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PROPERTIES OF EPITAXIAL $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -BASED
SUPERCONDUCTING SUPERLATTICES

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PROPERTIES OF EPITAXIAL $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -BASED SUPERCONDUCTING SUPERLATTICES

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ABSTRACT

Epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -based superconducting superlattices have been fabricated using pulsed laser deposition in which c -axis oriented $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ layers as thin as one unit cell thick are separated by relatively thick $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -based barrier layers. The superlattice T_c ($R = 0$) decreases rapidly with increasing barrier layer thickness, but then saturates at a finite T_c for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ layers as thin as a single c -axis unit cell. The superconducting properties of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -based superlattices are shown to depend strongly on the electronic properties of the barrier layers. The resistive transition width decreases significantly as the hole carrier density in the barrier layers is increased. However, T_c (onset) does not change, contrary to predictions of hole filling models. Theoretical analyses suggest that the broadening of the resistive transition for the thinnest $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ layers is most likely due to a crossover to 2D resistive behavior involving thermally-generated vortices. Scanning tunneling microscopy reveals that epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films grow unit cell-by-unit cell, by a terraced-island growth mode. This terraced microstructure explains the steps found in ultrathin $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ layers in these superlattices. These steps may act as superconducting weak links, providing support for 2D Josephson-coupled-array models of superconducting superlattices.

INTRODUCTION

High-temperature superconductivity is associated with layered, quasi-two-dimensional (2D) crystal structures and with carriers moving in CuO_2 planes. Because the c -axis superconducting coherence length is very short (e.g., $\xi_c \sim 3\text{--}6\text{\AA}$, vs a lattice constant $c \sim 11.7\text{\AA}$ in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$), questions arise whether isolated single-cell-thick layers of these materials are superconducting and, if so, how their superconductivity is affected by their extreme anisotropy (including possible reduced dimensionality) and by residual interlayer coupling or other interactions. Several groups recently have reported on the electrical transport properties of high temperature superconducting/semiconducting superlattices, including $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$,¹⁻⁴ $\text{YBa}_2\text{Cu}_3\text{O}_x/\text{Nd}_{1.83}\text{Ce}_{0.17}\text{CuO}_8$,⁵ and $\text{Bi}_2\text{Sr}_2\text{Ca}_{0.85}\text{Y}_{0.15}\text{Cu}_2\text{O}_8/\text{Bi}_2\text{Sr}_2\text{Ca}_{0.5}\text{Y}_{0.5}\text{Cu}_2\text{O}_8$ ⁶ structures. The

superconducting properties of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO/PrBCO) superlattices are sensitive functions of both the superconducting (YBCO) and the barrier (PrBCO) layer thicknesses, d . T_c decreases as the YBCO layer thickness decreases or as the PrBCO layer thickness increases, but for all YBCO layer thicknesses, including layers one unit cell thick, the superconducting transition temperature saturates at nonzero values; e.g., for YBCO layer thicknesses of one, two, and three unit cells isolated in a relatively thick PrBCO matrix, the zero resistance transition temperature, T_{c0} , is ~ 19 K, ~ 54 K, and ~ 70 K, respectively.³ Thus, there appears to be “coupling” between thin YBCO layers that are separated by PrBCO layers only a few unit cells thick. This is evident as, for a given YBCO layer thickness, T_c does not become independent of PrBCO layer thickness until $d_{\text{PrBCO}} > 5$ nm. In addition, the widths of the superconducting transitions are large, with $\Delta T_c \sim 37$ K for YBCO layers one unit cell thick isolated in a PrBCO matrix.

Recently, we have determined that the superconducting transition of YBCO-based superlattice structures depend on the electronic properties of the barrier layers, focusing specifically on the mobile hole concentration (resistivity) of the barrier layers.⁷ Superlattice structures were fabricated using three different barrier layer materials, namely $\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (PrBCO), $\text{Pr}_{0.7}\text{Y}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (PrYBCO), and $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (PrCaBCO). By systematically varying the hole concentration of the barrier layers used to isolate ultrathin YBCO layers, we find that the superconducting transition width depends on the carrier density in the barrier layer, while $T_c(\text{onset})$ does not.

Several explanations have been proposed for the resistive transitions observed in these superlattice structures, including proximity effect,⁸ localization effects,⁹ and hole-filling.¹⁰ Rasolt, Edis, and Tesanovic recently suggested¹¹ that these YBCO/PrBCO superlattices provide a 3D system in which the interlayer (c -axis) coupling can be weakened to zero, as indicated by the saturation of T_c values that occurs for large PrBCO thicknesses.³ Thus, a crossover to 2D behavior occurs, accompanied by characteristic 2D dissipation.¹¹ Minnhagen and Olsson also find that the resistive transition data for superlattices containing isolated YBCO layers (one or two cells thick) are well explained by the 2D Ginzburg-Landau Coulomb Gas model.¹² Finally, Ariosa and Beck have suggested¹³ that each superconducting CuO_2 bilayer in a superlattice structure may be modeled as a 2D array of ultrasmall Josephson junctions. Thus, several recent papers suggest that reduced dimensionality plays an important role in the resistive behavior of these structures.

RESULTS AND DISCUSSION

The c -axis perpendicular superlattice structures were fabricated using in situ pulsed-laser deposition, as has been described elsewhere.^{3,7} We

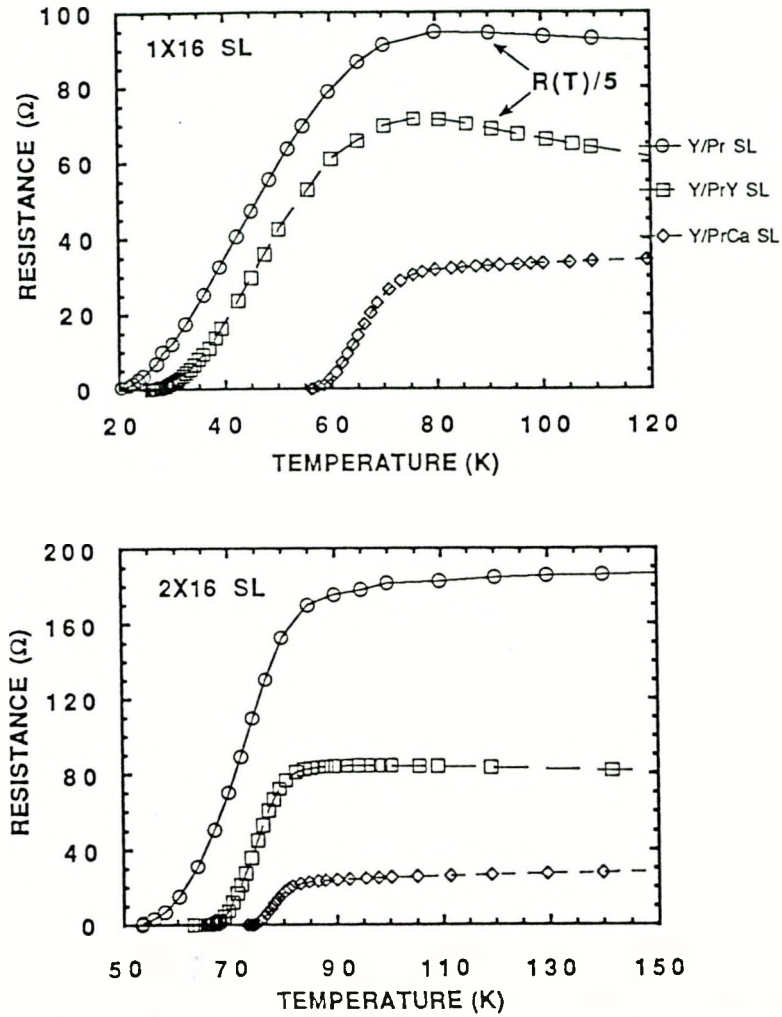


Fig. 1. $R(T)$ for 1×16 and 2×16 YBCO/PrBCO (O), YBCO/PrYBCO (\square), and YBCO/PrCaBCO (\diamond) superlattice structures.

describe them using the nomenclature " $N \times M$ ", where N and M are the numbers of YBCO and barrier layer unit cells per superlattice period, respectively. Most of the structures consist of 30 superlattice periods. Of the three barrier layer materials utilized, PrBCO is the least conductive with the lowest mobile hole concentration. PrYBCO is more conductive, but still demonstrates a divergent resistivity at low temperatures. The properties of the PrCaBCO thin films, in which divalent Ca is added to introduce holes into an otherwise semiconducting compound, are nearly metallic with very little temperature dependence in the resistivity. A description of the superconducting properties of the PrCaBCO thin film system has been reported elsewhere.¹⁴

Figure 1 shows the $R(T)$ behavior for 1×16 and 2×16 superlattices with either PrBCO, PrYBCO, or PrCaBCO utilized as the barrier layer material. A systematic dependence of the resistive transitions on the carrier density of

the barrier layers is observed. The most interesting effect is a significant increase in T_{c0} with increasing barrier layer carrier density. For the 1×16 superlattices, T_{c0} increases from ~ 20 K for the YBCO/PrBCO structure to > 50 K for the YBCO/PrCaBCO structure. However, note that $T_c(\text{onset})$ [defined as the initial inflection of $R(T)$] is not significantly influenced by the hole concentration in the barrier layers. This is more clearly seen in Fig. 2 where $T_c(\text{onset})$ and T_{c0} are plotted as functions of barrier layer thickness. Although T_{c0} increases (transition width decreases) significantly as the barrier layer carrier density is increased, $T_c(\text{onset})$ is insensitive to the barrier layer composition, depending only on the thicknesses of the YBCO and barrier layers.

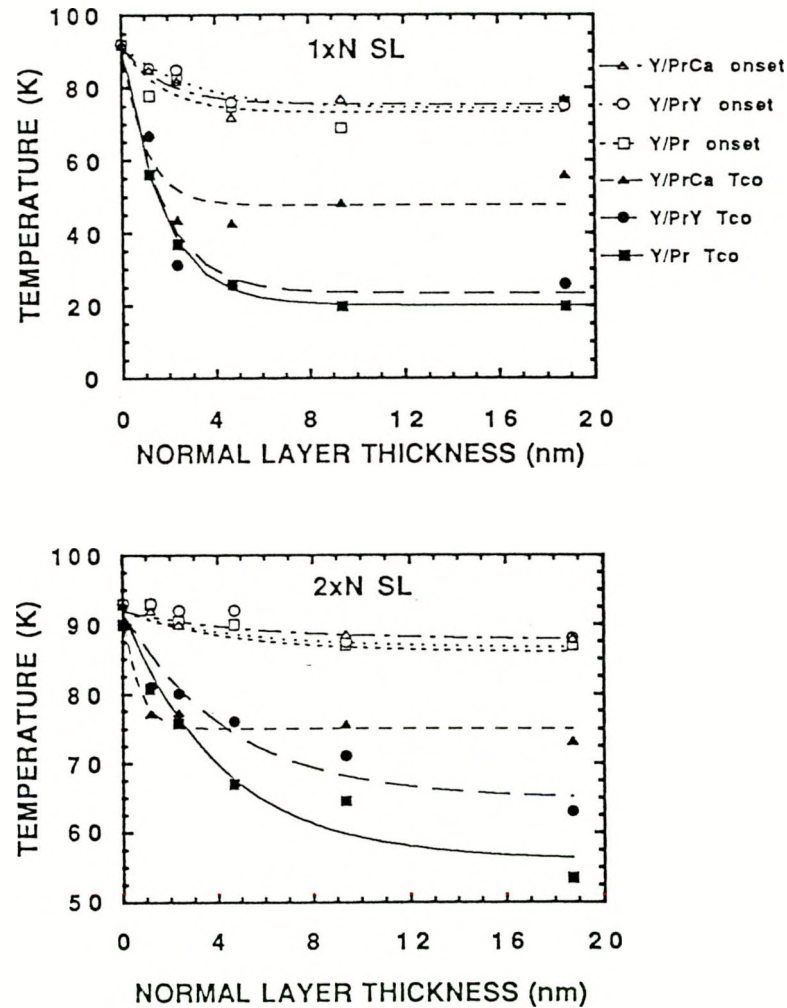


Fig. 2. $T_c(\text{onset})$ (open symbols) and T_{c0} (filled symbols) as a function of normal layer thickness for $1 \times N$ and $2 \times N$ YBCO/PrBCO (\square, \blacksquare), YBCO/PrYBCO (\circ, \bullet), and YBCO/PrCaBCO (Δ, \blacktriangle) superlattice structures.

It recently was proposed that electron transfer from the PrBCO layers into the YBCO layers (resulting in hole-filling in the YBCO layers) can explain the depression of T_c for YBCO/PrBCO superlattices.¹⁰ Hole-filling was previously used to explain the suppression of T_c as Pr is substituted into superconducting "123" oxide materials. In these alloyed systems, the mixed valence of the Pr leads to a reduction of mobile hole density on the CuO_2 planes, with a subsequent reduction in T_c .^{15, 16} In YBCO/PrBCO superlattice structures, ultra-thin (perhaps 1 unit cell thick) YBCO layers with a high mobile hole density are layered alternately with a low carrier density material (PrBCO) of comparable thickness. The possibility of electron transfer from the PrBCO to the YBCO therefore must be considered. If there is a significant reduction of the mobile hole density in the YBCO layers, then a reduction in the transition temperature, T_c (onset), should result. Conversely, by adding holes to the barrier layers (for instance, by doping the PrBCO with divalent Ca), the transfer of holes from the YBCO layers into the barrier layers should be reduced, and T_c (onset) should increase.

Thus, within the hole-filling model, it is difficult to explain why there is no significant change in T_c (onset) as the carrier density in the barrier layers is increased, as seen in Figs. 1 and 2. The hole-filling model predicts that an increase in the hole density in the barrier layers should lead to an increase in T_c (onset), with little effect on the transition width. One does not expect the transition width to be a strong function of the YBCO carrier density. Recent experiments in which the mobile hole concentration was varied directly, by removing oxygen from YBCO thin films and YBCO/PrBCO superlattice structures, showed that T_{c0} and T_c (onset) shift together as the mobile hole concentration is decreased, with little or no additional broadening of the transition.⁷ Figure 3 shows the $R(T)/R(100\text{ K})$ behavior of a 2×8 $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and a 2×4 $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{Pr}_{0.5}\text{Y}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ superlattice structure after partial removal of oxygen by means of low pressure oxygen annealing. As oxygen is removed, no significant broadening of the superconducting transition region is observed, while T_{c0} and T_c (onset) both shift to lower temperatures. Thus, the insensitivity of T_c (onset) to the barrier layer carrier density provides evidence against a simple change in the hole carrier density as the explanation for the depression of T_c in YBCO/PrBCO superlattices.

A reduction of T_c (onset) and/or an increased transition width has also been observed for conventional low T_c superconducting systems as the superconducting layer thickness is reduced.¹⁸⁻²¹ In these materials, the reduction of T_c (onset) has been attributed to a suppression of the superconducting pair wave function amplitude, whereas the broadening involves a lack of phase coherence. The reduction in T_c (onset) for low T_c ,

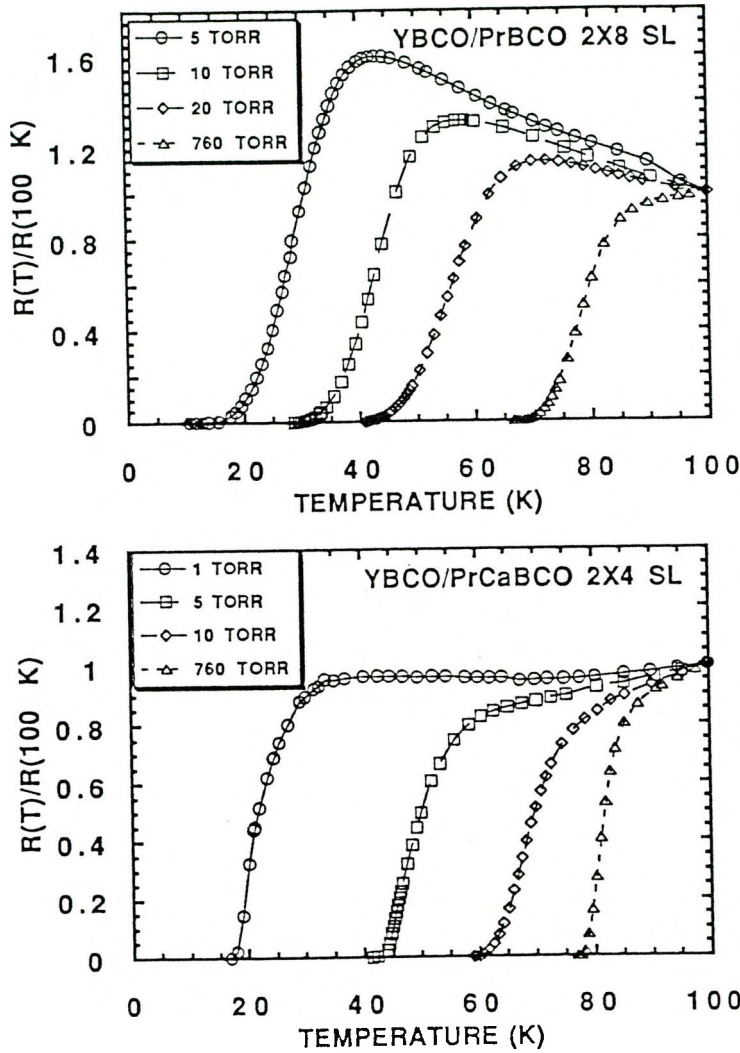


Fig. 3. $R(T)/R(100\text{ K})$ for 2×8 $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{PrBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and 2×4 $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$ superlattice structures after various low pressure oxygen anneals.

ultrathin films has been explained in terms of the proximity effect,⁸ in terms of localization in disordered 2D systems,⁹ and in terms of the boundary conditions for the order parameter within the Ginzburg-Landau free energy expression.²² Predictions based on proximity effect and order parameter boundary conditions agree qualitatively with the superconducting properties of YBCO/PrBCO superlattices. However, the length scale over which the YBCO layers become "decoupled", as the PrBCO barrier layer thickness is increased, is somewhat larger than the normal metal electronic coherence length in the semiconducting PrBCO. (This coherence length is expected to be less than the c-axis coherence length in metallic YBCO.) If localization in a disordered, 2D system, is responsible for

the reduction of T_c (onset), the resistivity of the superconducting layer should increase as T_c (onset) decreases, in agreement with our observations. For the YBCO/PrBCO superlattice system, the 1×16 ($\rho_{\text{YBCO}}(100 \text{ K}) = 400 \mu\Omega\text{-cm}$, T_c (onset) = 76 K) and the 2×16 ($\rho_{\text{YBCO}}(100 \text{ K}) = 300 \mu\Omega\text{-cm}$, T_c (onset) = 87 K) structures follow this trend.

Although some depression of T_c (onset) occurs as a function of layer thickness in the YBCO-based superlattice structures (Fig. 2), a more prominent feature is the broadening of the superconducting transition. In general, the transition width represents a lack of long-range phase coherence of the superconducting order parameter. Transition width broadening has also been observed in ultrathin films of low-temperature superconductors, and has been attributed to the presence of a 2-D array of weakly in-plane-coupled Josephson junctions and possibly to 2-D vortex-antivortex pair unbinding.²³⁻²⁶ Rasolt et al.¹¹ and Minnhagen and Olsson¹² have pointed out that a crossover to 2D resistive behavior occurs for sufficiently thin YBCO layers, as the PrBCO thickness is increased. When crossover occurs, characteristic 2D dissipation processes should be observed, specifically resistance in the 2D superconducting state due to free vortices that are produced by thermal unbinding of vortex-antivortex pairs.²⁷⁻³⁰ A low temperature Kosterlitz-Thouless (KT) transition also may occur, if a 3D superconducting transition caused by residual interplane coupling does not intervene first¹² (for intermediate PrBCO thicknesses).

One of the characteristic "signatures" of a 2D conductor is that in the temperature range well below the mean-field (bulk, or BCS) superconducting transition temperature, T_{cB} , but just above the Kosterlitz-Thouless temperature, T_c , where thermal unbinding of the vortex-antivortex pairs begins to produce free vortices,^{27,28} the thermally generated vortices produce a nonzero resistivity with temperature dependence^{29,30}

$$R(T)/R_N \sim \xi_+(T)^{-2} = \xi_{ab}(T_c)^{-2} \exp[-2b'(\tau_c/\tau)^{1/2}]. \quad (1)$$

In Eq. (1) R_N is the normal-state resistance, $\xi_+(T)$ is the phase correlation length for the superconducting order parameter,^{29,30} $\xi_{ab}(T)$ is the Ginzburg-Landau correlation length within the a - b plane, $\tau_c = (T_{cB} - T_c)/T_c$, $\tau = (T - T_c)/T_c$, and b' is a constant of order unity. From Eq. (1) it follows that $\log R(T)$ should scale as $\tau^{-1/2}$.

Figure 4 shows the result of plotting $\log R_n(T)$ vs $\tau^{-1/2}$, where $R_n(T) = R(T)/R(100 \text{ K})$, for the $M(\text{YBCO}) \times 16(\text{PrBCO})$ superlattice structures.^{3,31} These are the structures with the thickest PrBCO layers, for which the saturation of T_c suggests that a crossover to 2D resistive behavior has occurred. Good agreement with the expected scaling behavior is observed, and from these plots we extracted least-squares best-fit values for the KT transition temperature of $T_c = 14, 44, 70$, and 86.5 K, for the structures with 1-, 2-, 3-, and 8-cell-thick YBCO layers, respectively. We should note,

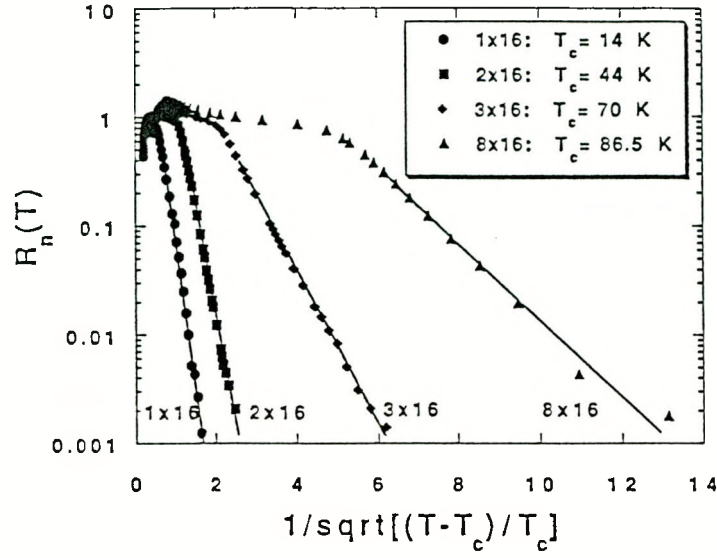


Fig. 4. Superconducting transitions for $M(\text{YBCO}) \times 16(\text{PrBCO})$ superlattices with $M = 1, 2, 3$, and 8 c -axis unit cells, plotted as $\log R_n(T)$ vs $\tau^{-1/2}$, where $R_n(T)$ is the normalized resistance $R(T)/R(100 \text{ K})$ and $\tau = (T - T_c)/T_c$. The T_c values determined by best least-squares fits to the data are given in the text.

however, that the $R(T)$ data are outside the region ($T_c < T \ll T_{cB}$) where Eq. (1) is strictly applicable.²⁹

Similar broadening of the superconducting transition, associated with a Kosterlitz-Thouless transition, is predicted for 2D films with a granular nature, consisting of an array of superconducting islands connected by weak links.^{24,32} As the temperature is decreased, the individual islands become superconducting at T_c (onset). However, long-range phase coherence is established by Josephson coupling between islands only at a lower temperature, leading to a broad transition. Although the YBCO-based superlattices are high quality, fully epitaxial structures, some evidence supporting a weak link description has recently been provided by Z-contrast transmission electron microscopy (TEM) images.^{7,33} These images indicate that the YBCO layers are not perfectly flat relative to the lattice structure; i.e., the YBCO layer shifts up (or down) by one c -axis unit cell increment as one progresses parallel to the a - b planes. These one-cell-thick "kinks" in the YBCO layers are due to steps on the growing film surface. Their significance for current flow in the YBCO layers is that conduction along the c -axis will be necessary at the kinks, in order for a continuous conducting path to be established. These kinks should influence the transport properties of these superlattice structures, contributing to the high resistivity seen in the YBCO layers, and introducing regions of weakened superconductivity. Since the

boundaries of these "kinks" are defined by the barrier layers, the properties of the weak links, in particular the perturbation of phase coherence across the region of weakened superconductivity, should be influenced by the electronic properties of the barriers.

We have also investigated the surface microstructure, determined by scanning tunneling microscopy (STM), in epitaxial YBCO thin films grown by pulsed-laser deposition.³⁴ STM images were obtained using a Nanoscope I STM. No special treatment of the film surface was required, and all of the images were obtained in air at room temperature. The samples were stored in a desiccator prior to being scanned. Figure 5 shows a STM image of a *c*-axis oriented epitaxial YBCO thin film grown at $T_{sub} \sim 680^\circ\text{C}$ on (100) SrTiO₃. Well-defined islands are clearly evident, with each island composed of stacks of terraces. The terrace step heights are multiples of the *c*-axis unit cell height (1.17 nm). The island-like appearance seen in Fig. 5 was common to *c*-axis oriented films, with the most symmetric, well-defined, and largest grains obtained at higher growth temperatures. The terrace width, defined as the distance between terrace steps, is ~ 10 nm, which is the same order of magnitude as the distance between "kinks" in the YBCO layers of YBCO/PrBCO superlattices, as observed by cross-section Z-contrast TEM.^{7,33}

The surface microstructure observed in the *c*-axis oriented thin films may have important implications concerning the transport properties of YBCO/PrBCO superlattice structures. The terraced "roughness" that is present on the growing surface (Fig. 5) will be translated into the individual layers in a multilayered structure. Based on the observed surface

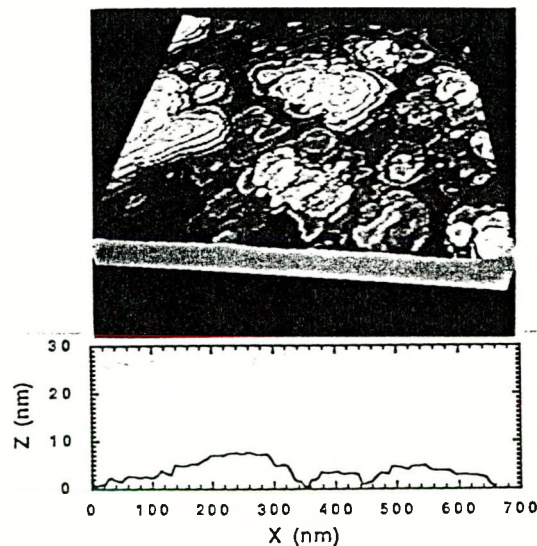


Fig. 5. STM image (upper) and line-scan profile (lower) of a *c*-axis oriented YBCO epitaxial thin film grown on (100) SrTiO₃ at $T_{sub} \sim 680^\circ\text{C}$. The dimensions of the STM image are $670 \times 670 \text{ nm}^2$.

microstructure,³⁴ together with Z-contrast TEM images of the superlattice structures,^{7,33} it appears that a complete theory of the transport properties of YBCO/PrBCO superlattice structures must include the effects of these "kinks," and that perhaps one should regard the YBCO layers as a 2D array of superconducting terraces connected by weak links.

In summary, we have found that T_c (onset) is relatively insensitive to the hole carrier density in the barrier layers of YBCO-based superlattices, suggesting that hole-filling is not a major contributor to the depression of T_c (onset) or T_{c0} , although it cannot be completely dismissed. T_c (onset) appears to depend intrinsically on the YBCO layer thickness, possibly determined by localization effects. On the other hand, the values of T_{c0} (and the transition widths) measured for these superlattice structures are not intrinsic to YBCO layers of a given thickness, but are highly dependent on the boundary conditions and the barrier layer material. Theoretical analyses show that the systematic depression of T_c and the broadening of the resistive transition for the thinnest YBCO layers may be related to a crossover from 3D to 2D behavior.^{11,12} The resistance in the region of the broadened superconducting transitions scales with temperature as expected for the (flux flow) resistance produced by thermally generated 2D vortices^{29,30}, or for a 2D array of superconducting weak links.³² The terraced-island morphology, as revealed by STM, implies that ultrathin YBCO layers in superlattice structures should contain a high density of "steps" or "kinks". These should play a significant role as superconducting weak links in broadening the resistive transition in such multilayered structures.^{7,31}

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