Interlayer Coupling Effect in High-T_c Superconductors Probed by $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ Superlattices

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(Received 17 January 1990)

The effect of coupling between layers of the high-T_c superconductors was studied by varying both the $YBa_2Cu_3O_{7-x}$ and $PrBa_2Cu_3O_{7-x}$ layer thicknesses in $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices. Superconductivity was found even when the $YBa_2Cu_3O_{7-x}$ layers had nominal thicknesses as small as 12 Å (one unit-cell thickness) but coupling between unit-cell layers is needed to achieve a T_c of 90 K. The decrease of T_c in isolated $YBa_2Cu_3O_{7-x}$ layers with increasing sheet resistance is on the same scale of about $\hbar/e^2$ as observed for ordinary superconductors.

PACS numbers: 74.70.Jm, 74.40.+k, 74.70.Vy, 74.75.+t

Superconductivity in the high-T_c superconductors (HTSC) has been identified with carriers in the copper-oxide planes of these compounds. The coherence length in the c direction is shorter than the distance between Cu-O planes in the adjacent unit cells.[1] The questions of whether an isolated single unit-cell layer could exhibit high-T_c superconductivity and what role the interlayer coupling plays in determining T_c have been considered theoretically[2] but remain controversial. Recent experiments on ultrathin $YBa_2Cu_3O_{7-x}$ (YBCO) films showed that there was a pronounced decrease of T_c when the film thickness was reduced below about 100 Å.[3,4] These results indicate the possibility of obtaining superconductivity in very thin YBCO films, but leave open the question of whether the observed T_c reduction with film thickness is intrinsic or due to materials problems, such as structural defects caused by the lattice mismatch between the film and the substrate and surface oxygen depletion.

In this Letter we present the results of a study of the superconductivity in $YBa_2Cu_3O_{7-x}/PrBa_2Cu_3O_{7-x}$ superlattices in which we have found that a YBCO layer of nominal thickness of only 12 Å is superconducting but that the coupling between layers is also important for achieving a high T_c. There are two advantages of using these artificial structures to address the coupling problem in HTSC. First, $PrBa_2Cu_3O_{7-x}$ (PrBCO) has an orthorhombic structure with lattice parameters very close to YBCO (with a maximum mismatch of 1.5%) thus providing a much better lattice match than most substrates to YBCO. Secondly, PrBCO is a nonsuperconductor with a very high resistivity at low temperatures. If there is any coupling effect between YBCO layers it is likely that we can tailor this coupling by varying the PrBCO layer thickness without changing the properties of the YBCO layers.

Superlattices consisting of high-T_c superconductors have been successfully made by both sputtering and laser deposition.[5,6] In our previous paper,[6] the preparation and characterization of high-quality $YBa_2Cu_3O_7/Y_{1-y}Pr_yBa_2Cu_3O_7$ superlattices on (100) $LaAlO_3$ substrates with total thickness of 2000 Å using a multitarget pulsed-laser deposition system have been reported. Briefly, x-ray-diffraction measurements showed that the superlattices had the c axis normal to the substrate surface and satellite peaks were observed even when each layer was as thin as 12 Å, clearly indicating the existence of superlattice modulation. A minimum yield of about 10% from 2.8-MeV He^{++}-ion-channeling experiments indicated good crystallinity of the superlattices. Cross-sectional transmission electron microscopy (TEM) showed that the layers had grown epitaxially on one another with an abrupt interface within one unit cell between YBCO and PrBCO layers. Cross-sectional TEM also provided a direct thickness calibration which is in good agreement with the estimates from x-ray satellite-peak spacing and Rutherford backscattering spectra. Even so the layer thicknesses quoted in this paper ought to be considered as nominal values.

The superlattice samples were patterned by pulsed-laser etching into bridges 300 μm wide and 2.5 mm long for four-point transport measurements. The contacts were made at the edges of the films so that all the layers were measured in parallel. Transition temperatures measured using an ac inductance method gave T_c values consistent with the resistance measurement. Besides the structural analysis mentioned above, the sheet conductance per YBCO layer as a function of PrBCO layer thickness was investigated to study further the integrity of the layered structure. Shown in Fig. 1 are the results for three YBCO layer thicknesses corresponding to one, two, and four unit cells of YBCO. At 100 K the resistivity of semiconducting PrBCO is much larger than that of YBCO and therefore the measured sheet conductance

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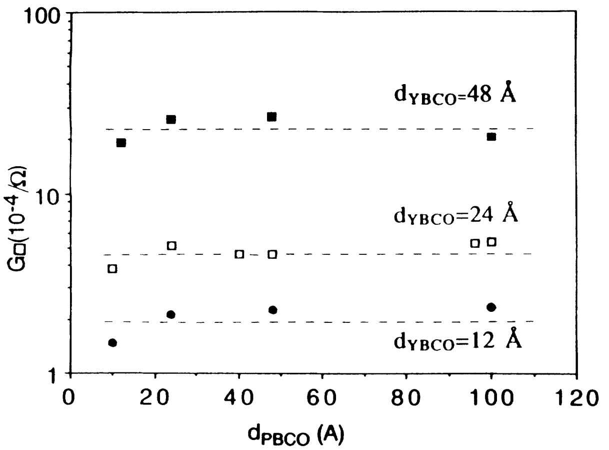

FIG. 1. Sheet conductance per YBCO layer of nominal thicknesses 12, 24, and 48 Å at 100 K as a function of PrBCO layer thickness.

divided by the number of periods gives the sheet conductance of a single YBCO layer. The results show that the sheet conductance per YBCO layer for a given thickness is essentially independent of PrBCO layer thicknesses in the range of 12 to over 100 Å. This demonstrates again that the layered structure was well formed and the normal-state transport properties of each single YBCO layer were not affected by the PrBCO layer thickness.

Superconducting transition curves for $YBa_2Cu_3O_{7-x}$/$PrBa_2Cu_3O_{7-x}$ superlattices with various YBCO layer thicknesses are plotted in Fig. 2 for a constant PrBCO layer thickness of 100 Å [except for samples of (6 Å)/(12 Å) and (12 Å)/(12 Å)]. It is seen that superconductivity occurs in YBCO films as thin as 12 Å, i.e., one unit cell thick. When the YBCO layer thickness decreases, the transition temperature decreases. When the PrBCO layer is very thin, both transition onset T_c^{on} and zero-resistance temperature T_{c0} increase [compare curves *f*, (12 Å)/(12 Å), and *d*, (12 Å)/(100 Å)]. The sample of (6 Å)/(12 Å) has semiconducting behavior as expected since 6 Å is shorter than the length of one unit cell of YBCO. We anticipate that for a YBCO layer thickness of 12 Å, we cannot rule out the possibility that the onset point is affected by microscopic variations in the layer thicknesses, whereas the zero point is affected by imperfections of the substrate, such as steps and scratches, and of the film itself. The broad transition in the (12 Å)/(100 Å) sample is a reflection of these effects. We base our later discussions on general features of the transition rather than exact values of T_c^{on}, T_{c0}, or the midpoint T_c^{mid}.

The transition temperatures T_c^{on} (defined as the temperature of 90% normal resistance) and T_c^{mid} for three YBCO layer thicknesses, 12, 24, and 48 Å, are shown in Fig. 3 as a function of PrBCO layer thickness. T_{c0} behaves in a similar way to T_c^{mid} and therefore is not shown here. For each YBCO layer thickness, T_c remains almost constant when the PrBCO layer thickness decreases from 200 to 30 Å. In this case the YBCO layers are believed to be well isolated from each other by intercalated PrBCO layers and the T_c value can be considered as that of a single YBCO layer sandwiched between PrBCO. It can be seen that T_c decreases as the

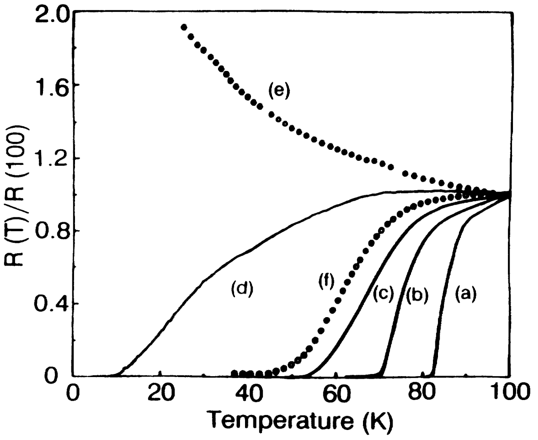

FIG. 2. Superconducting transition curves (normalized to resistivity at 100 K) for superlattices of various periodicities: curve *a*, (100 Å)/(100 Å); curve *b*, (48 Å)/(100 Å); curve *c*, (24 Å)/(100 Å); curve *d*, (12 Å)/(100 Å); curve *e*, (6 Å)/(12 Å); and curve *f*, (12 Å)/(12 Å). It shows that T_c decreases when the YBCO layer thickness decreases (from *a* to *e*). It also shows that T_c increases when the PrBCO layer becomes very thin (compare curves *d* and *f*).

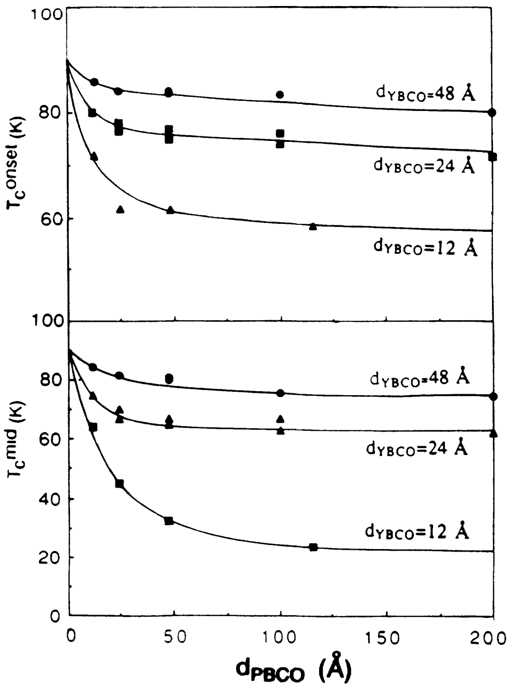

FIG. 3. T_c^{on} and T_c^{mid} vs PrBCO layer thickness for superlattices with YBCO layer thicknesses of 12, 24, and 48 Å.

YBCO layer thickness is reduced. When the PrBCO layer thickness is less than 20–30 Å, T_c starts to increase. The values of T_c^{on}, T_c^{mid}, and T_{c0} extrapolated to zero PrBCO thickness are all close to 90 K.

Many factors can affect the dependence of T_c on the layer thickness. While additional microscopic defects introduced by the multilayer growth were minimized, other factors, such as proximity effect, interdiffusion, and mechanical shorting between the conducting layers, are still possible. We will argue below why none of these effects can consistently explain the data.

In superconductor/normal-metal superlattices, the proximity effect can cause a reduction of T_c when the layer thickness of the superconductor is comparable to its coherence length. This reduction of T_c saturates when the normal-metal layer thickness is greater than the proximity coherence length of the normal metal and diminishes when the normal layer is thinner than that. However, two facts make it difficult to explain our T_c reduction with standard proximity-effect models. First, unlike the conventional normal metals, PrBCO has a conductivity at low temperatures orders of magnitude smaller than that of YBCO thus making the proximity coherence length very small and the proximity effect negligible. Second, the coherence length of YBCO in the c direction is only 3–5 Å and the proximity effect ought not to affect T_c in films much thicker than this.[7]

Our results also cannot be explained by interdiffusion between the YBCO and PrBCO layers. As shown for bulk materials[8] and thin films,[9] the T_c of $Y_{1-y}Pr_yBa_2Cu_3O_7$ decreases as the Pr concentration increases. When $y=0.5$, T_{c0} is about 15 K and for $y>0.5$ the system is semiconducting. If Pr interdiffused over a distance greater than 6 Å, the YBCO layers in (12 Å)/(100 Å) superlattices would be doped with a high concentration of Pr and we would not be able to observe superconductivity. In (12 Å)/(12 Å) superlattices we obtained $T_{c0}=44$ K which is much higher than the value of 15 K for $y=0.5$. These results suggest that the interdiffusion length of Pr and Y in our superlattices is less than half a unit cell. A similar argument applies to the reduction of T_c caused by hole absorption associated with the increased valence of Pr.[10] It also seems impossible that the pair-breaking mechanism discussed in Ref. 10 could be relevant since the Pr atoms of the PrBCO layers are too far from the Cu-O planes in the YBCO layers. Taking into account that PrBCO has similar thermodynamic properties and the same oxygen concentration as YBCO, oxygen depletion from the YBCO layers by PrBCO is also not likely.

Interconnection of layers by structural imperfections introduced during fabrication, particularly particles associated with pulsed-laser deposition,[11] is an important possibility which must be considered. However, from Fig. 1 we can see that the sheet conductance per layer for a given YBCO layer thickness is about the same regardless of the PrBCO layer thickness, showing negligible effects of possible shortings between layers on the transport properties. Furthermore, the sample of (12 Å)/(12 Å) has a lower T_c than that of (24 Å)/(24 Å), though they have the same average composition. Likewise the semiconducting behavior of the (6 Å)/(12 Å) superlattice contrasts strongly with the relatively high T_c in the (12 Å)/(12 Å) sample which has the identical isolating layer between the superconducting layers. These distinct transport results suggest that the effect of shorting by particles and its influence on the behavior of T_c is only secondary. This is further supported in Fig. 4 by comparing the superlattice data with those of single-layer ultrathin films.

Instead of any of the possibilities discussed above, we propose that the behavior of T_c in the superlattices indicates the importance of coupling effects in the c direction between the unit-cell layers of YBCO. Although a superconducting transition can be obtained in a YBCO layer with only one unit-cell thickness, several unit cells need to be coupled together to produce a high T_c. When the number of coupled unit-cell layers of YBCO is increased, the value of T_c increases until it saturates at 90 K. Similarly, when the PrBCO layer thickness is reduced to less than 20–30 Å, the neighboring YBCO layers are able to couple with each other and T_c increases. A similar enhancement of T_c with increasing coupling between superconducting layers has been observed in Nb/Ge multilayers, where the system changed from two- to three-dimensional because of tunneling-induced coupling.[12]

In Fig. 4, the onset and zero point of the supercon-

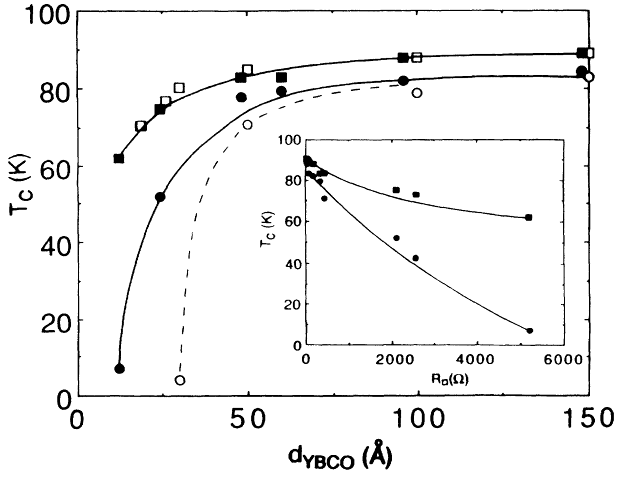

FIG. 4. T_c^{on} (squares) and T_{c0} (circles) as a function of the YBCO layer thickness (solid symbols). The PrBCO layer thickness is 100 Å which is sufficient to isolate adjacent YBCO layers. The results for ultrathin films sputtered on $SrTiO_3$ are also included (open symbols) for comparison. Inset: T_c^{on} and T_{c0} of isolated YBCO layer as a function of sheet resistance per YBCO layer.

ducting transition for multilayers with a PrBCO layer thickness of 100 Å are plotted as a function of YBCO layer thickness. For this PrBCO layer thickness, the superconducting layers have been decoupled from one another. The data from single ultrathin YBCO films sputtered on (100) $SrTiO_3$ are also shown for comparison. As can be seen, by improving the lattice match and hence reducing the structural defects, we can get full superconductivity in a thinner layer of YBCO in the superlattices than in the ultrathin single films. However, it should be noted that the onset of the transitions obtained from the two different experiments coincide with each other showing that even T_c^{on} decreases abruptly when the YBCO layer thickness is less than about 50 Å. Considering the differences between the two systems and the techniques used to produce them, this agreement suggests that the decrease of T_c with film thickness is indeed intrinsic to YBCO and can be understood by the coupling effect discussed above.

To further explore the nature of the T_c decrease with film thickness, the superconducting-transition onset and zero point of the decoupled YBCO layers are plotted as a function of sheet resistance per YBCO layer as shown in the inset of Fig. 4. The behavior is similar to that of two-dimensional films of the ordinary superconductors like Bi, Pb,[13] and amorphous Mo-Ge.[14] As can be seen from Fig. 4 the sheet resistance at which T_c is substantially reduced is about 5000 Ω/□. This is of the same order as the characteristic value $\hbar/e^2$ of the sheet resistance for which a similar reduction observed in conventional superconductors has been attributed to localization effects in two-dimensional systems.[15] It suggests that the reduction of T_c in HTSC might have the same origin as in the ordinary superconductors. However, it should be noted that there are substantial differences between the two cases. First, there is crystalline order in the YBCO films whereas the other systems are amorphous. The normal-state conduction mechanisms are very different. The YBCO films show a decrease of resistivity with decreasing temperature implying a dominance by inelastic scattering whereas in the conventional superconductors the resistivity is nearly temperature independent. The ordinary superconducting films are all in the "dirty limit," i.e., the electron mean free path is much less than the intrinsic coherence length, and the film thicknesses are smaller than the Ginzburg-Landau coherence lengths. In the YBCO, the mean free path and coherence length are comparable and the film thickness is greater than the intrinsic coherence length. Nevertheless, the similarity of the dependence of T_c on sheet resistance in the two types of systems gives another illustration of the universal nature of the relation between these two quantities in two-dimensional systems.

In summary, we have presented results which strongly suggest that superconductivity can occur in a layer of YBCO of thickness as small as one unit cell (12 Å in the *c* direction) but that coupling between the unit-cell layers is needed to achieve a T_c of 90 K. The dependence of transition temperature on sheet resistance as the layer thickness is changed suggests that T_c is depressed by mechanisms similar to those of ordinary superconductors in spite of differences in the electronic properties of the two systems.

We would like to thank C. T. Rogers, J. B. Barner, P. England, L. H. Greene, and J.-M. Tarascon for helpful discussions. We wish to acknowledge support from the National Science Foundation Grants No. ECS-8910668 and No. DMR-8511982.

Note added.—Since this manuscript was first submitted, a related study of YBCO/PrBCO superlattices has been published by Triscone *et al.*[16]

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