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Origin of the variation of T_c with superconducting layer thickness and separation in $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_7$ superlattices

R. G. Goodrich and P. W. Adams

Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001

D. H. Lowndes and D. P. Norton

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6056

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We have investigated the superconducting (SC) properties of ultra-thin $YBa_2Cu_3O_{7-x}$ layers in $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_7$ superlattices in which the coupling between adjacent $YBa_2Cu_3O_{7-x}$ layers is modulated by varying thicknesses of insulating $PrBa_2Cu_3O_7$. In particular, we have formulated a phenomenological model that accounts for the dependence of the $YBa_2Cu_3O_{7-x}$ transition temperature on both the thickness of and the separation between $YBa_2Cu_3O_{7-x}$ layers. The results of our analysis are compared with previous studies of conventional low-T_c thin film systems. [S0163-1829(97)51446-9]

It is now well known that the superconducting transition temperature, T_c, of thin amorphous, elemental films (conventional low-T_c superconductors) is suppressed as the film thickness is decreased. However, recent measurements of the T_c of isolated c-axis oriented $YBa_2Cu_3O_{7-x}$ (YBCO) layers, separated by insulating layers of $PrBa_2Cu_3O_7$ in $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_7$ (YBCO/PrBCO) superlattices also show this effect.[1–3] In fact, as shown in Fig. 1, the transition temperature of a single unit-cell-thick YBCO layer is more than a factor of 4 lower than that of bulk YBCO (or a thick YBCO film).

The resistivity of PrBCO is high along the c direction and increases with decreasing temperature.[4] Thus, PrBCO can be used to isolate multiple unit-cell-thick YBCO layers in epitaxially grown superlattice structures. Consequently, YBCO/PrBCO superlattices provide an ideal system to study both the dependence of T_c on superconducting (SC) layer thickness *and* the effects of varying the coupling between closely spaced SC planes. We believe that this interlayer coupling is essentially a proximity effect that is mediated by tunneling through the insulating PrBCO layers. In what follows, we report a systematic analysis of the dependence of T_c on YBCO film thickness and the film separation distance in YBCO/PrBCO superlattice samples comprised of 10–15 superlattice periods. We first analyze the dependence of T_c on YBCO layer thickness for well isolated YBCO layers (large SC layer separations). Then we examine the variation of T_c with interlayer separation at constant SC layer thickness. In both analyses we compare our results to earlier measurements on conventional thin, amorphous low-T_c materials.

Previously the variation of T_c in YBCO/PrBCO superlattices was interpreted using a number of theoretical models that included mechanisms such as the contribution of carriers from PrBCO to the YBCO layers,[3] loss of interlayer coupling,[5] and changes in anisotropy.[6] In contrast, we give here a more straightforward interpretation of the dependence of T_c on SC layer thickness and interlayer coupling that is based in part on recent theoretical studies of the suppression of T_c in thin films.[7–9] We demonstrate that our phenomenological model can account for all of the behavior in Fig. 1 as well as similar behavior observed in low-T_c/insulator superlattices solely in terms of dimensionality and interlayer coupling.

Early work on ultrathin films of amorphous Pb (Refs. 10 and 11), Bi (Ref. 10), Nb (Ref. 12), Sn (Ref. 13), and MoC (Ref. 14) showed that T_c is suppressed from its thick film value by an amount that is inversely proportional to the film thickness. In these measurements film thicknesses varying from hundreds of atomic layers to about one unit-cell thick were studied. To explain the suppression of T_c in the thinnest films, Simonin[7] added a term to the Ginzburg-Landau free energy equation that forces the order parameter to go to zero at the upper and lower surfaces of the film. Other models

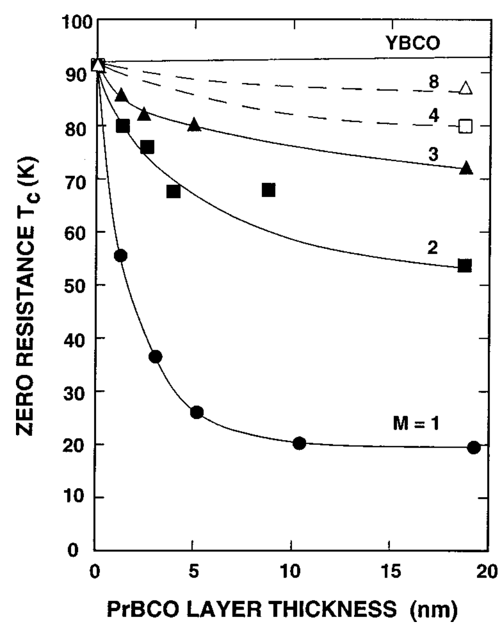

FIG. 1. Zero-resistance transition temperature as a function of PrBCO layer thickness for c-axis perpendicular YBCO/PrBCO superlattices with $M=1$, 2, 3, 4, and 8 cell-thick superconducting YBCO layers.

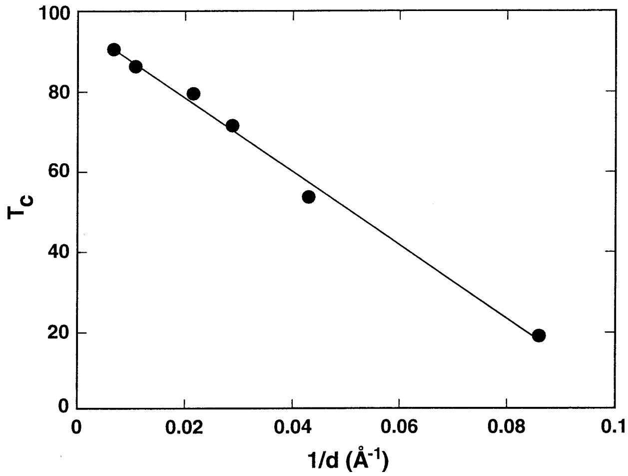

FIG. 2. Change in zero-resistance transition temperature, $\Delta T_c = T_c^{\text{bulk}} - T_c(d)$, of superconducting YBCO layers separated by 16 unit cells ($\sim$187 Å) of PrBCO, as a function of the reciprocal of the total YBCO layer thickness. The line is a linear fit to the data.

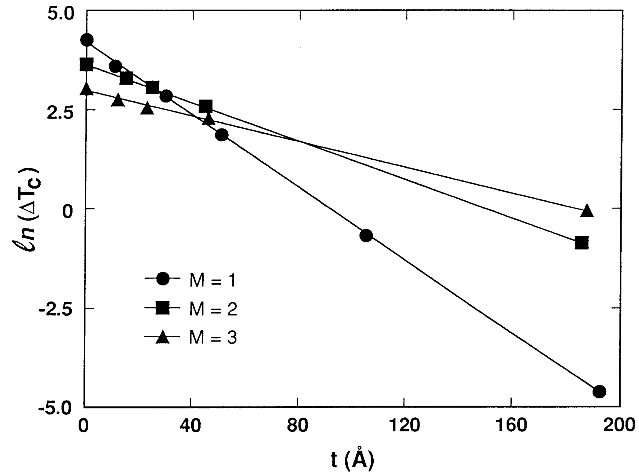

FIG. 3. Natural logarithm of $\Delta T_c = T_c(d,t) - T_c(d,t=\infty)$ for YBCO layers with thicknesses corresponding to $M=1$, 2, and 3 c-axis unit cells as a function of PrBCO separation layer thickness. The lines are linear fits to the data.

show that disorder induced electron correlation effects can lead to anomalous attenuation of the electronic density of states which in turn lowers T_c.[8,9,15] Notwithstanding these theoretical considerations, there is strong empirical evidence that T_c varies with film thickness d as,

$$T_c(d) = T_c^{\text{bulk}}[1-(d_m/d)], \tag{1}$$

where T_c^{bulk} is the bulk or thick film transition temperature of the material, and d_m is the critical film thickness below which superconductivity is lost, even at zero temperature. This critical thickness is related to the BCS total interaction potential, N_0V, where N_0 is the density of states at the Fermi energy and V is the constant Cooper pair interaction potential, by the relation $d_m = 2a/N_0V$, where a is the lattice parameter.[7]

Figure 2 shows the values of T_c for 1, 2, 3, 4, and 8 unit-cell-thick layers of YBCO separated by 16 c-axis unit cells ($\sim$187 Å) of PrBCO. At this separation the SC YBCO layers are essentially completely decoupled, as will be shown later. The transitions in the 1 unit-cell-thick samples were quite broad with 10% to 90% widths of order 30 K. Thicker layers of YBCO produced somewhat sharper transitions. For this reason we defined the transition temperature T_c by the onset of zero resistance. The data used in Fig. 2 were taken directly from the data shown in Fig. 1. After multiplying the YBCO's c-axis lattice constant of 11.7 Å and subtracting $T_c^{\text{bulk}} = 91$ K from each data point we obtain the results shown in Fig. 2 for the change in T_c, $\Delta T_c = T_c^{\text{bulk}} - T_c(d)$, as a function of $1/d$. The linear behavior in Fig. 2 demonstrates that Eq. (1) is valid for thin YBCO layers, and that YBCO behaves in essentially the same manner as conventional low-T_c superconductors when its thickness is varied. We point out that the $\Delta T_c(d) = 0$ intercept occurs at a film thickness of 145 Å which may be interpreted as the effective layer thickness of bulk YBCO. This value is in good agreement with the value of 155 Å determined independently by rf impedance measurements on thick-film YBCO samples.[16] The conventional behavior of the T_c of these YBCO layers as a function of their thickness strongly suggests that the observed T_c variation is a manifestation of the dimensionality of the YBCO and that more complicated explanations of this behavior, such as charged transfer effects,[3] are not required.

From a linear fit to the data in Fig. 2 we determine a value of $d_m = 10.1$ Å, using $T_c^{\text{bulk}} = 91$ K. This critical thickness is just slightly less than the thickness of one c-axis YBCO unit cell, but considerably greater than the thickness of the Cu-O bi-layer within the unit cell, and is consistent with the knowledge that a full unit cell of YBCO is required for superconductivity. From this critical thickness the bulk interaction potential, N_0V, is calculated to be 2.3 which is approximately seven times larger than the value inferred for Nb from similar measurements.[6] We point out that values of d_m, and

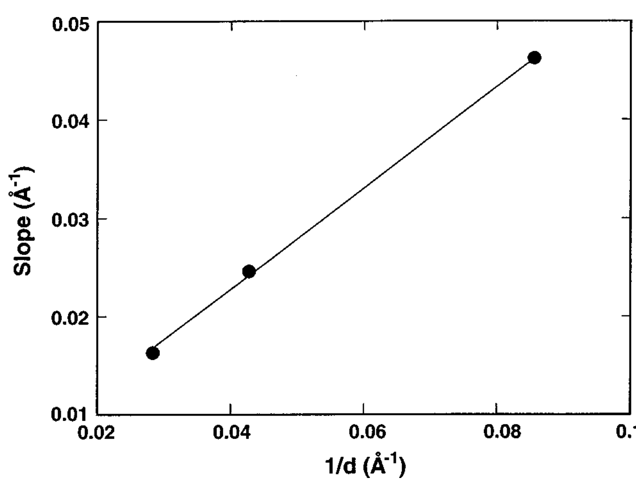

FIG. 4. Slopes of the linear fits in Fig. 3 as a function of the reciprocal of the distance between the superconducting layers. The line is a fit to the data.

hence N_0V, obtained in Pb in this manner[10] can vary by a factor of almost three depending on film deposition methods, or substrate materials.

From Fig. 1 it can also be seen that increasing the separation of constant-thickness YBCO layers also causes T_c to decrease. Thus in YBCO/PrBCO superlattices T_c is not only a function of d but also of PrBCO layer thickness, t. This observation suggests that adjacent YBCO layers interact through the PrBCO. To further test this hypothesis we have subtracted the extrapolated values of T_c at infinite separation, $T_c(d,t=\infty)$, from each of the measured critical temperatures for YBCO layers with thickness of $M=1$, 2, and 3 unit cells ($d=M\times 11.7$ Å). The resulting plot of the natural logarithm of the change in T_c, $\Delta T_c = T_c(d,t) - T_c(d,\infty)$, versus SC layer separation t is shown in Fig. 3 along with linear fits to each data set. The values of $T_c(d,\infty)$ were obtained by minimizing χ^2 in the linear fits and are less than 0.1 K below the transition temperatures measured in superlattice samples with the largest YBCO separations. Note that $T_c(d,0)$ corresponds to T_c^{bulk}. From Fig. 3 it is clear that ΔT_c decreases exponentially with increasing separation, t,

$$\Delta T_c = A\ \exp(-Bt), \tag{2}$$

where $A=[T_c^{\text{bulk}} - T(d,\infty)]$ is the total magnitude of the effect, and B is a constant that will be discussed below.

The overall dependence of T_c on separation of the YBCO layers is similar to that found earlier in Pb/Sb and Pb/Ge multilayers.[17] In addition, work on SC $Mo_{79}Ge_{21}$ ($T_c^{\text{bulk}} = 7.4$ K) sandwiched between $Mo_{1-x}Ge_x$, layers with variable conductivity (ranging from insulating to metallic depending on the percentage of Mo) in multilayer structures also showed these effects.[18] The interpretation of the behavior observed in these low-T_c systems is that adjacent SC layers are partially coupled by electron transmission through the thin separation layers. This coupling process is believed to relax the dimensionality restraints in the SC layers thereby increasing T_c when the SC film separation is decreased. It has been proposed that the electronic coupling between SC layers occurs either by electron tunneling or by diffusion through the insulating layer. In Ref. 17 it is argued that the charge transfer is largely due to electron tunneling processes, including both direct and multistep (indirect[19]) tunneling. The direct process is elastic and is believed to be the dominant coupling mechanism in $Mo_{1-x}Ge_x$ superlattices when the separation layers are metallic.[18] In contrast, multistep or indirect tunneling in which an electron tunnels from one SC layer into a localized state of the spacing insulator and then tunnels out to the next SC layer is inherently inelastic. This process is believed to dominate the coupling between adjacent SC layers in $Mo_{1-x}Ge_x$ superlattices with insulating separation layers.[18] Nevertheless, in both of these processes the tunneling probability decreases exponentially with spacing layer thickness.[19] It should be noted here that we are considering tunneling in the c-axis direction, and it is known that single particle tunneling from YBCO to a non-superconducting metal through insulating barriers can occur[20] along this direction. Conduction along the c-axis direction in insulating PrBCO layers such as those used in our YBCO/PrBCO superlattices has been shown to be dominated by variable range hopping.[21] This suggests that localized states in the insulating PrBCO barriers layers are present, and for the large PrBCO layer thicknesses indirect tunneling processes should dominate[19] the coupling between adjacent YBCO layers.

Finally, in Fig. 4 we show the slopes of each of the curves of Fig. 3 as a function of the reciprocal of YBCO thickness, $1/d$. From this data it can be seen that there is also a linear relationship between the constant B in Eq. (2) and $1/d$. This result, along with Eqs. (1) and (2), suggests the following universal functional dependence of T_c on d and t,

$$T_c = T_c^{\text{bulk}}\{1-(d_m/d)[1-\exp(-gt/d)]\}, \tag{3}$$

where g is a dimensionless constant which contains information both about the tunneling dynamics and the superconducting properties of the YBCO layers. Equation (3) accounts for all of the data in Fig. 1, keeping in mind that d must be greater than d_m. Note that Eq. (3) gives the proper value of $T_c = T_c^{\text{bulk}}$ in the limit of $t\to 0$ or $d\to\infty$. Also note that Eq. (1) is recovered in the limit of $t\to\infty$. Equation (3) also is consistent with the Xiong-Herzog-Dynes[17] model of the variation of T_c with Sb thickness in Pb/Sb multilayers. They suggested that the coupling between Pb layers can lead to an enhancement of the effective Pb thickness $d_{\text{eff}} = d/(1-T)$ where T is the electron transmission coefficient through the insulating Sb layers. In fact, the $\exp(-gt/d)$ term in Eq. (3) is a generalization of T in that it accounts for both the insulator thickness *and* the superconducting layer thickness. The ratio $d/[1-\exp(-gt/d)]$ in Eq. (3) also can be interpreted as an effective thickness.

In conclusion, we find that measurements of the zero-resistance transition temperatures of ultrathin YBCO layers in YBCO/PrBCO superlattices have many similarities to results that have been obtained earlier for ultrathin low-T_c conventional superconductors. The observed depression of T_c with decreasing SC layer is, in both our system and the low-T_c systems, primarily the result of reducing the dimensionality of the SC's from three dimensions to two dimensions. This degradation of T_c can, in part, be compensated by electron tunneling mediated coupling between nearby SC layers. Finally, we find a universal function describing the relationship between T_c and the thickness and separation of the SC layers.

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