On the large role of weak uncorrelated pinning introduced by BZO nanorods at low temperatures in REBCO thin films

A. Xu, V. Braccini, J. Jaroszynski, Y. Xin, and D. C. Larbalestier
National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32310
CNR-SPIN, Corso Perrone 24, I-16152 Genova, Italy
(Dated: November 8, 2011)

REBa$_2$Cu$_3$O$_x$ films can achieve remarkably high critical current density values by the incorporation of insulating nanoparticles. A particular interesting case concerns BaZrO$_3$ (BZO) nanorods, whose strongly correlated effect is seen at high temperatures. Here we investigate the field, temperature and angular dependence of the critical current density over a wide temperature range from 4.2 K to 77 K, and magnetic fields up to 31 T. We show that the correlated c-axis pinning of BZO nanorods becomes progressively less obvious at lower temperature. Indeed at 4.2 K and fields up to 31 T, the only correlated pinning is for fields parallel to the film plane. We interpret the change as being due to significant contributions from dense but weak pins that thermal fluctuations render ineffective at high temperatures but which become strong at lower temperatures.

PACS numbers: 74.25.Wx, 74.25.Sv, 74.78.-w, 74.72.-h

I. INTRODUCTION

REBa$_2$Cu$_3$O$_x$ (REBCO, where RE = rare earth) thin films are very promising high temperature superconductors for applications because of their high current-carrying capability in very strong magnetic fields over a wide operating temperature regime. Higher and less anisotropic critical currents $I_c$ still remain very desirable for widespread applications. Fortunately, it has been demonstrated that several approaches work for the $I_c$ improvement, including increasing the thickness of the REBCO layer mitigating the weak-link effect of grain boundaries and by better understanding and more effective enhancement of the pinning mechanisms.

Enhancement of flux pinning by optimization of nanoscale defects is fundamental to $I_c$ improvement. More specifically, it was first established that oxygen deficiencies were the governing pinning centers in high quality YBCO single crystals. In turn, dislocations and point defects were found to be strong pins at low temperatures and important for the typically observed one or two order of magnitude higher $I_c$ of YBCO thin films. Compared to the growth defects mentioned above, intentionally introduced pinning centers have captured more attention due to their high tunability. In early study of the pinning effects of heavy-ion irradiation on YBCO single crystals large $I_c$ increases at all temperature and magnetic fields, especially for magnetic field parallel to the irradiation direction was found. Later, self-assembled BaZrO$_3$ nanorods lying along the c-axis were incorporated into YBCO thin films and coated conductors. BZO has a high lattice mismatch, a high melting temperature, and it is insoluble in REBCO, all of which contribute to the substantial improvement of $I_c$ over a wide angular range far from the c-axis. A series of BZO (Ba metal oxide, where M = Sn, Hf and Ir) second phases acting as correlated c-axis pinning effects were explored thereafter in order to decrease the anisotropic $J_c$. Unfortunately, the additions of BZO are detrimental to $T_c$ and thus degrade the properties at liquid nitrogen temperature. However, Harrington et al. obtained excellent pinning properties while keeping $T_c$ at 92 K even with very high concentrations of self-assembled RE$_3$TaO$_7$ (RE = Er, Gd and Yb) nanorods, where, unlike the tensile stress-inducing BZO, the RE$_3$TaO$_7$ introduces a compressive stress on the REBCO. More recently, an 0.5 μm thick pulsed laser deposition (PLD) YBCO film with embedded double perovskite Ba$_2$YNbO$_6$ nanorods showed a maximum flux pinning force density $F_{p\text{ max}}$ in excess of 30 and 120 GN/m$^3$ at 75.5 K and 65 K. Such $F_{p\text{ max}}$ values exceed a long challenged benchmark, that of 18-20 GN/m$^3$ found in the best pinning-tailored NbTi wire at $T = 4.2$ K.

In general the maximum $J_c$ of any superconductor is limited by the depairing current density $J_d = \phi_0/3\pi\mu_0\lambda^2\xi$ according to anisotropic Ginzburg-Landau theory. Here $\phi_0$ is the flux quantum, $\lambda$ is the London penetration length, $\xi$ is the coherence length and $\mu_0$ is the vacuum permeability. In fact REBCO films can clearly exert strong pinning, as shown by estimates that ~30% of $J_d$ is achieved in YBCO thin films, at least at low fields. But high field magnets do require further $J_c$ improvement by flux pinning engineering of REBCO conductors. In spite of REBCO having by far the highest irreversibility fields $H_{irr}(T)$ of all cuprate superconductors, neither $H_{irr}(T)$ nor $J_c(H)$ are high enough for making magnets of 5–10 T unless the temperature is reduced below ~30–40 K. So far, most pinning studies have been concentrated on $J_c$ enhancement at high temperatures, 65 K to 77 K, and magnetic fields near self-field for power transmission. However, potential applications of REBCO conductors extend over much broader temperature and magnetic field regimes. Recently, thanks to the availability of REBCO conductor with suitable mechanical properties, the development of all-superconducting magnets above 25 T at 4.2 K became feasible, and some prototypes greater than 30 T were demonstrated.
However, coil design depends on understanding the detailed angular dependence of \( J_c \) over broad temperature and field ranges so that safe coil quench design can be predicted with confidence. Moreover, superconducting magnetic storage systems, motors and generators, working at intermediate temperatures \( \sim 30 \, \text{K} \), are important potential applications too.\textsuperscript{27-29} Thus, a systematic study of the pinning mechanisms over a broad range covering all applications regimes is indispensable for both practical and fundamental reasons.

BZO nanorods are well known to produce strong correlated \( c \)-axis pinning at high temperatures. Surprisingly, however, recent studies of samples containing BZO nanorods showed no signs of \( c \)-axis correlated pinning at 4.2 K, even though \( J_c(H) \) was strongly enhanced.\textsuperscript{30,31} Here we show that the dominant 4.2 K pinning characteristic, valid up to at least 31 T, can be fit to a standard anisotropic mass scaling except near the \( ab \)-plane where correlated pinning, probably by the CuO charge-reservoir layers enhances \( J_c \). By measuring the angular-dependent current density \( J_c(\theta) \) over a wide range of field and temperature, we observe that this uncorrelated pinning is valid only below \( \sim 30 \, \text{K} \). We conclude that these low temperature angular-independent pins are largely point pins induced by the strain fields of the BZO nanorods. Because they are point pins, they are easily de-pinned at higher temperatures, leaving the strong correlated \( c \)-axis pinning effects of the BaZrO\(_3\) nanorods then quite evident. At 4.2 K, however, the point pins contribute almost half of the \( J_c \).

**II. EXPERIMENTAL DETAILS**

We performed an extensive angular \( J_c(\theta, T, H) \) characterization of a recent REBCO thin film in fields up to 31 T and temperatures from 4.2 to 77 K. The 1.1 \( \mu \)m thick film was grown by metal-organic chemical vapor deposition (MOCVD) on a high strength metal alloy tape commercially available as Hastelloy, on which a buffer layer textured by ion-beam assisted deposition (IBAD) was deposited. A \( \sim 2 \mu \)m thick sputtered silver layer was deposited on the REBCO layer as protection and intermediate electrical contact layer for the \( \sim 50 \mu \)m thick copper layer, which was electro-plated on it.\textsuperscript{32} This sample is representative of the most advanced coated conductor made by SuperPower Inc. The Zr addition produces BZO nanorods with an equivalent flux density \( B_\phi \sim 3 \, \text{T} \). Earlier study has shown that such films have high \( J_c \) properties at both high and low temperatures.\textsuperscript{33}

The 4.2 K and high field four-probe critical current measurements were performed in a 52 mm cold bore 15 T superconducting magnet and the 52 mm warm bore 31 T Bitter magnet, fitted with a 38 mm bore liquid He cryostat. The 10 K to 77 K measurements were carried out in a 16 T Physical Property Measurement System (PPMS). Samples were rotated with respect to the external magnetic field around the axis parallel to the current direction to maintain a maximum Lorentz force configuration. The angle \( \theta = 0 \) is defined as the applied magnetic field perpendicular to the tape plane which is parallel to the crystallographic \( c \)-axis direction with a typical uncertainty of 1 – 4° caused by an offset caused by the IBAD process.\textsuperscript{30,32}

Due to the high critical current \( (I_c) \) values observed at lower temperatures, samples with different geometries were prepared in order to avoid harmful Joule heating and overstressing by the large Lorentz forces \( (I_c \times B) \) possible in different regimes of temperature and magnetic field. A 50 \times 500 \( \mu \)m bar was cut by Nd-YAG (yttrium aluminum garnet) laser for the \( J_c \) measurement at 77 K. Even narrower samples, \( \sim 10 \mu \)m wide and 200 \( \mu \)m long were patterned by SEM/FIB so as to restrict \( I_c \) to \( \leq 5 \) A when the sample was measured in helium gas between 10 and 70 K. In both cases, copper and silver layers were removed by wet-etching. Larger bridges about \( \sim 1 \) mm wide and 1 cm long were patterned leaving the silver and copper layers present for the 4.2 K measurements. Two different home-made \( I_c \) probes equipped with rotating sample platforms were used. One had a maximum current-carrying capability of \( \sim 500 \, \text{A} \) for high \( I_c \) measurement in liquid helium, while the second had \( \sim 5 \) A capability in the PPMS cryostat for studies at temperatures above 10 K.

TEM images were taken in a JEOL JEM 2011 transmission electron microscope. The critical temperature \( (T_c) \) is defined as the temperature where resistance \( R \) equals zero. The 77 K irreversibility field was determined from the field dependence of \( J_c \) with the criterion \( J_c = 100 \, \text{A/cm}^2 \). For lower temperatures where \( H_{irr}(T) \) is greater than magnetic field available \( H_{irr}(T) \) was assessed from the formula in.\textsuperscript{31}

**III. RESULTS**

The MOCVD sample under study has critical temperature \( T_c = 90.7 \, \text{K} \). The nominal composition of this sample is \( \text{Y}_{0.8}\text{Gd}_{0.2}\text{Ba}_2\text{Cu}_3\text{O}_y \) with 7.5 at. \% Zr doping, composition found to give the highest in-field \( J_c \) values at 77 K.\textsuperscript{32} The sample has \( J_c \) as high as 3.4 MA/cm\(^2\) at self-field and 1.0 MA/cm\(^2\)at 1 T, at \( \theta = 0^\circ \). Even more importantly, both the irreversibility field, \( H_{irr} = 10.2 \, \text{T} \), and the maximum flux pinning force, \( F_{p_{max}} = 12 \, \text{GN/m}^3 \) along the \( c \)-axis are substantially higher than that of other REBCO films with similar thickness, \( \sim 1 \, \mu \)m.

Figure 1 shows a typical cross-sectional, bright-field TEM image of this MOCVD sample. BZO nanorods, with \( \sim 8 \) nm diameter, with inclinations of \( \sim 5 - 20^\circ \) to the \( c \)-axis are quite obvious. Their density corresponds to a matching field \( B_\phi = \phi_0/\alpha^2 \approx 2.6 \, \text{T} \), where \( \alpha \approx 28 \) nm is the measured average distance between the BZO nanorods and \( \phi_0 = \hbar/2e \approx 2.1 \times 10^{-15} \, \text{Wb} \) is the flux quantum. Such nanorods are responsible for the broad \( I_c \) maxima observed at elevated temperatures when \( H \) is off the film plane. This strong correlated pinning pro-
FIG. 1: Cross-sectional TEM image of the MOCVD sample. Splayed BZO nanorods along c-axis and tilted RE$_2$O$_3$ precipitate arrays along ab-plane are the major visible correlated pinning centers. The RE$_2$O$_3$ precipitates are effective 3D pins that enhance $J_c$ at all orientations. A low density of threading dislocations that are effective pins along the c-axis are also visible in this image.

produces the outstanding superconducting performance observed at 77 K. Moreover, a high density of self-assembled RE$_2$O$_3$ precipitate arrays aligned along the ab-plane is another important source of pinning. Interestingly, TEM observation indicates that the ab-plane of REBCO tilts $\sim 2^\circ$ from the buffer layer because of the IBAD process while the RE$_2$O$_3$ precipitate arrays tilt away from the ab-plane by $\sim 5^\circ$, as previously reported. This tilting of the precipitate arrays is important for the pinning. The dislocations provide additional c-axis correlated pinning, although we believe that their contribution is negligible compared to BZO nanorods because of their low density.

Figure 2 (a) presents the c-axis field dependence $J_c(H||c)$ for magnetic fields up to 16 T or 31 T at various temperatures from 4.2 K to 77 K. For $T = 77$ K, only data below the irreversibility field, $H_{irr} = 10.2$ T are plotted. $J_c(H)$ shows less field dependence with decreasing temperature. At 10 K, $J_c$ at self-field reaches 43 MA/cm$^2$ which corresponds to $\sim 17 \%$ of $J_d$. $J_c$ at 16 T and 10 K is as high as 3.7 MA/cm$^2$ equal to the self-field $J_c$ at 77 K. At 4.2 K, $J_c$ decreases from 33.3 MA/cm$^2$ at 1 T to 2.9 MA/cm$^2$ at 31 T. These high current densities correspond to $J_c = 1.5$ kA at 1 T and 0.13 kA at 31 T for standard production 4 mm wide tape. Such high $J_c$ values make effective characterization of such conductors difficult. Notably, this MOCVD BZO-containing sample shows higher $J_c$ at all temperatures below $\sim 70$ K than optimized NbTi wire evaluated at 4.2 K. The power-law dependence of $J_c$ on magnetic field, $J_c \propto H^{-\alpha}$, is observed at low temperatures. At 4.2 K, the power-law exponent $\alpha = 0.7$ describes $J_c$ well in the whole 1–31 T range of magnetic field.

FIG. 2: (color online) (a) Field dependence of $J_c$ for $H$ parallel to the c-axis at temperatures between 4.2 K and 77 K and magnetic fields up to 31 T and (b) The corresponding flux pinning force calculated from $F_p = J_c \times \mu_0 H$. Only data below $H_{irr}$ is shown at $T = 77$ K in (a). $J_c \propto H^{-\alpha}$ is observed at temperatures below $\sim 30$ K. It can be seen that $F_{p,\text{max}}$ is trending to more than 1 TN/m$^3$ at the lowest temperature. Lines connecting data points are guides for the eyes.

Figure 2 (b) shows flux pinning force density $F_p = J_c \times \mu_0 H$ as a function of external magnetic field parallel to the c-axis at various temperatures. The superior $F_p$ values confirm the strong pinning provided by the defects existing in this sample. The highest measured $F_p$ at 4.2 K and 31 T almost reaches 1000 GN/m$^3$. This is the record high value observed in any superconductor so far. It is also striking that the pinning force above 4 T at 30 K barely depends on magnetic field. The maximum pinning force is $\sim 267$ GN/m$^3$ at 30 K while at lower temperatures $F_{p,\text{max}}$ almost reaches 1000 GN/m$^3$. This is the record high value observed in any superconductor so far. The pinning force above 4 T at 30 K barely depends on magnetic field. The maximum pinning force is $\sim 267$ GN/m$^3$ at 30 K while at lower temperatures $F_{p,\text{max}}$ almost reaches 1000 GN/m$^3$. This is the record high value observed in any superconductor so far.
The double-peaked $F_p(H)$ dependence shown by BZO-doped YBCO PLD thin films on SrTiO$_3$ single crystal substrates are not observed in the present work at any temperature.

The angular dependence of $J_c$ is a powerful tool for understanding pinning mechanisms and also crucial for magnet design. Figure 3 presents $J_c(\theta, 1$ and 4 T) of the MOCVD sample at 77 K, 50 K, 30 K and 10 K. $J_c$ maxima around the $c$-axis are clearly seen at 77 K and 50 K. These maxima do not occur exactly at 0° due to the splayed inclination of the BZO nanorods. At 1 T and 77 K, the $c$-axis $J_c$ reaches 1.1 MA/cm$^2$, about one third higher than the $J_c$ maximum value around the $ab$-plane and twice the minimum $J_c$ close to the $ab$-plane. As the magnetic field increases to 4 T, the $c$-axis $J_c$ peak becomes lower than the $ab$-peak, because the field exceeds the matching field of 2.6 T, corresponding to the BZO nanorod density. It is worth noting that both BZO nanorods and the RE$_2$O$_3$ precipitate arrays contribute to $J_c$ at 77 K, being responsible for the $c$-axis peak and for $J_c$ enhancement over the whole angular range especially around the $ab$-plane. Comparing the 77 K to lower temperature data at the same field, the $c$-axis peak becomes less distinct as temperature decreases, not being observable at all at temperatures below 30 K. This strongly suggests that the dominant pinning mechanism changes at lower temperatures and that the crossover temperature is $\sim$30 K. The ratio of $J_c||c$ and $J_c||ab$ decreases from 1.4 at 77 K to 0.9 at 50 K, showing the reversion to that expected by the mass anisotropy at low temperature. At lower temperature, the $J_c||ab$ peak becomes more evident, indicating a strengthening of the $ab$-plane correlated pinning. At low fields $J_c||ab$ varies from a cusplike dependence above 50 K to a smooth, Ginzburg-Landau (GL)-like peak at lower temperatures and at 10 K from GL-like at low fields to cusplike at high magnetic fields.

Ultra high-field magnet applications at 4.2 K are an important area for applications of YBCO conductors, for which we have performed studies of $J_c(\theta)$ up to 31 T. Figure 4 shows the angular dependence of $J_c$ of this MOCVD sample at 4.2 K up to 25 T. Evidently, the $c$-axis $J_c$ peak is totally washed out at 4 K at all fields from 3 T up to 25 T. As noted also in the contest of data taken at 10 K, the GL-like $J_c(\theta)$ dependence evolves towards a cusplike for magnetic fields above $\sim$5 T.

**IV. DISCUSSION**

The temperature and magnetic field dependence of the flux pinning force for many superconductors often obeys a scaling relation, as was first proposed by Fietz and Webb:

$$F_p(T, H) = \text{const} \times [H_{c2}(T)]^n g(h) \quad (1)$$

in which the exponent $n$ describes the temperature dependence of the upper critical field $H_{c2}(T)$ or in this case the irreversibility field $H_{irr}(T)$ and $g(h)$ is a pinning function which, in simple cases where the dominant pinning mechanisms are temperature-independent, depends only on the reduced field $h$ ($h = H/H_{irr}(T)$). We first address the extent to which this simple pinning assumption is valid for this strong pinning sample over such a broad range of $T$ and $H$.

The reduced pinning force density, $F_p/F_{p,max}$ as a function of reduced field $h$ is plotted in Fig. 5 (a) using a reduced field defined as $h = H/H_{irr}(T)$, instead of $h = H/H_{c2}(T)$ for low temperature superconductors.
In both cases the result is the same: the scaling field is defined for the field at which $J_c$ tends to zero. At $T = 77$ K, $F_p/F_{p,\text{max}}$ peaks at $h = 0.2$ and $H = 2.0$ T, which is close to the matching field of 2.6 T for BZO nanorods in this sample. On decreasing temperature from 77 K to 50 K, the peak $h_{\text{max}}$ shifts to lower reduced field, from 0.2 to 0.15, indicating that there is no simple correlation between $h_{\text{max}}$ and $B_0$. With further temperature decrease to 30 K, there is no obvious change of the peak position. However, the reduced peak width broadens as the temperature is lowered to 40 K and then to 30 K. Thus, exact temperature scaling does not occur in this sample, as is further demonstrated in Fig. 5 (b) by plotting log $F_p$ vs. log $H_{\text{irr}}(T)$ at varying reduced fields. The scaling exponent $n$ increases from 1.65 to 2.06 as the reduced field rises from 0.25 to 0.75. However, excellent temperature scaling is observed at constant reduced field, as shown by the linear dependence of log $F_p$ on log $H_{\text{irr}}(T)$. Taken together, both figures indicate that the dominant pinning mechanism(s) are varying with temperature.

More compelling evidence for the temperature dependence of the pinning mechanisms is provided by the temperature dependence of $J_c/J_d$, the ratio of the measured pinning critical current density to the calculated depairing current density, which may be used as a convenient measure of the effectiveness of the pinning defects in the phenomenology of type II superconductors. In this work, we use $J_c/J_d$ to track the pinning strength variation with temperature. Figure 6 presents $J_c/J_d$ for $H$ parallel to the $c$-axis as a function of temperature at several different magnetic fields, where $J_d$ is obtained from the following equation:\cite{38,39}

$$J_d(T, H) = J_d(0, 0) \left(1 - \frac{T}{T_c}\right)^{3/2} \left(1 - \frac{H}{H_{c2}(T)}\right)^{3/2}$$  

(2)

Here $J_d(0, 0) = 300$ MA/cm$^2$ (Ref. 38) and $H_{c2}(T) = H_{c2}(0)[1 - (T/T_c)^2]$. $T_c = 91$ K and $H_{c2}(0, H)|c| = 121$ T.\cite{40} In the more often used self-field limit, this expression reduces to the usual equation $J_d = \phi_0/3 \sqrt{2\pi\mu_0 \lambda^2 \xi}$.

Two evident features are observed in Fig. 6. First, it is clear that the ratio $J_c/J_d$ continuously declines with increasing temperature for all fields evaluated from 1–16 T, signaling that thermal fluctuations are important even in this strong pinning regime.\cite{11,12,13,24,25,26,32,42,43} However, the dominant regime for thermal fluctuations appears to vary with magnetic field. At the lowest field evaluated, 1 T, it appears that three temperature regimes can be observed. On raising the temperature from 4.2 K, there is an initially steep drop of $J_c/J_d$ up to about 30 K, the rate of decrease then flattening between about 30 and 65 K, before finally falling off more rapidly again at the highest temperatures. The transition to strong thermal fluctuations regime seems to occur at ~65 K at 1 T, 60 K at 4 T, 50 K at 8 T and 40 K at 16 T. The steep, low temperature fall in $J_c/J_d$ appears at all fields, which suggests that an additional pinning mechanism operating only at low temperatures is present. However, the contribution of this pinning mechanism is strongly suppressed by both increasing temperature and increasing field, so that at 16 T its effect is only weakly visible as a point of inflection at 10 – 15 K on the $J_c/J_d(T)$ plot.

More details of the low temperature pinning and high temperature pinning can be obtained by plotting log $J_c$ vs. $T$ and log $J_d$ vs. $T^2$. Flux pinning can be categorized as strong or weak based on the extent of the distortion of the flux line lattice by the pinning defects. It has been shown that the $J_c$ determined by weak pinning mechanisms decays as an exponential function of temperature while $J_c$ of strong pinning decays as an exponential function of $T^2$.\cite{13,26,42,43} Accordingly, Fig. 7 (a) plots log $J_c$ vs. $T^2$. $J_c$ indeed decays as an exponential function of $T^2$.

FIG. 5: (color online) (a) Reduced pinning force, $F_p/F_{p,\text{max}}$ for $H$ parallel to the $c$-axis as a function of reduced field $H/H_{\text{irr}}$ at temperature from 30 K to 77 K showing the lack of a perfect temperature scaling of $F_p$. (b) The scaling of $F_p$ as a function of irreversibility field at several reduced magnet fields shows good temperature scaling at constant reduced field, although the scaling exponent is not independent of reduced field.
K, as shown in Fig. 2 (a), and J

J

\varepsilon

precipitates provide strong pinning at \sim 30 K. Since we have observed that \varepsilon \text{ does not dominate from } \sim 30 K in YBCO single crystal samples.\textsuperscript{41} The BZO nanorods and RE\textsubscript{2}O\textsubscript{3} precipitates in this sample significantly raise the crossover temperature at which thermal fluctuations dominate from \sim 60 K, as compared to \sim 30 K in YBCO single crystal samples.\textsuperscript{41} The BZO nanorods and RE\textsubscript{2}O\textsubscript{3} precipitates provide strong pinning at \sim 30 – 60 K as clearly shown by multiple other studies.\textsuperscript{45,47,48}

Further information about the additional low temperature weak pinning can be obtained by analysis of the angular dependence of J\textsubscript{c} at 4.2 K using the anisotropic scaling approach proposed by Civale et al.\textsuperscript{49} If the pinning is due to uncorrelated defects randomly distributed over angular space, then J\textsubscript{c} should depend on H and \theta only through a single variable J\textsubscript{c}(H, \theta) = J\textsubscript{c}[\varepsilon(\theta)] where \varepsilon(\theta) = [\cos^2(\theta) + \gamma^{-2}\sin^2(\theta)]^{1/2}, where \gamma is the electronic mass anisotropy parameter. Since we have observed that J\textsubscript{c} follows J\textsubscript{c} \propto H^{-\alpha} at low temperature especially at 4.2 K, as shown in Fig. 2 (a), then J\textsubscript{c}(H, \theta) \propto [H\varepsilon(\theta)]^{-\alpha}.

Power law behavior with \alpha \approx 0.7 for BZO-containing samples and \alpha \approx 0.5 for non-BZO samples was reported by us previously.\textsuperscript{22} Figure 8 presents J\textsubscript{c}(\theta) at 5, 15 and 25 T using the anisotropic scaling parameters \alpha = 0.7 and \gamma = 3. These curves give a reasonable fit of the data in the low angle region, from 0\degree up to about 60 – 70\degree, leading us to conclude that, over this wide range, all around the c-axis where the correlated effects of the BZO nanoparticles are so evident at higher temperatures, the pinning is indeed uncorrelated and random. Clearly the most reasonable interpretation of this behavior is that the additional pinning that operates only up to 30 – 40 K

in the intermediate temperature regime of 30 – 65 K at 1 T, 30 – 60 K at 4 T, 30 – 50 K at 8 T and 12 T, but it is also clear that there is an “excess” J\textsubscript{c} at the lowest temperatures which quickly disappears at high temperature. Consistent with plot of J\textsubscript{c}/J\textsubscript{d} vs. T in Fig. 6, we also observe a sharp drop of J\textsubscript{c} at high temperature. At lower temperatures, as shown in Fig. 7 (b), the exponential decay of J\textsubscript{c}, shown by the linear dependence of log J\textsubscript{c} on T, is observed below \sim 40 K at 1 T and 4 T and below \sim 30 K at 8 T and 12 T. This analysis clearly suggests that additional weak pinning is present below \sim 30 K. In the \sim 30 – 60 K range, stronger pinning centers then control J\textsubscript{c} until even these strong pins are rendered ineffective by increasing thermal fluctuations at higher temperatures. The strong pinning provided by the BZO nanorods and RE\textsubscript{2}O\textsubscript{3} precipitates in this sample significantly raise the crossover temperature at which thermal fluctuations dominate from \sim 60 K, as compared to \sim 30 K in YBCO single crystal samples.\textsuperscript{41} The BZO nanorods and RE\textsubscript{2}O\textsubscript{3} precipitates provide strong pinning at \sim 30 – 60 K as clearly shown by multiple other studies.\textsuperscript{45,47,48}

Further information about the additional low temperature weak pinning can be obtained by analysis of the angular dependence of J\textsubscript{c} at 4.2 K using the anisotropic scaling approach proposed by Civale et al.\textsuperscript{49} If the pinning is due to uncorrelated defects randomly distributed over angular space, then J\textsubscript{c} should depend on H and \theta only through a single variable J\textsubscript{c}(H, \theta) = J\textsubscript{c}[\varepsilon(\theta)] where \varepsilon(\theta) = [\cos^2(\theta) + \gamma^{-2}\sin^2(\theta)]^{1/2}, where \gamma is the electronic mass anisotropy parameter. Since we have observed that J\textsubscript{c} follows J\textsubscript{c} \propto H^{-\alpha} at low temperature especially at 4.2 K, as shown in Fig. 2 (a), then J\textsubscript{c}(H, \theta) \propto [H\varepsilon(\theta)]^{-\alpha}.

Power law behavior with \alpha \approx 0.7 for BZO-containing samples and \alpha \approx 0.5 for non-BZO samples was reported by us previously.\textsuperscript{22} Figure 8 presents J\textsubscript{c}(\theta) at 5, 15 and 25 T using the anisotropic scaling parameters \alpha = 0.7 and \gamma = 3. These curves give a reasonable fit of the data in the low angle region, from 0\degree up to about 60 – 70\degree, leading us to conclude that, over this wide range, all around the c-axis where the correlated effects of the BZO nanoparticles are so evident at higher temperatures, the pinning is indeed uncorrelated and random. Clearly the most reasonable interpretation of this behavior is that the additional pinning that operates only up to 30 – 40 K
dominates over the strong pinning effect of the BZO nanorods at lower temperatures. It is interesting that the scaling suggests an effective anisotropy $\gamma = 3$ that is less than the $H_{c2}$ anisotropy $= 5$ expected from the intrinsic mass anisotropy $\gamma^2 = 25 - 30$. The reasons for this behavior are unclear at this time, but smaller anisotropy is positive in any case of applications. It is interesting that $\gamma = 3$ is also an excellent fit to the very broad study of the angular variation of $H_{irr}(T)$ for a very similar sample over the range 55-80 K and $H$ up to 45 T. In this case the effective $\gamma$ was evaluated in the limit that the pin strength goes to zero, rather than in the very finite pin strength limit assessed here, implying a rather consistent behavior over a wide range of the superconducting phase space.

The power law dependence of $J_c$ on $H$ has been reported in many works. However, the increase of $\alpha$ from 0.5 (no nanorods) to 0.7 at 4.2 K when BZO nanorods are present is in contrast to high temperature observations where the presence of BZO nanorods decreases $\alpha$. This is yet more evidence that different pinning mechanisms operate at high and low temperatures.

Based on the above analysis, we propose that BZO additions lead to a significant density of strain-induced, weak pins that cannot resist thermal fluctuation much above about 30 K. Since they do operate at all fields up to 31 T at 4.2 K, it is clear that they are much denser than the strong BZO and RE$_2$O$_3$ pins. Due to the strong effects of increasing field and temperature, we assume that these highly effective pins are dense but point pins (effective size of the order of $\xi^3$ or less, where $\xi$ is the coherence length). Our earlier 4.2 K comparison of non-BZO and BZO-containing samples up to 31 T showed two significant features: One was that the BZO samples have significantly higher $J_c$ while the second was that the enhancement disappeared by 30 − 35 T, certainly a high field but actually only about 25% of $H_{irr}$ or $H_{c2}$ at 4.2 K. Here our variable temperature examination of the BZO sample shows that the uncorrelated pinning effects produced by the point pins are only visible up to $\sim 30$ K. Both characterizations show that the pin strength decays rapidly with increasing $H$ and $T$, fluctuation with them being small and easily thermally de pinned. As Fig. 1 shows, only the larger BZO and RE$_2$O$_3$ strong pins are visible in TEM, so we do not yet have a measure of the point pin concentration. But we can reasonably infer that their density must be high, since they are able to completely hide the effect of the BZO nanorods, even at fields below $B_0$ (compare the $J_c$(1 T) data at 77 and 10 K in Figs. 3a and 3d). Indeed, the depressed $T_c$ of MOCVD and PLD YBCO samples induced by BZO nanorods has recently been attributed to oxygen deficiencies introduced by strain imposed by the lattice mismatch between BZO nanorods and the YBCO matrix. The specific supporting evidence was provided by atomic-resolution Z-contrast imaging and electron energy loss spectroscopy which showed oxygen deficiencies surrounding BZO nanorods, a finding which also supports our proposal for the presence of dense point pins. Thus the overall conclusion of our study is extremely positive: the splayed BZO nanorods found in this coated conductor provide strong correlated pinning to enhance $J_c$ around $c$-axis, a result shown in numerous studies in the 50-77 K range, but they also greatly add to the lower temperature $J_c$ by additionally inducing dense but weak isotropic pinning by strain-induced point defects that raise $J_c$ in the whole angular range at fields up to at least 31 T at 4.2 K where thermal depinming effects are small.

V. CONCLUSIONS

In this paper, we presented a very detailed $J_c(H, T, \theta)$ characterization of a modern, very high critical current density REBCO thin film containing $c$-axis oriented BZO nanorods and $ab$-plane RE$_2$O$_3$ pinning arrays over an exceptionally broad temperature ($4.2 - 77$ K) and magnetic field range ($0 - 31$ T). Analyzing the $J_c$ data we studied the pinning evolution on temperature associated with BZO nanorods. An important new conclusion is that weak isotropic pinning from point defects produced by the strain field around BZO nanorods dominates $J_c$ at low temperatures. More specifically, we observed that the usual $c$-axis $J_c$ peak caused by BZO nanorods disappears with decreasing temperature, and vanishes completely below $\sim 30$ K. At 4.2 K, we found that $J_c$ along the $c$-axis decays as $J_c \propto H^{-\alpha}$ with magnetic field up to 31 T. Although $J_c$ decays faster with magnetic field compared with samples without BZO nanorods at 4.2 K, it is still higher at field up to at least 31 T. At low temperatures, the $c$-axis $J_c$ peak is not seen at any magnetic field and the only correlated pinning present occurs around the $ab$-plane.

Acknowledgments

We are very grateful to colleagues in the HTS R&D group at the NHMFL who have provided many valuable comments and discussions, especially Alex Gurevich, David Hilton, Fumitake Kametani, and Chiara Tarantini. Some aspects of this work were supported by the Department of Energy, Office of Electric Delivery and Energy Research (grant number: DE-FC07-08ID14916) and some by the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement DMR-0654118 and by the State of Florida.
FIG. 8: (color online) Angular dependence of $J_c$ (symbols) and $J_c$ calculated from anisotropic G-L scaling model (thick lines) at 4.2 K and magnetic fields 5, 15 and 25 T. Anisotropic G-L scaling describes well the experimental data over a broad angular range up to, $\sim 0 - 60^\circ$ with $\alpha = 0.7$ and $\gamma = 3$. Thus, the weak uncorrelated pinning contributes to $J_c$ at 4.2 K except in the vicinity of the $ab$-plane where intrinsic pinning is important.

* Electronic address: aixiaxu@magnet.fsu.edu

1 D. C. Larbalestier, A. Gurevich, D. M. Feldmann, and A. Polyanskii, Nature 414, 368 (2001).
2 J. L. MacManus-Driscoll, A. Kursumovic, J. H. Durrell, S. Harrington, S. C. Wimbush, B. Maiorov, L. Stan, H. Zhou, T. Holesinger, and H. Wang, IEEE Appl. Supercond. 19, 3180 (2009).
3 V. Glyantsev, J. Huh, J. Dawley, P. Turner, C. Yung, B. Moeckly, B. Maiorov, Y. Coulter, C. Sheehan, and V. Matias (2011), proceedings of the MRS Spring 2011 Meeting, to be published.
4 G. A. Daniels, A. Gurevich, and D. C. Larbalestier, Appl. Phys. Lett. 77, 3251 (2000).
5 G. Hammerl, A. Schmehl, R. R. Schulz, B. Goetz, H. Bielefeldt, C. Schneider, H. Hilgenkamp, and J. Mannhart, Nature 407, 162 (2000).
6 D. M. Feldmann, T. G. Holesinger, R. Feenstra, C. Cantoni, W. Zhang, M. Rupich, X. Li, J. H. Durrell, A. Gurevich, and D. C. Larbalestier, J. Appl. Phys. 102, 083912 (2007).
7 G. Blatter, M. V. Feigel’man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. 66, 1125 (1994).
8 S. R. Foltyn, L. Civale, J. L. M.-D. Q. X. Jia, B. Maiorov, H. Wang, and M. Maley, Nat. Mater. 6, 631 (2007).
9 T. G. Holesinger, L. Civale, B. Maiorov, D. M. Feldmann, J. Coulter, D. J. Miller, V. A. Maroni, Z. Chen, D. C. Larbalestier, R. Feenstra, et al., Adv. Mater. 20, 391 (2008).
10 M. Daenumling, J. M. Seuntjens, and D. C. Larbalestier, Nature 346, 332 (1990).
11 J. Vargas and D. C. Larbalestier, Appl. Phys. Lett. 60, 1741 (1992).
12 B. Dam, J. M. Huijbregtse, F. C. Klaassen, R. C. F. van der Geest, G. Doornbos, J. H. Rector, A. M. Testa, S. Freisen, J. C. Martinez, B. Stauble-Pumpin, et al., Nature 399, 439 (1999).
13 T. L. Hylton and M. R. Beasley, Phys. Rev. B 41, 11669 (1990).
14 L. Civale, A. D. Marwick, T. K. Worthington, M. A. Kirk, J. R. Thompson, L. Krusin-Elbaum, Y. Sun, J. R. Clem, and F. Holtzberg, Phys. Rev. Lett. 67, 648 (1991).
15 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, Nat. Mater. 3, 439 (2004).
16 S. Yasunaga, M. Mukaida, A. Ichinose, S. Horii, R. Teranishi, K. Yamada, K. Matsumoto, R. Kita, Y. Yoshida, and N. Mori, Physica C 468, 1858 (2008).
17 J. Häniisch, C. Cai, R. Hühne, L. Schultz, and B. Holzapfel, Appl. Phys. Lett. 86, 122508 (2005).
18 S. Engel, T. Thersleff, R. Hühne, L. Schultz, B. Holzapfel, S. Engel, T. Thersleff, L. Schultz, B. Holzapfel, and L. Schultz, Appl. Phys. Lett. 90, 102505 (2007).
19 S. A. Harrington, J. H. Durrell, B. Maiorov, H. Wang, S. C. Wimbush, A. Kursumovic, J. H. Lee, and J. L. MacManus-Driscoll, Supercond. Sci. Technol. 22, 022001 (2009).
20 D. M. Feldmann, T. G. Holesinger, B. Maiorov, S. R. Foltyn, J. Y. Coulter, and I. Apodaca, Supercond. Sci. Technol. 23, 095004 (2010).
21 C. Meingast and D. C. Larbalestier, J. Appl. Phys. 66, 5971 (1989).
22 A. Gurevich, Supercond. Sci. Technol. 20, S128 (2007).
23 M. Miura, B. Maiorov, S. A. Baily, N. Haberkorn, J. O. Willis, K. Marken, T. Izumi, Y. Shiohara, and L. Civale, Phys. Rev. B 83, 184519 (2011).
