DC Electrical Resistivity Studies in bulk YBCO/Ag composites

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CERTIFICATE

This is to certify that the thesis entitled **"DC electrical resistivity studies of YBCO/Ag composite"** submitted by **Miss Banani Parida** in partial fulfillment for the requirement towards the award of Master of science degree in physics at **NIT, Rourkela** is an authentic work carried out by her under my supervision and guidance in low temperature lab of Department of physics.

To the best of my knowledge, the matter embodied in the thesis has not been submitted to any other University/Institute for the award of my degree.

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ACKNOWLEDGEMENT

ABSTRACT

Bulk YBCO sample is prepared using chemical route method. Ag is used as composite because of its advantageous properties. The samples are characterized by the ρ vs T measurement performed by FOUR PROBE METHOD. Data are collected by automatic programming and analyzed for the behaviour. SEM is done to study the grain morphology. It is found that by increase in the weight percentage of Ag in the composite sample the transition temperature decreases slightly. From SEM images, it is observed that the grains become smaller in sizes as compared to prestine.

CONTENTS

Chapter I

Introduction

1.1 General remarks

Superconductivity was discovered by Dutch Physicist Heike Kamerlingh Onnes in 1911 while working at low temperature. Superconductivity is a property of metal, alloys, and chemical compound at temperature called transition temperature or critical temperature where electrical resistivity vanishes and they becomes diamagnetic in nature due to Meissners effect.

For a material to be considered a superconductor it has to exhibit two distinctive properties:-

1.   The electrical resistivity in superconductors is zero for temperatures below a certain temperature T_c. So, one can apply dc electrical current without losses. In superconductors, the carriers are coupled forming Cooper pairs which are not scattered & therefore zero resistance is obtained. The condensate that results from this coupling is represented by a wave function (ψ) that varies in distances given by the coherence length ξ (T).

2.   The applied magnetic field is completely expelled from the interior of the superconducting specimen at temperature below the critical temperature T_c. Cooper pairs circulating at the surface of the sample that are able to screen the external magnetic field generate the expulsion. These currents penetrate into the sample in a distance characterized by the penetration length, λ (T), leading to an exponential decay of the magnetic field from the interior of a superconducting sample is known as the Meissner-Ochsenfeld effect. Temperature dependence of electrical resistivity of the oxide superconductor: $YBa_2Cu_3O_{7-\delta}$ exclusion of a weak, external magnetic field from the interior of the superconducting material has been shown in figure1(b).
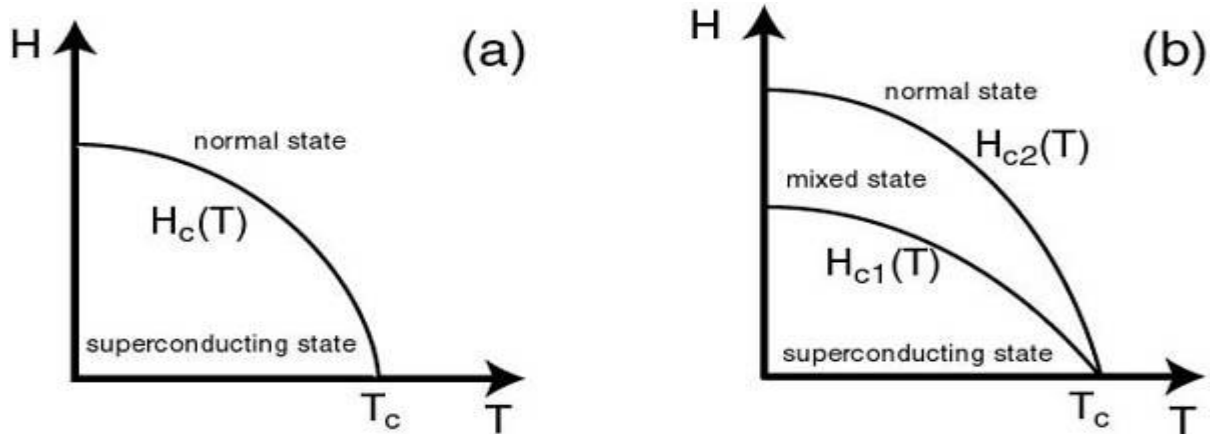
Fig.1: Magnetic phase diagram (H (T)) for a) Type–I superconductors: one critical field Hc exists; b) Type-II superconductors: where two critical fields exist (Lower critical field (Hc1) & upper critical field (Hc2))

 Some other p**roperties of superconductors:**

Most of the physical properties of superconductors vary from material to material, such as the heat capacity and the critical temperature, critical field, and critical current density at which superconductivity is destroyed. On the other hand, there is a class of properties that are independent of the underlying material. For instance, all superconductors have *exactly* zero resistivity to low applied currents when there is no magnetic field present or if the applied field does not exceed a critical value. The existence of these "universal" properties implies that superconductivity is a thermodynamic phase, and thus possesses certain distinguishing properties which are largely independent of microscopic details.

1. Zero electrical resistance- Some materials are placed in series with a current source *I* and measure the resulting voltage *V* across the sample. The resistance of the sample is given by Ohm's law as *R=V/I*. If the voltage is zero, this means that the resistance is zero and that the sample is in the superconducting state. Superconductors are also able to maintain a current

with no applied voltage whatsoever, a property exploited in superconducting electromagnets such as those found in MRI (magnetic resonance imaging).

2. Superconductors obey Meissners effect- A superconductor with little or no magnetic field within it is said to be in the Meissner state. Meissner and ochsenfeld measured the flux distribution outside tin and lead specimens which has been cooled below their transition temperature while in a magnetic field. They found their transition temperatures the specimens spontaneously became perfectly diamagnetic. Superconductor never has a flux density even when in applied magnetic field (B=0).

 3. Effect of magnetic field – The superconducting state of a metal exist only in a particular range of temperature and field strength. Superconducting state will appear in the metal is that some combination of temperature and field strength should be less then critical value. It will disappear if the temperature of the specimen is raised above its Tc or sufficiently strong magnetic field .is employed.

$$H_c = H_o \left[1 - (T/Tc)^2\right]$$

Hc is the maximum critical field strength at the temperature T. Ho is the maximum critical field strength occurring at absolute zero and Tc is the critical temperature the highest temperature of superconductivity.

4. Effect of current -The magnetic field that causes a superconductor to become normal from a superconducting state is not necessarily by an external applied field, It may arise as a result of electric current flow in the conductor. the minimum current that can be passed in a simple without disturbing its superconductivity, is called critical current I_c. if a wire of radius r of a type 1 superconductor carries a current I, there is a surface magnetic field, $H_i = I/2\pi r$, associated with the current. If H_i exceeds Hc, the materials go normal. If in addition a

transverse magnetic field H is applied to the wire, the condition for the transition to the normal state at the surface is that the sum of the applied field and the field due to the current should be equal to the critical field $2\pi r$.

$$H_c = H_i + 2H$$

This is called Silsbee's rule. The critical current I_c will decrease linearly with increase of applied field until it reaches zero at $H = H_c/2$. If the applied field is zero. $I_c = 2\pi r H_c$.

5. Isotope effect-The critical temperature of superconductors varies with isotopic mass. The observation was first made by Maxwell and others, who used mercury isotope s. To give an idea of magnitude of the effect, for mercury Tc varies from 4.185 K to 4.146 K as the isotope mass M varies from 199.5 to 203.4. The isotope mass can enter in the process of the formation of the superconducting phases of the electron states only through the electron - phonons interaction.

6. Penetration depth-F. London and H.London described the meissner effect and zero resistivity by adding the two condition E= 0 (from the absence of resistivity) and B= 0 (from meissners effect) to Maxwell electromagnetic equation. According to them the applied field does not suddenly drop to zero at the surface of the superconductor, but decays exponentially according to the equation,

$$H = H_0 \exp(-x/\lambda)$$

Where H_0 is the value of magnetic field at the surface and λ is a characteristics length known as the penetration depth; λ is the distance for H to fall from Ho to Ho/e.

Superconductors are of two types:

(1)   Type I

(2)   Type II superconductors

Whether the Ginzburg Landau parameter k is is smaller or larger than $1/\sqrt{\tfrac{1}{2}}$. The Ginzburg Landau parameter is defined as:

$$k = \lambda/\xi$$

Where λ describes the penetration depth of the magnetic field inside the superconductor and ξ is the characteristic length over which the Cooper pair density increases from to its maximum value n. For a type I superconductor $k < \sqrt{\tfrac{1}{2}}$ whereas for type II superconductor $k > \sqrt{\tfrac{1}{2}}$.

1.1.1 Type I superconductors

Type I superconductors possesses only one critical magnetic field $H_c(T)$, below which the superconductor produces shielding currents that flow on the surface of the material expelling the magnetic field from its interior. In this condition the superconductor is in the Meissner state. Above H_c, the applied magnetic field penetrates completely into the interior of the material, disrupting the superconductivity.

1.1.2 Type II superconductors

Type II superconductors are characterised by two critical fields: a lower critical field, H_{c1}, and an upper critical field, H_{c1} .Figure shows the magnetic phase diagram for a type II superconductor. Below H_{c1} the superconductor behaves likes a type I superconductor and it is the Meissner state, Meissner currents flowing at the surface disabling the magnetic field to penetrate into the sample. As the magnetic field is increased above H_{c1} (T) but still below the H_{c2}, the magnetic field penetrates the superconductor in the form of tiny quantized microscopic filaments called vortex. A vortex consists of a normal core where the cooper pair

density is zero and this core is surrounded by a superconducting region in which flows a persistent supercurrent.Further increasing the magnetic field, the number of vortices inside the sample increases and the upper critical field H_{c2}, the cores of the neighbouring vortex overlap and the sample goes to the normal state. Between H_{c1} and H_{c2} the superconductor is said to be in the mixed state.
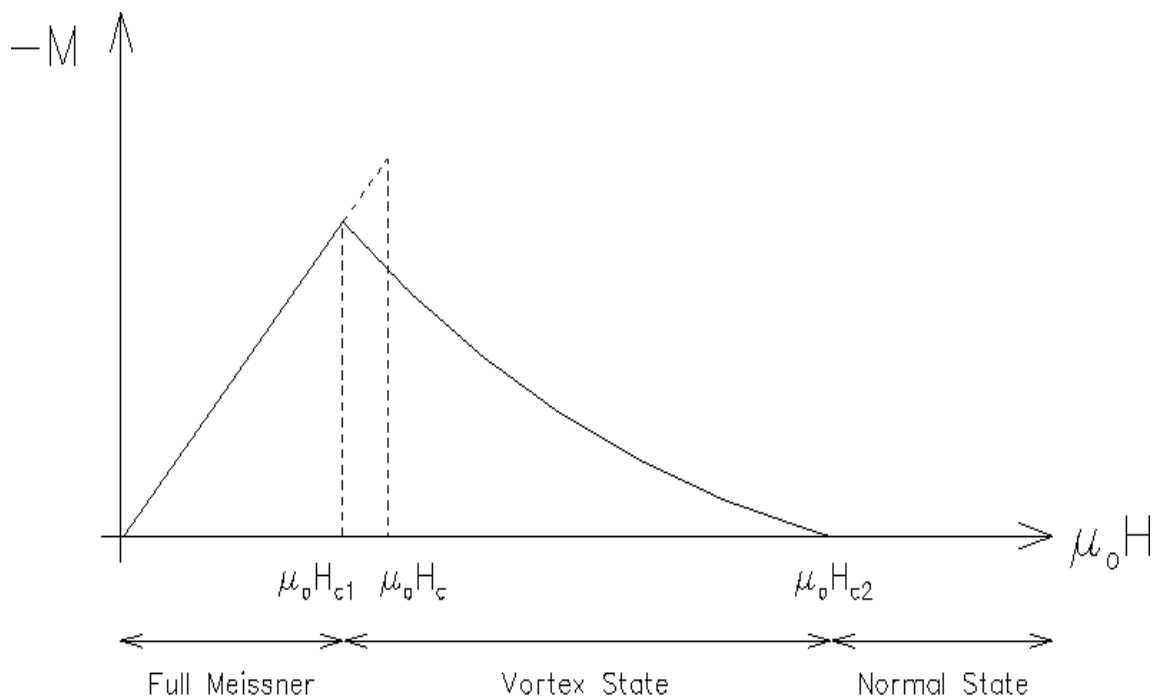

Figure 2. M – H graph showing the different state of a type-II superconductor

1.1.3 Critical current density

YBCO and Ag composite sample has been reported to show enhancement of critical current density. Addition of nanosized non-superconducting material into the matrix of YBCO brings about enhancement of critical current density (Jc) by flux pinning mechanism.

Ag nanoparticles embedded in the YBCO matrix induce flux pinning in the composites. It has been reported [1] that Ag mainly diffuses to the grain boundaries (GBs) of YBCO, and the Jc is improved significantly. The results indicate that Ag has played a role to repair the GBs in HTSs [2]. In addition, YBCO-Ag composites showed higher values of strength and fracture toughness when Ag was added [3]. In addition to Ag composite to thin film some published has been done in thick film of YBCO + Ag composites [4] where microstructural studies have been made. Efforts are ongoing to reduce the silver content and increase the Jc of present-generation BSCCO wire, but most effort is being expended to bring online the second-generation of coated conductors [5].

1.2 HIGH-T$_C$ SUPERCONDUCTOR: YBa$_2$Cu$_3$O$_{7-\delta}$ Material

Yttrium barium copper oxide, often abbreviated as YBCO, is a crystalline chemical compound with the formula YBa$_2$Cu$_3$O$_7$. This material, a famous "high-temperature superconductor", achieved prominence because it was the first material to achieve superconductivity above the boiling point of nitrogen. YBCO was the first material to become superconducting above 77 K, the boiling point of nitrogen. All materials developed before 1986 became superconducting only at temperatures near the boiling points of liquid helium or liquid hydrogen ($T_b = 20.28$ K) - the highest being Nb$_3$Ge at 23 K. The significance of the discovery of YBCO is the much lower cost of the refrigerant used to cool the material to below the critical temperature.
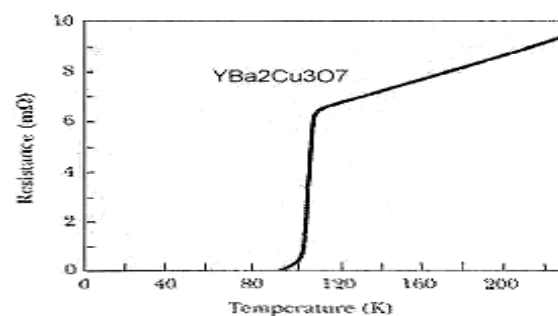

Figure 3. Temperature dependent resistance of YBCO Superconductor

1.2.1 Crystallographic aspects:

The structure of a high-T_c superconductor is closely related to perovskite structure, and the structure of these compounds has been described as a distorted, oxygen deficient multi-layered perovskite structure. One of the properties of the crystal structure of oxide superconductors is an alternating multi-layer of CuO_2 planes with superconductivity taking place between these layers. The more layers of CuO_2 the higher T_c. This structure causes a large anisotropy in normal conducting and superconducting properties, since electrical currents are carried by holes induced in the oxygen sites of the CuO_2 sheets.

Yttrium barium copper oxide, often abbreviated YBCO, is a crystallinechemical compound with the formula $YBa_2Cu_3O_7$. This material, a famous "high-temperature superconductor", achieved prominence because it was the first material to achieve superconductivity above the boiling point of nitrogen (77 K).YBCO crystallizes in a defect perovskite structure consisting of layers. The boundary of each layer is defined by planes of square planar CuO_4 units sharing 4 vertices (figure 2). The planes can sometimes be slightly puckered perpendicular to these CuO_2 planes are CuO_4 ribbons sharing 2 vertices. The yttrium atoms are found between the CuO_2 planes, while the barium atoms are found between the CuO_4 ribbons and the CuO_2 planes. This structural feature is illustrated in the figure below.

Although $YBa_2Cu_3O_7$ is a well-defined chemical compound with a specific structure and stoichiometry, materials with less than seven oxygen atoms per formula unit are non-stoichiometric compounds. The structure of these materials depends on the oxygen content. This non-stoichiometry is denoted by the $YBa_2Cu_3O_{7-x}$ in the chemical formula. When $x = 1$, the O (1) sites in the Cu (1) layer are vacant and the structure is tetragonal. The tetragonal form of YBCO is insulating and does not super conduct. Increasing the oxygen content

slightly causes more of the O (1) sites to become occupied. For $x < 0.65$, Cu-O chains along the b-axis of the crystal are formed. Elongation of the b-axis changes the structure to orthorhombic, with lattice parameters of $a = 3.82$, $b = 3.89$, and $c = 11.68$ Å. Optimum superconducting properties occur when $x \sim 0.07$ and all of the O (1) sites are occupied with few vacancies.

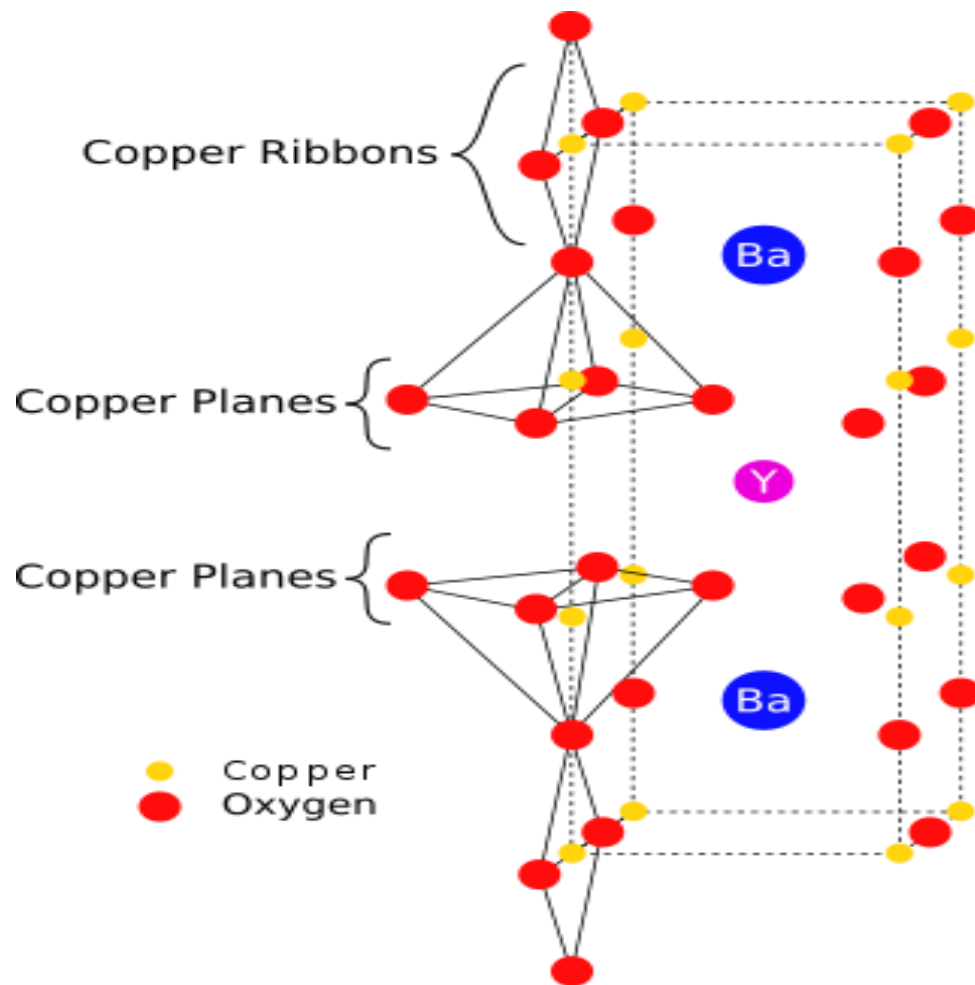**Figure 4. Structure of YBCO in tetragonal phase**

1.2.2 Anisotropy of YBCO materials:

Another important aspect of $YBa_2Cu_3O_{7-\delta}$ material is that, like the other cuprates has an isotropic behaviour, as a consequence of its crystalline structure that is reflected in the directional dependence of λ, ξ & H_{c2}. The anisotropy of these parameters is remarkable between the c-direction & the a or b direction, while the anisotropy between the a & b direction is small & can be neglected in most cases. The anisotropy of HTS materials can be described using Ginzburg- Landau theory that introduces a different effective mass of the hole carriers in different directions. The effective mass in the ab-plane is denoted m_{ab} & along c- axis m_c. The anisotropy is described by the parameter γ, defined as $\gamma = (m_c/m_{ab})^{1/2} = H_{c2}^{ab}/H_{c2}^{c}$ with $\gamma > 1$. The value for YBCO lies between 5 & 8. The large value of Ti & Bi compounds indicate a high anisotropy. As, YBCO has a relatively small γ value which means that it is less anisotropic.

The anisotropy of high-TC superconductors is related to their layered structure & long separation between the CuO2 planes when compared to ξ_c. YBCO is the less anisotropic high-T_c superconductor because on one side the distances between CuO2 planes is only $d \approx 8$ Å & on the other side the CuO chain have metallic conductivity & the superconductivity is induced by proximity effect. In contrast Bi, Ti compounds are more anisotropic because the change reservoir blocks are insulators & the distance is much larger than the respective value of $2\xi_c$. The anisotropy of the effective mass γ in these compounds drive, as a consequence, to an anisotropy in the critical current density, i.e. $j_c^{ab} \gg j_c^{c}$.

Chapter II

2. Experimental Procedure:

2.1 Sample preparation:

The bulk YBCO sample was prepared from the stoichiometric amounts of cationic ratio of Y: Ba: Cu=1:2:3 by the chemical route method. The mixture was stirred in 2-methylethanol and allowed for well mixing for about 12 hours. The solution was then dried and evaporated at 70 ^{0}C to 80 ^{0}C until the mixed powder was obtained. This powder was then calcined at 900 ^{0}C for 6 hours. This calcined YBCO powder was taken and mixed with the varying concentration of Ag_2O (i.e 1, 5 and 10 wt.%). This mixture of YBCO and Ag (varying different weights) was grinded for about 2 hours.bA series of polycrystalline samples of YBCO + Ag (of different concentration) were then pressed into pellets for final sintering at 920 ^{0}C and then cooled to 500 ^{0}C for 10 hours in an oxygen atmosphere for annealing. The temperature dependent resistance ρ (T) was measured using standard four-probe technique with a nanovoltmeter (Keithley-2182 A), Current source (Keithley 6221) and Temperature controller (Lakeshore 332). The surface morphology of the pellets was investigated by scanning electron microscope (Model No. JSM-6480 LV, Make JEOL).

2.2 Characterization:

(1) R-T Measurement by Four probe method

The determination of the resistance as the ratio of the voltage signal (measured with a nanovoltmeter) and the bias current (measured as voltage signal on a known shunt resistance in series with the sample). Therefore, the four-contact configuration is fundamental because it enables to minimize the contribution of spurious voltage signals due to current contacts. The error due to the voltage contact contribute is also minimized because the input

impedance of the nanovoltmeter is 1GΩ or higher, so the current flowing in voltage circuit between the sample and the nanovoltmeter is at least 10^{-9} times lower than the bias current. In a scheme of the current/voltage probes on the sample is reported. The voltage drop is measured between the two probes labelled by means of a digital voltmeter. Since, as mentioned above, the potential drop across the contact resistance associated with probes is minimized, only the resistance associated with the superconductor between probes is measured.

(2) Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM) is a type of electron microscope that images the sample surface by scanning it with a high-energy beam of electrons in a raster scan pattern. The electrons interact with the atoms that make up the sample producing signals that contain information about the sample's surface topography, composition and other properties such as electrical conductivity. The types of signals produced by an SEM include secondary electrons (Secondary electrons are electrons generated as ionization products. They are called 'secondary' because they are generated by other radiation (the *primary* radiation). This radiation can be in the form of ions, electrons, or photons with sufficiently high energy), back-scattered electrons (BSE), characteristic X-rays, light (cathodo luminescence), specimen current and transmitted electrons. Secondary electron detectors are common in all SEM. The signals result from interactions of the electron beam with atoms at or near the surface of the sample. In the most common or standard detection mode, secondary electron imaging or SEI, the SEM can produce very high-resolution images of a sample surface, revealing details about less than 1 to 5 nm in size. Due to the very narrow electron beam, SEM micrographs have a large depth of field yielding a characteristic three-dimensional appearance useful for understanding the surface structure of a sample. Back-scattered electrons (BSE) are beam

electrons that are reflected from the sample by elastic scattering. BSE are often used in analytical SEM along with the spectra made from the characteristic X-rays. Because the intensity of the BSE signal is strongly related to the atomic number (Z) of the specimen, BSE images can provide information about the distribution of different elements in the sample.

Chapter III

3. Results and Discussions

3.1 DC Electrical Resistivity

Figure 5 shows the temperature dependent resistivity for composite samples. It is observed that T_c is affected due to Ag_2O addition. The plot exhibits two different regimes. The one is corresponding to the normal state that shows a metallic behavior (above $2T_c$). The normal resistivity is found to be linear from room temperature to a certain temperature the other is the region characterized by the contribution of Cooper pairs fluctuation to the conductivity above Tc. This is mainly due to the increasing rate of pair formation on decreasing the temperature. From the ρ-T plot it is observed that the metallicity of the composites increases upto studied samples.

Figure 6 shows the temperature derivative of resistivity for composite samples. Differerent critical temperatures are observed from the different composites through the incorporation of nano-particles of Ag as listed in table below. The temperature derivative of resistivity curves shows a decreasing trend with increasing wt. % of Ag. The peak broadening is occurring due to addition of Ag which is affecting the intergranular weak link between grains.
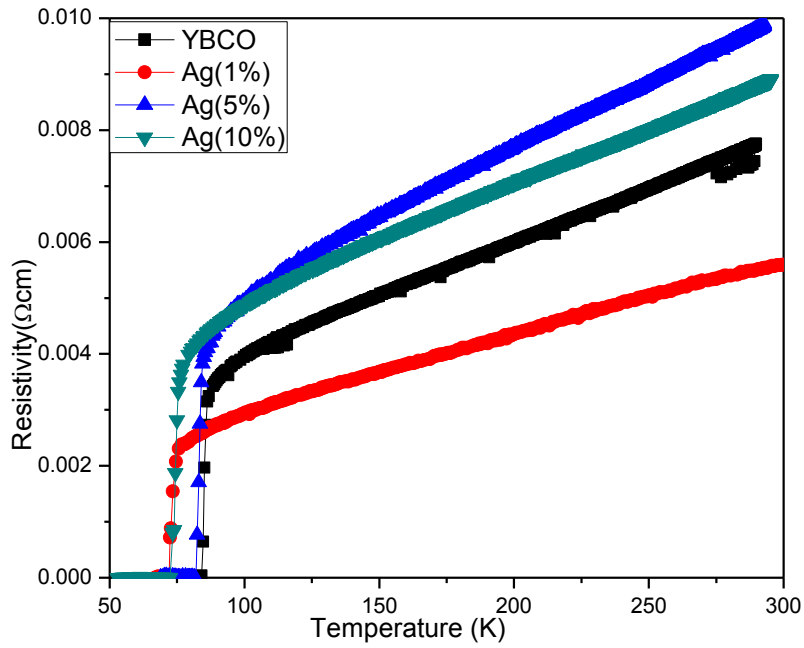
Figure 5: Temperature dependence of the resistivity for YBCO + xAg composites
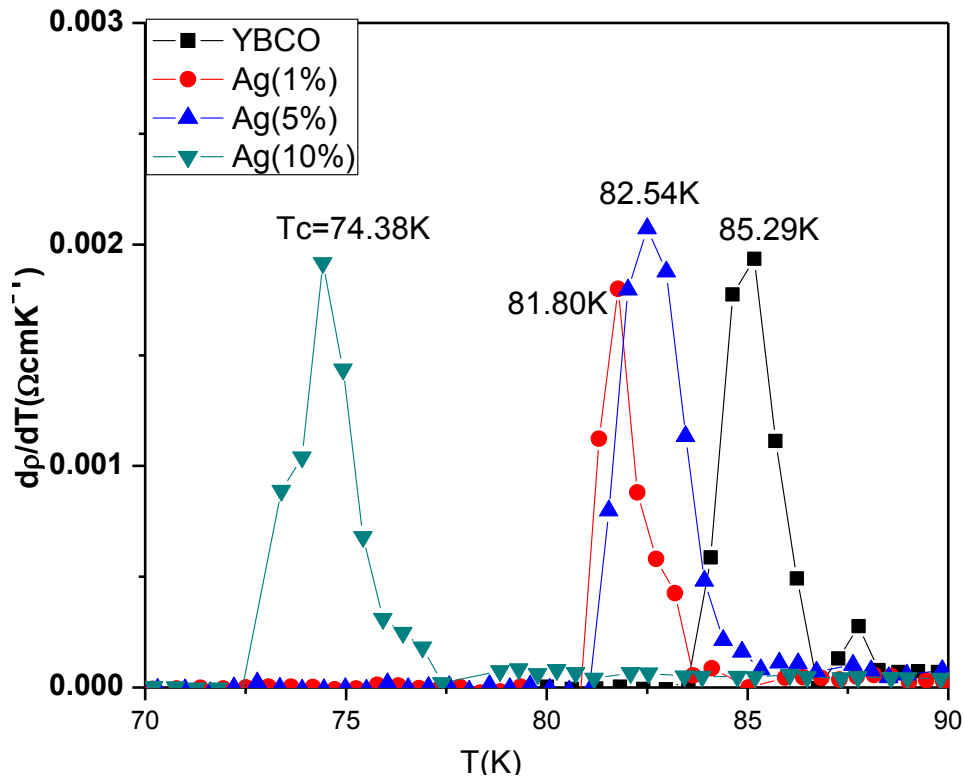
(x = 0, 1, 5 and 10 wt. %).

Fig 6: **Temperature derivative of resistivity of YBCO + xAg composites (x =0, 1, 5&10 wt. %).**
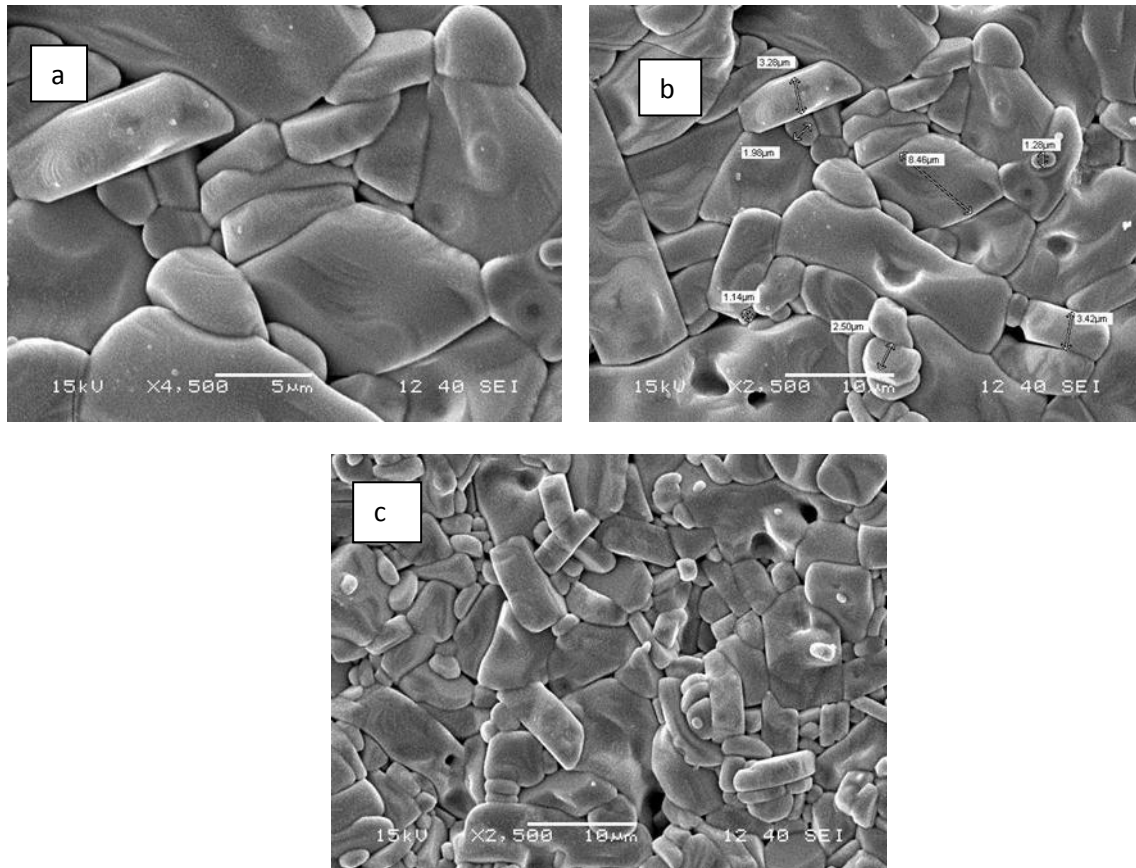
3.2 Grain morphology:

Figure 7 shows the morphology of YBCO/Ag composites.

The microstructural characterisation i.e., grain size distribution of the composites as shown in Fig. 7. The images display well developed grains of varying sizes oriented in all direction. Significant feature observed is the reduction of grain size after addition of Ag. Interlink between the grains is improved which is evident from fig 7(c).Grains become smaller with the addition of Ag.

Chapter IV

4. Conclusion:

From the R-T plot it is observed that T_c value decreases with the increase in Ag weight percentage in the composites which affects the transport property. And the grain size is also decreasing with the addition of Ag. As Ag goes to the grain boundaries the metallicity of the composies increases. The studies can be extended for measurement of critical current density. Though 5 wt.% composite shows slightly higher normal state resistivty still shows better superconducting properties as compared to others.

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