Growth of Tl$_2$Ba$_2$CaCu$_2$O$_7$ Thin Films on Curved Substrates for Metamaterial Applications

S C Speller$^1$, T Hao$^2$, C J Stevens$^2$, D J Edwards$^2$ and C R M Grovenor$^1$

$^1$ Department of Materials, University of Oxford, Parks Road, Oxford, OX1 3PH
$^2$ Department of Engineering Science, University of Oxford, Parks Road, Oxford, OX1 3PJ
E-mail: susannah.speller@materials.ox.ac.uk

Abstract. Recent developments in the field of magnetic metamaterials have shown that coupled split-ring resonator (SRR) structures can manipulate magnetic fields on a scale shorter than the wavelength of the electromagnetic radiation [1]. In particular, stacks of copper/dielectric SRR structures can be used as high Q filters or as waveguides in near field imaging applications [2]. However, major improvements in the sensitivity of such devices would be achieved by fabricating the SRR elements from high temperature superconducting (HTS) films on appropriate dielectric substrates. In order to produce a stack of superconducting/dielectric SRR structures, the ideal method involves patterning them into an HTS film grown on the curved surface of a dielectric cylinder. To our knowledge this has not previously been attempted on the small scale required for these applications. The desired feature sizes range from a few millimetres for filter applications down to about 10µm for high resolution imaging applications. We report here our initial attempts at the growth of Tl$_2$Ba$_2$CaCu$_2$O$_7$ (Tl-2212) thin films on the surface of LaAlO$_3$ and MgO single crystal cylinders of diameter 3mm and length 5mm. Scanning Electron Microscopy, Electron Backscatter Diffraction and a novel X-ray Diffraction technique have been used to study the microstructure and analyse the growth morphology of the Tl-2212 films in this unusual geometry. Simulations have been carried out on an axial array of SRR structures that could be fabricated from these HTS/dielectric cylinders.

1. Introduction
Metamaterials are artificial materials consisting of periodic arrays of conducting elements which behave as media with electromagnetic properties that do not exist in nature. The physical size of the individual resonators in a metamaterial (and their periodicity) is much smaller than the wavelength of the electromagnetic radiation they interact with, and hence the material is apparently homogeneous. The effective permittivity $\varepsilon_{\text{eff}}$ and magnetic permeability $\mu_{\text{eff}}$ of the medium can be precisely controlled, and it has even been shown theoretically and experimentally that it is possible to design materials which exhibit $\varepsilon_{\text{eff}} < 0$ and $\mu_{\text{eff}} < 0$ simultaneously [3, 4]. These double-negative materials have a negative refractive index, as originally predicted by Veselago in 1968 [5], and in 2000 Pendry proved that slabs of negative refractive index materials can act as perfect superlenses [6]. This stimulated the rapid development of metamaterials for a wide range of different applications.

High permeability metamaterials capable of guiding magnetic flux have applications in areas such as MRI imaging where RF signals from nuclear resonance need to be guided to a receiving coil. Metamaterials have an advantage over conventional magnetic materials for this application because they are non-magnetic in dc fields and hence do not disturb the homogeneity of the applied constant magnetic field. Various resonant elements have been proposed for controlling the permeability of metamaterials including the split ring resonator [7], the swiss roll (essentially a capacitor rolled on a dielectric core) [7]

© 2008 IOP Publishing Ltd
and the capacitively loaded loop [8]. Wiltshire et al have successfully demonstrated the use of a bundle of swiss rolls as an MRI flux guide to image the researchers thumb held 200mm away from the receiver coil [9], and further improvement of this material and its near field imaging performance have been made by Wiltshire, Edwards et al. [10].

1.1. Superconducting Metamaterials

The resonance strength of a metamaterial could potentially be improved by fabricating the resonant structures from high temperature superconducting (HTS) films grown on dielectric substrates because at frequencies below about 200GHz HTS materials have lower surface resistance (R_s) values than copper [11]. Producing superconducting metamaterials with the geometry required for magnetic flux guidance applications ideally involves patterning a stack of resonator structures (such as split pipes [12]) into an HTS film grown on the outside of a dielectric cylinder or cone. However, the growth of HTS material on curved substrates has not previously been attempted and the adaptation of techniques for depositing planar films to this more complex geometry is not trivial.

Low R_s values are achieved in planar HTS films by growing the superconducting phase epitaxially on lattice-matched single crystal substrates, ensuring good c-axis alignment with the surface normal and minimising the presence of weakly-linked high angle grain boundaries which degrade macroscopic critical currents in the superconductor [13, 14]. Single-crystal, cylindrical substrates present different crystal planes at different positions around the curved surface, with 4-fold rotational symmetry about the cylinder axis in cubic systems. This paper reports on the microstructure of the first Tl-based superconducting films grown on LaAlO_3 and MgO single crystal cylinders and discusses the feasibility of fabricating metamaterial devices from such composites.

2. Microstructural Characterization

2.1. Experimental Details

Tl_2Ba_2CaCu_2O_7 (Tl-2212) thin films were deposited on the curved surface of single-crystal LaAlO_3 and MgO cylinders (3mm diameter; 5mm length) using a two-stage process similar to the process we typically use for depositing planar films [15, 16]. An amorphous precursor film is deposited by rf sputtering from a Ba-Ca-Cu-O target. The cylindrical substrate is rotated on its axis at a speed of approximately 1 r.p.m. during deposition to achieve uniform coverage. An ex-situ thalliation anneal at 820°C for 1 hour in the presence of 0.8g of source powder is carried out to convert the Tl-free precursor film into the Tl-2212 superconducting phase. Deposition times of 3 hours have resulted in the growth of Tl-2212 films with a thickness of ≈500nm (measured using a MicroXAM optical profiler).

The surface morphology and crystallographic alignment of the Tl-2212 films have been investigated by Scanning Electron Microscopy (SEM) in a JEOL 6500F microscope equipped with TSL Electron Backscatter Diffraction (EBSD) system. A methodology has now been developed to carry out X-ray diffraction studies of these curved samples in a Philips MRD diffractometer to confirm macroscopically the high resolution EBSD orientation analysis.

2.2. Results

The surface morphology of Tl-2212 films grown on lattice-matched LaAlO_3 (LAO) cylinders is very dependent on its position with respect to the LAO [100] axis (see figure 1). At positions where the LAO [100] axis is normal to the specimen surface (at θ = 0°, 90°, 270° and 360°), the film has a flat, plate-like morphology as observed in films grown on planar [100] LAO substrates [17]. As the θ angle (i.e. the angle between LAO [100] and the surface normal) increases, the plates of TI-2212 are oriented at increasingly large angles to the cylinder surface (figure 2b). EBSD point analysis, summarised in 3a, has shown that the angle between the c-axis of the TI-2212 film and the cylinder surface normal increases with θ up to an angle of 45°. As θ approaches 45°, three different plate orientations can be seen in the SEM micrographs (figure 2c). Plates A and B are oriented with their a-axes parallel to the cylinder axis,
but with their c-axes tilted by $-45\pm5^\circ$ and $+45\pm5^\circ$ to the surface normal respectively. Plate C has its c-axis oriented along the axial direction of the cylinder.

Figure 1: SEM micrographs showing how the surface morphology of a Tl-2212 film grown on a LAO cylinder varies with radial position. The images were taken with the cylinder axis at $20^\circ$ to the incident electron beam.

Figure 2: Summary of EBSD analysis showing the radial misorientation of Tl-2212 c-axis to substrate [100]

Tl-2212 films can also be grown directly on MgO substrates without chemical interaction. However, the lattice mismatch of the Tl-2212 a-b plane with MgO (9%) is much larger than the mismatch with LAO (1.7%). Since the Tl-2212 phase grows much more rapidly in the a-b plane than along the c-direction, films on flat MgO substrates still have good c-axis alignment perpendicular to the substrate surface. However, the in-plane alignment is strongly dependent on the surface condition of the substrate, with at least two and often more in-plane orientations present [16]. In contrast to Tl-2212 films grown on LAO cylinders, the microstructure of films grown on single-crystal MgO cylinders is not position dependent. The surface morphology everywhere consists of flat, plate-like grains as shown in figure 4a. EBSD analysis has shown that Tl-2212 plates grow on MgO cylinders with their c-axes parallel to the substrate surface normal regardless of the angle made with the MgO [100] axis (see figure 3b). However, the films exhibit a range of in-plane alignments, with a preference for the a-axis of the Tl-2212 grains to be oriented with the axial direction of the cylinder (figure 4b).
2.3. Discussion

The SEM and EBSD analysis have clearly shown major differences between the microstructure of Tl-2212 films grown on cylindrical LaAlO$_3$ and MgO substrates. These results have been confirmed macroscopically by XRD experiments, and are shown schematically in figure 4. On LAO cylinders, the Tl-2212 phase grows epitaxially, with its c-axis aligned to the LAO [100] direction even when the angle between the LAO [100] axis and the surface normal is approaching 45°. This has been observed previously in Tl-2212 grown on vicinal LAO substrates for vicinal angles up to 20° [18, 19] and is well known to occur in YBa$_2$Cu$_3$O$_7$ (YBCO) films grown on lattice-matched vicinal SrTiO$_3$ substrates [20, 21, 22, 23]. At θ angles above 45°, the orientation of the plates rotates by 90° as expected from the 4-fold symmetry of the substrate. There is a region close to θ = 45° where both orientations are present together, along with some additional plates oriented with their c-axis along the cylinder’s axial direction. These are usually termed a-axis needles and have been frequently observed in Tl-2212 films [13].
In contrast, the Tl-2212 films grown on MgO cylinders are like films grown on flat MgO substrates at all positions on the surface. The c-axis of the film rotates around the cylinder to remain parallel to the surface normal. The large lattice mismatch between the HTS phase and the MgO substrate is responsible for this very different microstructure: the nuclei do not grow epitaxially on MgO because the energy required to form a semi-coherent interface is too high. Instead, the driving force for the c-axis alignment observed in planar Tl-2212 films is the minimization of surface energy (maximising the area of the low energy (001) surface). This microstructure is much more suitable for the fabrication of metamaterial devices because we need to maximise the critical current that can flow circumferentially around resonating structures patterned into the HTS film. However, in order to maximise the device performance, it is necessary to have in-plane alignment of the HTS film in addition to good c-axis alignment. In YBCO a unique in-plane alignment can be achieved on vicinal MgO by pre-annealing the substrate to generate a row of steps on the surface with which the YBCO nuclei align [24, 25, 26]. The only two studies of Tl-HTS growth on pre-annealed MgO substrates have not been successful in improving the degree of in-plane alignment, possibly due to differences in the growth mechanism of the HTS phases [27, 28], but further work on vicinal MgO is required to investigate this fully.

3. Filter Simulations
Initial simulations of an axial array of split ring resonators that could be fabricated from the Tl-2212/MgO cylinder have been carried out using Microstripes®. The array of 6 SRR structures, each 0.5mm wide with a 0.3mm gap, are spaced by 0.4mm as shown in figure 6a. When we model the excitation of this array by a coil at one end coupling to a receiver coil at the other end of the cylinder, figure 6b shows that the structure resonates at a frequency of 6.36GHz and has an increase in transmission at the resonant frequency of \( \approx 40\text{db} \). Previously 3D bandpass metafilters have been fabricated from stacks of SRR structures on planar glass resin substrates[29].

Figure 5: Simulation of an axial array of 6 split ring resonators on an MgO cylinder

(a) Geometry of the SRR array  
(b) Transmission characteristics

4. Conclusion
Tl-2212 films have been grown for the first time on curved substrates. Microstructural analysis has shown that MgO is a promising substrate for the fabrication of HTS/dielectric metamaterial devices, and initial simulations of axial arrays of SRR structures patterned in this composite show filter characteristics in the GHz range.
Acknowledgments

Dr Susannah Speller is supported under the RAEng/EPSRC Research Fellowship Scheme.

References

[1] Shamonina E, Kalinin V A, Ringhofer K H and Solymar L 2002 Journal of Applied Physics 92 6252
[2] Radovskaya A, Sydoruk O, Shamonin M, Stevens C J, Faulkner G, Edwards D J, Shamonina E and Solymar L 2007 Microwave and Optical Technology Letters 49 1054
[3] Smith D R, Padilla W J, Vier D C, Nemat-Nasser S C and Schultz S 2000 Phys. Rev. Lett. 84 4184–4187
[4] Shelby R A, Smith D R and Schultz S 2001 Science 292 77–79
[5] Veselago V G 1968 Soviet Physics Uspekhi 10 509
[6] Pendry J B 2000 Phys. Rev. Lett. 85 3966–3969
[7] Pendry J B, Holden A J, Robbins D J and Stewart W J 1999 IEEE Transactions on Microwave Theory and Techniques 47 2075
[8] Shamonina E, Kalinin V A, Ringhofer K H and Solymar L 2002 Electronics Letters 38 371
[9] Wiltshire M C K, Pendry J B, Young I R, Larkman D J, Gilderdale D J and Hajnal J V 2001 Science 291 849–851
[10] Wiltshire M, Hajnal J V, Pendry J B, Edwards D J and Stevens C J 2003 Optics Express 11 709–+ 2007
[11] Lancaster M J 1997 Passive Microwave Device Applications of High-Temperature Superconductors (Cambridge University Press)
[12] Hesmer F, Tatartschuk E, Zhuromskyy O, Radkovskaya A A, Shamonin M, Hao T, Stevens C J, Faulkner G, Edwards D J and Shamonina E 2007 Physica Status Solidi b 244
[13] Newman N and Lyons W G 1993 Journal of Superconductivity 6 119–160
[14] Dark C J, Speller S C 2005 Materials Science and Technology 19 269–282
[15] Emergo R, Wu J, Aytug T and Christen D 2004 Applied Physics Letters 85 618 – 620 ISSN 0003-6951
[16] Durrell J, Burnell G, Tsaneva V, Barber Z, Blamire M and Evetts J 2004 Physical Review B 70 214508 ISSN 1098-0121
[17] Chana O S, Kuzakhmetov A R, Warburton P A, Hyland D M C, Dew–Hughes D, Grovenor C R M, Kinsey R J, Burnell G, Booj W E, Blamire M G, Kleiner R and Muller P 2000 Applied Physics Letters 76 1098–1102
[18] Habermeier H 2000 Journal of Superconductivity 13 871 – 875 ISSN 0896-1107
[19] Hu W, Wang T, Shi D, Zhao X, Peng W, Lei C, Tao H, Chen Y and Li L 2001 Journal of Crystal Growth 231 129 – 135 ISSN 0022-0248
[20] Emergo R, Wu J, Aytug T and Christen D 2004 Applied Physics Letters 85 618 – 620 ISSN 0003-6951
[21] Durrell J, Burnell G, Tsaneva V, Barber Z, Blamire M and Evetts J 2004 Physical Review B 70 214508 ISSN 1098-0121
[22] Linhan D S and Kavv A P 1998 Journal of Materials Research 13 2791
[23] Degardin A, Bourg S, Castel X, Bodin C, Berthon J, Dolin C, Caristan E, Pontiggia F, Perrin A and Kreisler A 2000 Institute of Physics Conference Series 167 85
[24] Moeckly B, Russek S, Lathrop D, Buhrman R, Norton M and Carter C 1990 Applied Physics Letters 57 2951 – 2953 ISSN 0003-6951
[25] Badica P, Sundaresan A, Crisan A, Nie J C, Hirai M, Fujiwara S, Kito H and Ihara H 2003 Physica C 383 482
[26] Pal S, Stevens C and Edwards D 2005 Proc. of XXVIIth URSI General Assembly