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# Effect of granularity and inhomogeneity in excess conductivity of $YBa_2Cu_3O_{7-\delta}$+$x$$BaTiO_3$ superconductor

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ABSTRACT

Synthesis of polycrystalline YBCO+$x$$BaTiO_3$ ($x$=1.0, 2.5, 5.0) superconductor has been done and the effects of granularity and inhomogeneities due to inclusions of nano-$BaTiO_3$ in excess conductivity are reported in this work. The phase formation, texture and grain alignments were analyzed through XRD and SEM techniques. SEM results reveal that the grain size is reduced and morphology is improved with the incorporation of nano-$BaTiO_3$ particles. Superconducting order parameter fluctuation (SCOPF) studies on the electrical conductivity were investigated from the resistivity vs. temperature data in the experimental domain relatively above $T_c$. Log($\Delta\sigma$) vs. log($\varepsilon$) plots show that the 2D to 3D crossover temperature ($T_{LD}$) that demarcates dimensional nature of fluctuation inside the grains is influenced by $BaTiO_3$ incorporation in YBCO matrix. An upward shift of $T_{LD}$ in the mean field region has been observed as a consequent dominance of 3D region with increase in 1 wt% $BaTiO_3$ in the composites as compared to higher inclusions. It has been analyzed that microscopic inhomogeneities produced as a result of diffusion of a fraction of Ti ions into the grains affect fluctuations in the excess conductive region. The interplay of microscopic inhomogeneities produced inside the grains and mesoscopic inhomogeneities in the grain boundaries on the excess conductivity has been explained in terms of thermal fluctuations for the composites.



## 1. Introduction

One important phenomenological investigation of thermodynamic fluctuations in the transport properties of high $T_c$ superconductors (HTSC) has appeared to explain the cause of excess electrical conductivity ($\Delta\sigma$) related to the structural properties. In $YBa_2Cu_3O_{7-\delta}$ (YBCO) ceramics the structure can be viewed as constituted of superconducting layers composed of $CuO_2$ planes separated by isolating CuO chains with complex crystal chemistry and granularity. The principal aim of the studies is closely related to the inclusion of ferroelectric materials to change the chemical ambient in the reservoir layer in order to see how the superconducting properties of the material change. Since the discovery of HTSC, on-site substitution studies have proved very useful in understanding the mechanism of high-temperature superconductivity [1,2]. It has been known that when the ferroelectric materials are preferentially oriented in the (0 0 $l$) direction, they do not exhibit polarization hysteresis integrated with YBCO superconductors. The perovskite ferroelectric (Ba, Sr) $TiO_3$ and perovskite YBCO superconductor possess similar lattice structure (2–3% lattice mismatch in a–b planes) and crystal chemistry. These ferroelectrics embedded in HTSC have shown to generate a stress field and can therefore act as pinning centers [3,4]. Thus a composite of YBCO+$x$$BaTiO_3$ provides an ideal system for experimental study [1,5]. The widely undertaken analysis of excess conductivity in the composite system from different fluctuation theories [6–13] has concentrated mostly on investigating crossover [14–18] from a three-dimensional (3D) fluctuation (exponent, $\lambda$, approaching 0.5) at low temperatures to a two-dimensional (2D) one ($\lambda$ approaching 1.0) on heating across a crossover temperature ($T_{LD}$).

The inherent granular nature of YBCO has drawn attention for the intrinsic properties due to the grains and the extrinsic properties due to grain boundaries leading to strong structural disorder [19,20] at the microscopic and mesoscopic levels, respectively. The transition of the electrical resistance offered by these polycrystalline samples is a two-stage process [21–23]. As the temperature decreases, a pairing transition is first observed and then a coherence transition. At the pairing transition, the superconductivity stabilizes in homogeneous regions within the grains at a temperature that virtually coincides with the critical temperature ($T_c$) of the bulk. A large number of studies have been devoted to investigate the interplay between the superconducting order parameter fluctuation (SCOPF) and the inhomogeneities [24–28]. The mesoscopic inhomogeneities such as grain boundaries, cracks, voids, etc. having much larger length scale than the superconducting coherence length $\xi$ and being temperature independent are expected to influence the $R$–$T$ characteristics. The microscopic inhomogeneities such as structural (twin boundaries, stacking faults) and chemical

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imperfections (oxygen deficiencies, etc.) inside the grains occur in a length scale smaller than the mesoscopic inhomogeneities, but still larger than $\xi$. The temperature-dependent resistivity of the composite system depends on the connectivity of the grains. It has been claimed that for non-superconducting inclusions to be effective as pinning centers, their size should be of the order of coherence length [29]. These defects with size in the range of few times of coherence length have been found to act as a means to inhibit vortex motion, which results in the significant enhancement of critical current density ($J_c$) at high temperatures and applied magnetic fields. This aspect of $J_c$ enhancement is currently under our observation.

A relatively small number of investigations have been done on fluctuation conductivity along the *c*-axis of the HTSC. The pioneering work of Baraduc et al. [30] sets the basis for explaining results in the mean field region. The excess conductivity of the Aslamazov–Larkin (AL) type in the normal state, close to $T_c$ [31], follows a quasi-universal behavior, which is strongly related with Gaussian and genuinely critical fluctuations. Such studies are of interest and may give estimates of microscopic parameters such as the coherence lengths, phase breaking time and dimensionality of the order parameter; thus it could be possible to separate the fundamental disorders at microscopic and mesoscopic levels. In addition, theoretical concepts of the critical regime close to $T_c$ or of the pair formation may help to reveal the mechanism of superconductivity in YBCO.

In this paper, we describe fluctuation conductivity in polycrystalline samples of YBCO+$x$BaTiO$_3$ ($0 \le x \le 5$ wt%) having both inter- and intra-grain modifications. Samples were prepared through standard solid-state reaction route, with nano-BaTiO$_3$ inclusions. To identify power-law divergences in the conductivity the results were analyzed with the logarithmic temperature derivative of the conductivity. Nano-BaTiO$_3$ in comparison to micron size BaTiO$_3$ used for sample preparation has produced considerable modifications in the superconducting transition and consequently in the fluctuation regimes. The mesoscopic and microscopic inhomogeneities raised due to most of the BaTiO$_3$ residing at the grain boundaries and Ti ion diffusion into the grains, respectively, have significantly influenced the excess conductive region as well as the tailing region. The strong influence of BaTiO$_3$ content in the composites in the mean field region has been explained based on BaTiO$_3$ induced modifications on the overall electronic structure of grains.

## 2. Experimental

The YBCO sample was prepared from the stoichiometric compositions of high-purity powders of $Y_2O_3$, $BaCO_3$ and CuO by solid-state reaction route and synthesis of BaTiO$_3$ was carried out through chemical route. Superconductor YBCO+ferroelectric BaTiO$_3$ composites were prepared from a mixture of pre-reacted YBCO and BaTiO$_3$ powders. A series of polycrystalline composite samples of YBCO+$x$-BaTiO$_3$ ($x$=1.0, 2.5 and 5.0 wt%) have been prepared at a calcination temperature of 900 °C by standard solid-state reaction route. The products were then pressed into pellets for final stage of sintering at 920 °C for 12 h and then cooled to 500 °C at which they were 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 of ~1 μW in the measured resistance. A closed-cycle helium refrigerator (APD cryogenics—HC2) and a temperature controller (Scientific Co. 96001) were used for temperature variation. The temperature controller used a silicon diode sensor with 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 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. Material and methods

### 3.1. Synthesis and characterization of BaTiO$_3$

BaTiO$_3$ powder has been synthesized through a chemical route, where it has been calcined at 1250 °C for 2 h at a heating rate of 3 °C/min in a box furnace. The X-ray analysis revealed that BaTiO$_3$ is of single phase with a tetragonal structure and P4mm symmetry. Zeta particle analyzer was used to estimate the particle size and their distribution in the calcined powders. The results from particle size analysis shows that the average particle size of calcined powders of BaTiO$_3$ ranges between 100 and 400 nm (Fig. 1). These nanoparticles of BaTiO$_3$ at the grain boundaries form strong flux pinning centers and enhance some of the superconducting properties of YBCO. The idea of making composites of YBCO with BaTiO$_3$ is to strengthen the intergrain coupling to enhance the transport properties as fine BaTiO$_3$ acts as weak links in granular YBCO superconductors. Though composition variation at the grain boundaries is not expected to influence the onset of superconductivity inside the grains, these sources of fine particles through composite formation would increase the coupling between the grains and hence the transport properties of oxide superconductors.

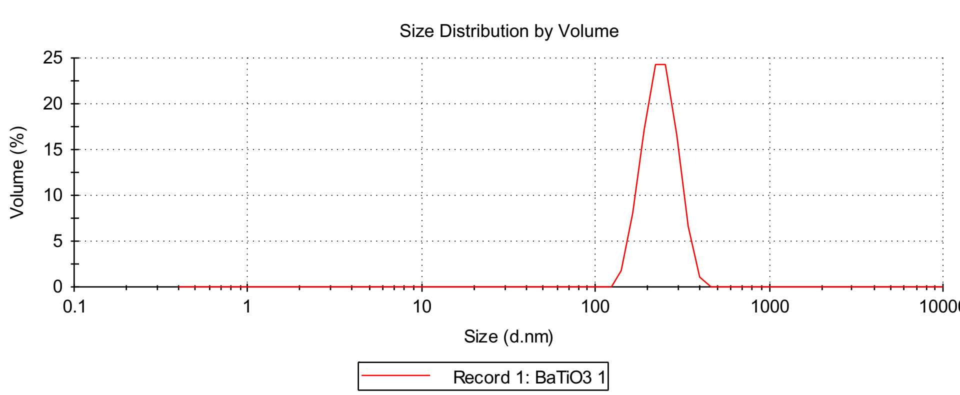


**Fig. 1.** Particle size analysis of BaTiO$_3$.