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YBCO films with Zr4+ doping grown by MOD method

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Abstract

YBa2Cu3O7-delta (YBCO) films with Zr doping have been prepared successfully by the trifluoroacetate metal-organic deposition (TFA-MOD) method through dissolving Zr acetylacetonate into the precursor solution. Yttria-stabilized zirconia(YSZ)nanoparticles were detected in the doped YBCO films by XRD. From the analysis of XRD omega and phi scans, the doped films have better out-of-plane and in-plane textures than those of the un-doped YBCO film. Although the doped YBCO films have lower T-c than that of the un-doped YBCO film, a very significant enhancement of normalized J(c) is displayed as compared to the undoped film at applied fields, indicating that an effective pinning force was created by Zr doping.

Keywords

YBCO, films, Zr4, doping, grown, MOD, method

Disciplines

Engineering | Physical Sciences and Mathematics

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***Abstract*—$YBa_2Cu_3O_{7-\delta}$ (YBCO) films with Zr doping have been prepared successfully by the trifluoroacetate metal-organic deposition (TFA-MOD) method through dissolving Zr acetylacetonate into the precursor solution. Yttria-stabilized zirconia(YSZ)nanoparticles were detected in the doped YBCO films by XRD. From the analysis of XRD ω and φ scans, the doped films have better out-of-plane and in-plane textures than those of the un-doped YBCO film. Although the doped YBCO films have lower T_c than that of the un-doped YBCO film, a very significant enhancement of normalized J_c is displayed as compared to the undoped film at applied fields, indicating that an effective pinning force was created by Zr doping.**

***Index Terms*—Nanoparticle doping, TFA-MOD, YBCO film.**

I. Introduction

FOR practical applications, especially power applications, high temperature superconductors (HTS) such as $YBa_2Cu_3O_{7-\delta}$ (YBCO) need to carry a high critical current density (J_c) under high magnetic fields. However, due to vortex motion, the capability of YBCO to carry currents is significantly reduced at higher applied magnetic fields. There is an effective way to improve the in-field performance of coated conductor by introducing various kinds of pinning centers using a variety of techniques, including interlayers of non-superconducting materials [1], [2], mixed rare-earth doping [3], [4], and doping with self-aligned $BaZrO_3$ (BZO) nanodots and nanorods [5]–[7], as well as the use of nanoparticle-modified substrate surfaces [8], [9]. Among these approaches, BZO nanoparticles [7], [9] grown heteroepitaxially within laser-ablated YBCO films are the most popular, because these particles are easily incorporated into the films from a source target composed of a ceramic BZO/YBCO mixture. Using a pulsed laser deposition (PLD) process, Goyal [7] *et al.* have succeeded in producing long, nearly continuous vortex pins along the c-axis in YBCO, in the form of self-assembled stacks of BZO nanodots and nanorods, which led to an improvement in J_c in magnetic fields. Recently, a new strategy for introducing BZO nanoparticles into YBCO films has also been developed by J. Gutierrez *et al.* [10], [11] on the basis of easily scalable chemical solution deposition techniques. The random crystalline orientation is the essential feature distinguishing chemically prepared nanocomposite films from those prepared through vacuum deposition methods. The quasi-isotropic character of the pinning has demonstrated that this new strategy is very effective in preventing vortex motion at high fields and high temperatures for all magnetic-field orientations. In this work, epitaxial YBCO/YSZ nanocomposite thin films were grown from a non-vacuum, low cost, and easily scalable metal-organic deposition method, and their microstructures and physical properties were investigated.

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II. Experimental

The trifluoroacetate precursor solution was prepared by a standard TFA-MOD process with a cation ratio of $Y : Ba : Cu = 1 : 1.5 : 3$. A Zr acetylacetonate precursor in stoichiometric proportions was added to obtain the TFA precursor solution with 6 mol.% Zr doping. The solution was coated on $10 \times 10\ mm^2$ $LaAlO_3$ single-crystal substrates with (001)-orientation by spin coating at a speed of 4000 rpm for 2 min at room temperature. The wet films were decomposed to amorphous precursor films by slowly heating them up to 400°C in a humid oxygen atmosphere. The amorphous precursor film was heated up to 800°C in humid Ar/O_2 (100 ppm O_2) atmosphere and held for 90 minutes. An oxygenation process was carried out at 550–450 °C for 90 min to achieve the superconducting phase.

X-ray diffraction measurements were carried out to examine the phase of the YBCO films. X-ray ω-scans and φ-scans were used to evaluate the out-of-plane and in-plane textures of the YBCO films, respectively. The film thickness after growth was determined, by profilometer analysis, to be 250–300 nm. The DC magnetization measurements were carried out with a physical properties measurement system (PPMS) in magnetic fields parallel to the c-axis of the specimens. The J_c values of the YBCO films in magnetic fields were determined by application of the Bean critical state model formula, $J_c(H) = 20\square M(H)/a(1-(a/3b))$, where $\square M$ is the vertical width of the magnetization hysteresis loop (emu cm^{-3}), and a and b (cm) are the cross-sectional dimensions of the sample perpendicular to the applied field, with $b \geq a$.

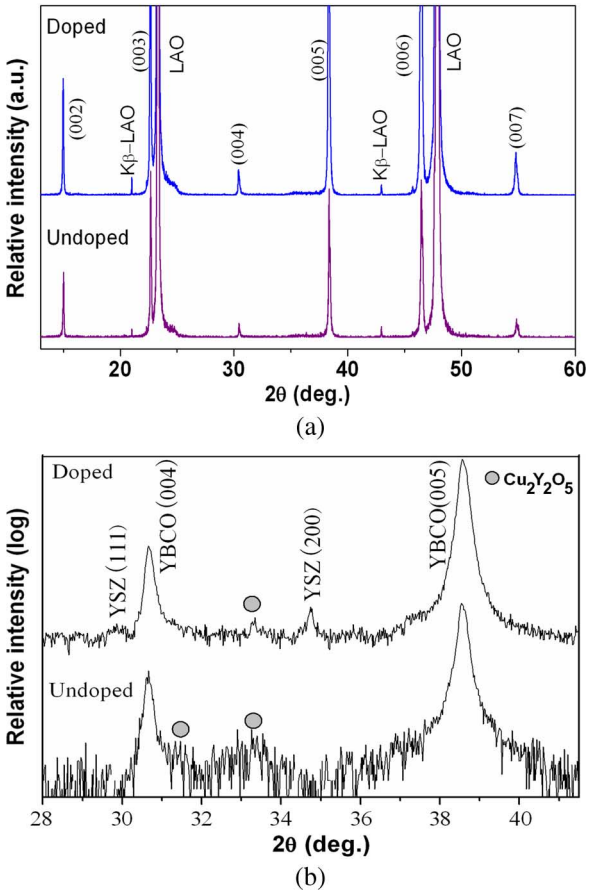

Fig. 1. (a) X-ray diffraction patterns of undoped YBCO film and YBCO films with different doping levels of Zirconium. (b) Enlargement of a section of Fig. 1(a) showing the presence of $Cu_2Y_2O_5$ impurity.

III. Results and Discussion

A. Analysis and Comparison of Phases

Fig. 1(a) shows X-ray diffraction patterns of the un-doped and doped YBCO films. Only (00l) diffraction peaks of YBCO were observed, which indicates a well-textured, c-axis oriented grain structure. In order to obtain further information on phases, XRD of the samples was conducted at a very low scan speed. As shown in Fig. 1(b), $Cu_2Y_2O_5$ impurity was detected in both un-doped and doped YBCO films because of the use of a Barium-poor starting solution, which is consistent with the results for K. Nakaoka [12], [13]. They found that highest J_c value was not obtained at a Ba/Y molar stoichiometric proportion of 2.0, but at a Ba/Y molar ratio of 1.4–1.6 owing to the flux pinning of nano-particles of CuO and $Cu_2Y_2O_5$ in the YBCO grains. So, we chose a Ba/Y molar ratio of 1.5 in both pure and Zr^{4+} doping YBCO coating solution.

We found that Yttria-stabilized Zirconia(YSZ)phase was detected in the YBCO films with Zirconium doping, which is not consistent with other reports [10], [11]. BZO phase was identified in Gutierrez J's YBCO films by using the YBCO precursor solution in stoichiometric proportions with 10%BZO doping [10]. However, the concentration of Ba in our experiment is much less than stoichiometric proportion to form BZO because of both using Barium-poor YBCO solution and only doping Zr salts (not BZO solution) into YBCO solution. So, the decomposition compounds of Zr precursor salt are more difficult to react with Ba compound than with the decomposition compounds of excess Y salt to form YSZ phase. From the full-width at half-maximum (FWHM) of the YSZ (200) and (111) peaks and the Scherrer formula, $(\mathrm{Dc} = 0.89\lambda/(\mathrm{B}\ \cos\ \theta)$; Dc is the size of the YSZ particle; λ is 0.15406 nm, which is the wavelength of the characteristic X-ray from a Cu target; B and θ are the full-width at half-maximum and the diffraction angle of the YSZ (002) and (110) peaks, respectively.) we estimated that the size of the YSZ nanodots was in the range of 10–20 nm. Their dimensions are the order of the coherence length of YBCO, and so they can act as flux pinning centers.

B. Analysis and Comparison of Texture

X-ray diffraction ω and φ scans were collected to determine the out-of-plane and in-plane alignments of the films, respectively. It can be clearly seen from Fig. 2, the FWHM values of the ω and φ scans for the un-doped YBCO films are 0.98° and 1.54°, respectively, which are larger than those of the doped YBCO film. The poor biaxial texture in the un-doped films may result from some $Cu_2Y_2O_5$. impurities which are not epitaxially grown. The $Cu_2Y_2O_5$ content in the doped YBCO films is less than that in the un-doped films so that they have very little effect on biaxial textures in the doped YBCO films. Furthermore, according to the x-ray patterns in Fig. 1(b), the intensity of the YSZ (111) peak is lower than that of the (200) peak, while the theoretical value of the YSZ (111) peak is much higher than that of the (200) peak, indicating that the presence of randomly oriented YSZ nanodots do not destroy biaxial textures in the doped YBCO films.

C. Analysis and Comparison of T_c

The critical temperature of samples was measured via a physical properties measurement system. As shown in Fig. 3, both un-doped and doped films have the same transition width. However, the $T_{c,onset}$ of the doped YBCO film is 1.5 K lower than that of the un-doped film. The lowering of T_c in the presence of the nanoparticles is consistent with other studies done on PLD YBCO films that were doped with Y_2O_3 nanoparticles [14]. The reason for the depressed T_c may possibly be Zr substitutions into Yttrium sites, which are able to locally depress T_c. The further researches are still needed to confirm the accurate reasons of the depressed T_c.

D. Analysis and Comparison of J_c

Devices with YBCO films are usually used at a temperature of 77 K, so we measured the J_c at 77 K using PPMS. Fig. 4(a) shows the field dependence of J_c/J_{c0} at 77 K, with the field parallel to the c-axis. It can be seen that the 6% Zr doped film has a very significant enhancement of normalized J_c values as compared to the un-doped film for all applied fields, indicating that an effective pinning force was created by 6% Zr doping. The overall increase in J_c at all magnetic fields can be more clearly seen when the magnetic-field dependence of the pinning force, $F_p = J_c(B) \times B$, is presented, as shown in Fig. 4(b). Although J_c decreases monotonically with increasing field, F_p increases to a maximum value $F_{p,max}$ at the field of 0.3 T field,

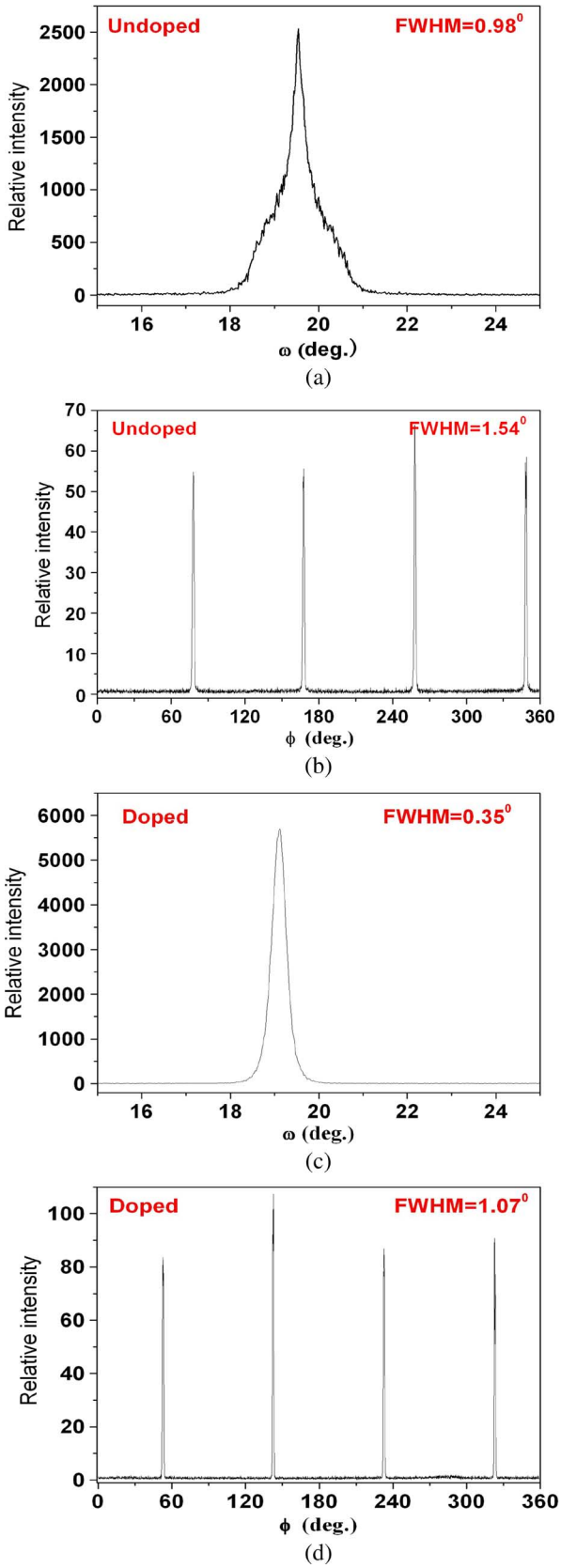

Fig. 2. FWHM values of the ω scan (a) and φ scan (b) for undoped YBCO films; FWHM values of the ω scan (c) and φ scan (d) for doped YBCO films.

then slowly decreases during further increase of the applied field. $F_{\mathrm{p,max}}$ of 3.0 GNm^{-3} in the 6% Zr doped YBCO film,

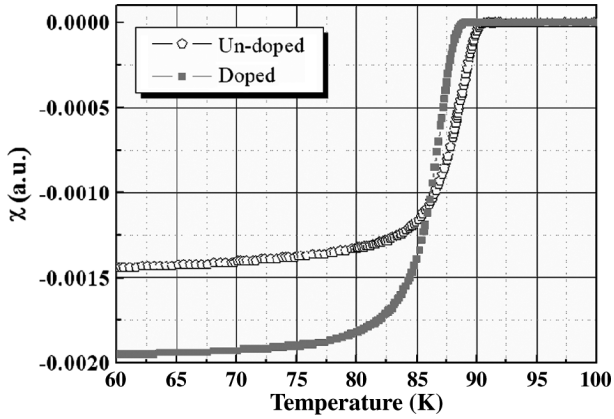

Fig. 3. The T_c of both doped and un-doped YBCO films.

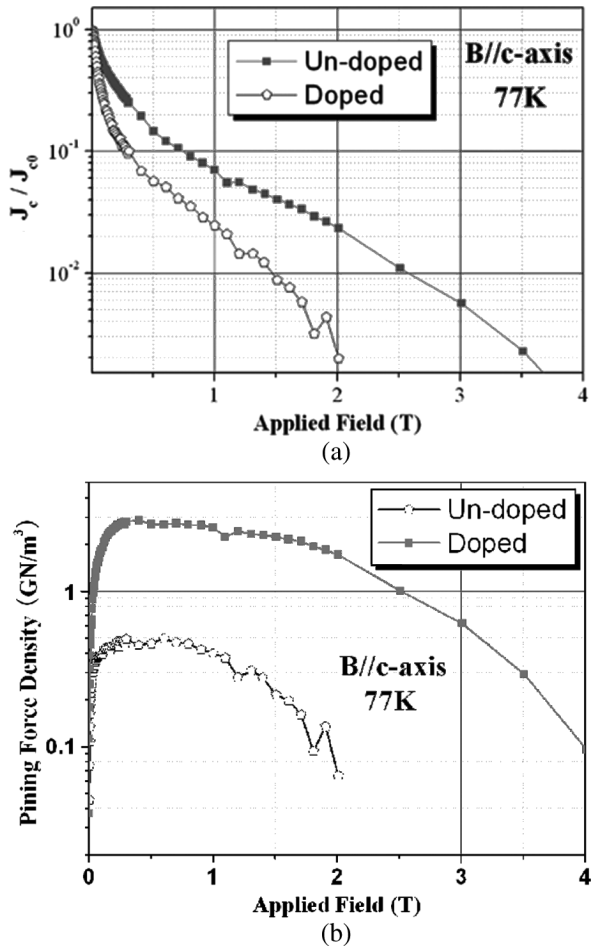

Fig. 4. The field dependence of J_c (a) and the pinning force (b) at 77 K, with the field parallel to the c-axis.

which is a close to 500% enhancement compared with that of the un-doped sample, clearly exemplifies the appealing prospects for the present nanocomposite films.

IV. Conclusion

In conclusion, we have shown that the metal-organic deposition technique can be modified to be used as a cost-effective and highly versatile method for introducing nanosized pinning centers into YBCO films. The TFA-YBCO route with Zr doped

precursor solutions was utilized to fabricate YSZ doped YBCO films through an *in-situ* crystallization process. Investigations into the behavior of the critical current density in magnetic fields reveal a strong enhancement of J_c at high magnetic fields in the YBCO film with 6% Zr doping. Pinning centers introduced by chemical solution deposition techniques show enhanced pinning properties in all magnetic fields.

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REFERENCES

[1] Q. X. Jia, S. R. Foltyn, P. N. Arendt, and J. F. Smith, "High-temperature superconducting thick films with enhanced supercurrent carrying capability," *Appl. Phys. Lett.*, vol. 80, no. 9, pp. 1601–1603, Mar. 2002.

[2] S. R. Foltyn, H. Wang, Q. X. Jia, and P. N. Arendt, "Overcoming the barrier to 1000 A/cm width superconducting coatings," *Appl. Phys. Lett.*, vol. 87, no. 16, pp. 162505-1–162505-3, Oct. 2005.

[3] J. L. MacManus-Driscoll, S. R. Foltyn, Q. X. Jia, H. Wang, A. Serquis, B. Maiorov, L. Civale, Y. Lin, M. E. Hawley, M. P. Maley, and D. E. Peterson, "Systematic enhancement of in-field critical current density with rare-earth ion size variance in superconducting rare-earth barium cuprate films," *Appl. Phys. Lett.*, vol. 84, no. 26, pp. 5329–5331, Jun. 2004.

[4] C. Cai, B. Holzapfel, J. Hänisch, L. Fernandez, and L. Schultz, "High critical current density and its field dependence in mixed rare earth $(\mathrm{Nd, Eu, Gd})\mathrm{Ba_2Cu_3O_{7-\delta}}$ thin films," *Appl. Phys. Lett.*, vol. 84, no. 3, p. 377, Jan. 2004.

[5] S. Kang, A. Goyal, J. Li, A. A. Gapud, P. M. Martin, L. Heatherly, J. R. Thompson, D. K. Christen, F. A. List, M. Paranthaman, and D. F. Lee, "High-performance high- T_c superconducting wires," *Science*, vol. 311, no. 5769, pp. 1911–1914, Mar. 2006.

[6] Y. Yamada, K. Takahashi, H. Kobayashi, M. Konishi, T. Watanabe, A. Ibi, T. Muroga, S. Miyata, T. Kato, T. Hirayama, and Y. Shiohara, "Epitaxial nanostructure and defects effective for pinning in $\mathrm{Y(RE)Ba_2Cu_3O_{7-x}}$ coated conductors," *Appl. Phys. Lett.*, vol. 87, no. 13, pp. 132502-1–132502-3, Sept. 2005.

[7] A. Goyal, S. Kang, K. J. Leonard, P. M. Martin, A. A. Gapud, M. Varela, M. Paranthaman, A. O. Ijaduola, E. D. Specht, J. R. Thompson, D. K. Christen, S. J. Pennycook, and F. A. List, "Irradiation-free, columnar defects comprised of self-assembled nanodots and nanorods resulting in strongly enhanced flux-pinning in $\mathrm{YBa2Cu3O7}$-δ films," *Supercond. Sci. Technol.*, vol. 18, no. 11, pp. 1533–1538, Nov. 2005.

[8] A. Crisan, S. Fujiwara, J. C. Nie, A. Sundaresan, and H. Ihara, "Sputtered nanodots: A costless method for inducing effective pinning centers in superconducting thin films," *Appl. Phys. Lett.*, vol. 79, no. 27, pp. 4547–4549, Dec. 2001.

[9] J. L. MacManus-Driscoll, S. R. Foltyn, Q. X. Jia, H. Wang, A. Serquis, L. Civale, B. Maiorov, M. E. Hawley, M. P. Maley, and D. E. Peterson, "Strongly enhanced current densities in Superconducting coated conductors of $\mathrm{YBa_2Cu_3O7 - x + BaZrO3}$," *Nat. Mater.*, vol. 3, no. 7, pp. 439–443, Jul. 2004.

[10] J. Gutierrez, A. Llordes, J. Gazques, M. Gibert, N. Roma, S. Ricart, A. Pomar, F. Sandiumenge, N. Mestres, T. Puig, and X. Obradors, "Strong isotropic flux pinning in solution-derived $\mathrm{YBa_2Cu_3O_{7-x}}$ nanocomposite superconductor films," *2007 Nature Materials*, vol. 6, no. 5, pp. 367–373, May 2007.

[11] Y. X. Zhou, S. Ghalsasi, I. Rusakova, and K. Salama, "Flux pinning in MOD YBCO films by chemical doping," *Supercond. Sci. Technol.*, vol. 20, no. 9, pp. S147–S154, Sept. 2007.

[12] K. Nakaoka, J. Matsuda, Y. Kitoh, T. Goto, Y. Yamada, T. Izumi, and Y. Shiohara, "Influence of starting solution composition on superconducting properties of YBCO coated conductors by advanced TFA-MOD process," *Physica C*, vol. 463–465, pp. 519–522, May 2007.

[13] J. Matsuda, K. Nakaoka, T. Izumi, Y. Yamada, and Y. Shiohara, "Effect of Ba/Y ratio in starting solution on microstructure evolution of YBCO films deposited by advanced TFA-MOD process," *Physica C*, vol. 468, pp. 997–1005, May 2008.

[14] P. Mele, K. Matsumoto, T. Horide, A. Ichinose, M. Mukaida, Y. Yoshida, and S. Horii, "Insertion of nanoparticulate artificial pinning centres in $\mathrm{YBa_2Cu_3O_{7-x}}$ films by laser ablation of a $\mathrm{Y_2O_3}$-surface modified target," *Supercond. Sci. Technol.*, vol. 20, no. 7, pp. 616–620, Jul. 2007.