

The effect of BaCeO₃ dopant concentration on magnetically defined B_{irr} and B_{c2} in YBa₂Cu₃O_{6+x} thin films deposited on SrTiO₃ substrates

H. Huhtinen¹, H. Palonen^{1,2}, M. Malmivirta¹, R. Jha³, V. P. S. Awana³ and P. Paturi¹

¹ Wihuri Physical Laboratory, Department of Physics and Astronomy, FI-20014 University of Turku, Finland

² The National Doctoral Programme in Nanoscience (NGS-NANO), Turku, Finland

³ The Superconductivity Division, National Physical Laboratory (CSIR), New Delhi 110012, India

E-mail: hannu.huhtinen@utu.fi

Abstract. The effect of BaCeO₃ (BCO) dopant concentration on magnetically obtained irreversibility field B_{irr} and upper critical field B_{c2} is investigated in YBa₂Cu₃O_{6+x} (YBCO) thin films prepared by pulsed laser deposition on SrTiO₃ (001) substrates. The unchanged values of B_{irr} and B_{c2} are connected to the structural properties where BCO has been observed to form non-elongated and randomly distributed particles within the whole concentration range of 0 – 10 wt.%. This indicates a different type of flux pinning mechanism for BCO-doped YBCO than for the widely studied BaZrO₃-doped YBCO where increased B_{irr} and B_{c2} values were observed.

1. Introduction

In order to optimize the high- T_c superconductors for large-scale applications, the requirement for optimized flux pinning in high magnetic fields is realized by organizing a network of self-assembled isotropic nanodots or anisotropic nanorods in the YBa₂Cu₃O_{6+x} (YBCO) matrix [1–5]. In addition to the improved in-field critical current j_c of YBCO thin films, its intrinsic high anisotropy has been radically modified by adding BaZrO₃ (BZO), as it creates film penetrating non-superconducting columnar defects with diameter equal of the vortex core [4, 6]. The situation changes dramatically, when the non-elongated and randomly distributed nanoparticles of *e.g.* BaCeO₃ (BCO) are used as a pinning centres [7].

For the undoped YBCO, the upper critical field B_{c2} is roughly six times larger in the field in-plane than in the field out-of-plane direction [8]. In spite of the intrinsic property of B_{c2} , it was also demonstrated that BZO doping clearly increases B_{c2} in all field directions, and the angular dependence of B_{c2} follows the Blatter scaling [9]. When compared to the BZO nanorods as a dopant, the enhancement of B_{c2} is more pronounced in the case of BaHfO₃ (BHO) which has almost similar in-plane lattice parameters than BZO, $a = b = 4.17 \text{ \AA}$, but forms slightly thinner but higher density of nanorods in YBCO matrix [10]. This leads to an assumption that BCO, which also has longer lattice parameters ($a = b = 4.40 \text{ \AA}$) than in BZO and BHO as well



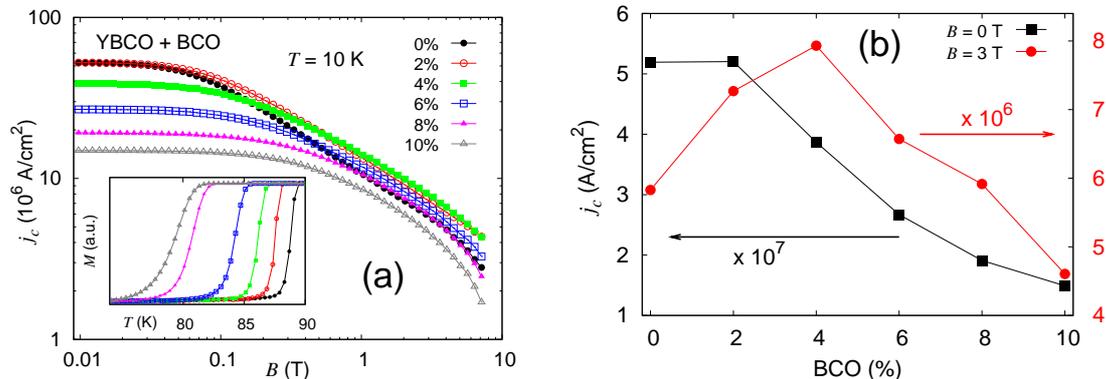


Figure 1. (a) Magnetic field dependence of j_c determined from the hysteresis loops at 10 K and the normalized ac magnetization as function of temperature (inset) for undoped and BCO doped YBCO films. (b) BCO content dependence of j_c in external fields of 0 and 3 T.

as reduced amount of strain, could produce completely different critical current properties in high magnetic fields, as well as different irreversibility field B_{irr} and B_{c2} .

Based on magnetic measurements, we have determined the temperature dependence of B_{irr} and B_{c2} for YBCO films doped with isolated BCO particles. The results are discussed with the conclusions obtained for BZO doped YBCO films having columnar defect network.

2. Experimental and basic magnetic properties

The films were prepared on (100) SrTiO₃ (STO) substrates by pulsed laser deposition (PLD) from YBCO targets with varied dopant concentrations containing 0, 2, 4, 6, 8 and 10 wt.% of BCO. Details of the preparation of targets and films as well their structural characterizations are described elsewhere [7]. Because the XRD (004) peak of BCO starting at $2\theta \approx 41^\circ$ is extremely wide, it was concluded that BCO is distributed into YBCO lattice as very small particles in (00 l) orientation. According to Scherrer formula, the width of the peak produces an average particle size of ≈ 2 nm. In addition, the $(2\theta, \phi)$ scans of YBCO (212)/(122) peaks indicate biaxial twins up to 10 wt.% of BCO, *i.e.* the strain is notably smaller than in the BZO-doped YBCO.

Magnetic measurements were made with a Quantum Design PPMS system, and the onset critical temperatures $T_{c,onset}$'s were determined with ac magnetization measurements in the range of 10 - 100 K (ac field of 0.1 mT). As can be seen from the inset of figure 1(a), $T_{c,onset}$ decreases systematically with increasing BCO concentration, having the maximum onset value of 89.5 K for 0 % and minimum of 82 K for 10 % BCO doped YBCO. In contrast with results for BZO doped YBCO where the width of the transition ΔT_c stays within 1.3 - 2.0 K [11], ΔT_c increases with increasing BCO content from 1.2 K (0 %) to 3.7 K (10 %), indicating increased amount of lattice mismatch induced strain and possible oxygen deficiency at the interface between dopants and superconducting matrix [7, 10]. The critical current densities j_c at 10 K were determined from the hysteresis loops with the Bean model for rectangular shape film: $j_c = 2\Delta m/[a(1 - a/3b)V]$, where a and b ($b \geq a$) are the width and the length of the sample, V is the sample volume and Δm is the opening of the hysteresis loop [12]. The shape of the $j_c(B)$ curve varies with BCO concentration, showing clearly different magnetic field dependence, as can be seen from figure 1(a). Figure 1(b) shows the BCO concentration dependent j_c data, shown at two different magnetic fields. In zero field, the maximum j_c can be achieved with low BCO content below 2 %, while the optimal concentration is increased up to ≈ 4 % in the applied field range of 3 T. These results are in line with our earlier observations for BCO doped YBCO films [7].

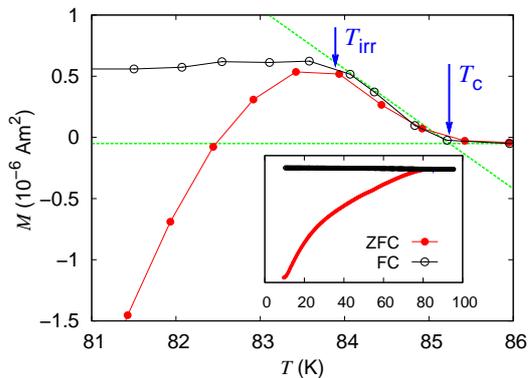


Figure 2. Determination of T_{irr} and T_c from the close-up of ZFC and FC curves as illustrated for the 4 wt.% BCO doped YBCO sample measured in 0.25 T field. Due to subtle diamagnetic signal of the substrate and the sample holder, the horizontal dashed line indicates the zero magnetization level of the sample and the descending dashed line the low temperature region as expressed in ref. [10].

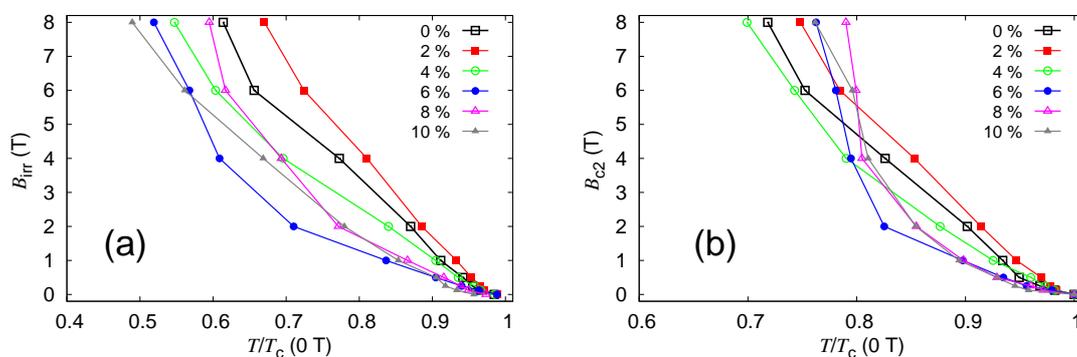


Figure 3. The reduced temperature dependence of B_{irr} (a) and B_{c2} (b) for YBCO thin films doped with various BCO concentration.

PPMS dc magnetization option is used to measure temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) magnetizations in fields of 0, 0.125, 0.25, 0.5, 1, 2, 4, 6 and 8 T. The irreversibility temperature T_{irr} for each measuring field is determined as a temperature where the ZFC and FC curves start to deviate from each other. The upper critical field B_{c2} is determined by using a method described in [10] where B_{c2} is equivalent to the magnetic field at which linear extrapolation of the FC magnetization from the low and high temperature regions meet (intersection of dashed lines), indicating T_c , as shown in figure 2.

3. Irreversibility field and upper critical field

When looking at the reduced temperature dependence of B_{irr} in figure 3(a), only the smallest amount of BCO (2 %) has even some kind of potential to increase the B_{irr} , while all the other BCO contents give smaller B_{irr} in the whole temperature range. The same situation occurs when B_{irr} is plotted as a function of absolute temperature values. However, when taking into account the sensitivity of the determination of T_{irr} , we can conclude that no clear difference between the curves can be observed and therefore the BCO doping does not have notable influence on B_{irr} . The behaviour is different what we have seen for BZO doped YBCO films, where clearly lower magnetically determined B_{irr} for undoped YBCO is explained by the strong pinning of BZO nanorods [11].

From the temperature dependence of B_{c2} in figure 3(b), we can observe that the curves with different BCO concentrations are situated close to each other without BCO dependent tendency, and therefore we conclude that BCO doping does not have effect on B_{c2} either. It is good to

notice that both B_{irr} and B_{c2} are observed from the high temperature range, close to onset T_c , where extremely small BCO pinning centers are not effective. This gives an explanation for the difference where increased $j_c(B)$ was seen in the data measured at 10 K. The behaviour is also in contradiction to the results, where BZO doping has been observed to cause a clear change in B_{irr} and B_{c2} , indicating a change in the coherence length and the band structure of YBCO [9, 11]. This is in agreement with our structural results where strain has been observed to be clearly smaller in BCO doped than in BZO doped YBCO films [7], and it is also in line with recent results where strained regions could prevent the Cooper pair formation thus becoming effective for pinning vortices [13]. However, an interesting detail can be seen that with high BCO content, between 6 and 10 % of BCO, at around $T = 0.85T_c$ increase of B_{c2} is much steeper with decreasing temperature when compared with the curves of low BCO concentration. In order to throw light on this phenomenon and assess the possible mechanism for it, the pulsed magnetic field measurements up to clearly higher fields are needed. In addition, it has been suggested earlier that the area of interface between dopant and the superconducting matrix plays an important role in the enhancement of B_{c2} [10], but according to our results, the shape and the distribution of the dopants seem to be the most critical and therefore the selection of suitable pinning material for different type of applications is the most important factor.

4. Conclusions

In this work, the irreversibility field and the upper critical field were determined from the magnetic measurements for YBCO thin films doped with various concentration of single BCO nanoparticles. Our results show that the temperature dependence of both B_{irr} and B_{c2} remained unchanged when BCO dopant was introduced into the YBCO lattice. The observation differs clearly from the results for YBCO with BZO nanorods, and the phenomenon is connected to the formation of isotropic BCO nanoparticles as well as to the lower strain caused by the BCO nanoparticles in the YBCO matrix.

Acknowledgments

The Wihuri Foundation, Finnish Foundation for Technology Promotion and Finnish Cultural Foundation are acknowledged for financial support.

References

- [1] Civale L, Marwick A D, Worthington T K, Kirk M A, Thompson J R, Krusin-Elbaum L, Sun Y, Clem J R and Holtzberg F 1991 *Phys. Rev. Lett.* **67** 648
- [2] Blatter G, Feigel'man M V, Geshkenbein V B, Larkin A I and Vinokur V M 1994 *Reviews of Modern Physics* **66** 1125
- [3] Polkovnikov A, Kafri Y and Nelson D R 2005 *Phys. Rev. B* **71** 014511
- [4] Maiorov B, Baily S A, Zhou H, Ugurlu O, Kennison J A, Dowden P C, Holesinger T G, Foltyn S R and Civale L 2009 *Nat. Mater.* **8** 398–404
- [5] Baca F J, Barnes P N, Emergo R L S, Haugan T J, Reichart J N and Wu J Z 2009 *Appl. Phys. Lett.* **94** 102512
- [6] MacManus-Driscoll J L, Foltyn S R, Jia Q X, Wang H, Serquis A, Civale L, Maiorov B, Hawley M E, Maley M P and Peterson D E 2004 *Nat. Mater.* **3** 439
- [7] Irjala M, Huhtinen H, Jha R, Awana V P S and Paturi P 2011 *IEEE T. Appl. Supercond.* **21** 2762
- [8] Wijngaarden R J, Heeck K, Griessen R, Menovsky A A and Menken M J V 1988 *Physica C* **152** 523
- [9] Palonen H, Huhtinen H, Shakhov M A and Paturi P 2013 *Supercond. Sci. Technol.* **26** 045003
- [10] Matsushita T, Nagamizu H, Tanabe K, Kiuchi M, Otabe E S, Tobita H, Yoshizumi M, Izumi T, Shiohara Y, Yokoe D, Kato T and Hirayama T 2012 *Supercond. Sci. Technol.* **25** 125003
- [11] Paturi P, Malmivirta M, Palonen H and Huhtinen H 2013 *Journal of Physics: Conference Series* submitted
- [12] Wiesinger H P, Sauerzopf F M and Weber H W 1992 *Physica C* **203** 121
- [13] Llordés A, Palau A, Gázquez J, Coll M, Vlad R, Pomar A, Arbiol J, Guzmán R, Ye S, Rouco V, Sandiumenge F, Ricart S, Puig T, Varela M, Chateigner D, Vanacken J, Gutiérrez J, Moshchalkov V, Deutscher G, Magen C and Obradors X 2012 *Nat. Mater.* **11** 329