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Variation of the transition temperature in strained epitaxial $Y_{1-x}Pr_xBa_2Cu_3O_7$ ultrathin layers

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Ultrathin films of $Y_{1-x}Pr_xBa_2Cu_3O_7$ ($x=0,0.2,0.3$) were deposited on perovskite $La_{1.95}Sr_{0.05}CuO_4$, $Pr_{1.95}Ce_{0.05}CuO_4$, and $Pr_xBa_2Cu_3O_7$ buffer layers for comparing the effects of epitaxial strain on the superconducting transition temperature (T_c). The resulting changes in T_c demonstrated that in-plane compressive strains improved the superconductivity. The T_c was higher for $Y_{1-x}Pr_xBa_2Cu_3O_7$ films grown on $Pr_xBa_2Cu_3O_7$ than for films grown on $Pr_{1.95}Ce_{0.05}CuO_4$, which indicates that the variation of T_c in perovskite superconductors is also attributable to factors other than strain, such as interlayer coupling, interlayer strain, and interface roughness.

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I. INTRODUCTION

Separated quasi-two-dimensional CuO_2 planes have been recognized as playing a crucial role in the high-temperature superconductivity of CuO_2-based compounds. As cation substitutions in the CuO_2 planes can produce significant changes in the superconducting transition temperature (T_c), it is believed that interplane coupling is very weak in cuprates. However, the variation in T_c in structures with similar CuO_2 planes—from 135 K in $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Ref. 1) to 20 K in $Bi_2Sr_{2-x}La_xCuO_{4+\delta}$ (Ref. 2)—indicates that other factors should also be considered. The pressure dependence of T_c represents a sensitive probe of superconducting states. A remarkable unexplained feature is the pressure-induced rise in T_c in optimally doped materials; for example, the T_c of Hg 1223 increases from 135 to 160 K upon the application of pressure.[3] High-pressure investigations provide useful information on changes in both coupling and charge-carrier density. However, the anisotropic T_c dependence on stresses along the in-plane and out-of-plane directions can make detailed analysis difficult.

The substrate-induced epitaxial strain in thin films offers a simple way of inducing a strain pattern that is not attainable under hydrostatic pressure.[4] Epitaxial films—especially ultrathin films—deposited on substrates with slightly different lattice parameters will experience in-plane strain. If the lattice parameter of the substrate is sufficiently small, the in-plane epitaxial strain is compressive, whereas a sufficiently large lattice in the substrate introduces tensile strain. For the former case, the in-plane lattice parameters of the film shrink while the out-of-plane lattice parameter c expands due to the Poisson effect (and vice versa for tensile epitaxial strain).

The strain effect on epitaxial $La_{2-x}Sr_xCuO_4$ films has been studied previously using $SrTiO_3$ (STO) and $SrLaAlO_4$ (SLAO) as substrates and buffer layers.[4–7] The lattice parameter of STO is $a=0.391$ nm, which is 3% larger than the in-plane lattice parameter of bulk $La_{1.85}Sr_{0.15}CuO_4$ ($a=0.378$ nm). Hence, $La_{2-x}Sr_xCuO_4$ films deposited on STO experience tensile strain and commonly have a T_c lower than that of bulk samples. In contrast, a higher T_c of 49.1 K was reported in epitaxial thin films of $La_{1.9}Sr_{0.1}CuO_4$ deposited on SLAO substrates with a lattice parameter of $a=0.376$ nm.[5] Recently, Bozovic *et al.* showed that T_c in the $La_{2-x}Sr_xCuO_4$ system was extremely sensitive to the oxygen intake,[6] which makes it difficult to characterize the effect of epitaxial strains on T_c in this system.

The epitaxial strain is more significant in very thin films. Because superconductivity can occur in $YBa_2Cu_3O_7$ (YBCO) layers that are only one unit-cell thick,[8] and the transition in YBCO films is not sensitive to oxygen doping and rare-earth-ion doping (e.g., Gd or Ho) on the Y site, the YBCO system presents a good candidate for fabricating ultrathin layers and investigating the effects of strain on T_c.[9,10] Zhai and Chu[11] compared the substrate-induced compressive and tensile strain effects on T_c in epitaxial YBCO films deposited on STO and $LaAlO_3$. Experimental and theoretical results suggest that reorganization of interatomic distances and charge transformations may underlie T_c variations in the thinnest YBCO layers.[9–12]

Chemical substitution of Y by most rare-earth elements leaves T_c almost unaffected at >90 K, except for $PrBa_2Cu_3O_7$ (PBCO) which has the same 1-2-3 structure. Despite extensive experimental and theoretical investigations, the origin of the suppression of superconductivity in PBCO has not been elucidated. In the $Y_{1-x}Pr_xBa_2Cu_3O_7$ (YPBCO) system, the T_c decreases with increasing Pr concentration. Neumeier *et al.*[13] investigated the effects of high pressure on YPBCO at different Pr concentrations. T_c increased with pressure for a Pr content of $x=0$, 0.1, and 0.2, with a much more complicated relationship between T_c and pressure observed for $x=0.3$: T_c decreased rapidly with pressure at higher Pr concentrations. So far no comparison of epitaxial compressive and tensile strain effects on T_c in a YPBCO system has been reported.

Perovskites $La_{2-x}Sr_xCuO_4$ and $Pr_{2-x}Ce_xCuO_4$ have 214-T and 214-T' structures and are known as p- and n-doped cuprate superconductors with CuO_2 planes.[14] In our experiments, 100 nm thick epitaxial $La_{1.95}Sr_{0.05}CuO_4$ (LSCO) and $Pr_{1.95}Ce_{0.05}CuO_4$ (PCCO) films deposited on (001) $LaAlO_3$

substrates had lattice parameters of $a=0.379$ and $c=1.320$ nm, and $a=0.396$ and $c=1.220$ nm, respectively. The *in-plane* lattice parameters of YPBCO ranged from $a=0.3821$, $b=0.3885$, and $c=1.1676$ nm for $x=0$ to $a=0.3882$, $b=0.3911$, and $c=1.1790$ nm for $x=1$. Therefore, ultrathin YPBCO layers deposited on LSCO and PCCO buffer layers will be expected to be under compressive and tensile epitaxial strains, respectively. YBCO deposited on PBCO is a widely studied system. Since the in-plane lattice parameter of YBCO is about 1% smaller than that of PBCO and the two compounds are chemically compatible, YBCO on PBCO would be an interesting system for performing the comparison. In this paper we describe an investigation of T_c in thin YPBCO layers (with $x=0,0.2,0.3$) with different thicknesses deposited on perovskite LSCO, PCCO, and PBCO buffer layers. These buffer layers are not superconductors, but they do contain CuO_2 planes.

II. EXPERIMENTS AND RESULTS

Both buffer layers and YPBCO thin films were grown *in situ* on (100) $LaAlO_3$ substrates by using pulse laser deposition, as described previously.[15] A KrF excimer laser (wavelength of 248 nm) provided an energy density of about 2 J cm^{-2} on the target surface. The targets were mounted on a multitarget holder that could be changed *in situ* as required. The deposition was performed under an oxygen pressure of 30 Pa. The substrate temperature was maintained at 820 °C during deposition of the LSCO and PCCO buffer layers and at 780 °C for growing YPBCO and PBCO. The thickness of the grown layers was controlled by varying the number of laser pulses. The LSCO, PCCO, and PBCO buffer layers were all 100 nm thick. Under these deposition conditions, the LSCO, PCCO, and PBCO single layers were not superconducting. After deposition, the samples were cooled down in 67 kPa oxygen and annealed at 450 °C for 10 min.

The grown films were examined using a Philips x-ray diffractometer with Cu $K\alpha$ radiation. Figure 1 shows a typical x-ray diffraction spectrum of an 80 nm YBCO layer deposited on a $LaAlO_3$ substrate with 100 nm LSCO buffer layers. Only the $(00l)$ peaks of YBCO and LSCO are evident in the θ-2θ pattern [see Fig. 1(a)], which demonstrates that both the LSCO and YBCO layers grew epitaxially with their c axis perpendicular to the substrate surface. Figure 1(b) shows the ϕ scan of a sample with sharp peaks at 90° intervals, indicating that the a-b planes of the YBCO and LSCO layers were well aligned with the substrate lattices. Similar x-ray diffraction results were obtained for an 80 nm YBCO film deposited on a $LaAlO_3$ substrate with a PCCO buffer layer (Fig. 2). The out-of-plane lattice parameters of YPBCO films clearly show expansion and compression when the YPBCO was less than 8 nm thick, as expected from the Poisson effect.

T_c decreased as the YPBCO layers became thinner. Figure 3 shows the resistivity-versus-temperature curves for YBCO layers with different thicknesses deposited on PBCO. The $T_c(R=0)$ values were 45 and 82.5 K for 2.4 and 8.4 nm thick YBCO layers, respectively, which are consistent with previously reported values for ultrathin YBCO films.[16–18]

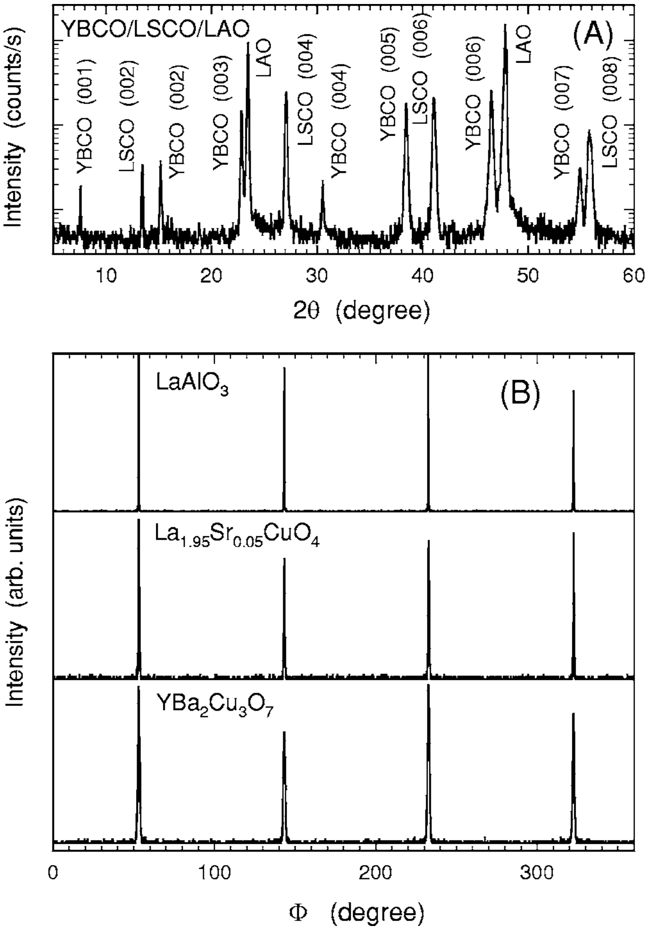

FIG. 1. The x-ray diffraction patterns of a θ-2θ scan (a) and a ϕ scan (b) for YBCO and LSCO epitaxial layers deposited on a $LaAlO_3$ substrate.

To study the epitaxial strain effect, YPBCO ($x=0,0.2,0.3$) layers with different thicknesses were deposited on 100 nm thick LSCO and PCCO buffer layers. For thick YPBCO layers, differences in T_c for films deposited on LSCO and PCCO buffer layers were difficult to distinguish. The thickness dependences of T_c for ultrathin films of YPBCO ($x=0,0.2,0.3$) are shown in Fig. 4. Although the T_c decreased when the YPBCO layer thickness was reduced, it is clear that T_c was higher for ultrathin YPBCO layers deposited on LSCO buffer layers than for those deposited on PCCO buffer. When the film was thicker than 12 nm, the T_c of YPBCO samples approached those found for thick (80 nm) films.

III. DISCUSSION

In-plane epitaxial strain is introduced into a grown film if a lattice mismatch exists, especially in ultrathin films.[11] Consistent with most thin films of cuprates, the superconductivity of the YPBCO system is degraded in thinner films, which is probably attributable to strain effects. We therefore focus our attention on comparing the changes in T_c for opposite strains in ultrathin layers.

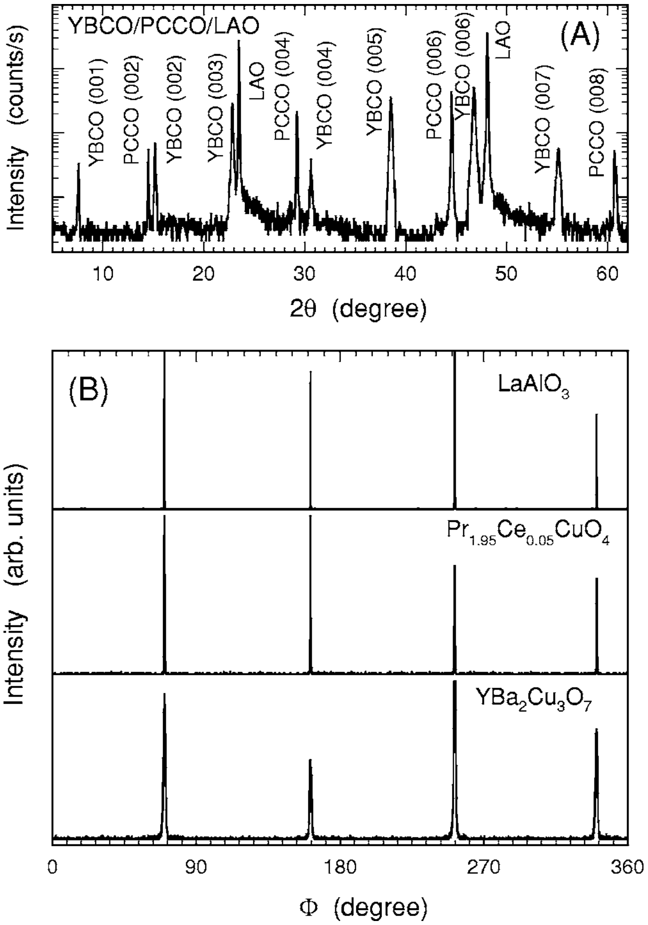

FIG. 2. The x-ray diffraction patterns of a θ-2θ scan (a) and a ϕ scan (b) for YBCO and PCCO epitaxial layers deposited on a $LaAlO_3$ substrate.

The in-plane lattice parameters for the LSCO and PCCO buffer layers were 0.379 and 0.396 nm, respectively. The YPBCO epitaxial films grown on LSCO buffer layers will experience a compressive strain, while films deposited on PCCO buffer layers experience tensile strain. Our x-ray diffraction investigations revealed that the out-of-plane lattice parameters for YPBCO films grown on the LSCO and PCCO buffer layers exhibited expansion and contraction when the YPBCO thickness was less than 8 nm. Both experimental and theoretical data suggest that reorganization of interatomic distances and charge transfer are responsible for T_c changes in strained ultrathin YBCO layers.[9–12] It is evident in Fig. 4 that T_c was higher for YPBCO layers deposited on LSCO than for those deposited on PCCO, although in both cases T_c decreased as the YPBCO layers became thinner. These suggest that a compression in the *a*-*b* planes accompanying an expansion in the out-of-plane direction is advantageous to the superconductivity of an YPBCO system. To test that the relationship between T_c and thickness was not attributable to diffusion across the buffer-film interfaces, $Nd_{1.85}Ce_{0.15}CuO_4$ ($a=0.395$ nm) was used as a buffer instead of PCCO. The lattice mismatch between YPBCO and $Nd_{1.85}Ce_{0.15}CuO_4$ is very close to that between YPBCO and PBCO, but the interface diffusion caused by $Nd_{1.85}Ce_{0.15}CuO_4$ would be more significant. We found that the changes in T_c of YPBCO/$Nd_{1.85}Ce_{0.15}CuO_4$ films were very similar to those in YPBCO/PBCO films, indicating that interface diffusion is not responsible for the variation in T_c demonstrated in Fig. 4.

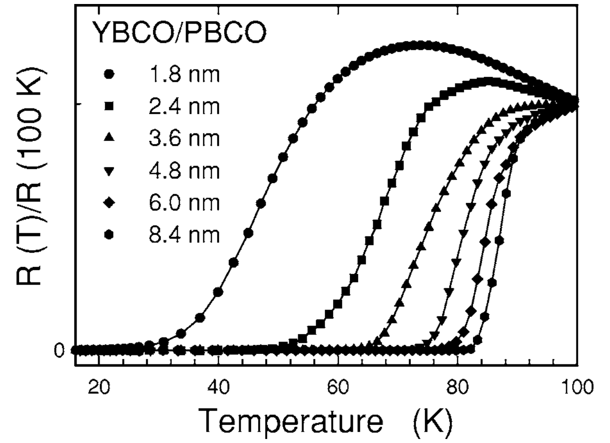

FIG. 3. The resistivity-versus-temperature curves of epitaxial YBCO layers with different thicknesses deposited on PBCO buffer layers.

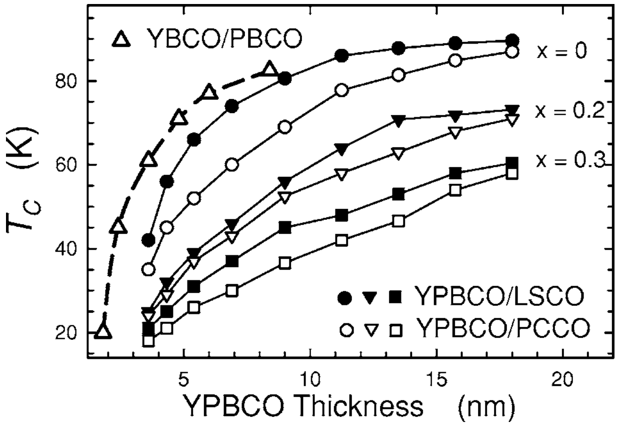

FIG. 4. The dependence of T_c on the thickness of a YPBCO ($x=0,0.2,0.3$) layer deposited on LSCO (solid symbols) and PCCO (open symbols) buffer layers. The open triangles connected with a dash line indicate the T_c of YBCO deposited on PBCO buffer layers.

The above observation is similar to that reported by Zhai and Chu in YBCO thin films deposited on $LaAlO_3$ and STO substrates.[11] In the YBCO system, the anisotropic-uniaxial stress dependence of T_c is given by $dT_c/d\varepsilon_a=-230$ K, $dT_c/d\varepsilon_b=220$ K, and $dT_c/d\varepsilon_c=-18$ K, where $d\varepsilon_a$, $d\varepsilon_b$, and $d\varepsilon_c$ refer to the strains along the *a*, *b*, and *c* axes, respectively.[19] For YBCO thin films deposited on $LaAlO_3$ substrates, the compressive strain is much greater along the *b* axis than along the *a* axis. The T_c of YBCO films deposited on $LaAlO_3$ is higher than those on STO, and the differences in T_c values increase with decreasing YBCO film thickness due to a substrate-induced strain effect. According to the anisotropic-uniaxial stress dependence of T_c, epitaxially strained YBCO films should always show an increased T_c on both STO and $LaAlO_3$ substrates, although such an enhance-

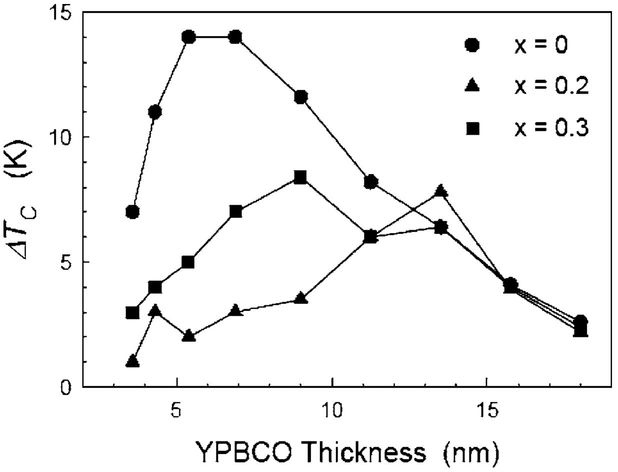

FIG. 5. The dependence of $\Delta T_c(\text{YPBCO}) = T_{c\ LSCO} - T_{c\ PCCO}$ on the layer thickness of YPBCO ($x = 0, 0.2, 0.3$).

ment in T_c was not significant in YBCO ultrathin films and the YBCO/PBCO superlattice. The degradation of T_c with decreasing YBCO film thickness can be attributed to a lack of interlayer coupling and cation disorder.

According to the lattice mismatch, the YPBCO epitaxial films deposited on LSCO or PCCO buffer layers are under compressive or tensile epitaxial strains, respectively. The T_c of ultrathin YPBCO films deposited on LSCO buffer layers are higher than those of films deposited on PCCO. The results demonstrate that in the plan compressive strains are favorite for the high T_c superconductivity. The strains in a film increase when the film thickness is reduced. However, the increase of $\Delta T_c(\text{YPBCO}) = T_{c\ LSCO} - T_{c\ PCCO}$ in YPBCO films is not monotonous (Fig. 5). The T_c of PBCO/YBCO/PBCO sandwiches are higher than that of YBCO films deposited on LSCO buffer, although in the sandwiches the YBCO layers will suffer epitaxial tensile stains in the *ab* plan. These results imply that, apart from the structural strain, other factors may also affect the superconducting transition for ultrathin YPBCO films.

Calculations of the uniaxial strain in YBCO suggested that charge transfer and charge rearrangements induced by the internal strain within the CuO_2 layers determine the strain dependence of T_c.[12] The differing pressure dependence of T_c for YPBCO with differing Pr concentrations may result from a shift of the anisotropic-uniaxial stress dependence caused by Pr substitution. It is interesting to note that although a higher T_c was observed in ultrathin YBCO films with compressive strain deposited on both $LaAlO_3$ substrates and LSCO buffer layers, the ultrathin YBCO films deposited on PBCO had an even higher T_c. It is obvious that the contribution of the interface will increase in importance as the film becomes thinner. Consideration of the lattice parameters of PBCO ($a = 0.3882$, $b = 0.3911$, and $c = 1.1790$ nm) suggests that while strain has an important effect on superconductivity, it is not the only one. Our experimental results suggest that the variation of T_c in high-T_c superconducting thin films is also attributable to factors other than the substrate-induced in-plane strain, such as interlayer coupling, interlayer strain, and interface roughness.

IV. SUMMARY

YPBCO ultrathin layers were grown epitaxially on LSCO, PCCO, and PBCO buffer layers so as to separate the effects of layer thickness and strain. The reduction in T_c with decreasing YBCO film thickness may be due not only to a strain effect, but also partially induced by a lack of interlayer coupling and cation disorder. The higher T_c for YPBCO films deposited on LSCO buffer layers demonstrates that in-plane compressive strain is beneficial to superconductivity. The ultrathin YBCO films deposited on PBCO exhibited a higher T_c than that deposited on PCCO buffer, which indicates that while strain has an important effect on superconductivity, it is not the only one. Our experimental results suggest that the variation of T_c in high-T_c materials is also attributable to factors other than epitaxial strain, such as interlayer coupling, interlayer strain, and interface roughness.

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