Depression and Broadening of the Superconducting Transition in Superlattices Based on $YBa_2Cu_3O_{7-\delta}$: Influence of the Barrier Layers

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The superconducting properties of $YBa_2Cu_3O_{7-\delta}$-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. T_c(onset) is apparently determined by the $YBa_2Cu_3O_{7-\delta}$ layer thickness, while the transition width, determined by long-range phase coherence of the superconducting wave function, depends on the electronic properties of the isolating barrier layers.

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Several groups recently have reported on the electrical transport properties of high-temperature-superconducting/semiconducting superlattices, including $YBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta}$ [1-4], $Nd_{1.83}Ce_{0.17}CuO_\delta/YBa_2Cu_3O_\delta$ [5], and $Bi_2Sr_2Ca_{0.85}Y_{0.15}Cu_2O_8/Bi_2Sr_2Ca_{0.5}Y_{0.5}Cu_2O_8$ [6] structures. Whereas the midpoint transition temperatures of Bi-2:2:1:2-based structures are essentially independent of the superlattice period, the superconducting properties of $YBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta}$ (YBCO/PrBCO) superlattices are a function of both the superconducting (YBCO) and the barrier (PrBCO) layer thicknesses d. T_c decreases as the YBCO layer thickness is decreased or as the PrBCO layer thickness is increased, but for all YBCO layer thicknesses, including layers one unit cell thick, the superconducting transition temperatures saturate 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 temperatures T_{c0} are ~ 19, ~ 54, 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 $d_{PrBCO} > 5$ 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.

Several explanations appear possible for the resistive transitions observed in these superlattice structures, including proximity effect [7], localization effects [8], and hole filling [9] (to be described later). However, fundamental questions concerning the role of the barrier layers in these structures must be answered experimentally before specific mechanisms can be considered. In particular, are the transition temperatures and transition widths for ultrathin YBCO layers, isolated by relatively thick PrBCO layers, intrinsically determined by the YBCO layer thickness, or are they largely determined by the electronic properties of the isolating matrix? In this study, we have determined that the superconducting transitions of YBCO-based superlattice structures indeed do depend on the electronic properties of the barrier layers, focusing specifically on the mobile hole concentration (resistivity) of the barrier layers. Superlattice structures were fabricated using three different barrier layer materials, namely, $PrBa_2Cu_3O_{7-\delta}$ (PrBCO), $Pr_{0.7}Y_{0.3}Ba_2Cu_3O_{7-\delta}$ (PrYBCO), and $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ (PrCaBCO). By systematically varying the hole concentration of the barrier layers used to isolate ultrathin YBCO layers, we have shown that, surprisingly, the superconducting transition width depends on the carrier density in the barrier layer, while T_c(onset) does not, contrary to predictions of the hole-filling model.

The c-axis-perpendicular superlattice structures were fabricated using *in situ* pulsed-laser ablation, as has been described elsewhere [3]. We 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. Figure 1 shows the resistivity of c-axis-perpendicular epitaxial films of the three barrier layer materials. 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 more complete description of the superconducting properties of the PrCaBCO thin film system has

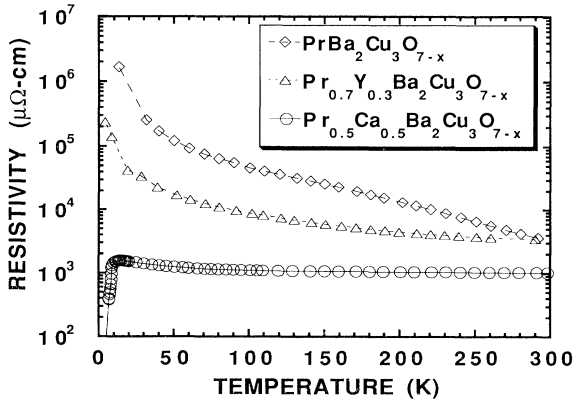

FIG. 1. Resistivity for c-axis-perpendicular, epitaxial thin films of $PrBa_2Cu_3O_{7-\delta}$ (◇), $Pr_{0.7}Y_{0.3}Ba_2Cu_3O_{7-\delta}$ (△), and tetragonal $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ (○) grown on (100) $SrTiO_3$.

been reported elsewhere [10].

The crystallinity of the superlattices was studied utilizing four-circle x-ray diffraction. The structures were grown on (100) $SrTiO_3$ and were fully epitaxial with c-axis-perpendicular orientation. For the YBCO/PrCaBCO structures, the degree of orthorhombic ordering tends to decrease as the thickness of the PrCaBCO barrier layers increases, in agreement with the near-tetragonal structure for c-axis-perpendicular PrCaBCO films [10]. For all of the structures, superlattice satellite peaks are present.

Figure 2 shows the $R(T)$ behavior for 1×16 and 2×16 superlattices with either PrBCO, PrYBCO, or PrCaBCO utilized as the barrier layer material. The resistive transitions of the superlattices clearly depend on the carrier density of the barrier layers. 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(onset) is not significantly influenced by the hole concentration in the barrier layers. This is more clearly seen in Fig. 3 where T_c(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(onset) is insensitive to the barrier layer composition, depending only on the thicknesses of the YBCO and barrier layers.

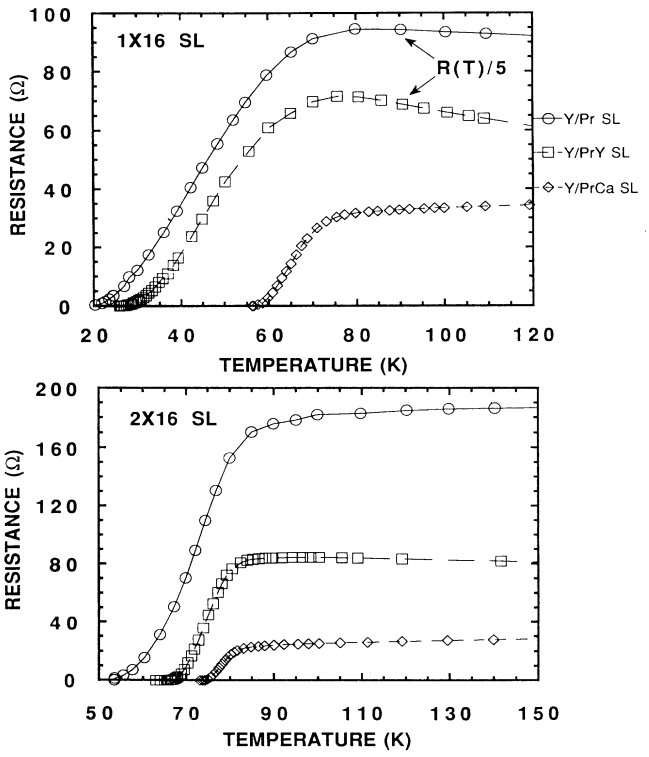

FIG. 2. $R(T)$ for 1×16 and 2×16 YBCO/PrBCO (○), YBCO/PrYBCO (□), and YBCO/PrCaBCO (◇) superlattice structures.

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 [9]. Hole filling was previously used to explain the suppression of T_c as Pr is substituted into superconducting "1:2:3" 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 [11,12]. In YBCO/PrBCO superlattice structures, ultrathin (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 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. 2 and 3. The hole-filling model predicts that a significant increase in the hole density in the barrier layers should lead to increased hole concentration in the

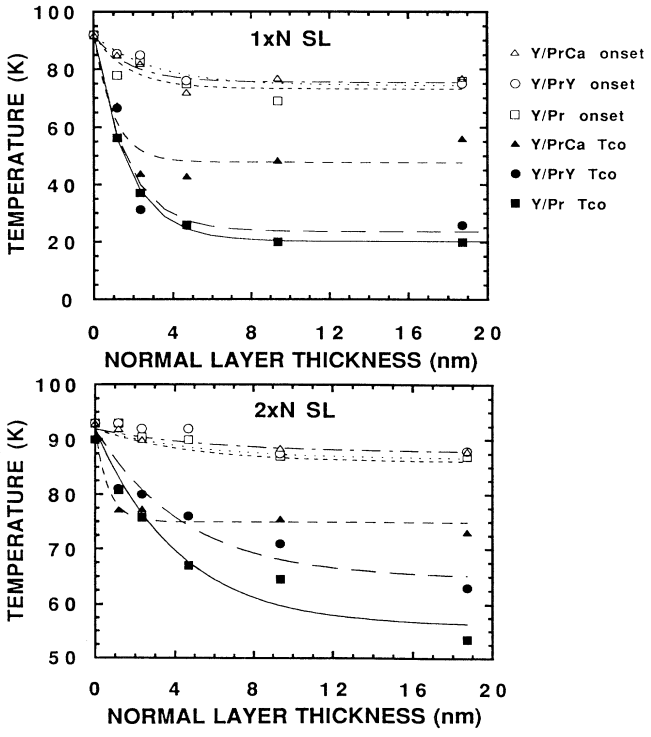

FIG. 3. T_c(onset) (open symbols) and T_{c0} (solid symbols) as a function of normal layer thickness for $1\times N$ and $2\times N$ YBCO/PrBCO (□,■), YBCO/PrYBCO (○,●), and YBCO/PrCaBCO (△,▲) superlattice structures.

superconducting YBCO layers and 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 [13]. 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.

In these superlattice structures, T_{c0} and T_c(onset) are both depressed with only T_{c0} (the transition width) sensitive to changes in the barrier layer carrier density. This observation suggests that one needs to consider separately the physics that determines a depression of T_c(onset) and a broadening of the transition width. A similar reduction of T_c(onset) and/or an increased transition width has been observed for many conventional low-T_c superconducting systems as the superconducting layer thickness is reduced [14–17]. The reduction of T_c(onset) is associated with a suppression of the superconducting pair wave function amplitude while the broadening involves a lack of phase coherence. The reduction in T_c(onset) for low-T_c ultrathin films has been explained in terms of the proximity effect [7], in terms of localization in disordered 2D systems [8], and in terms of the boundary conditions for the order parameter within the Ginzburg-Landau free-energy expression [18]. 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 significantly larger than the normal-metal electronic coherence length in the semiconducting PrBCO. (This coherence length should 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 [ρ_{YBCO}(100 K) = 400 $\mu\Omega$ cm, T_c(onset) = 76 K] and the 2×16 [ρ_{YBCO}(100 K) = 300 $\mu\Omega$ cm, T_c(onset) = 87 K] structures follow this trend. Strain effects could be important as well.

Although some depression of T_c(onset) occurs as a function of layer thickness in the YBCO-based superlattice structures (Fig. 3), significant broadening of the superconducting transition is observed as well. In general, the transition width represents a lack of long-range phase coherence of the superconducting order parameter. Transition-width broadening has been observed in ultrathin films of low-temperature superconductors, and attributed to the presence of an array of weakly in-plane-coupled Josephson junctions and possibly to 2D vortex-antivortex pair unbinding (Kosterlitz-Thouless transition) [19–22]. For very thin films with highly two-dimensional character, the formation of vortex-antivortex pairs and the breaking of these pairs lead to a broad superconducting transition. In order to observe this Kosterlitz-Thouless (KT) transition, the superconducting thin film should have a sheet resistance greater than 1 $k\Omega$/sq [23]. For the YBCO/PrBCO 1×16 superlattice structure, the sheet resistance for an isolated YBCO layer is R_{sq}(100 K) > 3.5 kΩ/sq. Rasolt, Edis, and Tešanović have recently shown that the $R(T)$ behavior expected for a KT transition correctly describes the measured $R(T)$ behavior for 1×16, 2×16, 3×16, and 8×16 YBCO/PrBCO superlattices [24]. Additional experiments (e.g., I-V measurements) are obviously needed to confirm a Kosterlitz-Thouless transition. If a 2D KT transition is used to describe these superlattice structures, one must resolve how the superconductor-normal interface boundary conditions, specifically adding carriers to the barrier layers, significantly affect the temperature T_{KT} where unbound vortices introduce a finite resistance. Note that evidence for a Kosterlitz-Thouless transition in YBCO thin films and single crystals has recently been reported [25,26].

Similar results would be expected for films with a granular nature, consisting of an array of superconducting islands connected by weak links [20,27]. 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 is provided by Z-contrast TEM images, as shown in Fig. 4. Figure 4(a) shows an image of a nominally 1×8 YBCO/PrBCO superlattice; the six dark vertical bands are the YBCO layers, and the wider, light bands are the PrBCO layers. Note that the YBCO layers are not perfectly flat relative to the lattice image; 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, as was shown in recent scanning tunneling microscopy images of YBCO thin films [28–30]. 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

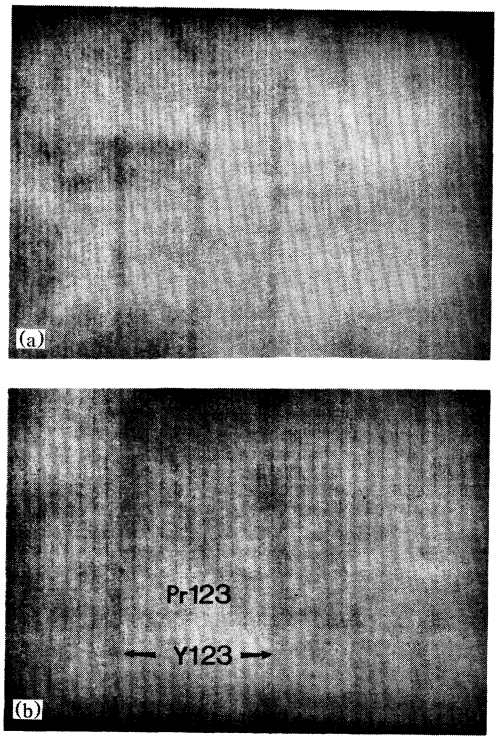

FIG. 4. Z-contrast TEM image of a 1×8 YBCO/PrBCO superlattice structure. The six narrow, dark bands in (a) are the YBCO while the wider, light bands are the PrBCO.

properties of the barriers.

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. Although additional measurements are needed, the results thus far for the broadening of the transition are consistent with a 2D vortex-antivortex unbinding (Kosterlitz-Thouless) formalism. However, the presence of kinks in the YBCO layers should be important in the broadening of the superconducting transition, especially for the 1×16 structures, and must be considered.

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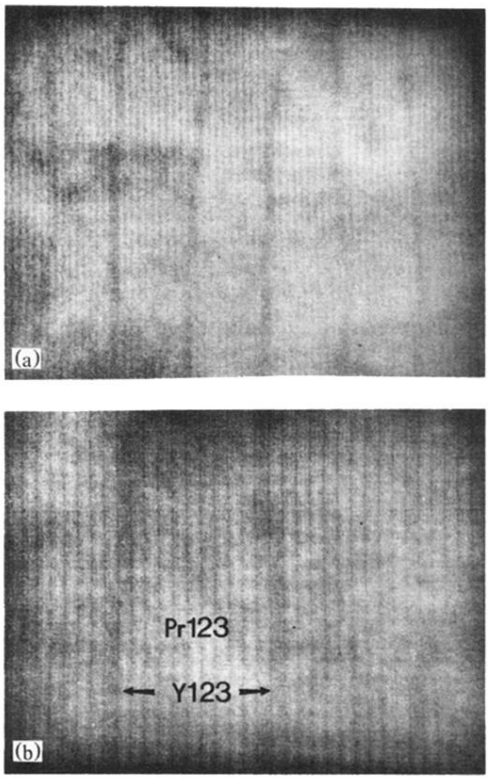

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