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Superconducting Properties of Ultrathin $Y(Dy)Ba_2Cu_3O_7$ layers in $Y(Dy)Ba_2Cu_3O_7$-$PrBa_2Cu_3O_7$ Heterostructures

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We have measured the superconducting properties of ultra thin layers of $YBa_2Cu_3O_7$ in c-axis $YBa_2Cu_3O_7$-$PrBa_2Cu_3O_7$ (YBCO-PrBCO) heterostructures. These structures contain one or a few layers of YBCO (or in some cases DyBCO). The critical temperature of *single layers* was investigated through a series of PrBCO-YBCO-PrBCO trilayers. The trilayers consist of d Å of YBCO sandwiched between a buffer and a capping PrBCO layer. The YBCO thicknesses were not chosen to be multiples of a unit cell (~12Å), but equal to n*1/3 of a unit cell. The series was built with nominal YBCO thicknesses of ~4Å, ~8Å, ~12Å, ⇒~40Å. Superconductivity is observed in all the samples, except in the 4Å one which displays a pronounced semiconducting behavior. We discuss these results and the differences between the properties of single and multilayer structures.

1. Introduction

Many different oxide combinations have been used to synthesize artificial high T_c superlattices, such as YBCO/DyBCO[1], YBCO/PrBCO[2-7], YBCO/NdCeCuO[8], or different combinations of BiSrCaCuO structures[9-11]. These materials offer the opportunity to modify the anisotropy of the oxide superconductors and allow therefore to observe the effect of the changing anisotropy on the superconducting properties. The role of the reduced dimensionality on the critical temperature has been the subject of many investigations[2-5,7,9,11,12]. One of the questions which is still under debate is whether or not an isolated single unit cell thick layer of cuprate is superconducting. This question has been addressed using superlattices of the "123" compound by progressively separating one unit cell thick YBCO layers from the neighboring layers, by inserting insulating PrBCO layers in between. The results show[2-5,13] that, for PrBCO separation layers larger than 60-70Å, T_c, defined as the temperature where the resistance drops below 1% of the normal state value, saturates at values ranging between 10-30K. This saturation value has been taken as the superconducting critical temperature of an isolated single unit cell thick layer of YBCO. Experiments on trilayers PrBCO/YBCO/PrBCO have been performed by Terashima et al.[7]. Using RHEED oscillations to monitor the growth rate, single unit cell thick layer of YBCO have been sandwiched between PrBCO. The superconducting critical temperature T_c of such a sandwich was about 28K. Although these different approaches seem to agree rather well in view of the complexity and differences between the experiments, it is still difficult to make a firm affirmation that a unit cell thick layer of YBCO is superconducting. The reasons are the following: on one hand it has not been possible to measure a single unit cell without PrBCO buffer-capping layers and the role played by the PrBCO conductivity (which can be very different depending on the preparation method) on the superconductivity is not yet clearly established. On the other hand the micro structure of these materials is complex as revealed by STM studies[14] and the characterization required to insure that the thickness of the film investigated is indeed one monolayer thick is very stringent. The growth mode of YBCO, unit cell by unit cell, is such that it is not obvious to guarantee perfect coverage and RHEED analysis certainly do not insure that a perfect layer by layer growth takes place, as detailed in a comment of Goldman[15]. Additionally, Chan et al. report recent experiments on trilayers were they did not observe superconductivity in one unit cell thick YBCO layers[16]. The critical temperature of a unit cell thick layer of YBCO is thus still an open question.

In this paper we report on the properties of c-axis PrBCO-YBCO-PrBCO trilayer structures with non-integer layers of YBCO. The idea was to see if signatures of the one, two, etc.. unit cells coverage could be observed in the transport properties, rendering easier the determination of the critical temperature of a

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single unit cell thick layer of YBCO. We also compare the properties of single and multilayer structures.

2. Experimental

The YBCO-PrBCO structures are prepared by magnetron sputtering onto (100) $SrTiO_3$ substrates heated at about 720°C. The sputtering gas is a mixture of Ar and O_2, $Ar/O_2=10$. The total pressure is 600mTorr. The rate calibrations are obtained using x-ray diffraction through finite size effects on single layers and by multilayer satellite peaks[17]. The calibrations agree to better than 10%. To produce the desired heterostructure a stepping motor positions the substrates alternatively in front of the PrBCO and YBCO guns.

3. Superconducting properties of PrBCO-YBCO-PrBCO trilayers.

Figure 1a and b shows the resistively measured transitions (using the van der Pauw technique) of single YBCO layers sandwiched between PrBCO buffer-capping layers. These trilayers were fabricated with a six unit cells PrBCO buffer layer, d Å of YBCO, and a protective, 2-8 unit cells, PrBCO capping layer. The YBCO thicknesses *were not chosen to be multiples of a unit cell* (~12Å), but equal to n∗1/3 of a unit cell. The series was built with nominal YBCO thicknesses of ~4Å, ~8Å, ~12Å, ⇒~40Å. These YBCO thicknesses are nominal thicknesses as determined through calibrations. The precision of the absolute values of the resistivity is not better than a factor of two since the contact geometry used was not ideal for the van der Pauw technique.

For such thin layers the precision on the thickness and the reproducibility of the results are a concern. According to the different calibrations the precision on the average thickness is better than 10%. To test the reproducibility of the results we prepared three 12Å samples, three 36Å, three 36Å/96Å (x3) (x3 means that the 36Å/96Å sequence is repeated three times), and three 36Å/96Å (x8) multilayers. Using the onset and the 10% point of the resistive transition (T_{onset},$T_{c10\%}$), we find that the reproducibility is better that 2.5K for both criteria except for the 12Å sample where, although the onsets agree within 2K, the bottom part of the transition is slightly different from sample to sample. Two samples had a $T_{c10\%}$ slightly below 4.2K whereas the 12Å sample of the series presented here has a $T_{c10\%}$ of 12.9K. Clearly, only the lowest part of the transition for very thin samples seems to show some scattering.

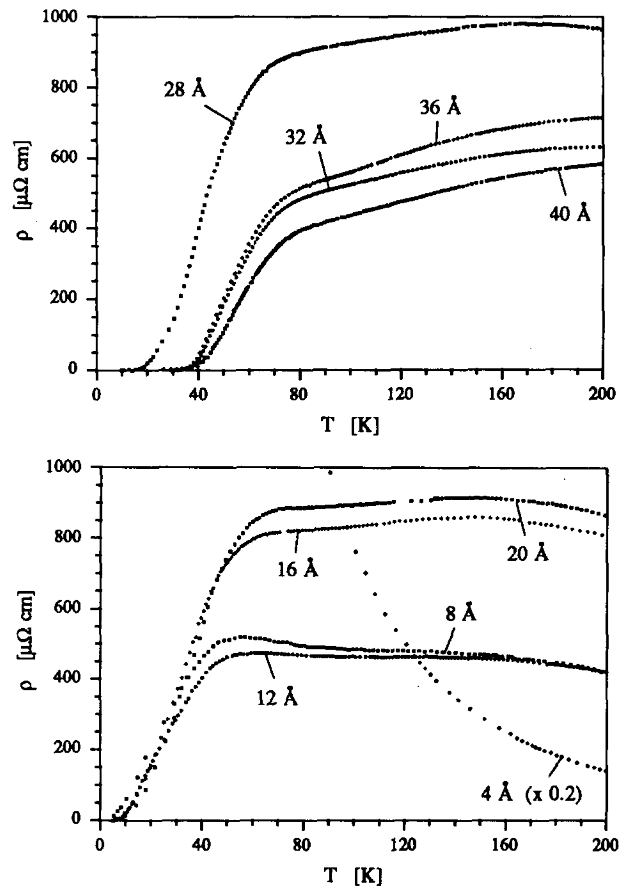

Figure 1a and b show the resistively measured transitions for the trilayer series. The indicated thicknesses are nominal YBCO thicknesses obtained through calibration runs.

Before discussing these transitions we would like to comment on the role played by the PrBCO on the superconducting temperature. Our PrBCO layers have resistivities of the order of 10^5-10^6μΩcm at 300K and 10^7-10^9μΩcm at 100K. Terashima et al. (who report higher T_c values) have reported that they use a much less resistive PrBCO with essentially no temperature dependence of the resistivity[18]. In our experiment, replacing PrBCO by a more conducting material such as YPrBCO alloys increases dramatically T_c. It is thus important when discussing T_c values to take into account the resistivity of PrBCO.

Figure 2 shows $T_{c,onset}$ and $T_{c10\%}$ for this set of samples. As can be seen, the 40Å, 36Å, and 32Å transitions are very similar with nearly identical onsets and $T_{c10\%}$. However, the 28Å sample, although having a similar onset to the 40-32Å samples, has a significantly lower $T_{c10\%}$. Simple percolation arguments can explain nicely these results. Figure 3 shows a schematic diagram of the 28 and 32Å samples assuming a perfect PrBCO buffer. Since YBCO grows unit cell by unit cell (UC), sputtering a layer whose average thickness is not a multiple of a unit cell will result in a partially filled top UC layer as illustrated in Fig. 3. The 28Å sample which corresponds to 2UC+1/3 of a UC has a two unit cell full coverage and a 33% coverage of the third UC. The 32Å has a top UC covered at 66% since 32Å corresponds to 2UC+2/3 of a UC. Let us now follow the simple idea that the onset of the transition will reflect the highest coverage while $T_{c10\%}$ will correspond to the highest coverage which reaches the percolation threshold for 2D, i.e. 50%. We thus conclude that the 32Å should reflect the onset and $T_{c10\%}$ of a three UC coverage whereas the 28Å sample should have the three UC onset but the $T_{c10\%}$ of the 2UC since the 3UC coverage is below 50%, i.e. the $T_{c10\%}$ should be different for these two samples. This is just what is observed on figure 2.

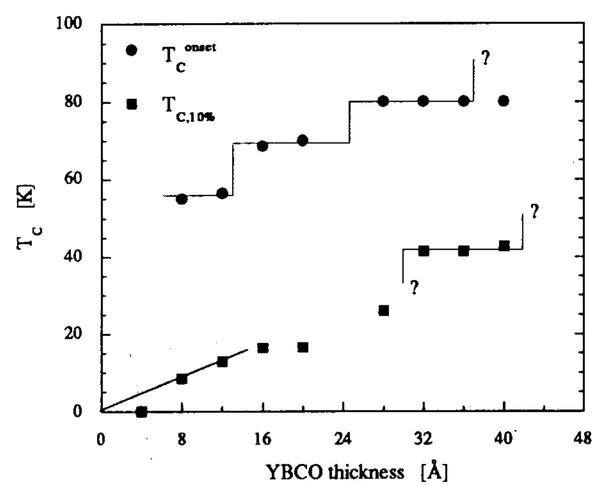

Figure 2 shows the T_{conset} and $T_{c10\%}$ for the trilayer series.

It is thus tempting to consider the plateaus at 80 and 42K to be the signature of a 3UC thick layer. As can be seen on Fig. 2 the onset values show two additional plateaus at 70 and 55K. The solid line is a prediction of the switching thicknesses according to the simple model discussed above and using the experimental plateau values. All the experimental points fall on the line except the 40Å sample which should show the 4UC onset.

Looking at figure 2 the plateau model is clearly less obvious for the $T_{c10\%}$ of the thinnest layers. However, the plateau argument may be too simple for

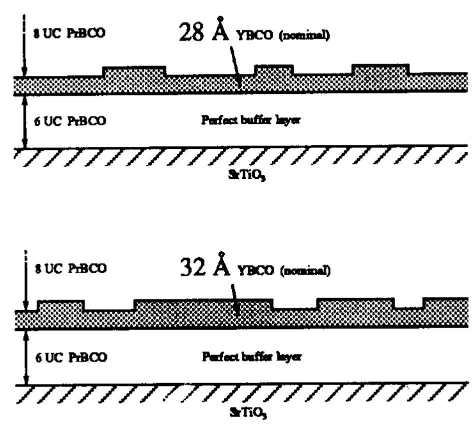

Figure 3 is a schematic diagram of a 28Å and of 32Å YBCO layers deposited onto an ideal buffer. The UC by UC growth results in a partially covered top layer.

such thin layers and depends very much on the mechanism and the origin of the broadening of the transitions. For example, if the broadening is related to a Kosterlitz-Thouless like transition, then the value of T_{KT} is proportional to the 2D superfluid sheet density n_s^{2D} which, in a simple model, will be proportional to the average coverage over a scale of the penetration depth λ. If the islands are small compared with λ one does not expect anymore plateaus but a linear increase of $T_{c10\%}$ with thickness for the thinnest samples. This is again close to what we observe. Although we may have important scattering for the $T_{c10\%}$ of the thinnest layers, the qualitative agreement between the results and the expected behavior through the simple model exposed above is striking.

Another important feature of these data is the fact that the 8 and 12Å films are both superconducting with similar transitions. If a one UC layer of YBCO would not be superconducting rendering necessary a two UC coverage to get superconductivity, the 8Å result would be difficult to understand. Effectively 8Å represents a one UC coverage of 66% but, if one requires a two UC coverage to explain superconductivity, 8Å corresponds to a two UC coverage of only 33%. Thus assuming a random distribution of the islands during growth, which would need 50% coverage for percolation, the superconducting transition in the 8Å sample can only be understood in terms of a 66% filled single UC layer. Of course, the edge growth mechanism of YBCO does not correspond to a completely random distribution of the islands and this mechanism may change the minimum coverage necessary for percolation. However, we nevertheless find that an explanation in terms of a two UC partial coverage is highly unlikely to be correct.

4. Superconducting properties of single layer to multilayers.

Typically, 24Å/96Å and 36Å/96Å multilayers have $T_{c10\%}$ of about 55 and 65K respectively while we find (see figure 2) only 13K and 41K for the 24Å and 36Å single layers, respectively. In this section we want to discuss the marked difference between the superconducting properties of single layers and multilayers. To understand the origin of this change in T_c we grew structures containing one or several superconducting layers separated by PrBCO. The idea was to understand whether it is the number of layers which plays a role or it is the position of the layer in the structure which is important.

Figure 4 shows the resistively measured transitions along with a schematic diagram of the samples investigated. First the structure A, containing two 36Å superconducting layers, was grown and compared to similar structures (B and C) but in which one of the superconducting layer is replaced by PrBCO. As can be seen on the top part of figure 4, A and C have very similar superconducting transitions whereas B has a lower T_c. Structure B corresponds to the trilayer series presented above but with a thicker PrBCO capping layer and, effectively, the R(T) curve for B and for the 36Å sample of figure 1 are very similar. The difference between B and C suggests that a buffer thickness of 6 or 8UC is not sufficient to get optimum quality layers. This, of course, also means that the T_c of the trilayer series presented on figure 1 and 2 has to be taken as a lower limit.

The bottom part of figure 4 illustrates the effect of increasing the number of layers in the structure on the superconducting properties. Going from two layers (sample A) to three layers with sample D does not change the superconducting transition. Astonishingly, going to five and six superconducting layers in the structure gives exactly the same result as for A (or D), but putting eight layers increases T_c by 15K. Preliminary Hall effect measurements indicate that the Hall coefficient of the (36Å/96Å)x8 multilayer is very similar to the one of multilayers with identical thicknesses of YBCO and PrBCO investigated in a previous work[19]. However, we measured a clear difference between the double layer and the multilayer indicating that the number of carriers is reduced in the double layer compared to the 8x-multilayer. The behavior in magnetic field and the values of the activation energies for flux motion, determined through Arrhenius plots[20] are also different between the double and the 8x-multilayer. We suspect these differences to be related to strains in the thin films due to substrate-film mismatch and that these are released at a thickness of about 800-900Å. Note that all our previous work was with multilayers thicker than 1000Å. Strain effects on T_c have also been observed in NdCeCuO/YBCO superlattices[8]. Hall effect measurements along with the magnetic field behavior of these samples will be reported in a forthcoming publication.

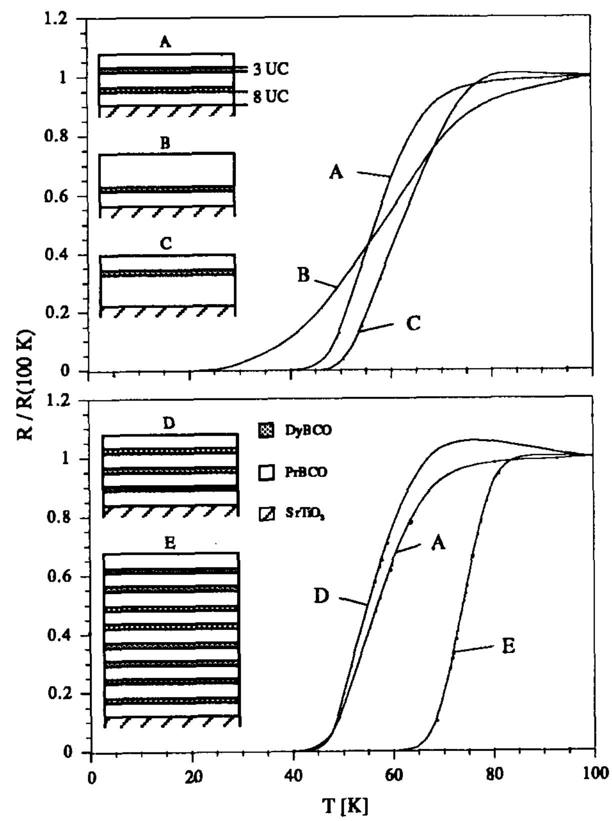

Figure 4 shows the zero field resistive transitions for different DyBCO-PrBCO structures. All the superconducting layers are 36Å thick. The PrBCO separation layers are 96Å thick..

5. Summary

We have measured the critical temperatures of YBCO-PrBCO heterostructures. We find that the critical temperature of PrBCO-YBCO-PrBCO trilayers built with non-integer YBCO layers display plateaus which may be indicative of the critical temperature of the one, two, and three unit cell thick layers. We find that the 8Å sample displays a full superconducting transition with T_c of about 4K. We have investigated the effect of increasing the number of layers in the structures on the superconducting transitions. In 36Å/96Å DyBCO/PrBCO multilayers we observe a sharp increase of T_c between six and eight layers which we suspect to be related to an effect of strain on the layers.

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