Superconductivity suppression and flux-pinning crossover in artificial multilayers of ternary $RBa_2Cu_3O_{7-\delta}$ (R=Gd, Nd, and Eu)

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Superlattices and trilayers consisting of three isostructured 123-type oxide superconductors, Gd123/Nd123/Eu123, were prepared on (100) $SrTiO_3$ using off-axis pulsed laser deposition. Superconducting transition temperatures (T_c) in the superlattices reduce monotonically with decreasing constituent thickness (d), while T_c and transport critical current density (J_c) of the trilayers show no such suppression, being as good as in pure 123 thin films. Individual flux pinning and collective thermal-activated flux motion characterized by log J_c vs log H reveal flux pinning crossovers with field, temperature and layer thickness. Epitaxial-strain-induced thickness effects and correlated defects are applicable to account for the suppressed T_c and flux pinning crossover in the present superlattices.

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All high-T_c superconductors with the exception of $(Ba_{1-x}K_x)BiO_3$ are highly anisotropic due to their natural layered structures.[1–3] An artificially layered system of oxide superconductors has been studied for a fundamental understanding of dimensionality, proximity effect and strain effects.[1–5] Apart from the wide interests in the superlattices of $RBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta}$ ($R$123/Pr123, R:Y or Gd, etc. rare earths),[2,3] superlattices with conducting, weak superconducting and ferromagnetic interlayers, such as Y123/$(Y_{1-x}Pr_x)$123,[1,4] $R$123/$(Y_{1-x}Ca_x)$123,[5] and $R$123/$(R_{1-x}Sr_x)MnO_4$,[6] attract much attention due to the possibility of the artificial modification of CuO_2-CuO_2 coupling. In most cases, these superlattices show a broadening of superconducting transition and a decrease in T_c, while a T_c enhancement effect was also reported.[7,8] Investigations into superlattices consisting of two isostructured high-T_c $R$123 are little explored after an early work with respect to Y123/Dy1123,[9] and no attempt at all has been made on artificial stacking structures of ternary $R$123 with nearly identical T_c, although a similar issue has been addressed for a Bi-based family with three succeeding members.[10]

In the present paper, we report on superconducting properties for the ternary system with sequenced stacks of Gd123/Nd123/Eu123. This selection is motivated by our latest work on the mixed rare earth $(Gd_{1/3}Eu_{1/3}Nd_{1/3})Ba_2Cu_3O_{7-\delta}$ thin film, which produces the enhanced flux pinning.[11] Further studies showed that the stress field is the dominating flux pinning mechanism in such a mixed rare earth thin film.[12] Moreover, we notice the newly reported J_c improvement and structural stability in the bilayers of Eu123/Y123[13] and Gd123/Y123,[14] and the grain-boundary J_c enhancement in the multilayer of Y123/$(Y_{1-x}Ca_x)$123.[5] Thus, in addition to the fundamental insight, we expect that the present study will be able to outline technology potentials, such as the application in coated conductors which emerge from epitaxial growth technology of $R$123 thin films.[15]

A series of multilayers named $(Gd123_d/Nd123_d/Eu123_d)\times n$ (d: identical constituent layer thickness, and n: periodic number), were prepared on single crystal (100) $SrTiO_3$, using off-axis pulsed laser deposition.[16] This technique is advantageous for the elimination of droplets, and the extremely smooth and uniform surfaces of the resultant thin films,[16] which are critically significant factors in achieving high quality superlattices. The details for sample preparation will be reported elsewhere.[17] All samples together with those for comparison (pure $R$123 thin films, and Eu123/Gd123 multilayers), have a similar total thickness of about 125 nm. They are checked by x-ray diffraction and inductive measurements before patterned. Good c-axis orientation and high in-plane texture were identified in all samples by observing the (005) $R$123 rocking curve (FWHM$\sim$0.6°) and (103) ϕ-scan (FWHM$<$2°). Resistivity and transport critical current (using a criterion of $E_c=1.25\times10^{-5}$ V/cm) were measured at various temperatures and magnetic fields (with vectors parallel to the c-axis and normal to current flowing directions) along a bridge patterned by photolithography. Scanning electron microscopy shows very smooth and feature-free surface, also evidenced by atomic force microscopy which gives the RMS roughness as low as 1–2 nm.

Figure 1 shows the x-ray diffraction peaks of various samples including pure Gd123 and Nd123 thin films, a Gd123/Eu123 bilayer, and multilayers of $(Gd123_d/Nd123_d/Eu123_d)\times n$ (n=1,5,10). Due to close lattice constants the diffraction peaks of pure Eu123 overlap with those of Gd123, further evidenced in the bilayer of Gd123/Eu123. In contrast, the trilayer of Gd123/Nd123/Eu123 clearly shows the separation of (007) peaks between Nd123 and Eu123 (or Gd123). Nevertheless, two such separated peaks are absent in Gd123/Nd123/Eu123 multilayers ($n>3$). As seen in the top two patterns, more than three satellite peaks marked with arrows appear as superlattice distinction. A calculation of the modulation wavelength by $\Lambda=\lambda_x/2(\sin\theta_i-\sin\theta_{i-l})$,[18] indicates that our multilayers have relatively large Λ, 1–2 times of each sequence layer thickness (3d). Schuller[18] found that the satellite peaks in Cu/Nb superlattices evolve from strong high-orders to weak low-orders, with increasing each layer thickness. Our results are very consistent with their observation.

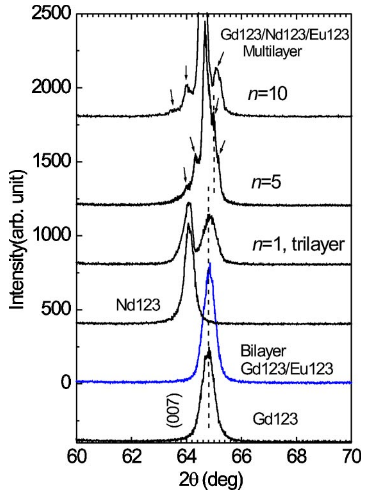

FIG. 1. X-ray diffraction for a series of multilayers $(Gd123_d/Nd123_d/Eu123_d)\times n$ $(n=1,5,10)$, and those for reference: Gd123/Eu123 bilayer and pure Gd123 and Nd123 thin films. Satellite peaks in superlattice are identified with arrows, and two dashed lines are illustrated for an eye-guide.

Figure 2 shows that the T_c varies with constituent layer thickness. Inductive T_c with 90% criterion generally corresponds to the zero resistance T_c appropriately within 2 K, while transport onset T_c is relatively high due to percolation effects. Clearly, both types of T_c reduce with decreasing constituent layer thickness. The transition width shows a slight increase as well. It is obvious that T_c is likely to drop lower than 77 K when $n>10$ (i.e., $d<4$ unit cells). Before a discussion with the mechanism of T_c suppression, we consider a special case, i.e., a trilayer at $n=1$, which gives onset $T_c>92$ K and $\Delta T_c<1.5$ K, as good as in pure *R*123 bulks. It is interesting to note that a number of the trilayers prepared by us appear more reproducible and stable in air than a single *R*123 thin film, easily achieving a higher T_c and narrower transition width. This may be due to the first layer of Gd123 acting as a buffer/seed to release the epitaxial tensile stress between the substrate and Nd123 which has larger lattice misfits. A similar effect in bilayer Eu123/Y123 has been addressed in detail in Ref. 13.

As mentioned above, the reduction in T_c is very common in *R*123/Pr123 superlattices, an *R*123/(LaSr)MnO_4 superlattice, and ultrathin *R*123 films. Several mechanisms have been proposed including the proximity effect,[1,19] Kosteritz–Thouless transition,[20] and interdiffusion and strain effect at interfaces,[3,21] etc. In our case, insulating, or ferromagnetic, or even weak superconducting interlayers are not involved. Modulation structure is stacked with periodic CuO chains-BaO–CuO_2 plane-*R*–CuO–BaO–CuO_2 chains. Only the isovalent *R* ion shifts sequentially. So, we can rule out the effects due to proximity and chemical diffusion. The suppression of T_c also seems unlikely to arise from dirty or highly defected interfaces as the off-axis deposition technique ensures extremely smooth and droplet-free film surface. This is confirmed by TEM observation.[17] In addition, T_c decreases monotonically with decreasing constituent layers from 35.6 u.c.$(n=10)$ to 3.6 u.c.$(n=1)$, regardless of whether it is close to an integer value or not. This implies that the imperfection introduced by the intergrowth is not the origin of T_c reduction. Otherwise, T_c for the interlayer with an integer unit cell (e.g., $d\approx 9$ u.c. at $n=4$) should have a higher T_c, leading to a stepwise change.

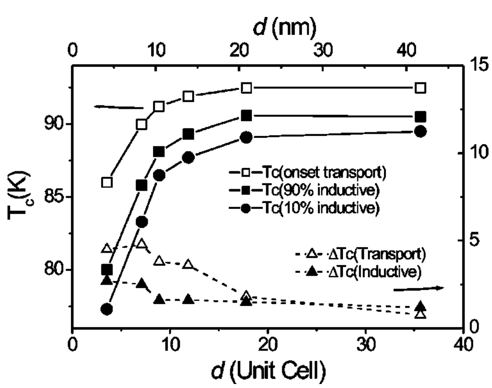

FIG. 2. The variation in transport and inductive T_c, and their transition width ΔT_c with constituent layer thickness d (top label in nm, bottom in number of unit cells).

We now evaluate the in-plane pressure effect caused by the epitaxial strain between stacked layers having different lattice constants. Since Gd123 and Eu123 have smaller lattice constants $(a=0.3897$, $b=0.3838$ nm$)$ than Nd123 $(a=0.3918$, $b=0.3861$ nm$)$,[22] in-plane biaxial tensile stress (to Gd123 or Eu123 layer) or compressive stress (to the Nd123 layer) is expected in our superlattices. Chen *et al.*[23] systematically studied the effect of pressure on T_c for almost all *R*123 superconductors. They found that the pressure derivative dT_c/dP is an increasing function of the R^{3+} radius, originating from the pressure-induced charge transfer from the charge reservoir to the conducting CuO_2 planes. Hydrostatic pressure on high T_c bulk does not affect T_c much because of the compensation of *c*-axis expansion due to the Poisson effect.[21] In contrast, epitaxial strain in thin films is intrinsic or so-called chemical stress, which may have various substantial effects including changes in the growth mode, holes density and related T_c.[3,19,21] Thin films of $(LaSr)_2CuO_4$ grown on different substrates, characterized by different stress tensors, are frequently reported with dramatic changes of T_c.[19,21] Recently, Cao *et al.*[24] studied the effect of epitaxial strain on Gd123 thin films, showing that the difference in T_c can be as large as 4–25 K between substrates of $SrTiO_3$ $(a=0.3906$ nm$)$ and $NdGaO_3$ $(a=0.5428$ nm$)$. One may consider two sheets in elastic contact to estimate the stress using elasticity theory. The tensile or compressive stress (σ) in an interlayer can be written as follows:[19] $\sigma=E_i(\alpha_1-\alpha_2)\delta T\ \delta l/d_i$, where E_i, α_i, δT, δl, d_i are the Young's modulus, thermal expansion coefficient, temperature variation, lattice mismatch, and layer thickness. In our case, $i=1$ (Gd123 or Eu123) and 2 (Nd123). Using $E_i=130$ GPa, $\alpha_1=13.0\times10^{-6}\ K^{-1}$, and $\alpha_2=12.1\times10^{-6}\ K^{-1}$,[23,25] $\delta T\sim 800$ K, and $d_i\sim 1.2$ nm, we reach $\sigma\sim 172$ KPa within this simple framework. Such an order of stress is hardly pronounced for bulk systems.[23] In the case of thin films, however, it may be rather effective to distort the CuO_2 plane as well as the charge reservoir CuO chain, and thus lead to the change of hole density and then T_c. As the spatial distribution of the stress field is attenuated with $1/r^6$, their influence range is rather

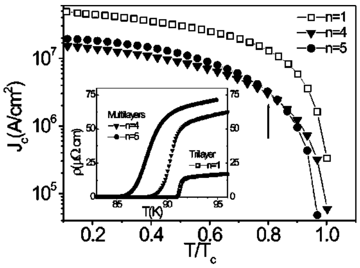

FIG. 3. Critical current density in the zero field vs reduced temperature for three multilayer samples (n=1, 4, and 5). Arrows indicate a crossover between the two superlattices. The inset shows the temperature dependence of the resistivity.

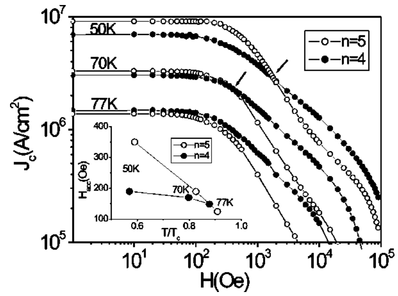

FIG. 4. Field dependence of J_c at 50, 70 and 77 K in a log–log plot. Arrows indicate the crossover of $J_c(H)$ between superlattice samples. The inset is the reduced-temperature dependence of accommodation field H_{acc}.

localized.[26] Accordingly, the T_c decrease directly induced by the stress is spatially limited, and even does not appear in macroscopic measurement when the interface number is low. This is the reason why no T_c suppression is observed in our trilayer sample. Similarly, one cannot attribute the entire T_c suppression directly to the strain effect. Note that the inductive measurement should indicate the superconducting responses of the whole sample. A reasonable scenario is that each constituent layer is electronically separated (depending on temperature) by a decoupling region resulting from epitaxial strains at interfaces. Such a region (rather narrow) makes each constituent layer behave like an ultrathin film. So, the major origin of T_c suppression is probably the thickness effect which can be understood by the Kosterlitz–Thouless theory,[20] or by finite-size scaling relation, $T_c(d) = T_c(\propto)(1-(d/\xi)^{1/\nu})$, where $T_c(\propto)$, ξ and ν are bulk superconducting transition temperature, coherent length and critical exponent, respectively.[27]

To check the above scenario, we prepared a series of single $R$123 thin films with different thickness from 10 to 200 nm. It was found that T_c drops with decreasing thickness (d'), consistent with the case of superlattices with d close to d'. Also the thickness dependence of T_c in Ref. 24 is supportive of our analysis. Moreover, compared with the Varela *et al.* Y123/Pr123 superlattices,[3] our multilayers have similar thickness dependences of T_c in the case of $d < 10$ nm. When the layer thickness is lower than 10 nm, however, T_c suppression in our multilayers is obviously weaker than in their Y123/Pr123 superlattices. This is understandable since the T_c for the latter is affected not only by expitaxial strain, but by superconductivity/anti-ferromagnetism proximity or chemical diffusion.

Next we turn to the flux pinning characteristics by studying the transport J_c at various temperatures and fields. As shown in Fig. 3, J_c for the trilayer sample is outstanding due to its high T_c, while J_c for two multilayers (n=4 and n=5) are nearly 2.5 times lower across a large range of temperatures. For all the three samples, log J_c drops linearly with T at the low-temperature region, consistent with collective-pinning theory.[28] There exists an entangled temperature dependency of J_c for two superlattices, characterized by a crossing at the reduced temperature of ~0.8 K, below which J_c for n=5 is higher regardless of its relatively low T_c. Similarly, there is a crossing $J_c(H)$ relationship. As shown in Fig. 4, the difference in J_c between n=4 and n=5 is not monotonous with temperatures and fields. At 77 K, a relatively high temperature, J_c for n=5 is lower at all range of fields, in agreement with its lower T_c. However, it increases at lower temperatures such as 70 and 50 K, becoming higher than that of n=4 in the regimes of low fields.

To understand the causes of the above crossover behavior, we notice that the individual and collective flux pinning behaviors vary with temperature and field. It is apparent that the field dependence of J_c in the log–log plot is divided into two regimes. At low fields, J_c is nearly independent of applied field characterized by a plateau. In the intermediate field regime, J_c decreases as a power law, and then drops sharply at high fields. A characteristic field termed the accommodation field, H_{acc}, marking the crossover from strong individual pinning to weak collective pinning, is determined from the kink of $J_c(H)$ as observed in numerous oxide superconductors with correlated disorders.[29] The inset in Fig. 4 illustrates the variation of H_{acc} (90% criterion) with reduced temperatures. It is clear that H_{acc} for n=5 increases from 125 to 350 Oe, while H_{acc} for n=4 does not change much when the temperature decreases from 77 to 50 K. The increase in H_{acc} implies the enhancement of correlated disorders (mainly of lines of edge dislocations[30]), which are the dominating pinning sources in the case of a noninteracting vortex at low temperatures and fields. As mentioned before, epitaxial strain at interfaces as well as layer thickness can trigger the variation of growth modes (either 3-D screw growth or 2-D block-by-block), and then the change in the screw or edge dislocations.[3,30] It is likely that due to the decreased layer thickness and increased number of interface, the sample with n=5 has the higher density of edge dislocations formed during the 2-D growth process. Thus, this sample provides a stronger individual pinning force,[31] then a higher J_c at low temperatures and fields than that of the sample with n=4. With increasing fields or temperatures, however, the pinning mechanism becomes vortex–vortex interactions observing the model of thermal activated collective flux motion characterized by an activation energy: $U(J,T) \sim (J_c/J)^{\mu}(1-t^2)^{1/3}$, where μ is a characteristic index of the collective pinning model,[28] and $t = T/T_c$. Due to the lower T_c, the sample of n=5 has a stronger thermal activation effect than that of the sample of n=4. Thus we see a sharper drop of J_c for n=5 at high fields, giving rise to lower J_c, regardless of its higher value at low fields.

In summary, we have prepared a series of $(\text{Gd123}_d/\text{Nd123}_d/\text{Eu123}_d)\times n$ multilayers with a similar total thickness. While the trilayer structure $(n=1)$ shows superconducting properties as good as in pure *R*123 thin films, T_c in superlattices $(n>1)$ decreases with increasing number *n*, i.e., decreasing constituent layer thickness. The T_c suppression is attributed to the thickness effect as the interlayer coupling is probably interrupted by a narrow weak superconducting region incited by epitaxial strain. Flux pinning in superlattices has a crossover feature, i.e., without monotonous dependence of J_c on *d*, or T_c, which can be explained by varying accommodation fields corresponding to different transitions from individual flux pinning at low fields to collectively activated flux motion at high temperatures or/and high fields. Finally it is interesting to note that the trilayer Gd123/Nd123/Eu123 is technologically superior to pure *R*123, which could be applicable to coated conductors where further improvement in processing techniques as well as J_c are being investigated worldwide.

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