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## Influence of matching field on critical current density and irreversibility temperature in $YBa_2Cu_3O_7$ films with $BaMO_3$ (M = Zr, Sn, Hf) nanorods

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The influence of the matching field  $(B_{\Phi})$  on critical current density  $(J_c)$  and irreversibility temperature  $(T_{irr})$  in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films containing BaMO<sub>3</sub> (M = Zr, Sn, Hf) nanorods was investigated. It was revealed that the irreversibility temperature normalized by the critical temperature  $(T_{irr}/T_c)$  was influenced by  $B_{\Phi}$ , for  $B > B_{\Phi}$ , but  $T_{irr}/T_c$  did not depend on which BaMO<sub>3</sub> material was used for  $B < B_{\Phi}$ , i.e., there was no dependence on nanorod density, diameter, interface sharpness, or  $T_c$  in the case of ideal nanorods. However,  $J_c/J_c(0 \text{ T})$  was found to decrease with increasing  $B_{\Phi}$  at low magnetic field strengths and to improve at high magnetic field strengths. In addition to  $J_c$  being dependent on  $B_{\Phi}$ , the  $T_c$  term in  $T_{irr}$  and  $J_c(0 \text{ T})$  were also found to have an effect on  $J_c$ . © 2016 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4942463]

Artificial pinning centers (APCs) significantly improve critical current density  $(J_c)$  in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO) films.  $BaMO_3$  (BMO; M = Zr, Sn, Hf) nanorods are one of the most effective APCs for obtaining a high  $J_c$  and global pinning force maximum  $(F_{p,max})$ .<sup>1–3</sup> A very high  $J_c$  was achieved with a high nanorod density at a high magnetic field and low temperature,<sup>4</sup> while shape control of the nanorods and hybrid APCs resulted in systematic variation of the vortex pinning.<sup>5–8</sup> These results show that BMO nanorods are promising for controlling the vortex pinning and enhancing  $J_{\rm c}$ . Elastic strain,<sup>9</sup> oxygen vacancies,<sup>10</sup> and defect-induced strain fields<sup>11</sup> change the matrix critical temperature  $(T_c)$  and elementary pinning force  $(f_p)$  in YBCO + BMO films. Geometric factors of the nanorods are also crucial to vortex pinning and dynamics: the straightness of the nanorods and their size determine the pinned volume, while the matching field  $(B_{\Phi})$  is proportional to their density. Although  $B_{\Phi}$  is believed to be one of the most important factors and many researchers observed  $B_{\Phi}$ -induced phenomena in YBCO that contained c-axis correlated pinning centers,<sup>6,12-16</sup> there is a lack of systematic studies on this topic, and therefore, the influence of  $B_{\Phi}$  on  $J_{c}$  remains unclear. To further understand the *c*-axis correlated pinning induced by BMO nanorods, a detailed analysis of vortex pinning is needed for YBCO + BMO films whose  $B_{\Phi}$  is systematically varied.

The dependence of  $J_c$  on BMO content was extensively studied to optimize  $J_c$  in YBCO + BMO films.<sup>17,18</sup> Because the microstructure depends on BMO content, YBCO + BMO films with varying BMO content are a well-defined system with which to study *c*-axis correlated pinning induced by BMO nanorods. However, no previous research has studied the dependence of  $J_c$  on BMO content to understand vortex behavior. In this study, therefore,  $B_{\Phi}$  was systematically controlled by varying BMO content and the M in BMO, while measuring  $J_c$  as a function of  $B_{\Phi}$ . Based on the results, the influence of  $B_{\Phi}$ ,  $J_c$  (0 T), and  $T_c$  on  $J_c$  is discussed to understand the mechanisms that determine  $J_c$  in YBCO + BMO films.

YBCO films were prepared on SrTiO<sub>3</sub> (100) single crystalline substrates at 830 °C and 0.26 mbar using pulsed laser deposition (PLD). YBCO + BZO, YBCO + BSO, and YBCO + BaHfO<sub>3</sub> (BHO) mixed targets were ablated, where BSO, BZO, and BHO content of the targets were 2.7–7.2 vol. %, 4.1-8.2 vol. %, and 3.1-6.3 vol. %, respectively; the films fabricated in this manner are referred to as YBCO + BZO(X), YBCO + BSO(X), and YBCO + BHO(X) in this letter, where X is the vol. % of BZO, BSO, or BHO. The resulting film thicknesses were 150-240 nm. The microstructure of the films was observed using transmission electron microscopy (TEM). Then, 1-mm long and  $100-\mu m$  wide bridges were formed using conventional photolithography and H<sub>3</sub>PO<sub>4</sub> etching to measure  $J_c$  with Physical Property Measurement System (PPMS). The magnetic field dependence of  $J_c$  was evaluated at 65-77 K under a magnetic field of 0-9 T, while the temperature (T) dependence of the resistivity was measured to obtain the irreversibility temperature  $(T_{irr})$ .  $J_c$  and  $T_{irr}$  measurements were performed mainly under a magnetic field parallel to the *c*-axis;  $J_c$  and  $T_{irr}$  were determined at an electric field strength of  $1 \,\mu$ V/cm. Table I summarizes basic parameters in the films.

Figure 1 shows cross-sectional bright-field TEM images of YBCO + BZO(4.1), YBCO + BSO(2.7), and YBCO + BHO (4.7) films. It is apparent from these images that BMO nanorods grow in the *c*-axis direction of YBCO, and ideal *c*-axis correlated pinning is, therefore, expected in these films. Nanorod diameter (*D*) was 6 nm, 10 nm, and 6 nm, and the spacing of nanorods (*d*) was  $\sim$ 20 nm, 25–33 nm, and  $\sim$ 20 nm in the YBCO + BZO(4.1), YBCO + BSO(2.7), and YBCO + BHO(4.7) films, respectively.

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TABLE I. Summary of basic parameters in the films.

Material	BMO content (vol. %)	Thickness (nm)	$T_{\rm c}({\rm K})$	$J_{\rm c}$ at 77 K, 0 T (MA/cm <sup>2</sup> )	$F_{p,max}$ at 77 K (GN/m <sup>3</sup> )	$F_{\rm p,max}$ at 65 K (GN/m <sup>3</sup> )
BSO	2.7	210	89.2	2.4	14.6	47.7
BSO	5.4	210	87.8		12.4	45.5
BSO	7.2	150	86.9	1.0	3.0	11.4
BZO	4.1	190	88.2	1.6	15.8	66.8
BZO	8.2	160	87.1	1.8	17.0	85.4
BHO	3.1	190	88.9	0.65	3.8	
BHO	4.7	160	87.5	0.95	8.1	60.4
BHO	6.3	180	85.9	0.82	6.9	50.8

Figure 2 shows the magnetic field dependence of  $T_{irr}$  in (a) the YBCO+BZO, (b) YBCO+BSO, and (c) YBCO + BHO films. For comparison,  $T_{irr}$  normalized by  $T_c$  ( $T_{irr}/T_c$ ) is also shown in Fig. 2(d).  $B-T_{irr}$  curves exhibit a shoulder at 1–5 T, and it is well known that the shoulder appears at

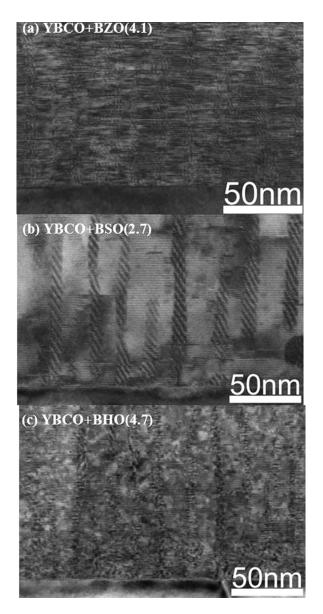


FIG. 1. Cross-sectional bright-field TEM images of (a) YBCO + BZO(4.1), (b) YBCO + BSO(2.7), and (c) YBCO + BHO(4.7) films. BMO nanorods grew in the *c*-axis direction of YBCO. *d* was ~20 nm, 25–33 nm, and ~20 nm in the YBCO + BZO(4.1), YBCO + BSO(2.7), and YBCO + BHO(4.7) films, respectively.

 $B_{\Phi}$ .<sup>13–16</sup>  $T_{\rm irr}/T_{\rm c}$  behavior was in good agreement in the  $B < B_{\Phi}$ range, regardless of nanorod structure, but it depended on nanorod structure in the  $B > B_{\Phi}$  range. From the shoulder that corresponds to the crossover between structure-independent  $T_{\rm irr}/T_{\rm c}$  and structure-dependent  $T_{\rm irr}/T_{\rm c}$ ,  $B_{\Phi}$  was estimated to be  $3.75 \pm 0.5$  T for YBCO + BZO(4.1),  $1.5 \pm 0.5$  T for YBCO + BSO(2.7), and 4.5  $\pm$  0.5 T for YBCO + BHO(4.7). Because  $B_{\Phi}$  is given by  $n\phi_0$  (n: nanorod density,  $\phi_0$ : magnetic flux quantum =  $2.07 \times 10^{-15}$  Wb), we used the TEM results to calculate  $B_{\Phi}$  as ~5 T for YBCO + BZO(4.1), ~2.5 T for YBCO + BSO(2.7), and  $\sim$ 5 T for YBCO + BHO(4.7).  $B_{\Phi}$  values obtained from the  $T_{irr}$  and TEM measurements were thus in good agreement. Figure 2(e) shows the dependence of  $B_{\Phi}$ on BMO content that was estimated from the  $B-T_{irr}$  curves;  $B_{\Phi}$  and BMO volume fraction are given by  $\phi_0/d^2$  and  $\pi D^2/d^2$  $4d^2$ , respectively. The results show that for small or moderate BMO content  $B_{\Phi}$  increased with increasing BMO content. However,  $B_{\Phi}$  decreased in the YBCO + BSO(7.2) and YBCO + BZO(8.2) samples, which have high BMO content, suggesting that an increase in nanorod diameter was to blame, especially because it has been previously reported that too high BMO content may result in degradation of the nanorod structure.<sup>18</sup> These results suggest that ideal *c*-axis correlated pinning was not achieved when the BMO content was too high, and therefore, we exclude the results in which BMO content was too high from our discussion of ideal nanorod pinning.

Figure 2(f) shows  $T_{irr}/T_c$  at 1 T and 7 T as a function of  $B_{\Phi}$  for ideal *c*-axis correlated pinning.  $T_{irr}/T_c$  did not depend on BMO content at 1 T ( $\langle B_{\Phi} \rangle$ ) owing to the strong Bose glass.<sup>19</sup>  $T_{irr}/T_c$  increased with increasing  $B_{\Phi}$  at 7 T ( $\rangle B_{\Phi} \rangle$ ), indicating that  $T_{irr}$  behavior in the  $B > B_{\Phi}$  range was determined only by  $B_{\Phi}$ , not by BMO selection. A similar tendency was also observed for SmBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> + BHO,<sup>20</sup> SmBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> + BSO,<sup>20</sup> and GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> + BHO<sup>21</sup> even if Y in YBCO was changed to other rare earth elements.

Figure 3(a) shows  $J_c-B$  curves at 77 K for YBCO + BZO(4.1), YBCO + BSO(2.7), YBCO + BHO(4.7), and pure YBCO films. BMO incorporation improved  $J_c$ , especially at high magnetic field strengths. Figure 3(b) shows the temperature and field angle dependence of the  $J_c-B$  curve for the YBCO + BSO(2.7) film. The magnetic field was applied along the *ab* plane (*B*//*ab*) and along the *c*-axis (*B*//*c*). Clear shoulders were observed in the  $J_c-B$  curves only for *B*//*c*; additionally,  $J_c$  was much higher for *B*//*c* than for *B*//*ab* at 1–3 T. In contrast, the pure YBCO films do not exhibit such strong *c*-axis correlated pinning.<sup>22</sup> Figures 3(c)–3(f) show  $J_c$ 

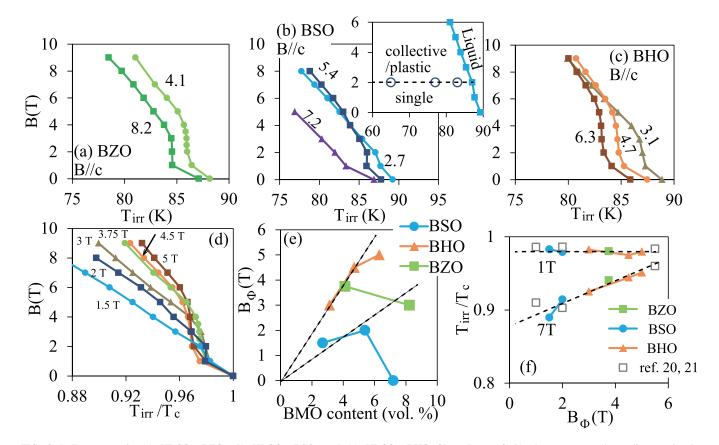


FIG. 2.  $B-T_{irr}$  curves in (a) YBCO + BZO, (b) YBCO + BSO, and (c) YBCO + BHO films. Inset of (b) shows vortex phase diagram in the YBCO + BSO(2.7) sample, where the field of shoulder in  $J_c-B$  and  $T_{irr}$  (~the Bose glass temperature) defines the regions for single vortex pinning (the strong Bose glass), collective/plastic vortex pinning (the weak Bose glass), and vortex liquid. (d)  $B-T_{irr}/T_c$  curves as a function of  $B_{\Phi}$  for the YBCO + BMO films with ideal nanorod pinning. The same symbols as used in (a)–(c) are used in (d). (e) Dependence of  $B_{\Phi}$  on BMO vol. % content. The lines show the  $B_{\Phi}$ -BMO content relationship with constant nanorod diameter. (f) Dependence of  $T_{irr}/T_c$  on  $B_{\Phi}$  at 1 T and 7 T. The results from Refs. 20 and 21 are also plotted. All data were obtained with B//c.

as a function of BMO content at various temperatures and magnetic field values. YBCO + BSO(2.7) exhibited the highest  $J_c$  at a low magnetic field of 1 T, while increasing BSO concentration to 7.2 vol. % did not improve  $J_c$ , regardless of applied magnetic field and temperature. BZO and BHO incorporation did not increase  $J_{\rm c}$  in the low magnetic field range of 0-1 T but significantly improved  $J_c$  at higher magnetic field strengths.  $J_c$  was higher in YBCO+BZO than in YBCO + BHO, regardless of temperature and applied magnetic field, and large  $J_{\rm c}$  was achieved at high magnetic fields at both 4.1 vol. % and 8.2 vol. % BZO content. It is difficult to achieve a high density of BSO nanorods because of their large diameter; however, because of their smaller diameters, the density of BZO and BHO nanorods can be increased. The present results are consistent with previous reports: heavy Zr doping into (Gd,Y)BCO significantly improved  $J_c$  at a low temperature and a high magnetic field,<sup>23</sup> while high  $J_c$  in films with a high BSO content has not yet been reported.<sup>17</sup>

Similar to the influence of  $B_{\Phi}$  on  $T_{irr}/T_c$ ,  $B_{\Phi}$  is expected to have a significant effect on  $J_c$ . To illustrate the influence of  $B_{\Phi}$  on  $J_c$ , Fig. 4(a) shows  $J_c/J_c(0T)$  at 77 K for the YBCO + BMO films. The highest  $J_c/J_c(0T)$  values in the 0.5–2 T and 7–9 T range were obtained for the YBCO + BSO(2.7) and YBCO + BHO(6.3) films, respectively. Conversely, the lowest  $J_c/J_c(0T)$  values in the 0.5–2 T and 7–9 T range were obtained for the YBCO + BHO(6.3) and YBCO + BSO(2.7) films, respectively. The YBCO + BSO(2.7) and YBCO+ BHO(6.3) films had the lowest and the highest  $B_{\Phi}$  in this study, respectively. With increasing  $B_{\Phi}$ ,  $J_c/J_c(0T)$  decreased at low magnetic field strengths but increased at high magnetic field strengths, demonstrating a systematic variation of the  $J_c$ -B curves with  $B_{\Phi}$ . Behavior of  $J_c$  similar to that observed at 77 K was also observed at 65 K (shown in Fig. 4(b)).  $F_{\rm p}$ /  $F_{\text{pmax}}$ -B curves at 77 K and 65 K are shown in Figs. 4(c) and 4(d), where  $F_{pmax}$  was obtained at the peak magnetic field  $(B_{\rm p})$ .  $B_{\rm p}$  was 2T and 6T in the YBCO + BSO(2.7) and YBCO + BHO(6.3) films, respectively, and  $B_p$  increased from 2 T to 6 T with increasing  $B_{\Phi}$  at 77 K.  $B_{p}$  ranged from 3 T for YBCO + BSO(2.7) to 8T for YBCO + BHO(6.3) at 65 K, depending on  $B_{\Phi}$ . The vortex phase diagram of the YBCO + BSO(2.7) sample is shown in Fig. 2(b). Strong nanorod pinning and a temperature-independent shoulder-field  $(\sim B_{\rm p})$  indicate that the shoulder in the  $J_{\rm c}$ -B curve did not result from the transition from Bragg glass to vortex glass,<sup>24</sup> but from the  $B_{\Phi}$  effect. Vortices are pinned by nanorods for  $B < B_{\Phi}$  and by elastic interactions for  $B > B_{\Phi}$ .  $J_{c}$  for  $B < B_{\Phi}$  is constant in the single vortex pinning region without thermal fluctuation, which is achieved at a low temperature.<sup>15,25,26</sup> As shown in Fig. 4, in the single vortex pinning region,  $J_c$  gradually decreases with the magnetic field because of thermal fluctuations at high temperatures such as  $65-77 \text{ K.}^{14,26} J_c/J_c(0 \text{ T})$ values for the YBCO + BSO(2.7) sample were 0.75, 0.41, and

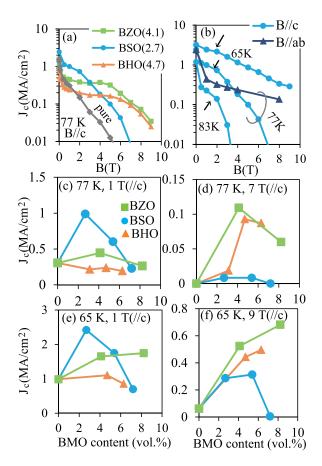


FIG. 3. (a)  $J_c$ -B curves with B//c at 77 K in the YBCO+BZO(4.1), YBCO+BSO(2.7), and YBCO+BHO(4.7) films. (b) Temperature and field angle dependence of  $J_c$ -B curves for the YBCO+BSO(2.7) films. Magnetic field was applied for B//ab and B//c at 77 K. Clear shoulders in the  $J_c$ -B curves (indicated by arrows) were observed only for B//c.  $J_c$  as a function of BMO content at (c) 77 K, 1 T, (d) 77 K, 7 T, (e) 65 K, 1 T, and (f) 65 K, 9 T. The same symbols are used in (c)-(f). YBCO+BSO(2.7) exhibited the highest  $J_c$  at low magnetic field strengths, while the highest  $J_c$  at high magnetic field strengths was achieved with 4.1 vol. % and 8.2 vol. % BZO content.

0.19 at 65 K, 77 K, and 83 K under a magnetic field of 1 T, while  $J_c/J_c(0T)$  values were 0.35 and 0.18 at 65 K and 77 K under a magnetic field of 3 T for the YBCO + BSO(4.7) film.  $J_c/J_c(0T)$  decreased with increasing temperature, indicating a thermal fluctuation effect on  $J_{\rm c}$  in the single vortex pinning region ( $B < B_{\Phi}$ ) at high temperatures. The motion of thermally fluctuating vortices is accelerated by neighboring unoccupied nanorods for  $B < B_{\Phi}$ , and the acceleration becomes significant when nanorod spacing is small. As neighboring nanorods are occupied by vortices at  $\sim B_{\Phi}$ , thermally assisted vortex motion to unoccupied nanorods becomes difficult. Thus, Jc rapidly decreases with increasing magnetic field strengths around ~0 T for small nanorod spacings, i.e., for large  $B_{\Phi}$ . Because the elastic interaction of vortices is weaker than direct pinning of nanorods, and high-density nanorods can accommodate many vortices,  $J_{\rm c}$  was improved through a high density of nanorods at high magnetic field strengths.

Figures 5(a)–5(d) show  $J_c$  as a function of  $B_{\Phi}$  at 77 K and 65 K under magnetic fields of 1 T, and 7 T and 9 T, respectively, where  $B_{\Phi}$  was obtained from  $T_{irr}$ . Although large  $J_c$ was obtained at low magnetic field strengths of 1 T for the films with small  $B_{\Phi}$ , large  $B_{\Phi}$  enhanced  $J_c$  at high magnetic field strengths of 7 T and 9 T. However,  $B_{\Phi}$  accounted only, in

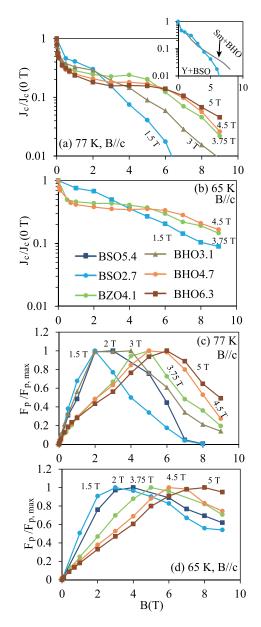


FIG. 4.  $J_c/J_c(0 T)$ –*B* curves for the films for B//c at (a) 77 K and (b) 65 K. Inset of (a) compares  $J_c/J_c(0 T)$ –*B* curve of the YBCO+BSO(2.7) film with that of the SmBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>+BHO sample reported in Fig. 5 of Ref. 20.  $F_p/F_{p,max}$ –*B* curves for the films at (c) 77 K and (d) 65 K. The same symbols and  $B_{\Phi}$  values are used in (a)–(d).  $J_c/J_c(0 T)$ –*B* curves and  $F_p/F_{p,max}$ –*B* curves systematically vary with  $B_{\Phi}$ .

part, for the magnetic field dependence of  $J_c$  that is visible in Fig. 5, because a single function of  $B_{\Phi}$  cannot describe  $J_c$ , demonstrating that other factors also affect  $J_c$ .

Although  $J_c(0 \text{ T})$  may depend on  $B_{\Phi}$ , its effect on  $J_c$  is worth discussing separately to understand the mechanisms that influence  $J_c$ . The effect of  $J_c(0 \text{ T})$  is discussed for the YBCO+BHO(4.7) and YBCO+BZO(4.1) films, where  $J_c$ was different despite having almost the same  $B_{\Phi}$  of ~4–5 T.  $J_c(0 \text{ T})$  is dependent on the current flow path, film homogeneity,  $f_p$ , matrix crystallinity, and  $T_c$ .  $J_c(YBCO + BZO(4.1))/$  $J_c(YBCO + BHO(4.7))$  at 3 T was 2.1 and 1.6 at 77 K and 65 K. The  $J_c$  ratio depended on temperature, suggesting that  $f_p$ , matrix crystallinity, and  $T_c$  were the dominant factors causing the difference in  $J_c(0 \text{ T})$  between the YBCO + BHO(4.7) and YBCO + BZO(4.1) samples.

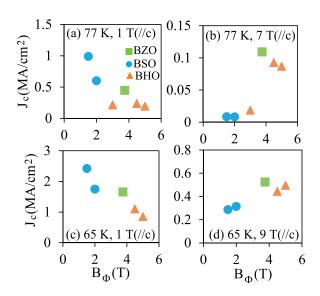


FIG. 5.  $B_{\Phi}$  dependence of  $J_c$  at (a) 77 K, 1 T, (b) 77 K, 7 T, (c) 65 K, 1 T, and (d) 65 K 9 T for B//c. The same symbols are used in (a)–(d). A single function of  $B_{\Phi}$  (unlike for  $T_{irr}/T_c$ ) cannot explain the behavior of  $J_c$ .

Although  $T_c$  was not significantly different in the films presented in this manuscript,  $T_c$  enhancement is effective in improving  $J_c$ , and therefore, the effect of  $T_c$  should also be discussed.  $T_{irr}$  is a function of both  $T_c$  and  $B_{\Phi}$  because:  $T_{irr}$  $= T_c \times T_{irr}/T_c = T_{irr}(T_c, T_{irr}/T_c(B_{\Phi})) = T_{irr}(T_c, B_{\Phi})$ .  $B_{irr} = 15$  T at 77 K in SmBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> + BHO with  $B_{\Phi}$  of ~1 T owing to the large  $T_c$  of 92.3 K;<sup>20</sup> but  $B_{irr} = 8.6$  T at 77 K in our YBCO + BSO(2.7) sample with a  $T_c$  of 89.2 K and  $B_{\Phi}$  of ~1.5 T—the difference in  $B_{irr}$  between the two samples originates from their differing  $T_c$  values. As shown in the inset of Fig. 4(a),  $J_c/J_c(0 \text{ T})$  for  $B > B_{\Phi}$  was larger for the SmBa<sub>2</sub> Cu<sub>3</sub>O<sub>7</sub> + BHO sample than for our YBCO + BSO(2.7) sample, owing to the SmBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> + BHO sample's larger  $B_{irr}$  or, to be more precise, owing to the effect of the  $T_c$  term in  $T_{irr}$ ( $B_{irr}$ ). This indicates that enhancing  $T_c$  improves  $J_c/J_c(0 \text{ T})$  for  $B > B_{\Phi}$ .

In summary, the microstructure,  $T_{irr}$ , and  $J_c-B$  curves of YBCO + BZO, YBCO + BSO, and YBCO + BHO films were analyzed.  $B_{\Phi}$  of the films ranged from 1.5 T to 5 T, and the films exhibited ideal nanorod pinning. Regardless of BMO selection and vol. % content,  $T_{irr}/T_c$  curves depended only on  $B_{\Phi}$ , while  $J_c/J_c(0 \text{ T})-B$  and  $F_p-B$  curves varied systematically with  $B_{\Phi}$ : with increasing  $B_{\Phi}$ ,  $J_c/J_c(0 \text{ T})$  decreased at low magnetic field strengths, but increased at high magnetic field strengths.  $B_{\Phi}$  determined  $T_{irr}/T_c$  or  $J_c/J_c(0 \text{ T})$ , while the  $T_c$  term in  $T_{irr}$  and  $J_c(0 \text{ T})$  also had an effect on  $J_c$ .

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