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# $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})$ $Cu_2O_z$ 1212 superconducting films on lanthanum aluminate

**H Sudra[1], G Gritzner[1], N Hörhager[2] and H W Weber[2]**

[1] Institute for Chemical Technology of Inorganic Materials, Johannes Kepler University, A-4040 Linz, Austria
[2] Atomic Institute of the Austrian Universities, Stadionallee 2, A-1020 Wien, Austria

E-mail: gerhard.gritzner@jku.at

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**Abstract**
Superconducting films of $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ 1212, approximately 3 $\mu$m in thickness, were fabricated on single-crystalline lanthanum aluminate. The films were practically phase pure and showed excellent in-plane and out-of-plane texture. Critical temperatures of up to 96 K were recorded. Critical current densities of the order of 40 MA $m^{-2}$ at 77 K in self-field were measured both by transport and by magnetic studies. Magnetic measurements were made between 5 and 77 K. Microstructure studies were carried out to learn about the effects influencing the critical current density.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Tl-1223 superconducting films on single-crystalline lanthanum aluminate were reported to display excellent critical currents, even in high external magnetic fields [1, 2]. Very little information, however, is available on the superconducting properties of the Tl-1212 phase. Yet this phase is supposed to show less anisotropy, better magnetic flux pinning and better performance in strong magnetic fields due to the short insulating distance between the superconducting Cu–$O_2$ layers. Following some preliminary reports soon after the discovery of the Tl-based superconducting families [3–5], compositional effects of Sr/Ba and Tl/Pb substitutions in Tl-1212 superconductors were investigated [6]. Detailed studies on the superconducting properties of bulk phase material of rare-earth-doped Tl-1212 superconductors were published recently [7–9].

The fabrication of doped Tl-1212 films employed vacuum techniques. Chromium containing (thallium, bismuth)-based 1212 films were made by pulsed laser deposition [10, 11]. The preparation of $TlBa_2CaCu_2O_7$ films via MOCVD synthesis [12] and the enhancement of superconductivity in $TlBa_2CaCu_2O_x$ thin films prepared by magnetron sputtering and by varying the oxygen content [13] have been reported as well.

In this paper we investigate the possibility of preparing gadolinium-doped (Tl, Pb)-1212 superconducting thick films on single-crystalline lanthanum aluminate by applying a precursor layer followed by vapor phase thallination.

## 2. Experimental details

### 2.1. Preparation of superconducting films

Superconducting films of the composition $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ were prepared on single-crystalline (100) lanthanum aluminate in a two-step procedure. A Tl-free precursor was prepared via the malic acid gel technique following a published procedure [4–6, 14]. The calcined oxidic precursor powders were dispersed in acetone and ground in a ball-mill. After evaporation of the acetone, a suspension of the precursor material in terpineol was applied to single-crystalline lanthanum aluminate by screen printing. Thallination was carried out by vapor transport from unfired sources prepared by compacting precursors mixed with $Tl_2O_3$ to achieve an overall composition of $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$. The substrate with the precursor and the sources were wrapped in silver foil and sintered at temperatures between 880 and 920 °C for 30 min to 30 h. The best results were achieved for a sintering temperature of 895 °C for 2 h. The surface

 

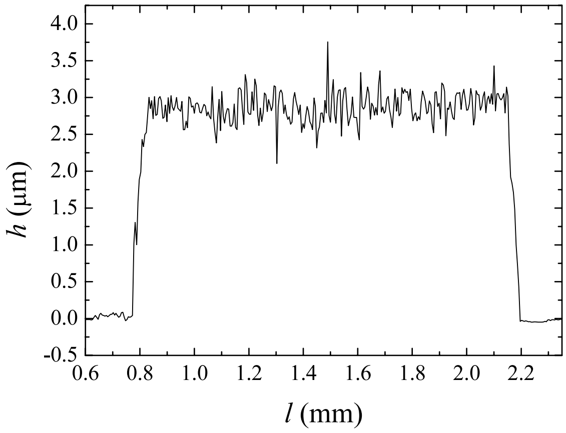


**Figure 1.** Cross section of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C.

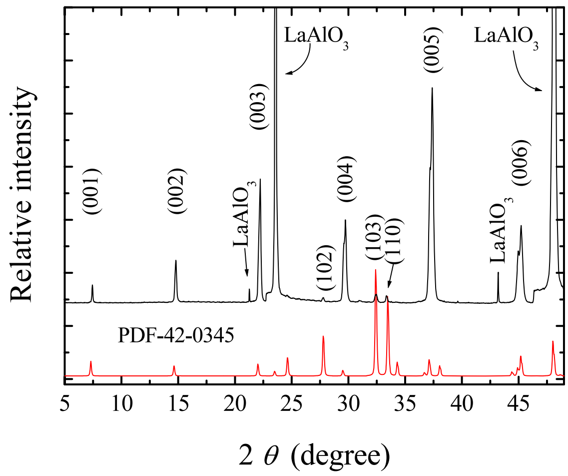


**Figure 2.** X-ray diffraction of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C (Ni-filtered Cu K$\alpha$ radiation).

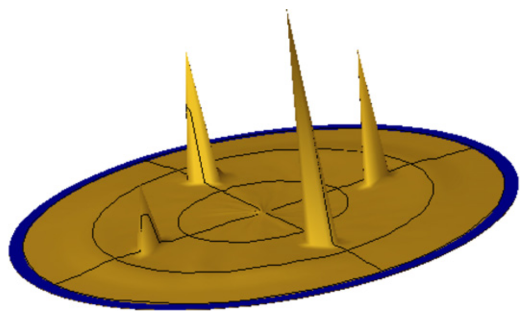

**Figure 3.** Pole figure of the (103) plane of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C ($2\theta$: 32.42°).

of the superconducting film was polished with a 1 $\mu$m $Al_2O_3$ suspension to a thickness of about 3 $\mu$m.

### *2.2. Characterization equipment*

X-ray diffraction (XRD) measurements with Ni-filtered Cu K$\alpha$ were carried out on an X'Pert instrument (Panalytical, Netherlands). Pole figures were derived with an Eulerian cradle. Optical microscopy was carried out on a Zeiss Axioplan in polarized light with an Ernst–Laves compensator (gypsum, first order). Micrographs were obtained on a scanning electron microscope (Zeiss 1540XB Gemini, Germany). Energy-dispersive x-ray fluorescence analysis (EDX) was performed on an Inca system (Oxford Instruments, Great Britain). Critical temperatures were obtained from the temperature dependence of the resistance employing the 1 $\mu$V cm$^{-1}$ criterion with a current of 1 mA and by SQUID studies. Critical current densities at 77 K were determined from dc transport studies. Critical current densities in the temperature range from 5 to 77 K were derived from the magnetic moments obtained from vibrating sample magnetometry. AC magnetic measurements were carried out with an amplitude of 0.3 mT for $H \parallel c$. The thickness of the films was measured with a profilometer (Perthometer C5D, Mahr, Germany).

## 3. Results and discussion

A typical profilometer scan across the film surface is presented in figure 1. It shows considerable roughness of the surface. The films were generally about 3 $\mu$m thick.

X-ray diffraction spectra of the Tl-1212 films showed that the superconducting layer was both phase pure and $c$-axis oriented (figure 2). Figure 2 includes the reference spectrum for $TlSr_2CaCu_2O_7$ [15]. Secondary phases could not be detected in the XRD spectra.

The pole figures as well as the $\varphi$ and $\psi$ scans demonstrate that the superconducting film was biaxially aligned (figures 3 and 4).

The FWHM value for the $\varphi$ scan of the (103) peak ($2\theta$: 32.42°) was 2.2°, and for the $\psi$ scan of the (005) peak ($2\theta$: 37.13°) 3.8°.

Critical temperatures ($T_{c(0)}$) between 92 and 96 K were found from transport measurements (figure 5) with a transition width ($\Delta T_{(90-10\%)}$) of around 2 K. The critical temperature of 96 K is only slightly lower than the highest critical temperature of 101 K observed in the best samples of the bulk material [9].

The magnetic measurements resulted in comparable $T_{(onset)}$ values. A typical result of the magnetic measurements is presented in figure 6.

The critical current densities in self-field obtained from transport measurements at 77 K were around 40 MA m$^{-2}$. Magnetic measurements in the temperature range from 5 to 77 K were taken in a vibrating sample magnetometer, where the magnetic moments were recorded as a function of the applied magnetic field ('field loops'). The results of the critical current densities calculated from the loops using the Bean model are shown in figure 7. Agreement between the critical current densities measured by transport in self-field with the respective data from the magnetic measurements was found.

The surprising, albeit not necessarily satisfying, information obtained in this study is the low critical current density. The critical currents of the thick films are considerably

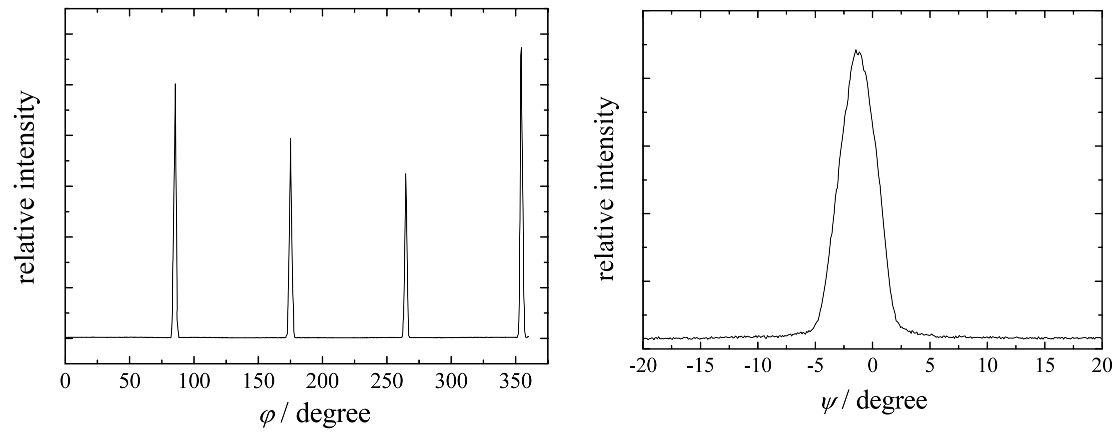


**Figure 4.** $\varphi$ and $\psi$ scans of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C.

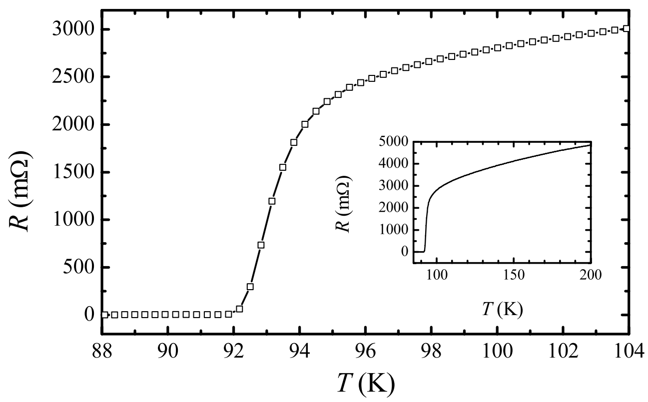


**Figure 5.** Temperature dependence of the resistance of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C.

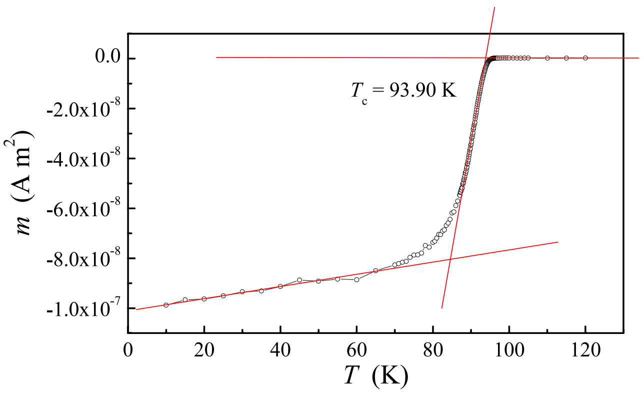


**Figure 6.** Magnetic moment ($m$) of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C.

lower than those achieved for thick Tl-1223 films [1, 2]. For such films critical currents of the order of 12 GA m$^{-2}$ have been published. The critical current densities observed for our thick Tl-1212 films are also much smaller than the critical currents reported for thin Tl-1212 films prepared by vacuum techniques, where values of 10–19 GA m$^{-2}$ have been measured [10, 11, 13]. Yet x-ray diffraction shows very good in-plane and out-of-plane alignment of the film. Detailed optical and electron microscopy offer an explanation for the low critical current densities.

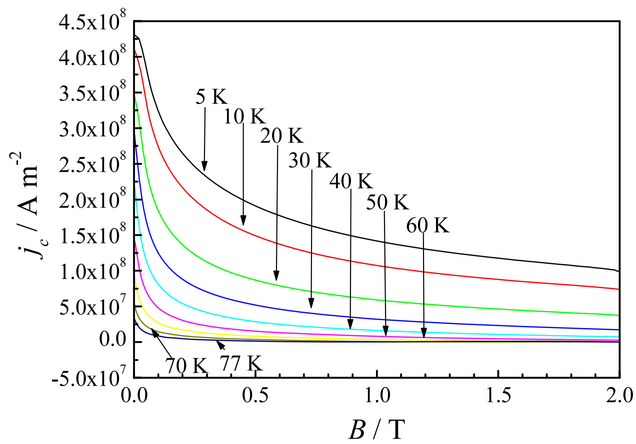


**Figure 7.** Field dependence of critical current densities at different temperatures of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on single-crystalline (100) $LaAlO_3$ sintered for 2 h at 895 °C.

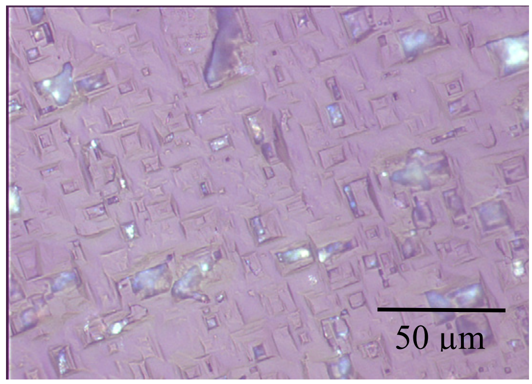


**Figure 8.** Optical micrograph in polarized light with an Ernst–Laves compensator of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ layer very close to the mono-crystalline $LaAlO_3$ sintered at 895 °C for 2 h.

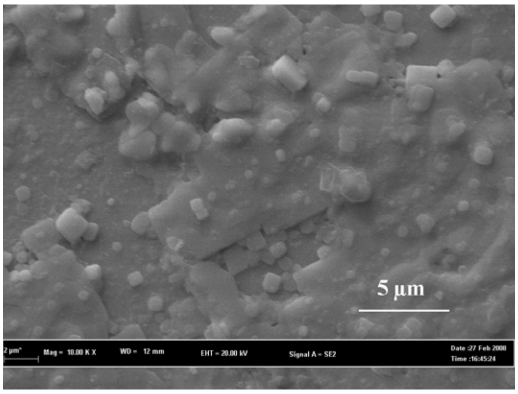


**Figure 9.** Electron micrograph of the surface of a $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on mono-crystalline $LaAlO_3$ sintered at 895 °C for 2 h.

Optical microscopy in polarized light with an Ernst–Laves compensator of a layer very close to the substrate showed excellent alignment and good connection of the superconducting crystallites (figure 8). An electron micrograph of the surface of a 3 $\mu$m thick $(Tl_{0.5}Pb_{0.5})(Sr_{0.95}Ba_{0.05})_2(Ca_{0.8}Gd_{0.2})Cu_2O_z$ film on mono-crystalline $LaAlO_3$ sintered at 895 °C, however, illustrated that crystallites formed further away from the substrate did not show good contacts (figure 9). Thus the low values of the critical current densities can be explained by the very poor interconnectivity of the grains further away from the lanthanum aluminate substrate and, to a lesser degree, by the very weak flux pinning within the grains.

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