Magneto-transport Characteristics of Superconducting $R$Ba$_2$Cu$_3$O$_7$-delta Multilayers and Quasi-multilayers

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Abstract: To study flux pinning modulation due to intentional adjustments of rare earth cations, layer sequences and island-like secondary phases, we built up a series of artificial multilayers consisting of binary rare earth $R$123 as well as quasi-multilayers consisting of Y123/M (M = oxides such as Yttria Stabilized Zirconia) on single crystalline SrTiO$_3$ by pulsed laser deposition. The field dependence of transport critical current density $J_c$ was measured at various temperatures and directions of the applied magnetic field. Comparing with pure Y123 films, almost all present multilayers and quasi-multilayers showed a crossover behaviour of flux pinning, evidenced by an existing medium field ($H_{cro}$) below which $J_c$ is lower than of pure Y123 films, whereas above $J_c$ is higher. Furthermore, a strong temperature dependence of such a crossover field was observed. The higher the temperature, the higher $H_{cro}$, implying that an artificial tailoring of flux pinning may appear in a narrow range of fields as the mixed vortex state moves towards to the region of weak vortex glass and vortex liquid. However, the improvement of flux pinning is hardly observed in the state of strong vortex glass.

1. Introduction

In the last years, great effort had been made to overcome the weak linking at grain-boundary networks of coated conductors based on epitaxial $R$Ba$_2$Cu$_3$O$_{7-\delta}$ ($R$123, $R$ = Y and Nd etc., rare earth) films. The weak linking now, however, appears no longer a major obstacle to achieve high in-field critical current density ($J_c$). It is reported that $J_c$ in high magnetic fields is mainly limited by the intragrain properties, rather than of grain boundaries [1]. Improving flux pinning within the $R$123 grains is therefore becoming significant issue for the development of conductor technology [2-7].

Columnar defects produced by irradiation and antiphase boundaries produced by miscut substrates provide extended linear defects as strong artificial pinning centers [8, 9]. Recently several other artificial routes to increase flux pinning have been realized for $R$123 films, including chemical doping (e.g. to change the initial compositions of PVD targets) [2-4], growth control (e.g. to build up multilayers or incomplete multilayers) [5-7], and substrate decoration (e.g. to introduce nano-scale islands of a second phase onto the substrate) [10-12]. Among them, the growth controlling route is very attractive as it can achieve a high density of second-phase defects up to $10^1$ cm$^{-2}$ [7]. By building up quasi-multilayers of Y123/M (M = single element metal such as Ir and Hf, or oxides), we also achieved the pronounced increase of $J_c$ in high fields [13, 14].

In the present work, artificial quasi-multilayers consisting of Y123/M (M is Y$_2$O$_3$ and Yttria Stabilized Zirconia, YSZ) as well as the multilayers consisting of binary $R$123 are prepared for the investigation into flux pinning modulation due to intentional adjustments of rare earth cations, layer
sequences and island-like secondary phases. The structural and flux pinning properties are investigated in comparison with that of pure Y123 thin films.

2. Experimental

The samples were prepared on (100) SrTiO$_3$ single crystal using pulsed laser deposition with a KrF excimer laser of $\lambda=248$ nm, and a repetition rate of 5Hz. For quasi-multilayers of Y123/M(M=YSZ and Y$_2$O$_3$), YSZ or Y$_2$O$_3$ target was set into position and a certain number of pulses, $n$ (=2 and 10) were done after every $m$ pulses on the Y123 target ($m$ is fixed at 40 for the present study). Other deposition conditions can be found in Ref. 13. This was repeated 70 times. Multilayers of Nd123/Y123 are prepared as well by a similar processing with the target alternations of two targets.

The bilayers and superlattices of Nd123/Y123 are named with NY1 and NdY2, respectively. Corresponding laser pulses and repetition for them are done as typical as 2x(750/750) and 30x(75/25). The growth rates for Nd123 and Y123 are around 1 Å per pulse, higher than the 0.4 Å per pulse for YSZ and Y$_2$O$_3$. Hence, 40 pulses of YBCO give roughly 3.4 unit cells while 2-10 pulses of YSZ and Y$_2$O$_3$ are assumed as incomplete layers of 0.075-0.37 unit cells. The total thickness of superconducting layers is around 280-300 nm for all the present samples.

Microstructure, texture, surface morphology and superconducting transition temperatures are checked by four-circle X-ray diffraction (XRD), atomic force microscopy (AFM) and inductive measurements, respectively. Resistivity and critical current density were measured in various magnetic fields up to 9 T with a Quantum Design PPMS system by standard four-probe method on a bridge of 0.8 mm length and 50 $\mu$m width, patterned by photolithography. The current flowing and external magnetic field were applied always normal to $c$-axis of the samples. The critical current density was determined by using an electric field criterion of $E_c = 1 \mu$V/cm.

3. Results and Discussion

3.1. Structural characterization and heterogeneous phases

As shown in Fig. 1, X-ray $\theta$-2$\theta$ diffraction patterns indicate the c-axis orientation of Y123, not only for two types of quasi-multilayers, but also for the bilayers of Nd123/Y123. In the case of Y123/YSZ, a heterogenous perovskite BaZrO$_3$ is present instead of the original doping phase due to a solid state reaction. This is similar to the case of metallic Ir doped Y123 films, where a heterogeneous BaIrO$_3$ appears [14]. With increasing YSZ pulse number, the intensity of BaZrO$_3$ is obviously enhanced while the $c$-axis texture of Y123 degrades. The quasi-multlayer of Y123/Y$_2$O$_3$, however, has no additional phase emerged except for the original secondary phase of Y$_2$O$_3$. This together with good epitaxial growth and lattice matching, allow Y123 textures hardly affected by Y$_2$O$_3$ doping. For NY2, the XRD patterns show satellite peaks aside the main (00$l$) peaks, which clearly demonstrates the superlattice features in the sample.

As shown in Fig. 2, surface morphology for both pure Y123 films and quasi-multilayers is characterized by scattered large particles with the size over than 50 nm. These large particles are identified as CuO$_x$ by EDX analysis in SEM, consistent with previous observation done by other groups [15, 16]. In contrast, finer particles as small as 10 nm only appear in the quasi-multilayers. They are presumed to be the precipitates of homogeneous Y$_2$O$_3$ or heterogeneous BaZrO$_3$ due to their absence in pure YBCO films. This is further evidenced by the direct observation into SrTiO$_3$ substrate decorated with 2-50 pulses of Y$_2$O$_3$ or YSZ, where finer particles are visible in the size range of 10 nm, nearly same as in the right images of Fig. 2. This is similar to the case of Ir doped multilayers, in which the second-phase BaIrO$_3$ particles have the size of around 13 nm given by a Scherrer estimation [14].
3.2. Superconducting and magneto-transport performance
There are significant differences in \( T_c \) variation between Y123/YSZ and Y123/Y\(_2\)O\(_3\). \( T_c \) slightly changes with \( Y_2O_3 \) doping content, while it decreases greatly with the doping content in the case of Y123/YSZ. As well, the transition width for Y123/YSZ is broadened from 1.5 K to 2.7 K when the pulse number increases to 700, unlike the case of Y123/Y\(_2\)O\(_3\) where the \( ?T_c \) (\( \leq 1.5 \) K) nearly remains as narrow as in pure Y123 films, even for the sample with the highest amount of \( Y_2O_3 \) (i.e. the sample with total pulse number of 1400). Bilayers of Nd123/Y123 show a \( T_c \approx 87.2 \) K, \( ?T_c \approx 1.5 \) K, 1-2 K lower than that of multilayers (NY2), but both have a quite better \( T_c \) than the pure Nd123 film prepared with the same conditions. This may result from the degradation of \( T_c \) in Nd123 sequence of the bilayers due to the elemental exchange between Nd and Ba sites, which seem to be suppressed in the present multilayers,
but emerges in the present pure Nd123. This is obviously interesting, and worthy being further investigated.

![Figure 4](image1.png)

**Figure 4.** Magnetic field dependence of transport $J_c$ of two types of quasi-multilayers, showing the temperature related crossover of $J_c$ with respect to the pure Y123 film prepared with a similar processing condition.

![Figure 5](image2.png)

**Figure 5.** the $J_c$ vs. $H$ of Fig. 4 are plotted in a log-log scale, making crossover behaviour clearer (see the arrows).

![Figure 6](image3.png)

**Figure 6.** Crossover of $J_c$ are also shown in the bilayers and multilayers of Nd123/Y123.

Figure 4 shows the field dependence of critical current density $J_c$ for the Y$_2$O$_3$ and YSZ doped films as well as a pure Y123 film ($n = 0$) at three temperatures, 50 K, 70 K, and 77 K. The sample of $n = 2$ is selected due to its less decrease in $T_c$ compared with the pure Y123. In zero or low fields, $J_c$s for both doped films are lower than that of the pure Y123 films at all given temperatures, which is reasonable due to the decreased $T_c$ in the doped films. In intermediate and high fields, however, $J_c$ is enhanced, giving rise to a crossover field ($H_{cro}$) which becomes lower as the applied temperature goes down. It is more apparent on a log-log scale shown in Fig. 5. Clearly the flux pinning is enhanced in high fields, and such an improvement arises in a wider region of fields as the temperature decreases. For other samples with higher doping contents, the flux pinning enhancement is hardly observed, especially in the case of Y123/YSZ where $T_c$ and c-axis orientation degrade so much due to a chemical reaction. As shown in Fig. 6, one can find the similar crossover of $J_c$ between the bilayers and multilayers of Nd123/Y123.

3.3. Understanding of tailorable flux pinning with respect to vortex diagram
To further understand the flux pinning modification, we recollect the temperature dependence of crossover fields mentioned above into the vortex diagrams, previously reported by us [3]. With the measured irreversibility line $H_{\text{irr}}(T/T_{\text{irr}})$ and accommodation fields, $H_{\text{acc}}(T/T_{\text{irr}})$, the diagram simply divides vortex state of RE123 thin films into three regions, i.e., strong glass, weak glass and vortex liquid. The temperature dependence of crossover fields spits the weak glass region of vortex diagrams, into two parts of with and without the enhanced $J_c$, subject to the type of heterogeneous additions. In the case of $Y_2O_3$, there appears a wide region of $H-T$ with the enhancement of flux pinning. The higher the temperature, the higher $H_{\text{cro}}$, implying that an artificial tailoring of flux pinning may appear in a narrower range of fields as the mixed vortex state moves towards to the region of weak vortex glass and vortex liquid. However, the improvement of flux pinning is hardly observed in the state of strong vortex glass. This needs more investigations to clarify possible mechanism and to suggest potential application for artificial flux pinning.

4. Summary
A series of artificial multilayers and quasi-multilayers are prepared on crystalline SrTiO$_3$ by pulsed laser deposition. Quasi-multilayers of Y123/YSZ and Y123/Y$_2$O$_3$ show different heterogeneous precipitates and superconducting behaviors. Unlike Y$_2$O$_3$, the doping of YSZ results in a pronouncedly decrease of $T_c$ as well as the broadened transition width since a chemical reaction take places, leading to a heterogeneous perovskite phase of BaZrO$_3$. The present multilayers consisting of Nd123/Y123 show a clear characterization of superlattices and a good superconducting transition performance. Both Y$_2$O$_3$ and YSZ doping may lead to a lower $J_c$ in low field, but higher $J_c$ in high fields. Typically a crossover field is observed when they are compared with pure Y123 films. The higher the temperature, the higher $H_{\text{cro}}$, implying that an artificial tailoring of flux pinning may appear in a narrow range of fields as the mixed vortex state moves towards to the region of weak vortex glass and vortex liquid. These results are helpful for the understanding of fundamental pinning properties as well as potential application of artificial flux pinning.

5. References
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