Physica C 470 (2010) 295-303

Contents lists available at ScienceDirect

Physica C

journal homepage: www.elsevier.com/locate/physc

Fluctuation induced magneto-conductivity studies in $YBa_2Cu_3O_{7-\delta} + xBaZrO_3$ composite high- T_c superconductors

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ARTICLE INFO

Article history: Received 7 November 2009 Received in revised form 27 December 2009 Accepted 13 January 2010 Available online 20 January 2010

Keywords: YBCO superconductor Fluctuation effects Magneto-transport Critical parameters

ABSTRACT

Fluctuations on the electrical conductivity of polycrystalline YBa₂Cu₃O_{7- δ} + xBaZrO₃ (*x* = 1.0, 2.5, 5.0 and 10.0 wt.%) superconductors were investigated from the resistivity vs. temperature data for zero field and 8 T (Tesla) external magnetic fields. Attempts have been made to identify the optimum inclusion of BaZrO₃ (BZO) in YBa₂Cu₃O_{7- δ} (YBCO) superconductors. The phase formation, texture and grain alignments were analyzed by XRD and SEM techniques. Then the effects of superconducting fluctuations on the electrical conductivity of granular composite superconductors were studied for zero field and 8 T external magnetic fields. Though inclusions of BZO sub-micron particles are not expected to influence superconducting order-parameter fluctuation (SCOPF) much, the transition from 2D to 3D of the order parameter in the mean-field region depends on the BZO content in the composites. It has been observed that BZO residing at the grain boundary of YBCO matrix influences the tailing region without having significant change in the mean-field critical temperature. In the present work, attention has been focused mostly in the experimental domain relatively above the *T_c*. It reveals that, 1 wt.% composite exhibits a better superconducting property in comparison with pure YBCO.

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1. Introduction

The enhancement of flux pinning by defect engineering in the nano-scale to maintain high current densities at high magnetic fields has drawn a lot of attention from the scientific community. Barium zirconate (BZO) is the most attractive material to induce artificial pinning centers in $YBa_2Cu_3O_{7-\delta}$ (YBCO) bulk, melt textured and thin films in order to increase the critical current density [1]. Gutierrez et al. [2] found that non-coherent interfaces are the driving force for generating nano-structured superconductors. The main reasons for using BZO as artificial pinning center in YBCO are: (i) BZO has a high melting temperature with respect to YBCO so that the growth kinetics should be slow leading to small particles, (ii) zirconium does not substitute in the YBCO structure and (iii) although BZO can grow epitaxially with YBCO, it has a large lattice mismatch (approximately 9%), therefore strain between the phases could introduce defects for enhanced pinning. It was shown by Macmanus et al. [3] that, inclusions of BZO particles of dimensions in the 10-100 nm range could result in a significant improvement of dc properties of YBCO. Very steep pinning potentials of BZO inclusions and an improvement of the critical current density, J_c have been registered by Ciontea et al. [4] and Pompeo et al. [5]. Generation of this sub-micron sized defect BZO has many favourable properties like toughness enhancement towards the composite formation with YBCO. When BZO is made to form composite with YBCO, it develops strain between the phases that introduces enhanced pinning due to a large lattice mismatch (~9%) of BZO. These sub-micron sized BZO particles at the grain boundaries form strong flux pinning centers enhances some of the superconducting properties of YBCO [6]. The effect of adding nanocrystalline BZO powders to YBCO target on the flux pinning properties of YB-CO + *x*BZO films has been extensively investigated by Padma Kumar and Vijay Kumar [7].

In recent years great strides have been made in order to increase the pinning of vortex lines, which is an essential achievement for useful applications of YBCO superconductors in general, and of cuprate superconductors in particular. The mixed-state flux dynamics in high temperature superconductors (HTSCs) is an interesting and thrust area for contemporary research. Several models [8–10] have been proposed to describe the complete shape of resistive transition in presence of magnetic fields. However, Aslamazov–Larkin (AL) approximations are valid in the effect of fluctuations in superconducting order parameter. It gives rise to the effective existence of order parameter and the renormalization of relaxation time of electronic excitations. The magneto-resistance and broadening in the resistive transition of these new compounds caused by both electrical current and magnetic field [11–15] have been investigated extensively.





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^{0921-4534/\$ -} see front matter @ 2010 Elsevier B.V. All rights reserved. doi:10.1016/j.physc.2010.01.038

It is believed that the broadening behavior in the low-resistance region near T_c ($\rho = 0$) is a consequence of the progressive decoupling of the superconducting grains with increasing BZO content at the grain boundaries. The broadening excites many researchers to suggest different interpretations for the origin of this unusual observation. The existence of an intrinsic double superconducting transition in the analysis of temperature derivative of $\rho(T)$ for YB-CO + xBZO suggests that: (i) within a temperature resolution of 20 mK the intrinsic resistive transition of the HTSC does not exhibit any double transition anomaly and (ii) the double peak structure observed in $d\rho/dT$ by some authors [16–21] is associated with stoichiometric inhomogeneities and experimentally developed microvoids. It is assumed that a superconducting percolation cluster is formed in the plane where the electric current flows. Such a structure provides an effective pinning since the magnetic field is trapped in finite clusters in normal phase. The non-zero dissipation is thought to be the motion of flux lines resulting from the melting (or de-pinning) of the vortex solid near the onset of finite resistance [22-24]. It is interesting to find the fluctuation induced conductivity smaller in superconductor-dielectric composites, corresponding to their counterparts in pristine YBCO for both 3D and 2D regimes.

In this paper high resolution data of the electrical resistivity $\rho(T)$ around the superconducting transition of YBCO + *x*BZO (*x* = 1.0, 2.5, 5.0, 10.0 wt.%) is presented. Analysis for the agreement of the experimental data with the predictions of different theoretical models has been made. The results obtained from this investigation show that the fluctuation conductivity has better Gaussian behavior as a consequence of the inclusion of sub-micron sized BZO particles. It resolved an important issue regarding the slight increase of critical temperature in the superconducting regime. The origin of the enhanced transport properties of composites has been correlated with the microstructure at the sub-micron scale induced by non-coherently oriented BZO particles.

2. Experimental

The YBCO sample was prepared from the stoichiometric compositions of high-purity powders of Y₂O₃, BaCO₃ and CuO by solidstate reaction route and synthesis of BZO was carried out by employing nitrate precursors like Barium nitrate, Ba(NO₃)₂ and Zirconyl nitrate hydrate, ZrO(NO₃)₂·xH₂O in (1:1 M ratio) with a calcination temperature of 900 °C. Superconductor YBCO + dielectric BZO composites were prepared from a mixture of pre-reacted YBCO and BZO powders. A series of polycrystalline composite samples of YBCO + *x*BZO (*x* = 1.0, 2.5, 5.0 and 10.0 wt.%) have been prepared at a calcination temperature of 880 °C by standard solidstate reaction route. The products were then pressed into pallets for final stage of sintering at 900 °C for 12 h and then cooled to 500 °C where they kept for 12 h in an oxygen atmosphere for annealing. All the samples were characterized by X-ray powder diffraction technique (PW 3020 vertical goniometer and 3710 X' Pert MPD control unit, $CuK\alpha$) and temperature dependent resistance $\rho(T)$ was measured using standard four-probe technique with a Nanovoltmeter (Keithley-181) and an indigenously developed constant current source. With the voltage resolution of 10^{-8} V of the Nanovoltmeter, a constant current source of 10 mA flowing through the samples gives a resolution ${\sim}1\,\mu W$ in the measured resistance. A closed cycle Helium refrigerators (APD cryogenics-HC2) and a temperature controller (Scientific Co. 96001) were used for temperature variation. The temperature controller used a silicon diode sensor having a temperature resolution of ±0.1 K. A Computer controlled data acquisition system was used to acquire the resistance data from 40 K to room temperature. The resistance data were required during the heating cycle with heating rate confined to 3 K-min⁻¹. The grain morphology of the samples was analyzed by scanning electron microscope (Model No. JSM-6480 LV, Make JEOL) and the compositional analysis was determined by energy dispersive X-ray analysis (EDX) using an INCA Oxford Analyzer attached to a scanning electron microscope.

3. Materials and methods

3.1. Synthesis and characterization of BZO

Barium nitrate Ba(NO₃)₂ and Zirconyl nitrate hydrate ZrO(-NO₃)₂·xH₂O were employed for the synthesis of BZO powder, 1:1 M ratios of two precursors were taken to a polystyrene bottle and ball-milled for 4 h in ethanol using zirconia balls as milling medium. The ball-milled mixture was thoroughly mixed at ambient temperature, dried and calcined at 900 °C for 2 h at a heating rate of 3 °C/min in a box furnace. The X-ray analysis (Fig. 1) revealed that, BZO is of single phase having a cubic structure with Pm-3 m symmetry. At a sintering temperature of 900 °C the peaks corresponding to barium carbonate and zirconia disappeared while corresponding BZO crystalline phases are observed similar to the results obtained by Ciontea et al. [4]. Zeta particle analyzer was used to estimate the particle size and their distribution in the calcined powders. The micro-structural features of the sintered powders were determined with a scanning electron microscope (SEM). The results from SEM shows that the average particle size distribution (Fig. 2) in the calcined mixture ranges between 500 and 800 nm and can also be better confirmed by the results from Zeta particle analyzer (Fig. 3). These sub-micron sized BZO particles at the grain boundaries form strong flux pinning centers, enhances some of the superconducting properties of YBCO. The above microstructure has been further discussed in connection with the analysis of electronic transport measurements in absence and presence of an externally applied magnetic field.

3.2. Phase formation of YBCO + xBZO

The diffraction pattern analysis of the composite samples of YB-CO + xBZO were indexed using Chekcell software and the phases were confirmed to be orthorhombic at room temperature with a space group Pmmm. Appearance of peaks (0 0 3), (0 0 4), (0 0 5) and (0 0 6) in the XRD pattern reveals (0 0 *l*)-orientation of YBCO.



Fig. 1. XRD patterns of YBCO, BZO and YBCO + *x*BZO composites with different BZO wt.%.

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