Supplementary Information: Enhanced flux pinning in YBCO multilayer films with BCO nanodots and segmented BZO nanorods

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XRD characterizations

The phase purity was checked by measuring $\theta - 2\theta$ scans and no impurities were found (Fig. S1). The strongest peak of BZO is seen around 43° which confirms the crystallinity of BZO. Furthermore, the BCO (002) is seen around 41° but since the particles are very small, only a wide and weak peak is seen around that value. Furthermore, the measurement was used to define the $c$ axis parameter of YBCO (Table S1). The dopant increases the length of the $c$ axis, 11.68 Å on the undoped sample and at least 11.71 Å on all the doped samples. Since there is no clear correlation with the number of layers in the film, it is highly possible that the differences between samples other than the undoped Y are caused by insignificant sample-to-sample variation. The correlation length (Table S1) in the samples, signifying the order of the lattice, is between 80 and 100 Å, only Y and Z are better ordered. This seems to be in line with the FWHMs of the YBCO (005) as well, that are below 0.30° on all samples, but Y and Z have significantly smaller FWHM, 0.15° and 0.10°, respectively. Furthermore, according to the $2\theta - \phi$ scans of YBCO (212), the films are biaxially twinned, apart from the CZ which has a distorted twin structure. The scan of YBCO (102) showed that there were no $a$ axis oriented grains.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Figure_S1.png}
\caption{The $\theta - 2\theta$ measurements of all samples.}
\end{figure}
Table S1. The most important structural parameters measured by XRD and the resistive critical temperatures, $T_c$ and the widths of the transitions, $\Delta T_c$.

| sample | FWHM YBCO (005) ($^\circ$) | $c$ axis length (Å) | $r_c$ (Å) | $T_c$ (K) | $\Delta T_c$ (K) |
|--------|-----------------------------|---------------------|-----------|-----------|------------------|
| Y      | 0.15                        | 11.680              | 124       | 88.4      | 1.4              |
| C      | 0.28                        | 11.722              | 74        | 87.5      | 2.1              |
| Z      | 0.10                        | 11.727              | 154       | 87.8      | 2.2              |
| CZ     | 0.26                        | 11.729              | 76        | 85.0      | 3.9              |
| m1000CZ| 0.29                        | 11.719              | 88        | 87.5      | 1.9              |
| m1000ZC| 0.29                        | 11.730              | 96        | 88.1      | 2.0              |
| m500   | 0.29                        | 11.713              | 98        | 88.4      | 1.9              |
| m250   | 0.29                        | 11.724              | 88        | 88.0      | 2.0              |
| m100   | 0.25                        | 11.721              | 95        | 88.4      | 1.9              |

Figure S2. (a) The BCO particles seen in the m1000ZC sample, indicated by white arrows. (b) Other defects in the same sample seen in the TEM images.
Figure S3. The critical current densities at 40 K (a) in the case $B \parallel c$ axis of YBCO and (b) $B \parallel ab$ plane, extracted from the angular dependencies. The part of the data for m100 is missing due to current limitations of the measurement system.

**TEM characterizations**

The TEM image (Fig. S2 (a)) shows BCO particles in m1000ZC. The diameter of the particles is roughly $(3 \pm 1)$ nm. Other defects seen in the BCO layer of 1000ZC (Fig. S2 (b)) are strained zones, dislocations, basal dislocations and Y124 intergrowths. Additionally, the BCO layer contains very long stacking faults that are seen as white lines in the images. The BZO layer has also stacking faults, although shorter ones. The interface between the BZO layer and the BCO doped layer is good, although there are some dislocations to accommodate strain changes between a BCO and BZO doped layers. On the STO substrate/film interface, there is a $15~\text{–}~20$ nm thick layer that is not fully crystallized. On the BZO/BCO interface, there can be seen accumulation of BCO, sometimes above BZO nanorods. As a whole, the effect of these defects on pinning is smaller as compared to correlated pinning centres.$^{34,35}$

As measured by TEM, the thickness of m250 sample is 310 nm, somewhat less than the 380 nm of the m1000ZC. The m250 has a 6 nm disordered layer at the film/substrate interface. The nanorods in m250 are similar to 1000ZC, although shorter because of the smaller layer thickness. The twin regions are small which means that there are also correspondingly large number of twin boundaries. The sample m100 is 305 nm thick with a 5 nm layer of unordered YBCO next to the substrate interface. The BZO here is not splayed, but straight and the BZO layer thickness is on average only 16 nm. The thickness and distance of the nanorods is the same as on samples with thicker layers. The CZ sample, on the other hand, has both good and strained areas, and altogether the sample is very strained. There are no BZO columns but some nanodots have formed nanocolumn-like features.

**Superconducting properties**

To get a good picture of the pinning properties of the samples, the measurements of $J_c(\theta)$ were made at 10, 40 and 77 K in 1, 2, 4, 6 and 8 T. The magnetic field dependencies of $J_c$ at 40 K, extracted from the angular data, show clear differences between samples. In the case $B \parallel c$ axis of YBCO the (Fig. S3 (a)), the samples having a $c$ peak due to BZO have a high $J_c$ within the deposited samples. Also, samples with nanorods but no $c$ peak have a rather high $J_c$ value. Only the m250 is deteriorated more in increasing field than other samples with similar self-field $J_c$. In the $ab$ direction (Fig. S3 (b)) the Z sample with a monolayer of BZO is not any more among the highest $J_c$ samples. Here, especially the high $J_c$ values of samples with a large number of layers can be most clearly seen. Only the $J_c$ of the C sample decreases faster under the external magnetic field than other samples with similar value of $J_c$ in the self-field.

**Simulation details**

All vortices in the simulation were divided into 40 vertical parts, i.e. the simulation was formed of stacked layers. All the layers were subject to periodic boundary conditions in the $ab$ plane. Thus the vortices leaving the samples due to the Lorentz force re-enter from the other side. Inside each layer, a vortex can interact with other vortices in the same layer. Between the layers, the line tension of a vortex acts as a binding force. Because of the layered structure, the simulation cannot be used to describe the situations close to case $B \parallel ab$ plane of YBCO. The only defects that were introduced to pin vortices were either nanorods (radius of 3 nm) or nanodots (radius 1.5 nm). The distribution of the nanorods was taken from previous
but for computational reasons their mutual distances were doubled. If there were another layer of nanorods, like in “m250 equivalent” sample, the position of the nanorods at another layer was varied slightly. This was done to avoid having nanorods on top of each other, as suggested by the experimental results in this work. The positions of the nanodots were random. No other typical defects for YBCO were introduced because we wanted to see the effect of these defects in particular. The defects were divided so that there are first nanorod layers with certain thickness then equally many nanodot layers. This was repeated until the total thickness of simulation is 40 layers. To speed up the stabilization of the vortex lattice, initially the vortices were set into hexagonal lattice. In the model, the temperature is not implemented i.e. it is 0 K. The simulation was written in Python using molecular dynamics with velocity Verlet algorithm.

The $J_c$ was iterated by bisection method. The simulation was run with one current and the next current was adjusted according to the stability of the found state. The solution was considered stable if the vortices were moving less than double the coherence length of YBCO in the $ab$ plane. This was done by comparing the position of the vortex at 1,000 and 500 steps earlier with the present coordinates. Also, if this condition of stability was not fulfilled, the stability was determined based on the speed of vortices. If it was below 200 m/s, the simulation was considered stable. Both stability conditions were checked at the same time, with regular intervals.

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