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Flux pinning of stress induced magnetic inhomogeneity in the bilayers of YBa₂Cu₃O_{7-δ}/La_{0.67}Sr_{0.33}MnO_{3-δ}

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Flux pinning of stress-induced magnetic inhomogeneity in the bilayers of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_{3-\delta}$

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Elaborately designed bilayers consisting of epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) and $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_{3-\delta}$ (LSMO) films were fabricated by pulsed laser deposition with respect to the investigation into magnetic-dependent vortex pinning effect. The improvement in the critical current density and a pronounced upward shift in the superconducting irreversibility line based on magnetotransport measurements are observed when compared to the pure YBCO film, suggesting the enhancement in flux pinning in the studied bilayer. It is believed that the improved flux pinning for YBCO arises from the magnetic inhomogeneity of the underlying LSMO. Magnetization measurements show a nonuniform magnetic state in the LSMO film, most probably being macroscopically phase-separated clusters with ferromagnetic (FM) and anti-FM domains, which can be caused by the epitaxial strain due to the lattice mismatch. Such a magnetic disorder is hardly affected by the external magnetic field, unlike the magnetic disorder induced by the domain structures of FM materials where the magnetic domains naturally disappear as the applied field exceeds the coercive field. Hence, the present magnetic flux pinning may be stiff in the large magnetic fields, showing good prospects for artificial flux pinning enhancements. © 2009 American Institute of Physics.

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I. INTRODUCTION

The enhancement in vortex pinning in epitaxial $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (RBCO; *R*: rare earth Y, Gd, etc.) films is very significant not only for basic understanding but also for the technological applications of RBCO coated conductors. In the past few years, several artificial routes to increase flux pinning have been realized for RBCO films, including chemical doping (e.g., to change the initial compositions of physical vapor deposition targets),^{1,2} growth control (e.g., to build up multilayer or incomplete multilayers),^{3,4} and substrate modulation (e.g., to introduce nanoscale islands of a second phase on the substrate).⁵ Despite the diversity in form, all the schemes of pinning enhancement above are based on the idea that the nanoscaled defects or precipitates destroy the superconductivity locally, acting as the attractors for the normal vortex core. The maximal energy per unit length of the vortex is the condensation energy of Cooper pairs in the volume of the vortex core, $U_{\text{cp}} \sim (\Phi_0/8\pi\lambda_L)^2$, where Φ_0 is the flux quantum and λ_L is the London penetration length, $\lambda_L \sim 150$ nm, for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) at 0 K. The intrinsic limitation for vortex core pinning is the rapidly decreasing pinning energy when the temperature approaches T_c due to the increase of λ_L as $\lambda_L(T) = \lambda_L(0)(1 - T/T_c)^{-1/2}$.

Bulaevskii *et al.*⁶ proposed a different flux pinning mechanism by using magnetic domain structures in which the main idea is to pin the whole magnetic flux of the vortex

rather than its core. It is anticipated that the maximum pinning energy for single vortex in superconductor (SC)/ferromagnet multilayer systems may be two orders magnitude larger than the pinning energy by columnar defects.⁶ There have been several attempts on the artificial bilayers or multilayers consisting of SC/FM.^{7,8} Obvious flux pinning improvement is normally observed, but its validity is limited only in very low fields due to the shrinking of magnetic domains as the external field exceeds the coercive field.

In the present work, a different magnetic pinning scheme is proposed. A rather thin film of heterogeneous $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_{3-\delta}$ (LSMO) underlies in the superconducting YBCO film, which probably induces a nonuniform magnetic state due to epitaxial strain. Such a magnetic disorder may be the ferromagnetic (FM)/anti-FM (AFM) phase separation with FM and AFM clusters. Of most importance is that the present magnetic inhomogeneity is an intrinsic disorder, which is insensitive to the external field, and thus the enhanced magnetic pinning may be effective in the large magnetic fields. This is unlike the normal magnetic disorders induced by the domain structures of FM materials where the magnetic domains naturally disappear as the applied field exceeds the coercive field.

II. EXPERIMENTAL

A pure YBCO film of 50 nm in thickness and a YBCO (50 nm)/LSMO (40 nm) bilayer were fabricated on the substrates of single crystal (001) LaAlO_3 (LAO) by using pulsed laser deposition. The laser was fired, respectively, on the

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FIG. 1. (Color online) X-ray diffraction patterns for the bilayer and the pure YBCO film.

stoichiometric targets of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_{3-\delta}$ with a repetition rate of 2 Hz and an energy density of about 2.0 J/cm^2 . The details for sample preparation can be found elsewhere.^{9,10}

The film thickness controlled by the pulse number was measured by UVISSEL/460-VIS-AGAS ellipsometer, indicating that the deposition rates for LSMO and YBCO are about 0.5 and 1 Å/pulse, respectively. Structural measurements were carried out on a four circle x-ray diffractometer (XRD) with $\text{Cu } K_\alpha$ radiation. Magnetization and magnetotransport measurements were performed in a cryostat equipped with a 9 T magnet (Quantum Design PPMS-9T).

III. RESULTS AND DISCUSSION

Figure 1 shows the $\theta \sim 2\theta$ XRD patterns for the bilayer and the pure YBCO film. It is observed that only (00 l) diffraction peaks of YBCO, LSMO, and LAO are present without the peaks corresponding to random crystallographic orientations or secondary phases. This suggests that the LSMO and YBCO layers are c -axis oriented without any impurities.

To give a direct observation for flux pinning performance of a SC, it may be essential to clarify the critical current density (J_c) and its dependences of field and temperature. Figure 2 shows the field dependences of the critical current density for the present two samples at 10 and 77 K, respectively, where the J_c is magnetically evaluated by applying the modified critical state model¹¹ based on the measured magnetic hysteresis loops [see the insets of Figs. 2(a) and 2(b)]. It is revealed that at a relatively high temperature such as 77 K, the bilayer shows a larger J_c in the intermediate region of magnetic field, reflecting the presence of an additional pinning mechanism in the studied bilayer, although such an improvement is absent at a low temperature such as 10 K due to the weak pinning ability and the reduced critical temperature.

The enhanced flux pinning occurring at high temperatures is actually in agreement with the scenario of magnetic pinning. It is expected that the further improvement is feasible at higher temperatures and magnetic fields. However, it is much complicated to understand the extracted J_c and its field dependence behavior based on the magnetic measurements due to the possible vortex liquid driven by thermal

FIG. 2. (Color online) Field dependence of the critical current density. The inset shows the hysteresis loops for both samples at two different temperatures, respectively. (a) 10 K and (b) 77 K.

fluctuation and the entangled contributions of three magnetic signals, which come from the SC YBCO, ferromagnet LSMO, and substrate LAO, respectively. To obtain more useful message on flux pinning and vortex dissipation, the electric resistance broadening effect at external magnetic fields was performed. Figure 3 shows the temperature dependences of electric resistance in various magnetic fields, which are always directed parallel to the c axis of YBCO and perpendicular to the current flowing direction. Compared to the

FIG. 3. (Color online) Temperature dependence of the normalized resistance at various applied magnetic fields. (a) The bilayer. (b) The pure YBCO.

FIG. 4. (Color online) IRLs with the zero resistance criterion of $\rho/\rho_n = 1/10\,000$ in a reduced temperature representation. Solid lines are a fit to $H_{irr} = \alpha(1 - T/T_c)^\beta$.

pure film, the present bilayer exhibits a reduced transition width in the magnetic field, although it shows a larger zero-field transition width of about 1.9 K. Note that the superconducting width ΔT_c is taken by the criterion of 10%–90% ρ_n , where ρ_n is the resistivity in the normal state just above the transition. In the high magnetic fields such as 5 T, the ΔT_c for the bilayer is about 7.4 K, 0.6 K lower than that in the pure YBCO film.

For high temperature SCs, it is generally believed that the broadening of magnetoresistance in magnetic fields arises from the thermal dissipation due to the motion of vortices.¹² The suppressed broadening of magnetoresistance mentioned above implies that the enhanced flux pinning exists in the bilayer of YBCO/LSMO. This is further demonstrated by the irreversibility field and its temperature dependence. Figure 4 shows the irreversibility lines (IRLs) in a reduced temperature scale, determined from the R - T measurements with the criterion of “so-called” zero resistance ($\rho/\rho_n = 1/10\,000$). The IRLs can be well fitted (solid line) by a power law of $H_{irr} = \alpha(1 - T/T_c)^\beta$, with the exponent β of 1.35 for the bilayer and 1.59 for the pure film, respectively. This value is lower than that of $\beta = 2$ predicted by the melting theory,^{13,14} but it can be well understood in the framework of giant flux creep, which gives $\beta = 1.5$.¹⁵ Another parameter α , which in fact denotes the irreversibility field at zero temperature, is given as well. At low temperatures, the H_{irr} for the bilayer appears smaller than that of the pure YBCO. However, the former may increase over more than the latter as the temperature increases, leading to a pronouncedly characteristic, i.e., a crossover behavior existing at a certain reduced temperature such as $T/T_c \sim 0.80$, over which the IRL shifts upward obviously for the bilayer, indicating the enhancement in vortex pinning occurring at high temperatures, as concluded from the magnetic J_c - H performances. The present crossover behavior of IRLs is actually very different from those in the cases of artificial pinning due to nanoparticle additions, where the lower the temperature, the higher the H_{irr} , showing an obvious upward shifting of IRL at lower temperature.^{16,17}

Enhanced flux pinning at high temperatures for the bilayer is considered to come from the magnetic pinning, which will be discussed later. However, it must be pointed

FIG. 5. (Color online) Dependence of the zero temperature activation energy U_0/k_B on applied magnetic field. The solid lines are the best fit with power law relation $U_0 \propto H^{-\alpha}$. The inset is the Arrhenius plot from which the values of the activation energy U_0 are obtained.

out that at low temperatures, the vortex pinning may be still dominated by natural pinning sites originating from the growth-induced threading dislocations, either edge dislocations or screw dislocations.¹⁸ Due to the large lattice mismatch between YBCO and LAO ($a_{\text{LAO}} = 0.3790$ nm and $a_{\text{YBCO}} = 0.3850$ nm), more dislocations are expected in the pure YBCO film when compared with the bilayer, where the misfit between YBCO and LSMO is small ($a_{\text{LSMO}} = 0.3873$ nm). It is supposed that the lower pinning centers and the reduced critical temperature lead to the reduction in the critical current density of the bilayer at low temperatures. In addition, this assumption may be evaluated by the change in pinning energy. Beyond IRL is the region of thermally activated flux flow (TAFF) where dissipative resistance appears. The TAFF theory predicts that in the low current limit, the resistivity obeys the Arrhenius relation $R(H, T) = R_0 \exp[-U(H, T)/k_B T]$, where $U(H, T)$ is the activation energy (i.e., the pinning energy) for the hopping process of a flux line or flux bundle, which depends on the applied magnetic fields and temperatures. If the temperature dependence of the activation energy is linear with the expression of $U(H, T) = U_0(1 - T/T_c)$, where U_0 is the activation energy at 0 K, then $R = R_0 \exp(U_0/k_B T_c) \exp(-U_0/k_B T)$ will be achieved, which means that in the Arrhenius diagram, $\ln R - 1/T$ represents a linear relation in the TAFF region with the slope of U_0/k_B .

For the present cases, the linear relation of $\ln R - 1/T$ dependence is indeed observed below 1% ρ_n ,¹⁹ as shown in the inset of Fig. 5. This suggests that the TAFF model mentioned above is applicable to study the flux motion for the present samples, with which the activation energy at 0 K (U_0) can be extracted. Figure 5 shows the field dependence of the zero temperature activation energy, and the solid line is a fit with the formula $U_0 \propto H^{-\alpha}$. It is revealed that the bilayer exhibits a lower U_0 in all applied fields, demonstrating the much weaker pinning of the sample at low temperatures again. Note that the obtained α value for two types of samples are both near 1/2, consistent with the prediction of Geshkenbein and Vinokur,²⁰ in which a mechanism of plastic

flux creep is proposed to ascribe the dissipation to the plastic shear of dislocations in weakly pinning vortex liquid.

Up to now, we have not yet looked into the enhanced flux pinning at high temperatures as well as the pinned origin in the bilayers. As the temperature increases, the main flux pinning mechanism dominated by vortex core interaction for the studied bilayer at low temperatures is probably replaced by the flux pinning mechanism controlled by magnetic disorders. This may arise from the competitions of three characteristic energies, i.e., the energy of core pinning, the energy of magnetic pinning, and the thermal energy. In the framework of magnetic pinning suggested by Bulaevskii *et al.*,⁶ the estimated maximum pinning energy of a single vortex line is $U_{mp} \sim \Phi_0 M_0 d_s$, where M_0 is the magnetization of FM domain and d_s is the thickness of SC. Suppose $M_0 = 500$ emu, a pinning energy as large as 10^5 K can be obtained. This value is comparable to the pinning energy of the vortex core at 0 K but 2~3 orders larger than that at the temperature close to T_c . For a real system, the magnetic pinning energy is actually smaller than the value estimated as above due to the random distribution of FM domains. This makes the magnetic pinning hardly appear, especially at low temperatures where the vortex pinning is controlled by vortex core pinning. The magnetic pinning, however, may take place at higher temperatures, being remarkable as the temperature approaches T_c because of the insensitive temperature dependence of such a magnetic disorder.

In the conventional SC/FM bilayer or multilayer reported before,⁶⁻⁸ magnetic pinning is caused by the magnetic inhomogeneities such as domain structures, which needs the FM layer(s) exhibiting a strong perpendicular magnetic anisotropy, subject to the coercive field.⁶ Below the coercive field the magnetic domain walls may be pinned, giving rise to the magnetic pinning effect. As the field exceeds the coercive field, however, the pinning may disappear at all. Even for the very hard FM materials such as $\text{BaFe}_{12}\text{O}_{19}$ (the coercive field is 6000 Oe at the direction of the easy axis), the effective improvement in pinning is present only in the magnetic fields of as high as 1 T.⁷ It is believed that the magnetic inhomogeneity in the present bilayer is not caused by the above-mentioned domain structure because the studied LSMO layer is a typical soft magnetic material with the coercive field of only 100~200 Oe even at the low temperatures.

In the following, a macroscopic scenario with the phase separation consisting of FM and AFM clusters is proposed to understand the experimental results achieved. It is well known that the perovskite compound of LSMO exhibits its magnetic performances with a typical double exchange mechanism, in which phase separation is not expected.²¹ For thin films, however, the epitaxial strain induced by the lattice mismatch plays an important role in determining the orbital order of manganite, which may drive the phase away from the usual FM state due to the strong coupling of the orbit and spin dynamics.²² According to phase diagram theories proposed by Konishi *et al.*²³ and Fang *et al.*,²⁴ the LSMO film state may be driven from a FM metallic phase to a C-type AFM insulating phase with the increase in epitaxial stress. Considering the effect of disorder, the complete transition

FIG. 6. (Color online) M vs H curves at 10 K for several LSMO films with different thicknesses. The inset shows the temperature of FC magnetization for a 40 nm thick LSMO film measured at the magnetic field of 2000 Oe, and a magnetization step is illustrated by an arrow.

from the FM to AFM phase may not occur. Instead, the phase separation or coexistence of FM-AFM may appear dominant, especially in the proximity of the transition.²⁵ Recently, Tebano *et al.* reported the strain-induced phase separation indeed occurs in LSMO/LAO films with the thickness range from 100 to 30 unit cells.^{26,27} Their results suggest a coexistence of two stable phases: the orbital ordered chain-type insulating antiferromagnetic (C-type AFM) and the orbital disordered metallic ferromagnetic. As well, our latest studies show the evidence concerning about the nonuniform stress state and high disorders in thin LSMO films not only from x-ray diffraction but also from Raman measurements.²⁸ Moreover, it was observed that the epitaxial strain in thin films may produce substantial effects on various aspects including the growth mode and defect distribution as well as the associated structural and physical properties. In terms of magnetic properties of thin LSMO films, at least two factors should be considered, i.e., the rotation of the easy magnetization axis and magnetic inhomogeneity.

In the framework of magnetic pinning suggested by Bulaevskii, to obtain perpendicular magnetic anisotropy is very indispensable for effective magnetic interaction between vortices and the local field.⁶ To test the anisotropy, magnetization is carried out at the field parallel and perpendicular to the film plane. Figure 6 shows the virgin magnetization curves for the LSMO films with different thicknesses, clearly demonstrating the rotation of easy axis from the direction of in-plane to that of out-of-plane, as the film thickness decreases from 150 to 25 nm. For the 150 nm thick LSMO film, the saturation field in out-of-plane (c axis) is about 8000 Oe, much larger than 2000 Oe for that of in-plane, indicating that the easy axis of the film lies in ab plane. As the thickness reduces, however, it is difficult for in-plane magnetizing. For example, the in-plane saturation field of a 25 nm thick LSMO film may increase to 6000 Oe. The rotation of easy axis of magnetic films, induced by the epitaxial strain, is testified by other's torque magnetometry.²⁹ It has

been shown that LCMO thin films grown under compressive strain show an out-of-plane orientation of the magnetization³⁰ and the out-of-plane components of moments, which are absolutely necessary for magnetic flux pinning.

A clear evidence of FM/AFM phase separation can be seen from the inset of Fig. 6. The inset shows the temperature dependence of the magnetization, measured under the condition of field-cooling (FC) in a field of 2000 Oe. The magnetization step occurs at about 150 K is consistent with AFM transition, over which the AFM order disappears. Thus, as the temperature exceeds 150 K, a much steeper decline in the magnetization is observed as a result of the loss of the pinning on FM moments along the field direction. It is worth noting that similar $M(T)$ behaviors were observed in spin-glass systems³¹ and samples involving a spin-glass phase (FM/SG),³² respectively. However, since the presence of AFM phase in the compressively strained LSMO films was supported by both experiments and theory, it is reasonable to use the phase separation of FM and AFM to explain our results. According to the magnetic force microscope observation done by Sun *et al.*,³³ the coexistence of FM and non-FM regions may be randomly distributed with the typical size of the magnitude of microns similar to the domain width. In reality, it is the stress-induced phase separation that leads to magnetic inhomogeneity and in turn produces the remarkable magnetic pinning effect.

IV. CONCLUSIONS

In summary, we have studied the magnetization and transport behaviors for the bilayer of YBCO/LSMO prepared by the technique of pulsed laser ablation. Compared to the pure YBCO film, the pinning mechanism of the bilayer is dominated by magnetic interaction in the high temperature regime, which gives rise to remarkable enhancement in vortex pinning. The magnetic pinning is believed to arise from the magnetic disorder, which is actually a macroscopically FM/AFM phase separation induced by the epitaxial stress. The magnetic disorder is similar, in some extent, to the decoration by the magnetic dots on YBCO surface but unlike that induced by domain structures, which may disappear at all as the field exceeds the coercive field. This implies that the present improvement in vortex pinning can take place at larger magnetic field and thus may be promising and applicable by using a simple underlying heterogeneous.

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