High- T_c superconductivity of YBa₂Cu₃O₇/PrBa₂Cu₃O₇ superlattices: An interlayer-coupling model

A. K. Rajagopal

Naval Research Laboratory, Washington, D.C. 20375-5000

S. D. Mahanti

Department of Physics and Astronomy and Center for Materials Research, Michigan State University, East Lansing, Michigan 48824 (Reserved 15 October 1000) arrived measuremint received 10 February 1001)

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A simple model of intralayer and interlayer couplings based on a generalized BCS pairing theory of superlattices of layered superconductors is used to correlate the recently observed trends in the T_c 's of YBa₂Cu₃O₇/PrBa₂Cu₃O₇ superlattices. These observed trends in the T_c 's are obtained in this model if certain inequalitities are satisfied among the direct and that mediated by the PrBa₂Cu₃O₇ intercell-interlayer, intracell-interlayer couplings. These inequalitities among the couplings may be reconciled in terms of the chemical structure of these systems.

Almost all high- T_c superconductors with the exception of $(Ba_{1-x}K_x)BiO_3$ have layered crystal structure and possess electronic and magnetic anistropy.¹ The active building blocks of these materials, as far as superconductivity is concerned, are CuO₂ layers with strong intralayer and weak interlayer interactions. A large number of theoretical papers²⁻⁷ have been written addressing this question of anisotropic coupling between the layers and its effect on the chemical structure dependence of the superconducting transition temperature T_c , and the gap anisotropy.

A fundamental question about these layered superconductors is whether a single CuO_2 layer is in fact superconducting or whether one needs interlayer coupling for driving the system to a superconducting state.² To address this question and to see how T_c changes with crystal structure, i.e., number of CuO₂ layers per unit cell, several groups have $^{8-10}$ recently reported the fabrication, structure, and superconducting properties of epitaxial, nonsymmetric $M \times N$ superlattices of M YBa₂Cu₃O₇ multilayers of M = 1, 2, 3, 4, and 8 unit cells thick separated by insulating $PrBa_2Cu_3O_7$ multilayers of N unit cells with N ranging from 1 to 16. Unfortunately it has not been possible to isolate a single CuO₂ layer (with finite hole density) separated by sufficiently large number of insulated layers. The system that comes closest is CuO₂ bilayers, i.e., one unit cell of YBa₂Cu₃O₇ separated by several (maximum 16) insulating PrBa₂Cu₃O₇ layers. These experiments have shown that the superconducting transition temperatures $T_c(M,N)$ of these systems, depend on the YBa₂Cu₃O₇ layer thickness and the interlayer separation. Figure 1 displays the experimental findings. The following inequalities summarize these observed trends:

(i)
$$T_c(M,N) > T_c(M,N+1)$$

 $(M = 1,2,...;N = 0,1,2,...),$ (1)

(ii)
$$T_c(M+1,N) \ge T_c(M,N) \ge T_c(M-1,N)$$

 $(M=1,2,\ldots;N=0,1,2\ldots)$, (2)

(iii)
$$T_c(M,N) - T_c(M,N+1) > T_c(M+1,N)$$

$$-T_{c}(M+1,N+1)$$
 (3a)

as well as

$$T_c(1,1)/T_c(1,\infty) > T_c(2,1)/T_c(2,\infty)$$
 (3b)

The equality sign in Eq. (2) holds for N = 0.

One interesting observation (see Fig. 1) is that a single $YBa_2Cu_3O_7$ unit cell (BaO-CuO₂-Y-CuO₂-BaO-CuO)



FIG. 1. Experimental superconducting transition temperatures of an $M \times N$ supercell of YBa₂Cu₃O₇/PrBa₂Cu₃O₇ taken from Ref. 3.

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separated by sixteen $PrBa_2Cu_3O_7$ unit cells has $T_c = 20$ K whereas two $YBa_2Cu_3O_7$ unit cells again separated by the same number of $PrBa_2Cu_3O_7$ unit cells has $T_c = 60$ K. This raises some interesting questions regarding the nature of the coupling between two CuO_2 layers which are in the same unit cell and those belonging to different unit cells, questions which we address in this paper.

There have been several attempts to correlate the T_c 's of a large number of high- T_c systems in terms of a generalized BCS pairing theory of layered superconductors,^{3-7,11} by invoking interlayer and intralayer couplings. The purpose of the present work is to adopt the theory in Refs. 5 and 6, to develop a model of superconductivity of superlattices of layered superconductors of the type YBa₂Cu₃O₇/PrBa₂Cu₃O₇ investigated recently. This system has the significant feature that PrBa₂Cu₃O₇.

The critical temperature T_c with Cooper pairing interactions among carriers within the conduction layers is determined by the maximum value of T_c obtained from the solution of the determinantal equation^{3,5,6}

$$\det |I_J(T_c)\delta_{J,J'} + \lambda_{J,J'}| = 0.$$
⁽⁴⁾

Here

$$I_J^{-1}(T_c) \cong \ln(\Theta_J^*/T_c) \tag{5}$$

with Θ_I^* a cut-off parameter for the Jth layer and $\lambda_{II'}$ are the dimensionless generalized Cooper pair coupling parameters associated with the planes. The structure of the determinant depends on the nature of the layers in the superlattice reflecting the features of the YBa₂Cu₃O₇/PrBa₂Cu₃O₇ system. We employ a nearest layer interaction model. We take two Cu-O₂ layers of YBa₂Cu₃O₇ as the relevant conducting planes and call them as a unit -(AB)-. The PrBa₂Cu₃O₇ layer although nonsuperconducting by itself is however taken as an important layer denoted by -C. We introduce an intralayer coupling strength $\lambda_{AA} = \lambda_{BB} = -\lambda$ for YBa₂Cu₃O₇ with $x = I(T_c) - \lambda$, and $[\lambda_{cc} + I_c(T_c)]$ is taken to be a positive parameter, z, because an isolated PrBa₂Cu₃O₇ layer is nonsuperconducting. The supercell of $[(AB)_M - C_N]$ then has further coupling parameters associated with interlayer interaction; intracell interaction between A and B is taken to be λ' , intercell interaction between A and B is taken to be λ'' , and the interaction between A and C or C and B is taken as ω , and the interlayer interaction between two C's is taken to be ω' . It turns out in the analysis of T_c , one obtains an indirect



FIG. 2. Schematic Diagram of the $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattice, intercell- and intracell- and interlayer- and intralayer-coupling parameters.

intercell interaction between A and B via C denoted by $\lambda^* = \omega^2/z$. It is convenient to introduce a dimensionless parameter $\gamma = \omega'/z$. In Fig. 2, we exhibit these couplings pictorially to facilitate visualizing the various couplings in the supercell.

Before discussing the solutions of Eqs. (4) and (5) we would like to make a digression and point out how the interlayer intracell coupling λ' and interlayer intercell coupling λ'' can depend on how the unit cells grow on different substrates. From the geometrical arrangement of CuO₂ layers we can easily see that there are basically two types of interlayer coupling. One is between two CuO_2 layers separated by Y layer (denoted by λ_Y) and the other separated by BaO-CuO-BaO layers (denoted by λ_{CuO}). Clearly λ_{Y} and λ_{CuO} are different, the latter presumably larger than the former due to the presence of the bridging oxygens. Also which of these two will be λ' and λ'' will depend upon how the unit cells grow on the substrate. For example, for the $(YBa_2Cu_3O_7)_2(PrBa_2Cu_3O_7)_{16}$ system if the arrangement of (YBa₂Cu₃O₇)₂ is (BaO-CuO₂-Y-CuO₂-BaO-CuO;BaO-CuO₂-Y-CuO₂-BaO-CuO) then $\lambda' = \lambda_{Y}$ and $\lambda'' = \lambda_{CuO}$. If, on the other hand, the arrangement is (CuO₂-BaO-CuO-BaO-CuO₂-Y;CuO₂-BaO-CuO-BaO-CuO₂-Y), then λ' $=\lambda_{CuO}$ and $\lambda''=\lambda_{Y}$. Thus we need to find out how T_c depends on λ' and λ'' for the multilayer systems which we discuss next.

The structure of the determinant, Eq. (4), for the $..[(AB)_M - (C)_N].$ supercell containing R units of $[(AB)_M - (C)_N]$ is of order $(2M+N)R \times (2M+N)R$, where

(6)

$$D_{(2M+N)R}(M,N) = \det \begin{bmatrix} D_{2M+N} & \Omega_{2M+N} & 0_{2M+N} & \dots & 0_{2M+N} \\ \Omega_{2M+N}^{\dagger} & D_{2M+N} & \Omega_{2M+N} & \dots & 0_{2M+N} \\ 0_{2M+N} & \cdots & \Omega_{2M+N}^{\dagger} & \dots & D_{2M+N} \end{bmatrix}$$

where 0_{2M+N} is the $(2M+N) \times (2M+N)$ null matrix, D_{2M+N} is a square matrix with the structure

$$D_{2M+N} = \begin{bmatrix} A_{2M,2M} & \Omega_{N,2M} \\ \Omega^{\dagger}_{2M,N} & C_{N,N} \end{bmatrix}, \qquad (7)$$

and Ω_{2M+N} and Ω_{2M+N}^{\dagger} are the intersupercell coupling matrix of the form

$$\Omega_{2M+N} = \begin{bmatrix} 0 & \dots & 0 \\ \vdots & \ddots & \vdots \\ \omega & \dots & 0 \end{bmatrix},$$

$$\Omega_{2M+N}^{\dagger} = \begin{bmatrix} 0 & \dots & \omega \\ \vdots & \ddots & \vdots \\ 0 & \dots & 0 \end{bmatrix}.$$
(8)

In Eq. (7) D_{2M+N} is a square matrix which is partitioned into a $2M \times 2M$ square matrix, $A_{2M,2M}$, representing the $(AB)_M$ complex and $N \times N$ square matrix $C_{N,N}$ representing the interactions in the (C_N) complex a rectangular matrix of order $N \times 2M$, $\Omega_{N,2M}$ and another rectangular matrix of order $N \times 2M$, $\Omega_{2M,N}^{-1}$ representing the coupling between $(AB)_M$ and $(C)_N$ complex, given by

$$A_{2M,2M} = \begin{cases} x \quad \lambda' \quad 0 \quad 0 \quad \dots \quad 0 \\ \lambda' \quad x \quad \lambda'' \quad 0 \quad \dots \quad 0 \\ \ddots & & \ddots & \ddots \\ 0 \quad \dots \quad & \lambda' \quad x \end{cases} , \qquad (9)$$

$$\Omega_{N,2M} = \begin{bmatrix} 0 & \dots & 0 \\ \ddots & \ddots & \vdots \\ 0 & \dots & 0 \end{bmatrix} , \qquad (10)$$

$$\Omega_{2M,N}^{\dagger} = \begin{bmatrix} 0 & \dots & \omega \\ \vdots & \ddots & \vdots \\ 0 & \dots & 0 \end{bmatrix} , \qquad (10)$$

$$C_{N,N} = \begin{bmatrix} z \quad \omega' \quad 0 \quad 0 \quad \dots \quad 0 \\ \omega' \quad z \quad \omega' \quad 0 & \ddots & 0 \\ 0 \quad \omega' \quad z \quad \omega' & 0 & \ddots \\ 0 \quad \omega' \quad z \quad \omega' & 0 & \ddots \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 \quad \ddots \quad \ddots \quad \omega' \quad z \end{bmatrix} . \qquad (11)$$

We have analytically computed the T_c 's for the simple supercells given in Table I in the limit $R \to \infty$, which represents the system under investigation. In this model, the bulk pure YBa₂Cu₃O₇ result is obtained for N = 0, for all M. When there are a large number of PrBa₂Cu₃O₇'s [or $(C_N), N \to \infty$] we expect the single supercell of $-(AB)_M$ to become isolated in which case the solution is also relatively simple, because we only need to obtain the zeros of the determinant of $A_{2M,2M}$. The general case is a little more complicated and so we will here illustrate the result for a representative case of $\cdots [(AB)_1 - (C)_2] \cdots$. In Table I, $T_c(N,M)$'s are given in terms of the standard effective coupling strength $\lambda_{\text{eff}}(M,N)$ via

$$\{\ln[(\Theta^*/T_c(M,N))]\}^{-1} = \lambda_{\text{eff}}(M,N) .$$
(12)

Here Θ^* is an appropriate cutoff energy parameter, unspecified in the present sequel.

In the pure bulk YBa₂Cu₃O₇ case, we have $D_{2MR}(M,0)$ in place of Eq. (6) given by a simple expression

$$D_{2MR}(M,0) = \begin{vmatrix} x & \lambda' & 0 & 0 & \dots & 0 \\ \lambda' & x & \lambda'' & 0 & 0 \\ 0 & \lambda'' & x & \lambda' & \\ \vdots & & & \ddots & \vdots \\ 0 & \cdot & \cdot & \cdot & \lambda' & x \end{vmatrix} .$$
(13)

It is easy to evaluate this determinant by construction a pair of simple difference equations by direct expansion of the determinant:

$$D_{2MR} = x U_{2MR-1} - \lambda'^2 D_{2MR-2} , \qquad (14a)$$

$$U_{2MR-1} = x D_{2MR-2} - \lambda'^2 U_{2MR-3} .$$
 (14b)

We then seek a solution of the form

$$D_{2MR} \sim \alpha^{MR} , \qquad (15)$$

where α is to be determined selfconsistenly. We find then

$$\alpha^2 + (\lambda'^2 + \lambda''^2 - x^2)\alpha + \lambda'^2\lambda''^2 = 0.$$
 (16)

The solution is then written in terms of the two roots α_{\pm} of Eq. (16) with the conditions that $D_0=1$ and $D_2=(x^2-\lambda'^2)$ and so

$$D_{2MR} = \left[\frac{\lambda^{\prime\prime}}{\lambda^{\prime}} \frac{\sin(2MR\theta)}{\sin(2\theta)} + \frac{\sin[2(MR+1)\theta]}{\sin(2\theta)}\right] (\lambda^{\prime}\lambda^{\prime\prime})^{MR},$$
(17)

where 2θ is defined by

$$2\lambda'\lambda''\cos(2\theta) = x^2 - \lambda'^2 - \lambda''^2$$
(18)

from which it follows that

$$\lambda_{\text{eff}}(M,0) = \lambda + [\lambda'^2 + \lambda''^2 + 2\lambda'\lambda''\cos(2\theta_c)]^{1/2} , \qquad (19)$$

where $2\theta_c$ is determine by the condition that $D_{2MR} = 0$ for $R \to \infty$. From (17), it follows that $(MR + 1) \to MR$ for large R and so

$$2\theta_c \rightarrow \frac{\pi}{MR} \rightarrow 0$$

and thus

$$\lambda_{\text{eff}}(M,0) = \lambda + \lambda' + \lambda'' \tag{20}$$

independent of M, as expected.

The case of infinite number of $PrBa_2Cu_3O_7$ and finite number of $YBa_2Cu_3O_7$ layers is equivalent to an isolated stack of *M* layers of -(AB)-. The λ_{eff} is contained in

TABLE I. Model of $M \times N$ superlattices where M is the number of YBa₂Cu₃O₇ (AB) and N is the number of PrBa₂Cu₃O₇ (C) layers in one supercell. The transition temperature $T_c(M,N)$ is related to the effective coupling constant $\lambda_{\text{eff}}(M,N)$ by $\lambda_{\text{eff}}(M,N) = [\ln(\theta^*/T_c(M,N))]^{-1}$. λ is the intralayer coupling; in the superconducting (A and B) layer, λ' is the effective coupling between A and B mediated by a single nonsuperconducting (C) layer and the dimensionless parameter γ is a measure of the C-C coupling. See text for details.

| (M , N) | Superlattice with interlayer coupling specified | $\lambda_{	eff}$ |
|--------------------------------|--|--|
| (1,0) | $[\cdot(AB)\cdot][\cdot(AB)\cdot]$ | $\lambda + \lambda' + \lambda''$ |
| (1,1) | $[\cdot(AB)\cdot(C)_1\cdot][\cdot(AB)\cdot(C)_1\cdot]$ | $\lambda + \lambda' + 2\lambda^*$ |
| (1,2) | $[\cdot(AB)\cdot(C)_2\cdot][\cdot(AB)\cdot(C)_2\cdot]$ | $\lambda + \lambda' + \lambda^* (1 - \gamma)$ |
| (1,3) | $[\cdot(AB)\cdot(C)_{3}\cdot][\cdot(AB)\cdot(C)_{3}\cdot]$ | $\lambda + \lambda' + \lambda^* / (1 - 2\gamma^2)$ |
| (1,4) | $[\cdot(AB)\cdot(C)_4\cdot][\cdot(AB)\cdot(C)_4\cdot]$ | $\lambda + \lambda' + \lambda^* (1 - Y) (1 - \gamma - \gamma^2)$ |
| Phys. Rev. Lett. $(1, \infty)$ | $[\cdot (AB) \cdot]$ | $\lambda + \lambda'$ |
| (2,0) | $[\cdot(\widehat{AB})_2\cdot][\cdot(\widehat{AB})_2\cdot]$ | $\lambda + \lambda' + \lambda''$ |
| (2,1) | $[\cdot(AB)_{2}\cdot(C)\cdot][\cdot(AB)_{2}\cdot(C)\cdot]$ | $\lambda + \lambda^{\prime\prime}/2 + \lambda^*$ + $[\lambda^{\prime 2} + (\lambda^{\prime\prime} - 2\lambda^*)^2/4]^{1/2}$ |
| (2,2) | $[\cdot(AB)_2\cdot(C)_2\cdot][\cdot(AB)_2\cdot(C)_2\cdot]$ | $\lambda + \lambda''/2 + \lambda^*/2(1-\gamma) + [\lambda'^2 + \{\lambda''-\lambda^*/(1-\gamma)\}^2/4]^{1/2}$ |
| (2,∞) | $[\cdot (AB)_2 \cdot]$ | $ \lambda + [\lambda'^2 + \lambda''^2/2 \\ + \lambda''(4\lambda'^2 + \lambda''^2)^{1/2}/2]^{1/2} $ |

the above analysis by putting R = 1 in which case we have

$$\lambda_{\text{eff}}(\boldsymbol{M}, \infty) = \lambda + [\lambda'^2 + \lambda''^2 + 2\lambda'\lambda'' \cos[2\theta_c(\boldsymbol{M}, \infty)]^{1/2}, \quad (21)$$

where $2\theta_c(M, \infty)$ is determined from the condition $D_{2M} = 0$ or

$$\frac{\lambda''}{\lambda'}\sin[2M\theta_c(M,\infty)] + \sin[2(M+1)\theta_c(M,\infty)] = 0.$$
 (22)

Clearly this depends on the number of $YBa_2Cu_3O_7$ layers. For M = 1,2 the explicit answers are given in Table I.

From Table I, it is interesting to note that the introduction of an intervening C layer leads to an indirect interplanar coupling λ^* between A and B and subsequent addition of C layers renormalize λ^* by a "propagator" parameterized by γ which depends on the number of C layers added. Clearly an infinite YBa₂Cu₃O₇ does not care what we define as intercell interlayer coupling λ' and intracell interlayer coupling λ'' since we can always redefine our unit cell. Thus $\lambda_{\text{eff}}(M,0)$ is symmetric under the interchange of λ' and λ'' . Also $\lambda_{\text{eff}}(M,0)$ is independent of M because if we take M unit cells of YBa₂Cu₃O₇ and repeat in space we get the same thing as one unit cell of YBa₂Cu₃O₇ which repeats in space.

To further illustrate the procedure, we consider here in detail the calculation of $T_c(1,2)$. The determinant $D_{4R}(1,2)$ is expanded to obtain a set of 4R coupled difference equations of the form:

$$D_{4R} = x U_{4R-1} - \lambda'^2 V_{4R-2} , \qquad (23)$$

$$U_{4R-1} = x V_{4R-2} - \omega^2 W_{4R-3} , \qquad (24)$$

$$V_{4R-2} = x W_{4R-3} - \omega'^2 D_{4R-4} , \qquad (25)$$

$$W_{4R-3} = z D_{4R-4} - \omega^2 V_{4R-5} . (26)$$

We seek a solution of the form

$$D_{4R} \sim \alpha^R , \qquad (27)$$

where α is determined self-consistently by solving the above set of difference equations. In the present case α obeys a quadratic equation

$$\alpha^{2} + \alpha [x^{2}(\omega^{\prime 2} - z^{2}) + 2xz\omega^{2} - \omega^{\prime 2}\lambda^{\prime 2} + z^{2}\lambda^{\prime 2} - \omega^{4}] + \omega^{4}\omega^{\prime 2}\lambda^{\prime 2} = 0.$$
(28)

Following the same procedure as before we obtain

$$\lambda_{\text{eff}}(1,2) = \lambda + \lambda' + \lambda^* / (1-\gamma) , \qquad (29)$$

where

$$\lambda^* = \omega^2 / z \text{ and } \gamma = \omega' / z$$
 (30)

By employing similar procedures, the results in Table I were derived.

To understand the observed trends in T_c in these superlattice systems, Eqs. (1)-(3), we deduce several inequalities among $\lambda_{\text{eff}}(M,N)$. These in turn imply certain constraints on the basic inter- and intralayer-coupling constants introduced in the model:

$$\lambda_{\text{eff}}(1,N) > \lambda_{\text{eff}}(1,N+1) > \lambda_{\text{eff}}(1,N+2) > \cdots$$
(31)

$$if \quad \lambda^{\prime\prime} > 2\lambda^* \tag{32}$$

and
$$0 < \lambda < 1/2$$
. (33)

From Eq. (12) this implies the inequalities for T_c 's in Eqs. (1) and (2). It is also found that

$$\lambda_{\text{eff}}(1,1) - \lambda_{\text{eff}}(1,\infty) > \lambda_{\text{eff}}(2,1) - \lambda_{\text{eff}}(2,\infty)$$
(34)

provided

$$\lambda' < (\lambda'' - 2\lambda^*)/2 . \tag{35}$$

This may be seen to be consistent with the observed inequalities given in Eqs. (3a) and (3b).

We will now give some physical significance to the inequalities among the basic coupling strengths, $\lambda', \lambda'', \lambda^*$, and γ given in Eqs. (32), (33), and (35). Equation (33) simply implies that the coupling between the insulating PrBa₂Cu₃O₇ layers should be expected to be weak. From these inequalities, it is found that the dominant superconducting coupling is λ'' . This is the intercell interlayer AB coupling. Equation (32) is easily understood by the observation that λ^* is the PrBa₂Cu₃O₇ mediated effective coupling between A and B. From Eqs. (32) and (35), we see that the intracell interlayer coupling λ' must be smaller than intercell interlayer coupling λ'' . This may seem puzzling. However if we identify the A and B layers in a given cell as being separated by an Yttrium layer and associated λ' with this interaction then it seems reasonable that $\lambda'(=\lambda_{\rm Y})$ is smaller than the intercell coupling λ'' $(=\lambda_{CuO})$ mediated by the Cu-O chains, BaO networks, etc. Since these superlattices are deposited on SrTiO₃ or MgO substrates, it appears chemically advantageous to form the first unit cell of YBa₂Cu₃O₇ with the following ordering: SrO-CuO₂-Y-CuO₂-BaO-CuO- \cdots provided of course the surface of the substrate is a SrO or MgO layer. This argument also suggests that if the YBa₂Cu₃O₇ layers start growing on the TiO₂ layer of SrTiO₃, one might get a stacking of the type TiO₂-BaO-CuO-BaO-CuO₂-..., in which case one would have obtained $\lambda' = \lambda_{CuO} > \lambda'' = \lambda_{Y}$.

In summary, we have developed in this paper a model for a qualitative understanding of the superconducting transition temperature of $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattice systems, based on the relative strengths of the interlayer couplings between superconducting (A and B) and nonsuperconducting (C) layers.

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