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High T_c films on silicon and sapphire substrates

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Abstract: High T_c thin films of $YBa_2Cu_3O_7$ were deposited on single crystal substrates of silicon and sapphire using both the multilayer and bulk deposition techniques. Silicon substrate showed promising results with an onset transition temperature of 80K.

Key words: high T_c thin films; $Yba_2Cu_3O_7$; single crystal substrate, deposition techniques
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1 Introduction:

Since the first report in 1986 on the superconductivity in ceramic oxides of the Ba-La-Cu-O system, with transition temperature in the 30 ~ 40 K range, a large family of copper oxide based superconductors with T_c between 90 and 140 K have been reported[1-10]. The use of now accessible T_c's in the development of high-speed microelectronics, however, requires preparation of these materials in thick- and thin-film forms of specified samples.

The thin film synthesis got an impulse after the discovery of high transition rare earth oxide superconductors in 1986. It was soon established that very high critical current densities can be obtained in thin films at the liquid nitrogen temperature. Naturally the attainment of high current densities depends strongly on the growth quality of the films. The polycrystalline films did not exhibit high T_c, due to the granular nature of the films. In such cases the high T_c grains are surrounded by deteriorated oxygen deficient materials. Thus all attempts were concentrated on the fabrication of highly oriented single crystalline films. Resistivity measurements for single crystal have confirmed that Cooper pairs take part in conduction along the ab-plane. This suggests that the c-axis oriented films are crucial for practical applications. The deposition parameters and the choice of the substrate material are the two most important features for the growth of high quality superconducting films, beside the substrate temperature during film deposition. The main interest in high transition superconducting films is to prepare the perovskite structure like films, as the new oxides exhibit this structure.

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There are numerous applications of superconducting thin films. Prospects for practical electronic applications of superconductivity have driven the development of high T_c superconductor thin film technology at an unprecedented pace. The applications of superconducting thin films range from those requiring only single layer of superconductor (e. g. simple interconnect, infrared sensors and many passive microwave devices) to those based on complex multilayer circuits processes (e. g. sophisticated interconnect applications; superconducting quantum interference devices, magnetometers and Josephson integrated circuits).

The high T_c materials are frequently denoted by the elemental initials and the number of cations in the chemical formula, e. g. $YBa_2Cu_3O_7$ is referred as 123 phase of YBCO, $Bi_2Sr_2Ca_2Cu_3O_{10}$ is referred as 2223 phase of BSCCO and $Tl_2Ba_2Ca_2Cu_3O_{10}$ is denoted by TBCCO. Nearly all the high T_c superconductors have been based on copper and oxygen. The first compound found to be superconducting above 77K and the most widely studied material is $YBa_2Cu_3O_7$ (YBCO). Researchers have deposited YBCO thin films using every available means of physical vapor deposition as well as a variety of chemical vapor-phase and liquid-phase techniques. The properties of YBCO films are slowly converging toward what may be their practical limits. High-quality films are nearly single crystal in character as a result of epitaxial growth on lattice-matched substrates. Such films have critical temperatures as high as 92K and critical current densities of $5x10^6$ amps/cm^2 at 77K and $2-4x10^7$ amps/cm^2 at 4K. These critical currents exceed those obtained for single crystal specimens, perhaps because of the presence of point defects in the films, which enhance flux pinning. YBCO is not the only oxide superconductor being developed in thin film form. Cuprates based on bismuth and thallium exhibit superconductivity above 100K. However, the bismuth and thallium cuprates have even more elements and a greater number of secondary phases than YBCO. As a result it is very difficult to produce films with nearly ideal properties[11].

A number of deposition techniques have been used for the preparation of thin films. Some popular techniques are laser ablation[12,13], sputtering[14,15], electron-beam evaporation[16], thermal evaporation[17], etc. The technique chosen should be one which is both reliable and reproducible and allows the fabrication of smooth, highly oriented single crystal films. Since our laboratory had experience in electron-beam and thermal evaporation these two techniques were selected for the work reported in this paper.

The structure of the deposited film is the same as that of the substrate. Hence, generally the substrate chosen are single crystals which are normally expensive and not suitable for various applications[18]. Wide scale commercial application of thin-film technology, however, requires the use of relatively economical substrates of desired dimensions.

Thin films of 123 and 2223 compounds were also prepared on metal coated glass slides. Films of silver, gold and copper of thickness 100 were evaporated on strips cut from ordinary microscope glass slides. We deposited superconducting thin films of YBaCuO compound on single crystal substrates of silicon and sapphire. Not only are these substrates relatively cheap but they are also easily available. Silicon also has a big market in the electronic industry.

Thin films of YBCO were deposited on single crystal substrates of silicon and sapphire and the electrical properties of the films were studied. We used both the multilayer and bulk deposition techniques. The experimental techniques are described in Section (II). Results and discussions are dealt in Section (III). We finally conclude our work in Section (IV).

2 Experimental techniques

High T_c superconducting thin films of 123 compound were deposited on precleaned substrates at pressure better than 10^{-5} mbars. Edwards E306A unit was used for deposition of films by thermal evaporation and electron-beam bombardment technique. Substrates were mounted 20 cms above the source. The substrates could be heated to high temperatures, without breaking the vacuum. The film thickness could be controlled and measured with the FTM4 quartz crystal monitor.

Stacks of multilayers of Y, Ba, Cu films of appropriate thicknesses were "insitu" deposited on top of each other from either three different filaments (thermal deposition) or three different cermats (electron-beam evaporation). Films were also deposited from the prepared YBaCuO compound which exhibited superconducting behavior in bulk.

The films were "insitu" annealed at different substrate temperatures, in the presence of oxygen. Some films were also annealed in a temperature controlled furnace. Temperature was raised in steps of 50℃ up to a maximum of 500℃. At each step the temperature was maintained long enough to attain equilibrium between the film, the substrate and the furnace temperature. This precaution was taken to avoid any risk of cracking or sliding off of the film due to different coefficients of thermal expansion of the film and the substrate.

For the resistivity measurements of the fabricated films pressure contacts were avoided. The substrates used were fragile and pressure contacts can destroy the films. Condensation is another problem which is encountered during the cooling process of the films. We modified the sample holder accordingly and the collinear four-probe technique, was employed. This technique requires a precise voltmeter and a constant current supply. Contacts on the films were made with silver paint (P700 JM) using fine copper wires (0.07 mm in diameter). A four-pin IC stand served to bridge the contacts between the film and a sample holder. The film sample was then placed in a specially designed liquid nitrogen cryostat.

A microprocessor based data acquisition system, capable of recording a minimum signal of 2 micro volts, designed and constructed in the lab, was used for the measurement of change of film resistance corresponding to the change in temperature. A software converted the electromotive force (emf) generated across the thermocouple into temperature. Resistivity of the film could be computed from the emf generated across the four-probes if the dimensions of the sample were entered in the computer code.

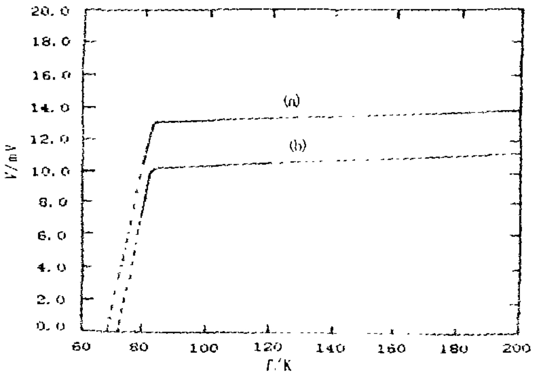
3 Results and discussions:

High T_c superconducting thin films were deposited on silicon and sapphire substrates using the thermal deposition unit. During film deposition the substrates were maintained at room temperature. The pressure was better than 10^{-5} mbars. Stacks of multilayers of Y, Ba, and Cu were deposited in the proper stoichiometric ratios to give a composite thickness of ~ 1018. The results of electrical measurements on as grown films (Figure 1) indicate an onset at 82 and 84 K for silicon and sapphire substrates, respectively. In this figure we plot the emf generated across the thermocouple (mV) as a function of the substrate temperature (K).

A set of so deposited films were annealed in vacuum chamber at 100℃ for at least one hour and the second was annealed in a furnace in the presence of air. The results are shown in Figures 2 and 3, for sapphire and silicon substrates, respectively. Although the film resistance showed a decrease, but the onset remained around 82 K and 84 K as in the previous case (Figure 1).

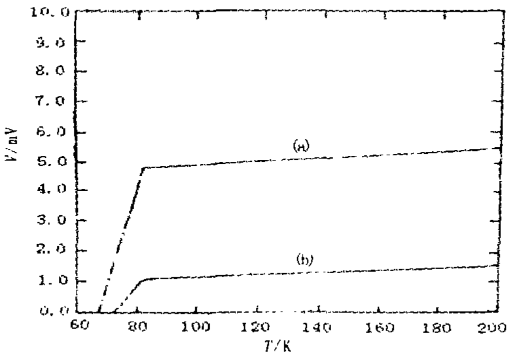
Next thin films were deposited on substrates maintained at 100℃ during deposition. Films were furnace annealed in the presence of oxygen for more than 2 hours at 150℃. The offset temperature was increased but there was no appreciable change in the onset temperature. Further annealing at 150℃ for 5 hours decreased the film resistance and improved the offset (Figure 4).

Films were also deposited on silicon substrates using the electron-beam evaporation technique. The ele-



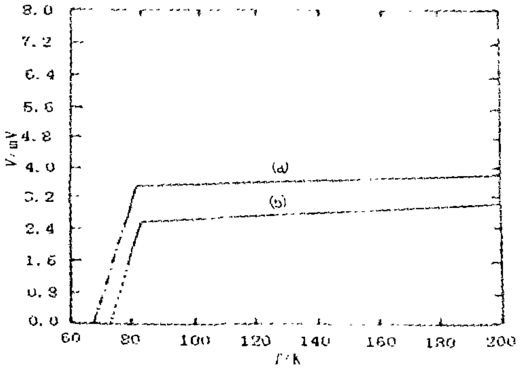
(a) silicon, and, (b) sapphire substrates. The films were deposited at room temperature.

Figure 1 Voltage-temperature characteristics of thermally deposited multilayer films of YBaCuO of average thickness 1018Å



(a) insitu annealing at 1000 C for one hour, and, (b) insitu and furnace annealing at 100^0 C for one hour each

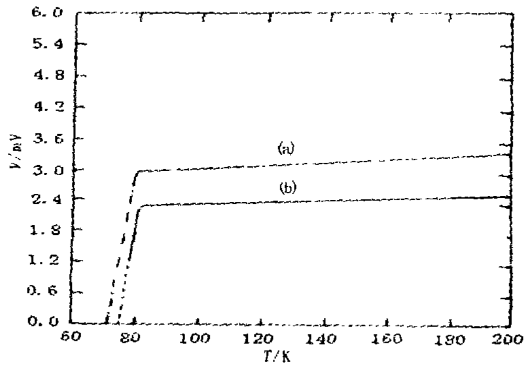
Figure 2 Voltage-temperature characteristics of thermally deposited multilayer films of YBaCuO on sapphire substrates, maintained at room temperature, of average thickness 840Å



(a) insitu annealing at 100^0 C for one hour, and, (b) insitu and furnace annealing at 1000 C for one hour each.

Figure 3 Voltage-temperature characteristics of thermally deposited multilayer films of YBaCuO on silicon substrates, maintained at room temperature, of average thickness 840Å

ments Y, Ba, and Cu were placed in three different cermats of the electron-beam coating unit. They were then deposited in stoichiometric ratio of 1:2:3; a thin film of one of the elements was deposited on the substrate which followed the insitu deposition of the other two elements in the sequence of the compound. This procedure was repeated a number of times to get a final composite thickness of ~ 1000. The pressure was maintained at ~ 10^{-5} mbars. The electrical measurements show a clear onset at 80 K (Figure 5) and an extrapolated offset at ~ 76.4 K. Figure 5 and Figure 6 depict the resistivity-temperature characteristics where the resistivity (mohms-cm) of the film is plotted as a function of substrate temperature (K).



(a) silicon substrate, and, (b) sapphire substrate.

Figure 4 Voltage-temperature characteristics of thermally deposited multilayer films of YBaCuO deposited at 100°C and furnace annealed. The average thickness was 1028Å and the annealing temperature was 150℃ and time 2.5 hours

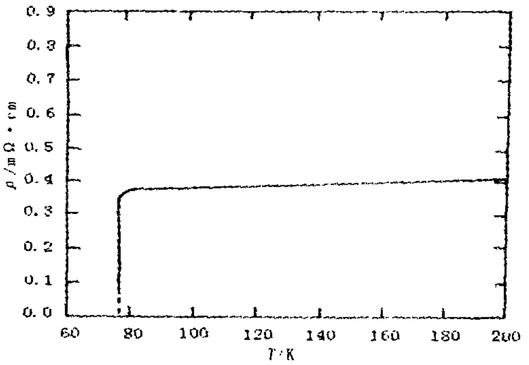


Figure 5 Resistivity-temperature characteristics of multilayer deposition on silicon substrate by electron-beam evaporation technique. The average film thickness was around 1000Å.

Since films deposited on the electron-beam unit were more promising than those on the thermal unit, the next set of films was also prepared on this unit. However, instead of multilayer deposition, bulk evaporation was tried. Prepared 123 compound was palletized with the help of a lab-designed hydraulic press. The pellets were then used as the target for the electron-beam. Films thus prepared showed an onset at 80 K and a much better offset at 77.5 K (Figure 6). The behavior was sharply metallic with a narrow transition width.

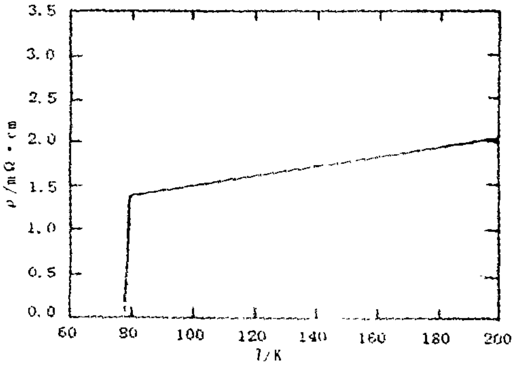


Figure 6 Resistivity-temperature characteristics of 123 deposition on siliconsubstrate by electron-beam evaporation technique.

4 Conclusions

High T_c films were deposited on silicon and sapphire substrates. The films had metallic behavior and the onset temperatures were around 80 K, which is lower than for bulk materials. It was also found that longer annealing did not change the onset temperature appreciably, but the transition width did decrease. The film deposition by electron-beam technique, both by multilayer and bulk deposition techniques gave almost identical results. Films deposited on silicon substrates showed better results. Although the zero resistance could not be attained in any film, but the voltage-temperature characteristics do show an onset clearly, indicating that the offset temperature is below the boiling point of liquid nitrogen (77.3 K). In future we plan to study in detail the effect of annealing temperature on the quality of high T_c superconducting films.

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硅和蓝宝石基底上的高 T_c 薄膜

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摘要：使用多层沉积和大块沉积两种技术，在硅和蓝宝石的单晶基底上沉积了 $Yba_2XCu_3O_7$ 高 T_c 薄膜. 实验显示：在硅基底上得到了转变温度为 80K 的期待结果.

关键词：高 T_c 薄膜；$Yba_2XCu_3O_7$；单晶基底；沉积技术