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David P. Norton and Douglas H. Lowndes

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Solid State Division
Oak Ridge National Laboratory
P. O. Box 2008
Oak Ridge, Tennessee 37831-6056

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David P. Norton and Douglas H. Lowndes
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ABSTRACT

We report on the superconducting properties of ultrathin *c*-axis-oriented $YBa_2Cu_3O_{7-\delta}$ (YBCO) layers grown by pulsed-laser deposition. For single YBCO layers embedded in a $PrBa_2Cu_3O_{7-\delta}$ matrix, with a YBCO layer thickness as small as one unit cell, there is evidence of a superconducting transition. We also find that the superconducting properties of ultrathin YBCO layers can be significantly enhanced by use of more conductive buffer and cap layers. In particular, $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$/YBCO/$Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ ultrathin structures have transport properties superior to $PrBa_2Cu_3O_{7-\delta}$/YBCO/$PrBa_2Cu_3O_{7-\delta}$ including an increase in T_c and a narrower superconducting transition.

INTRODUCTION

It has been recognized that the superconducting properties of $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films are altered as the film thickness is decreased. In particular, isolated YBCO layers in trilayer and superlattice structures show a significant departure from bulk properties, with a decrease in the superconducting transition temperature, T_c, as well as broadening of the transition, as the YBCO layer thickness approaches a single *c*-axis unit cell.[1–10] Transmission electron microscope and x-ray diffraction data for YBCO-based superlattices suggest sharp interfaces between the adjacent layers, with little or no evidence for interdiffusion, making it possible to study the properties of YBCO layers as thin as a single *c*-axis unit cell (1.17 nm).[11-13] However, unit-cell-high steps do result from the terraced island growth mechanism, and is believed to affect the electrical continuity of the thinnest YBCO layers.[12, 14–16] Recent studies of the effects of an applied electric field on ultrathin YBCO layers suggest that device structures based on ultrathin YBCO layers may be of interest.[17–19]

Thus, it is important to understand, as well as to improve, the superconducting properties of these ultrathin structures.

RESULTS AND DISCUSSION

In this paper, we report on the superconducting properties of ultrathin YBCO layers that are sandwiched between $PrBa_2Cu_3O_{7-\delta}$ (PBCO) cap and barrier layers, as well as on the enhancement of superconducting properties that occurs when more conductive buffer and cap layers are utilized. In particular, structures in which $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ (PrCaBCO) is used for the buffer and cap layers, surrounding YBCO layers as thin as a single unit cell (1.17 nm), have superconducting properties superior to similar structures employing PBCO for the buffer and cap layers, with T_c(onset) increased and the transition width narrowed. An interpretation of these results is given. In addition, the stability of ultrathin YBCO single-layer structures is addressed via observations of the degradation of their superconducting properties with time.

Ultrathin, single-layer YBCO structures were grown by pulsed laser deposition (PLD) as has been described elsewhere.[3,5] A pulsed KrF excimer laser beam [~350 mJ, 38ns full-width half-maximum (FWHM) pulse duration] was focused to a horizontal line on a ~25 mm diameter rotating target. The focused energy density was 2.5–3.0 J/cm^2. The line focus was scanned vertically to yield uniform films on vertically mounted substrates. The heated substrates were placed 6.5 cm from the pressed targets. Film growth was carried out at a substrate temperature of 680°C (calibrated by infrared thermometry) in an oxygen pressure of 200 mTorr. After growth, the samples were cooled at 10°C/min in 1 atm O_2 with 30 min anneals at 625 and 550°C. No in situ monitoring, such as RHEED, was utilized in the growth process. Nominal film thicknesses were determined by calibrating the growth rate using profilometer measurements of thick film structures. (100) $SrTiO_3$ substrates were utilized. Dc transport measurements were made utilizing a standard four-point technique. The samples were stored in a dry dessicator between measurements.

Several groups have studied the growth and properties of ultrathin YBCO layers with varied results, depending on the growth technique and the degree to which in situ monitoring of the growing surface was performed.[6–10] There is general agreement that a superconducting transition can be observed in a two unit cell thick YBCO layer with PBCO buffer and cap layers. However, a complete superconducting transition has been observed in such trilayers with a YBCO layer thickness of one unit cell only when in situ monitoring of the growing crystal surface was performed, usually employing

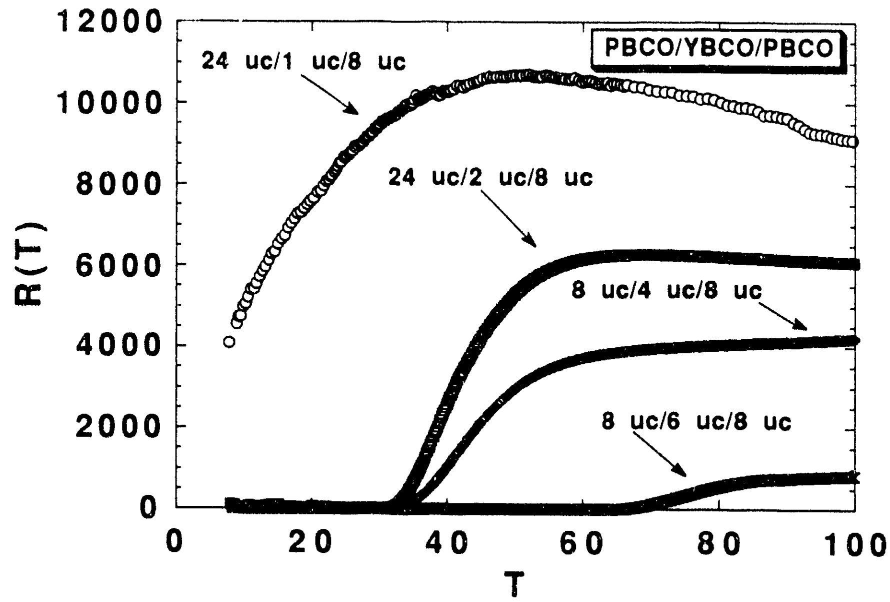

Fig. 1. R(T) data for PBCO/YBCO/PBCO ultrathin structures containing 1, 2, 4, and 6 unit cell (uc) thick YBCO layers grown by pulsed laser deposition.

Reflection High Energy Electron Diffraction (RHEED).[9] Figure 1 shows results that we obtained for ultrathin YBCO structures grown by pulsed laser deposition using PBCO buffer and cap layers. T_c decreases as theT_c(onset) ~55 K and T_c(R=0) ~30 K. For the one unit cell thick YBCO structure, a clear transition onset is observed, but the transition is incomplete down to 8 K. This probably indicates defects in the YBCO layer. One possible origin for the incomplete transition is that the growing surface is not atomically flat, but consists of terraces with *c*-axis unit-cell-high-steps. These steps are seen in both Z-contrast scanning transmission electron micrographs of YBCO/PBCO superlattices, as well as in scanning tunneling microscope images of epitaxial YBCO films.[12,14–16,20,21] Such a "step" in a one unit cell thick YBCO layer can be thought of as a defect which is 1.17 nm in height, larger than the *c*-axis coherence length (~2–3 Å). A high density of these steps could lead to a lack of phase-coherence across the sample, even though the individual YBCO layer thickness is decreased, with a complete superconducting transition observed for a YBCO layer as thin as two unit cells with terraced islands (grains) may be superconducting. This would result in a transition onset with no zero-resistance state, as is observed macroscopically.

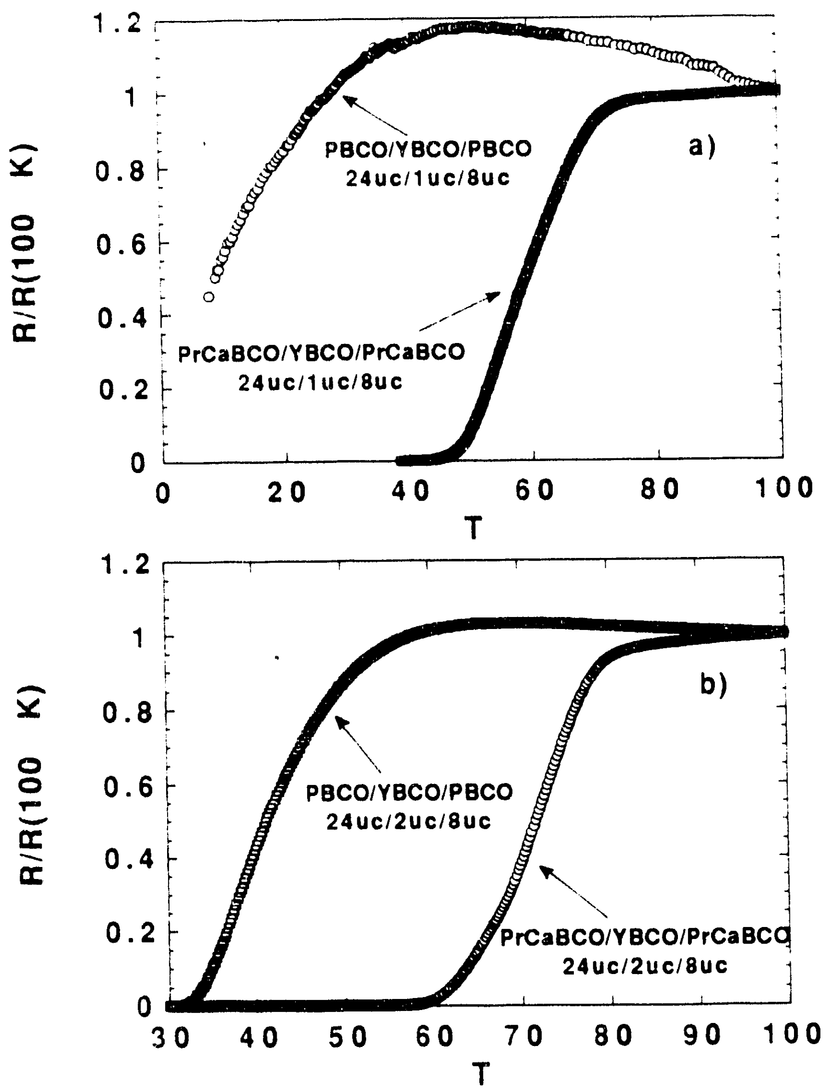

Fig. 2. R(T)/R(100 K) data for ultrathin structures containing a) 1 unit cell thick and b) 2 unit cells thick YBCO layers. This clearly shows the enhancement in the superconducting properties when $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ is used instead of $PrBa_2Cu_3O_{7-\delta}$ as the buffer and cap layer material.

A significant enhancement of superconductivity in ultrathin YBCO layers is observed when the PBCO buffer and cap layers are replaced by Ca-doped PBCO layers. Ca-doping of PBCO introduces additional hole carriers, resulting in higher conductivity and, in certain cases, superconductivity.[22] By replacing the PBCO with $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ in ultrathin trilayer structures, we observe a significant improvement in superconducting properties, as seen in Fig. 2. For the two unit cell thick YBCO structure, T_c(onset) increases

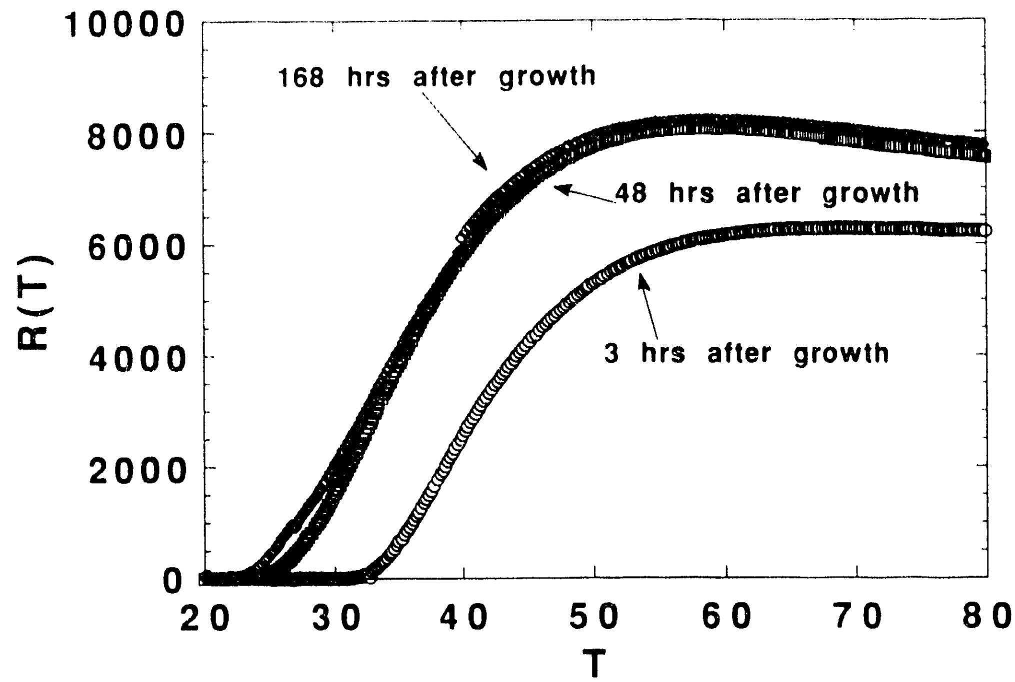

Fig. 3. R(T) for a 24 uc/ 2 uc/ 8 uc PBCO/YBCO/PBCO structure showing the aging effects for ultrathin YBCO layers.

from 55 K to 75 K and T_c(R=0) increases from 30 K to 57 K. For the one unit cell thick structure, T_c(onset) increases from 32 K to 66 K and a full superconducting transition occurs at T_c(R=0) ~ 45 K. It is interesting to note that these results differ somewhat from similar experiments on YBCO/PrCaBCO superlattices in that an increase in T_c(onset) is observed for the single layer structures, but not for the superlattices.[5] We also note that the *c*-axis PrCaBCO layers used as the buffer and cap layers are not superconducting, as the growth conditions used produce metallic but non-superconducting PrCaBCO layers.[22]

As mentioned earlier, Z-contrast STEM and STM studies show that a PLD YBCO film surface is not atomically flat, but consists of terraces with step heights which are, in most cases, a *c*-axis unit cell high. An ultrathin YBCO film deposited on this surface contains steps that become "kinks" in the continuity of the ultrathin YBCO layer. Conduction at these "kinks" requires transport along the *c*-axis in order to establish electrical continuity between adjacent terraces. One would expect that a weak link is formed at these "kinks" since the length of the "kink" is larger than the *c*-axis coherence length. The boundary conditions at the "kink" are determined by the buffer and cap layers. One might view the "kink" as the weak link of a S-N-S junction, with the properties of the normal (N) region determined by the buffer and cap layers. For such a junction, it is known that the coherence length in the normal region increases with the carrier density in the normal region. As more carriers are added to the normal region, the normal-metal coherence becomes larger, and the perturbation the superconducting order parameter by the weak link diminishes. This is consistent with what we observe for ultrathin YBCO layers: The "kink" defects become less effective as more carriers are added to the normal buffer and cap layers. If the density of hole carriers is sufficiently high in PrCaBCO, then this normal metal becomes a superconductor itself.[22] Thus, adding carriers to the weak links enhances the superconducting order parameter in the weak links, resulting in more bulk-like properties.

In addition to studying the effects of the buffer and cap layers on the superconducting properties of ultrathin YBCO layers, we also have investigated the stability of these structures against degradation of their electronic properties with time. Figure 3 shows the R(T) behavior for a 24 uc/2 uc/ 8 uc PBCO/YBCO/PBCO ultrathin structure 3 hrs, 48 hrs, and 168 hrs after film growth. The sample was stored in a dry dessicator between measurements. A 20% increase in the resistivity at 80 K and a decrease in T_c(R=0) from 30 K to 22 K were observed after aging in a dessicator for 48 hrs. However, additional aging appears to have little effect on transport properties. The initial resistivity increase, coupled with the decrease in T_c, suggests a decrease in the hole carrier density on the CuO_2 planes. Whether this is associated with some loss or ordering of the oxygen on the oxygen plane or chain sites is unclear. Applications seeking to exploit the properties of ultrathin YBCO layers in device structures may need to consider whether this instability is intrinsic, or can be circumvented. It is interesting to note that the opposite effect (increase in T_c and decrease in resistivity) has been observed upon aging of YBCO single crystals.

In conclusion, we find that the use of conductive $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ buffer and cap layers significantly enhances the properties of ultrathin YBCO layers, with T_c(R=0) ~ 45 K for a one unit cell thick YBCO layer grown by pulsed laser deposition with no in situ diagnostics of the growing film surface. This T_c enhancement is attributed to an increase of the carrier density within the weak link "kinks" in the continuity of ultrathin YBCO layers. These "kinks" result from unit-cell steps on the growing surface of the PBCO buffer layer. The increased carrier density results in a diminished perturbation of the superconducting order parameter at the weak links, thus enhancing phase coherence across the sample.

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