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Fabrication and microwave properties of double side $Tl(Ba,Sr)_2Ca_2Cu_3O_y$ films on LSAT substrate

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Epitaxial thin films of $Tl(Ba,Sr)_2Ca_2Cu_3O_y$ (Tl-1223) superconductor were grown on both sides of a moderate dielectric constant (ε=23.8) substrate $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (LSAT), towards the study of miniaturization of superconducting thin film microwave filter. The superconducting transition temperatures (T_c) are in the range 105 - 108 K. The critical current density is greater than 1 MA/cm^2 at 77 K. At low temperatures, the values of microwave surface resistance measured on both sides of the film are close to that of the commercially available YBCO thin films. The value of surface resistance (R_s) at 90 K and at 10 GHz is about 1.2 mΩ.

1. INTRODUCTION

Tl-based high-T_c superconductor (HTS) thin films based $Tl_2Ba_2CaCu_2O_y$ (Tl-2212) phase (T_c ~ 110 K) have been used in microwave devices because of their high T_c and low R_s. The later mainly depends on J_c, which in turn is related to microstructure of the films. For high values of J_c, first of all, the film should be epitaxial (in-plane aligned) and free from weak links (i.e. high-angle grain boundaries). Mis-oriented grains such as a-axis oriented and large amount of in-plane rotated are known to affect J_c drastically. Then, a well-connected microstructure without defects that dissipates microwave is required. The defects such as point defects, dislocations, impurities, size disorder and other defects whose size is comparable to the coherence length are believed to contribute to high flux pinning and thus high J_c. The microstructure is strongly related to the film processing conditions. Tl-films are normally made by a two-step process involving the preparation of amorphous film with or without the incorporation of thallium followed by crystallization at high temperatures, in the presence of thallium source. In contrast to in-situ process, this method produces films with varying microstructure depending on the preparative conditions such as temperature, time, thallium, and oxygen partial pressures. Therefore, it is essential to control the preparative conditions to the maximum extend possible to achieve the reproducibility of the films.

We have been working on the development of Tl-1223 films for microwave device applications, because of its high T_c ~ 133.5 K and low anisotropy compared to Tl-2212. In view of miniaturization of HTS thin films microwave filter, we have chosen a well lattice matched and moderate dielectric constant substrate, (100) LSAT. In contrast to $LaAlO_3$ (LAO), LSAT does not have twinning but the dielectric loss tangent seems to be higher than LAO. The loss of LSAT is related to the oxygen content, which has not yet been fully established. In this article, we report the preparation of double sided $Tl(BaSr)_2Ca_2Cu_3O_y$ (Tl-1223) thin films on 1 inch LSAT substrate and the results of the investigation on the surface morphology, T_c, J_c and microwave surface resistance.

2. EXPERIMENTS

Tl-1223 films were prepared by Amorphous Phase Epitaxy (APE) method in which an amorphous phase of composition $Tl(Ba,Sr)_2Ca_2Cu_3O_y$ was deposited on both sides of 1 inch diameter (100) LSAT substrate at room temperature by rf magnetron sputtering.[6] The amorphous films were kept in a circular shaped silver capsule containing a pre-reacted powdered thallium compound, sealed and annealed at temperatures in the range 840 - 860° C for several hours. The in-plane and out-of-plane orientation of films with respect to the substrate was investigated by X-ray diffraction (XRD) measurements. The microstructure was investigated using Scanning Electron Microscope (SEM). The superconducting transition temperature and critical current density was measured by ac-susceptibility method. The microwave surface resistance was

determined by a dielectric resonator technique at 22 GHz.[12]

3. RESULTS AND DISCUSSION

XRD pattern, as shown in Fig. 1(a), obtained by a θ - 2θ scan revealed that the films have a nearly pure Tl-1223 phase with c-axis oriented perpendicular to the plane of the substrate. The secondary phase was identified to be Tl-1212 as shown in the figure by an asterisk. The results of x-ray ϕ scan around (102) reflection are shown in Fig. 1(b). It can be seen that there are four narrow reflections at a regular interval of 90° with almost equal intensity, indicating a good in-plane alignment of Tl-1223 film with the substrate. The full width at half maximum (FWHF) of these reflections is between 0.68° and 0.72°, indicating a high degree of in-plane alignment which is very important to get high J_c and thus a low R_s [4].

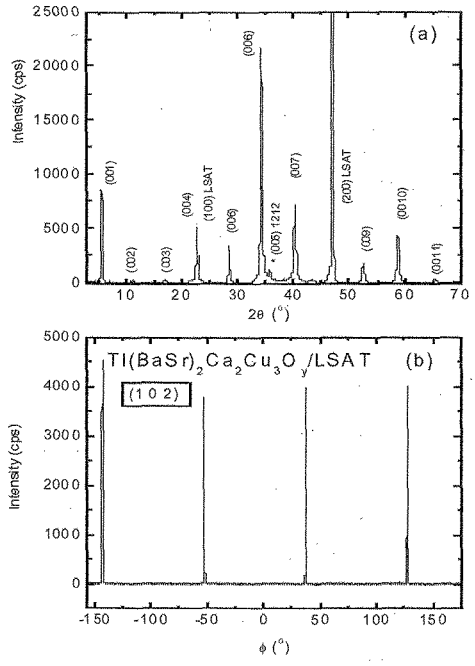

Fig. 1 (a) Out-of-plane (θ-2θ) XRD pattern of Tl-1223 film, showing the c-axis orientation. The reflection at 2θ=35.78° indicates the presence of small amount of Tl-1212 phase. (b) ϕ scan about (102) reflection revealing a good in-plane orientation.

SEM analysis revealed two kinds of surface morphology, as shown in Fig.2a and 2b, depending on the annealing conditions. Fundamentally, both images exhibit a well connected, smooth plate-like morphology with a similar density of pits as seen in other Tl-films [11]. But, the image shown in Fig. 2b exhibits, in addition, a needle-like grains and few particles on the surface. The needles appear when the silver capsule is not sealed completely i.e when the partial pressure of thallium decreases. The thickness of the needle is about 1μm and the maximum length can be 20 μm. Energy Dispersive X-ray (EDX) analysis revealed that both the plate-like surface and the needle have the same chemical composition, $Tl(Ba,Sr)_2Ca_2Cu_3O_y$. The particles were identified to be CuO. The needles seem to be a-axis oriented grains. As discussed below, the presence of such needles results in high R_s values although the value of J_c is comparable to that of the film without any needles.

The superconducting transition temperature and critical current density of Tl-1223 films were determined by ac susceptibility measurements using Physical Property Measuring System (PPMS), Quantum Design, USA. These measurements were made only on films with the size 5 mm × 5 mm because of limitations in our instrument. The superconducting diamagnetic onset was as high as 108 K, irrespective of films with or without needle shaped grains. Figure 3 shows the magnetic field dependence of J_c of a typical film without any needle at various temperatures. The J_c values were determined from the measurements of out-of-phase susceptibility, χ'' in various dc field as a function of the amplitude of ac excitation field, h_{ac} at different ac frequencies, 97, 997, 9997 Hz [12]. At 77 K, the value of J_c at 0.2 T is 0.2 MA/cm^2. We could not determine the value of J_c at zero-field because the maximum in χ'' occurs above the maximum limit of the ac amplitude of 15 Oe in our experiments.

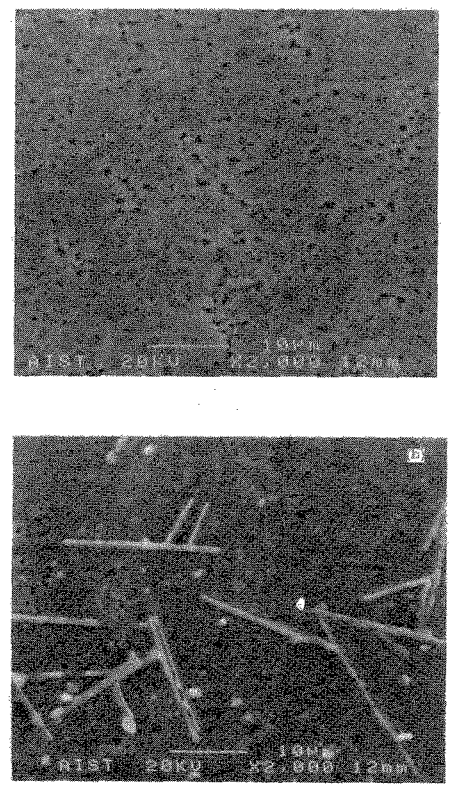

Fig. 2 SEM image of Tl-1223 films; (a) A smooth plate-like morphology with pits and (b) Plate-like morphology with pits and needle shaped grains on the surface.

However, a comparison with the behavior of J_c versus dc field in other HTS films, where the J_c drops about one order in small applied fields < 0.2 T), it is expected that at 77 K and zero field the J_c should be ~ 2 MA/cm^2. With increase of magnetic field J_c decreases much slowly compared to that in Tl-2212 system [13]. This behavior is could be due to lower anisotropy of Tl-1223 system [8].

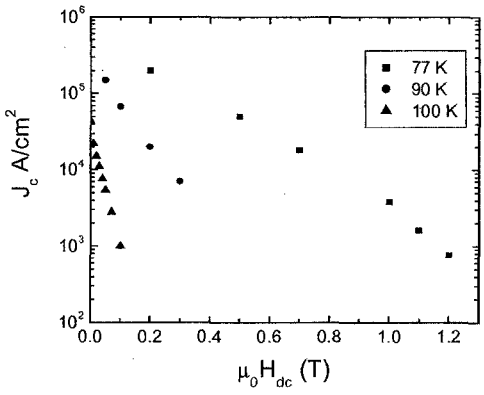

Fig. 3 The J_c versus applied magnetic field at temperatures 77, 90 K, and 100 K. At 77 K, J_c at zero field can be > 1MA/cm^2 at 77 K.

Therefore, for applications under magnetic field, Tl-1223 system should be better than Tl-2212 system. At 90 K, the value of J_c at zero field is ~ 0.8 MA/cm^2. It should be emphasized that the films with needle shaped grains also showed J_c values comparable to those of films without any needles. This confirms that the needles are present only on the surface. If it were present in the bulk one would expect a relatively smaller J_c. The high value of J_c in these films may be due to the chemical disorder at the Ba/Sr site, which are believed to act as pinning centers.

As mentioned earlier, we could not measure T_c and J_c of Tl-1223 films with the size 10 mm × 10 mm, because of the non-availability of experimental facilities to measure large area thin films without any contact. However, we have identified the films with high J_c qualitatively by a simple testing method, fishing HTS films out of liquid nitrogen bath by a permanent magnet (having a field of ~ 0.5 T) due to the effect of high flux pinning and hence high J_c [14]. By this method we could screen low J_c films which are not suitable for R_s measurements. In order to estimate J_c of fished out films quantitatively, the large area films were cut into the size 5 mm × 5 mm after the R_s measurement, and measured by ac susceptibility method as discussed above. The J_c of these films are estimated to be ~ 1 MA/cm^2 at 77 K and zero field.

Fig. 4 shows the temperature dependence of R_s for two Tl-1223 films, one without any needle and the other with needle-like grains on the surface of the film, along with the data of excellent $YBa_2Cu_3O_y$ (YBCO) films commercially available [15]. It is clear from this figure that the R_s of Tl-1223 films without needle is superior to YBCO film particularly at temperatures

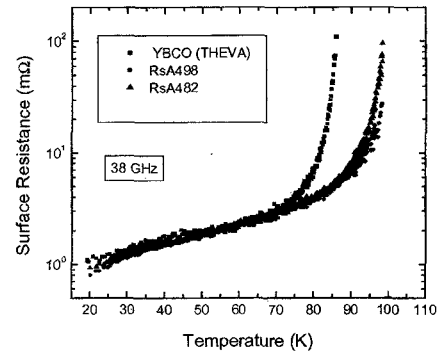

Fig. 4 Temperature dependence of R_s measured on 10 mm × 10 mm Tl-1223 films and in YBCO film at 38 GHz.

above 75 K. On the other hand, the R_s of film with needle-like grains are about one order higher than the former. This may be due to the absorption of microwave by the tail of damped Josephson Plasma caused by the supposedly a-axis oriented needles [16-18].

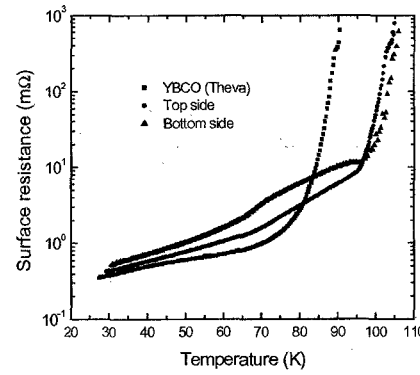

Fig. 5 Temperature dependence of R_s in double sided Tl-1223 films on 1 inch LSAT substrate. For comparison the values of Rs for YBCO is also shown.

The R_s values for the best films at 77 K and 10 GHz are in the range 237 - 245 μΩ according to the f^2 relation. These values are lower than that (317 μΩ) of the YBCO film. It is remarkable that the R_s at 90 K and 10 GHz is about 507 μΩ. Although this value satisfies the requirement for the best performance of many microwave devices, we believe that it should be possible to lower R_s values further by increasing T_c by at least 5 K, as the maximum achievable T_c for Tl-1223 phase is 133 K [6]. A typical example of the temperature dependence of R_s in double sided 1 inch

Tl-1223 films is shown in Fig. 5. It is important to note that at low temperatures the R_s on both side of the films are comparable to YBCO film. It is essential to see the performance of the filter with the use of Tl-1223 films on LSAT substrate to decide suitability of this component as microwave filter and the efforts are under way to design and pattering the films.

4. CONCLUSION

We have prepared high quality epitaxial double-sided Tl-1223 films with a thickness of 5000 Å on LSAT substrate. Two kinds of microstructures were observed, a plate like morphology and needle-like grains on top of the plate like morphology depending on the preparative condition. The latter gives high R_s values compared to the former. The T_c and J_c values of those films are ~108 K and > 1 MA/cm^2 at 77 K, respectively. The R_s of on both side of the Tl-1223 films are comparable to YBCO film at low temperatures suggesting that Tl-1223 film is a good candidate for microwave filter application at temperatures higher than 77 K.

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