)+l_1^2y^2\phi . \tag{10}$$

The obvious solutions are $\psi(x)=\psi_0\exp(-ex^2H/\hbar c)$ and $\phi(x)=\phi_0\exp(-ex^2H/\hbar c)$, with

$$(T/T_c-1+\lambda_1+h_2)\psi_0=\lambda_1\phi_0 , \tag{11}$$

$$(2\lambda_1+h_1)\phi_0=2\lambda_1\psi_0 , \tag{12}$$

where $h_n=2eHl_n^2/\hbar c$ ($n=1,2$). These last two equations have nonzero solutions only when

$$1-T/T_c=h_2+\lambda_1h_1/(2\lambda_1+h_1) . \tag{13}$$

This is our *exact* result for $H_{c2}^{\perp}$. We note that in this expression $T\to\infty$, if h_1 approaches $-2\lambda_1$ from above. Therefore $H_{c2}^{\perp}(T)$ has a horizontal asymptote at some negative value of H, but it becomes a straight line with negative slope as $H\to\infty$. It must then have positive curvature in between these two limits.

As $H\to\infty$, we see that this equation becomes

$$1-T/T_c\approx\lambda_1+h_2 . \tag{14}$$

Therefore at large fields our $H_{c2}^{\perp}$ becomes a straight line that intercepts the T axis at $T=(1-\lambda_1)T_c$. This is precisely the behavior of $H_{c2}^{\perp}$ in the experiment of Ref. 4 (see their Fig. 4). In fact, their $H_{c2}^{\perp}$ for $H\geq 0.5$ Tesla is given by[19] $H_{c2}^{\perp}\approx 173.1689-1.8919T$, and the T_c is 92.4375 K. We can identify therefore $l_2=13.725$ Å and $\lambda_1=0.0098$. Our Fig. 1 shows the agreement between our fit and the experimental data.

We can now proceed with finding H_{c2} for an arbitrary field orientation. We shall make use of the fact that the functional of Eq. (8) should be zero at the transition from the superconducting to the normal state. Indeed, we can easily verify that the equations that minimize it make it zero. The upper critical field can be obtained then by extremizing $G_{\rm eff}$ and setting its extremum equal to zero. We use this result in a perturbative calculation that will give us some intuition about H_{c2}.

If we set $\lambda_1=\lambda_2=0$, $G_{\rm eff}$ is extremized when $\psi(x)=\psi_0\exp(-ex^2H\cos\theta/\hbar c)$ and $\phi=0$. The upper critical field is given then by $1-T/T_c=h_2\cos\theta$. So, if we assume that λ_1 and λ_2 are small, we can get the correction to the H_{c2} by evaluating $G_{\rm eff}$ for the above unperturbed functional forms of ϕ and ψ, and setting the result equal to 0.

We insert $\phi=0$ and $\psi(x)=\psi_0\exp(-ex^2H\cos\theta/\hbar c)$ into Eq. (8), and we set the resulting $G_{\rm eff}$ equal to 0, obtaining thus

$$1-T/T_c=h_2\cos\theta+\lambda_1+\lambda_2-\lambda_2\exp[-h_2(d_2/2l_2)^2\sin^2\theta/\cos\theta] \; . \tag{15}$$

A similar expression has been obtained in Ref. 20, in a similar problem with only one order parameter. Clearly, if we set $H=0$ in Eq. (15) we get $T\neq T_c$. So the above result cannot be valid too close to T_c. Indeed, setting $\theta=0$ gives us Eq. (14), the large -H limit of $H_{c2}^{\perp}$. So Eq. (15) should be valid for $H>0.5$ tesla.

Furthermore, it cannot be relied upon for angles very close to 90°, since the unperturbed $\psi(x)$ would be a constant in that case. In fact, we know that for $\theta=90°$ the solutions in similar problems are oscillatory Mathieu functions,[7] and therefore the transition from a confined order parameter, such as the unperturbed choice above, to one of infinite extent proceeds rapidly.[20] Such a rapid evolution would be reflected in a similarly rapid change in H_{c2}. Since an order parameter of increasing spatial extent implies one of decreasing energy, the approach of $\theta=90°$ would lead to a *sharp* increase in H_{c2},peaking at $\theta=90°$. Symmetry about $\theta=90°$ then implies a cusp in H_{c2} at $\theta=90°$. The point where this cusp would become noticeable can be ascertained from the exponent in Eq. (15). It is zero at $\theta=0$, and infinite at $\theta=90°$. However, the angular resolution in the experiment of Welp *et al.*[5] is about one degree. Thus, even for their highest field (5 Tesla), and $\theta=89°$, the exponent would be of the order of 0.025, very far from the $\theta=90°$ value of ∞. In fact, this exponent becomes 1 at 5 tesla only for $\theta=89.97°$. Such fine experimental resolution is impossible. Clearly then, the case $\theta=90°$ cannot be seen experimentally in $YBa_2Cu_3O_{7-\delta}$ for fields as low as 5 tesla. Therefore *all* angles are qualitatively similar to $\theta=0$, since the exponent is so small. Thus, Eq. (15) is reliable for fields >0.5 tesla and practically for all angles, as long as λ_1 and λ_2 are small.

We can obtain a better approximation to H_{c2} variationally. The perturbative argument above demonstrates that the order parameters are not oscillatory for basically all θ. We shall then adopt the ansatz

$$\psi(x)=e^{-\gamma x^2/2}, \quad \phi(x)=be^{-\gamma x^2/2} \; , \tag{16}$$

where γ and b are variational parameters. We calculate $G_{\rm eff}$ using this ansatz. Setting the result equal to 0 will give $T_c(H)$ as a function of b and γ. Maximization of $T_c(H)$ with respect to b and γ will give the final variational approximation to $T_c(H)$ (or, equivalently, H_{c2}).

The evaluation of $G_{\rm eff}$ and the maximization with respect to b can be performed analytically, giving $T_c(H)$ as a function of γ,

$$T/T_c-1=-\lambda_1+\lambda_2(e^{-f}-1)-2gl_2^2+\lambda_1^2e^{-2d_1^2f/d_2^2}/(\lambda_1+gl_1^2) \; , \tag{17}$$

where

$$g=\gamma/4+e^2H^2\cos^2\theta/\hbar^2c^2\gamma \tag{18}$$

and

$$f=d_2^2\sin^2\theta e^2H^2/\hbar^2c^2\gamma \; . \tag{19}$$

Maximization of the right-hand size (rhs) of Eq. (17) with respect to γ will yield the final result. When $H=0$, then $\gamma=f=g=0$ and $T=T_c$, as it should. When $\theta=0$, then Eq. (17) is maximized for $\gamma=2eH/\hbar c=2g$. In that case we obtain the exact result of Eq. (13). Therefore our variational result is very good for small angles. Since the perturbative arguments indicated that the case $\theta=89°$ is not really that much different from the case $\theta=0$, we expect it to be equally reliable at all angles.

In the general case we have to maximize the expression in Eq. (17) numerically, thus obtaining results that can be compared with experiment. Given though the small value of λ_1 found earlier, we shall neglect the last term of Eq. (17), which is of order λ_1^2. We cannot do that very near T_c, because there $g=0$, and that term becomes of order λ_1. But we can do it when $gl_1^2\gg\lambda_1$, i.e., at sufficiently high fields. We shall show later that this means $H>0.5$ tesla.

So, for sufficiently high fields

$$T/T_c-1\approx-\lambda_1+\lambda_2(e^{-f}-1)-2gl_2^2 \; . \tag{20}$$

We now note that if $\lambda_1=\lambda_2=0$, i.e., $\gamma=2eH\cos\theta/\hbar c$, then the exponent f appearing in Eq. (17) is really the same as the exponent in Eq. (15), and therefore about 0.025 or less, for the range of fields in Refs. 4 and 5. We expect thus the exponents in Eq. (17) to be small. So the expression of Eq. (20) may be expanded to first order in f, and then maximized. We obtain

$$\gamma\approx(2eH/\hbar c)(\cos^2\theta+\lambda_2d_2^2\sin^2\theta/2l_2^2)^{1/2} \tag{21}$$

and

$$1-T/T_c\approx h_2(\cos^2\theta+\lambda_2d_2^2\sin^2\theta/2l_2^2)^{1/2}+\lambda_1 \; . \tag{22}$$

This has the usual θ dependence of the GL anisotropic theory, and it is precisely of the form found experimentally in Ref. 5. Note that for $\theta=0$ this reduces to Eq. (14), which was found to be very reliable for $H\geq0.5$ tesla, as emphasized earlier.

Equations (13) and (22) are the basic results of this paper. It is these that we use to fit the data. It must be emphasized that, according to Eq. (22), at fields larger than 0.5 tesla, H_{c2} is a straight line that intercepts the T axis at

$(1-\lambda_1)T_c$, for *any* θ. This agrees with what was observed in Ref. 4. Furthermore, the angular dependence of Eq. (22) is precisely the one seen experimentally in Ref. 5.

In detail, the experimental data for the field direction parallel to the layers[19] can be fitted to the straight line $H_{c2}^{\parallel}=960.5433-10.4921T$, where the field is measured in teslas and temperature in Kelvins. Comparison with Eq. (22) for $\theta=90°$ gives $\lambda_1=0.0096$, in full agreement with the value 0.0098 obtained for λ_1 from the $H_{c2}^{\perp}$ data, and $\lambda_2=1.0704$. Thus the large-H limits of $H_{c2}^{\perp}$ and $H_{c2}^{\parallel}$ have been used to determine $\lambda_1=0.0098$, $\lambda_2=1.07$, and $l_2=13.725$ Å. The last unknown parameter l_1 is determined by the curvature near T_c.

In Fig. 1 we show the fit of our Eqs. (13) and (17) to the experimental data,[4] for the choice of parameters $\lambda_1=0.0098$, $\lambda_2=1.07$, $l_1=400$ Å, and $l_2=13.725$ Å. We see that there is very good agreement between the theoretical fit and the data points. For these values of the parameters, and for $H=0.5$ tesla, $gl_1^2/\lambda_1\approx 11$. Therefore we see that indeed $gl_1^2 \gg \lambda_1$, for fields greater than 0.5 tesla, and thus the approximations made in obtaining Eq. (22) were justified.

We have thus shown that the inclusion of NSC layers in the free energy produced predictions that explain all the features of the experimental data. We should note that the nonsuperconducting order parameter is very quickly suppressed far from T_c. For example, at 1 Tesla, $\theta=0$, we have $\phi/\psi=0.004$, as can be deduced from Eq. (12). That is why the curvature dies away so quickly. The region of linear H_{c2} is also the region of zero nonsuperconducting order parameters (see Fig. 2). The linear regions in H_{c2} are described by Eqs. (13) and (22), which are the key analytic results of this section.

IV. $La_{2-x}Ba_xCuO_{4-y}$

In this section we calculate $H_{c2}(T)$ in $La_{2-x}Ba_xCuO_{4-y}$. No dc-magnetization measurements have been performed on single crystals of this material yet, thus we shall not be able to determine the numerical values of the parameters of our model.

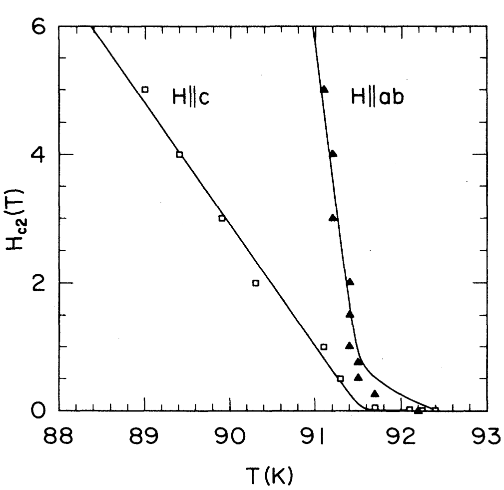

FIG. 1. Fit of our model to the data of Ref. 4 for $\lambda_1=0.0098$, $\lambda_2=1.07$, $l_1=400$ Å, $l_2=13.725$ Å.

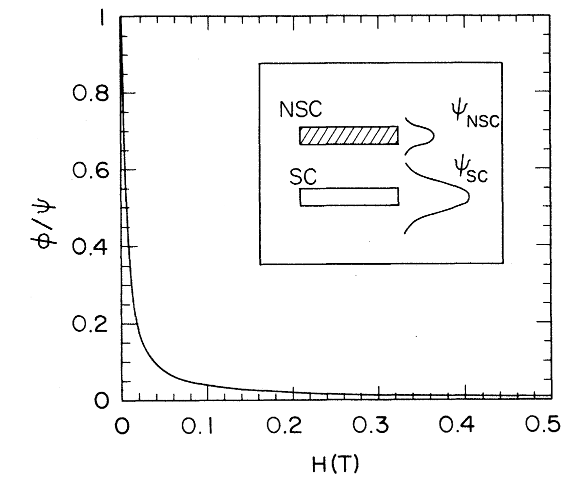

FIG. 2. Ratio of the nonsuperconducting to the superconducting order parameter for small fields parallel to the c axis, according to our model. The inset shows schematically the relationship between the superconducting order parameter on the SC layer and the order parameter induced by the proximity effect on the NSC layer.

In $La_{2-x}Ba_xCuO_{4-y}$, the superconducting CuO_2 layers are those associated with the subscript 1, while the nonsuperconducting LaO layers are associated with the subscript 2. We chose this convention here, in contrast to the convention used in Sec. III, because every *two* nonsuperconducting LaO layers are followed by *one* superconducting CuO_2 layer. In the previous case of $YBa_2Cu_3O_{7-\delta}$, *two* superconducting Cu(2) layers are followed by *one* nonsuperconducting Cu(1) layer. In this latter case, the subscript 2 had been associated with the superconducting layers, while here it will denote the nonsuperconducting layers.

We assume therefore that $a_1(T)=a_1(T-T_0)/T_c$ and $a_2(T)=a_2$, where T_0 is some phenomenological temperature, and a_1,a_2 are positive constants. Note that we may not neglect the LaO layers here, since they, and only they, will be responsible for any positive curvature that might exist.

We can rewrite the effective functional of Eq. (7) in terms of $\lambda_1=\eta_1^2/a_1(a_2+\eta_1)$, $\lambda_2=\eta_2\lambda_1/(a_2+\eta_1)$, $l_1^2=\hbar^2/2m_1^*a_1$, $l_2^2=\lambda_1\hbar^2/2m_2^*(a_2+\eta_1)$ and T_c, by using the rescaled order parameters $\phi'=a_1\phi$ and $\psi'=\eta_1\psi/\lambda_1$, and noting that $T_0/T_c=1+(2\eta_1/a_1)-2\lambda_1$. Then our effective functional becomes

$$G_{\text{eff}}=\int dx\,\{(T/T_c-1)\phi^2+2\lambda_1|\phi-\psi e^{id_1y\sin\theta}|^2$$
$$+\lambda_2|\psi-\psi^* e^{id_2y\sin\theta}|^2$$
$$+l_1^2[y^2\cos^2\theta\phi^2+(\partial\phi/\partial x)^2]$$
$$+2l_2^2(y^2\cos^2\theta|\psi|^2+|\partial\psi/\partial x|^2)\}\ , \quad (23)$$

where we dropped the primes, for brevity. This is our basic expression for the free energy of $La_{2-x}Ba_xCuO_{4-y}$, and all our subsequent work will be based on it. Remember that ψ and ϕ are the order parameters for the NSC and SC layers, respectively. There appear again four parameters only: λ_1 (the Josephson coupling between neighboring LaO and CuO_2 layers), λ_2 (the Josephson coupling between neighboring LaO layers), l_1 and l_2 (the phenomenological lengths associated with the CuO_2 and LaO layers). These are parameters that will be determined by fitting our H_{c2} predictions to the experimental data.

Let us begin by finding the upper critical field $H_{c2}^{\perp}$ parallel to the c axis ($\theta=0$), in which case both ψ and ϕ are real. Using the techniques of Sec. III, we get the following *exact* result for H_{c2}:

$$1-T/T_c=h_1+2\lambda_1 h_2/(\lambda_1+h_2)\ , \tag{24}$$

where again $h_n=2eHl_n^2/\hbar c$ ($n=1,2$). Note that $T\to\infty$ as h_2 approaches $-\lambda_1$ from above. Thus $H_{c2}(T)$ has a horizontal asymptote at some negative H, and it becomes a straight line with negative slope as $H\to\infty$. Consequently it must have positive curvature between these two limits. Note that as $H\to\infty$, this equation becomes

$$1-T/T_c\approx h_1+2\lambda_1\ . \tag{25}$$

Therefore we predict that at large fields $H_{c2}^{\perp}$ becomes a straight line that intercepts the T axis at $(1-2\lambda_1)T_c$, while it has positive curvature near T_c. Furthermore, the linear regime of $H_{c2}^{\perp}$ is precisely the regime where ψ/ϕ has become 0 , i.e., the region where the proximity effect we advocate has vanished, the order parameter on the NSC layers having become zero.

We now proceed to examine the case of a general field direction. A perturbative treatment similar to that of Sec. III yields, for small λ_1 and λ_2,

$$1-T/T_c\approx 2\lambda_1+h_1\cos\theta\ . \tag{26}$$

This is obviously valid only at large fields, in which case it agrees with the approximate limit for $\theta=0$ [see Eq. (25)].

We shall evaluate H_{c2} more generally by using a variational method once again. We adopt the ansatz $\psi(x)=be^{-\gamma x^2/2}$, $\phi(x)=e^{-\gamma x^2/2}$. This will be good only if the order parameter remains confined. Therefore it will not be good right at $\theta=90°$, but we hope that the region where it breaks down is within at most one degree from $\theta=90°$. We evaluate the $G_{\rm eff}$ of Eq. (23), using this ansatz, and set $G_{\rm eff}=0$. This equation gives $T_c(H)$, after maximization with respect to b and γ.

The evaluation of $G_{\rm eff}$ and the maximization with respect to b can be performed analytically, giving $T_c(H)$ as a function of γ,

$$1-T/T_c=2\lambda_1+2gl_1^2-2\lambda_1^2e^{-2d_1^2f/d_2^2}/(\lambda_1+\lambda_2+2gl_2^2-\lambda_2e^{-f})\ , \tag{27}$$

where g and f are defined exactly as in Eqs. (18) and (19). Minimization of the right-hand side with respect to γ yields the final answer. This can be done numerically. We can, however, obtain analytic results in some limits.

For $H=0$, $g=0$, $\gamma=0$, $f=0$, and $T=T_c$, as expected. In $La_{2-x}Ba_xCuO_{4-y}$, just as in $YBa_2Cu_3O_{7-\delta}$, f is very small (at most 0.012), and *if* λ_1 is as small as it seems to be in the case of $YBa_2Cu_3O_{7-\delta}$, then we may neglect the last term of Eq. (27), at least for sufficiently high fields (such that $2gl_2^2\gg\lambda_1$).

Then the maximization of the rhs of Eq. (27) leads trivially to our final result,

$$1-T/T_c\approx 2\lambda_1+h_1\cos\theta\ . \tag{28}$$

This is precisely the same result as the one obtained perturbatively in Eq. (26).

We conclude then that $H_{c2}(T)$ has positive curvature in $La_{2-x}Ba_xCuO_{4-y}$ as well, but it becomes a straight line at sufficiently high fields, intersecting the T axis below T_c. This intercept is the same for *all* field orientations. The angular dependence of the linear part of H_{c2} is once again of the GL form $(\cos^2\theta+\epsilon\sin^2\theta)^{1/2}$, but the anisotropy factor ϵ here is of order λ_1^2, compared with an anisotropy factor of 0.0325 in $YBa_2Cu_3O_{7-\delta}$. Therefore $La_{2-x}Ba_xCuO_{4-y}$ may be more anisotropic than $YBa_2Cu_3O_{7-\delta}$, if its λ_1 is comparable to the λ_1 of $YBa_2Cu_3O_{7-\delta}$.

If no other layers but CuO_2 layers contribute to the conductivity, there will be no dynamicaly independent order parameters on NSC layers. Even in this case, there could be a small positive curvature in H_{c2} due to some slight inequivalence of the two CuO_2 layers in the unit cell (see Schneider, Ref. 15).

V. CONCLUSION

We have presented a physical picture based on a proximity effect that gives rise to the generic positive curvature observed in layered superconductors. Due to their proximity with superconducting layers, nonsuperconducting layers acquire a nonzero order parameter near T_c, and a positive curvature appears. The curvature vanishes further away from T_c, when the order parameter on the nonsuperconducting layers has become effectively zero. In that regime of sufficiently high fields, $H_{c2}(T)$ is a straight line intersecting the T axis at the same point below T_c for *all* orientations of the field. We have been mostly concerned with high-T_c copper oxide superconductors and have found our predictions to be in very good agreement with the experimental results of Ref. 4. We find that the angular dependence of the linear part of H_{c2} is given by the GL anisotropic form $(\cos^2\theta+\epsilon\sin^2\theta)^{1/2}$, where θ is the angle between **H** and the c axis. The anisotropy factor is almost zero for $La_{2-x}Ba_xCuO_{4-y}$ while it is 0.0325 for $YBa_2Cu_3O_{7-\delta}$.

We should note that a really thorough treatment of $YBa_2Cu_3O_{7-\delta}$ should have included the BaO and Y layers. However, we have obtained agreement with the data even in the context of our "truncated" plane-chain model. Thus most of the curvature is presumably due to the inclusion of the Cu(1) chains in the free energy, the other

NSC layers being less important. Of course, in $La_{2-x}Ba_xCuO_{4-y}$ the LaO layers are all important, since there is no curvature if they are neglected. Our mechanism is universal to all layered superconducting systems that have NSC layers in the proximity of SC layers, and is thus applicable to the low-T_c layered superconductors.[6]

An alternative mechanism has been proposed recently[21] for the positive curvature in $YBa_2Cu_3O_{7-\delta}$, in which the curvature of $H_{c2}(T)$ can be interpreted as evidence for *d*-wave pairing or for a mixture of *s* and *d* wave pairing. This work uses six fitting parameters, compared to our four parameters, and it is applicable to high-T_c superconductors only, while our mechanism is generic to *all* layered superconductors. The case of arbitrary field orientation is not examined in Ref. 21. It would be interesting to see if their interpretation agrees with the experimental results of Ref. 5. It would be also very interesting if dc-magnetization measurements were performed on $La_{2-x}Ba_xCuO_{4-y}$ to determine the existence of positive curvature there. If the LaO layers are insignificant, then there will be none. Otherwise, H_{c2} will be again a straight line at sufficiently high fields, that intersects the T axis at about $0.98T_c$, for *all* field orientations, assuming $\lambda_1 \sim 0.01$.

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