CONF-9203110--2

THE VARIABILITY RESIDUAL STRESSES OF THICK SUPERCONDUCTOR FILMS DURING ORTHORHOMBIC TO TETRAGONAL TRANSFORMATION

ANL/CP--75687

DE92 010499

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1992

Manuscript will be submitted to the *1992 International Conference on Advanced Science and Technology*, Argonne, IL, March 28, 1992

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*This work was supported by the U.S. Department of Energy, Conservation and Renewable Energy, as part of a program to develop electric power technology, under Contract W-31-109-Eng-38.

DISCLAIMER

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Abstract-- $YBa_2Cu_3O_x$ thick films have been deposited by spray pyrolysis of a sol-gel on 10 cm diameter polycrystalline MgO wafers. The film thickness was built up in layers of approximately 1 µm thick. The in-plane residual stresses were measured by an optical interferometry (shadow moiré) method as a function of film structure. In-plane residual stress maps over the area of the wafer have been obtained. The average stress of the 5 µm orthorhombic phase was 0.84 GPa. As the film transforms from the orthorhombic to the tetragonal structure, the tensile stresses decreased by 0.5 GPa.

Introduction

Superconductor thin films have been found to have a much higher critical current density (J_c) than bulk materials. However, there are several problems which may occur in thin film deposition processes. Epitaxial growth can be used to attain a high critical current density, but it is limited by the substrate crystal structure and deposition technique. Microstructure control can improve the mechanical properties and limit the weak link problem. The other problem is the residual stress of the film. These stresses can be a result of the difference in lattice expansion between the substrate and the film or phase transition during annealing. These two effects are especially important in the residual stresses which develop in thin films of YBCO deposited on polycrystalline MgO.

The residual stresses in thin films may be out-of-plane which would cause peeling and loss of adhesion, or in-plane which may result in film cracking and deflection of the substrate. The effect of residual stresses is more severe for thin films of large area and limits the applications.

There are several techniques for measuring strain. The general techniques include: the use of strain gauges, optical methods, and X-ray analysis. Optical techniques are unique since they are non-destructive, easy to use, and provide measurements over large spatial areas.

Spray pyrolysis of metalorganic compounds or complexes is a simple nonvacuum thin film deposition method, which is an inexpensive and convenient method for producing oxide films on large area substrates. This paper examines the sol-gel deposition technique for superconducting films and the effect of phase transformation on in-plane residual stress for only the 5 μm thick film.

Experimental Procedure

Film Fabrication

The YBCO precursor solution is prepared from the metal acetate salts dissolved in distilled water and gelled in adding monoethanolamine [1].

The sample deposited at 260°C was crack free. Heat treatment at high temperature was carried out after spray deposition. Specimens were heated at 3°C/min to 900°C and held for 1 hour. Initially, the furnace atmosphere was 1% O_2 in N_2, and the atmosphere was changed to 100% O_2 as the furnace cooled. Specimens were cooled at 0.5°C/min to 450°C and held for 10 hours to ensure full oxydation. The phase development during heat treatment was examined by X-ray diffraction. The coating and heat treatment process was repeated until the desired thickness was obtained. Each layer was approximately 1 μm thick.

Orthorhombic and tetragonal transformation

Orthorhombic $YBa_2Cu_3O_x$ thick films were converted back to the tetragonal state by annealing in flowing nitrogen. The specimens were heated at 3°C/min to 700°C and held for 1 hour; then they were cooled slowly to 600°C and held for 24 hours.

Residual stress measurement

In-plane residual stress measurement was previously decribed in Andonian and Danyluk [2], Wu et al. [3]. Shadow moiré fringes give the deflection of specimen and are converted to the strain. From measuring the strain, the residual stresses were calculated by comparison to substrate.

Results and Discussion

In the YBCO films, the tetragonal to orthorhombic phase transition and lattice expansion mismatch between the substrate and the film were responsible for the residual stresses.

In-plane residual stress developed from lattice expansion mismatch can be calculated from the following equation

$$\sigma_{rs} \propto \varepsilon \propto \left(\frac{\Delta a_{YBCO}}{a_{YBCO}} - \frac{\Delta a_{MgO}}{a_{MgO}}\right) = \varepsilon_{YBCO} - \varepsilon_{MgO}. \qquad (1)$$

σ_{rs} is the residual stress between YBCO and MgO, and ε is the expansion strain. a_{YBCO} and a_{MgO} are the lattice parameter of YBCO and MgO at 900°C, and Δa_{YBCO} and Δa_{MgO} are the lattice parameter change of YBCO and MgO between 900°C and 25°C. ε_{YBCO} and ε_{MgO} are lattice expansion of YBCO and MgO between 900°C and 25°C. If ε_{YBCO} is larger than ε_{MgO}, the ε will be positive and the film will be in tension. The lattice expansion of YBCO orthorhombic phase is 0.0145 and tetragonal phase is 0.00954 [4].

The orthorhombic 123 deposited on the MgO substrate will induce the tensile stress. The phase transition from tetragonal to orthorhombic phase will reduce the tensile stress if film is constrained by the substrate, shown in Figure 1. The average stress of orthorhombic to tetragonal phase transformation was decreased from 0.84 GPa to 0.34 GPa. Annealing a YBCO sample in nitrogen will lead to oxygen stoichiometry decrease. If the stoichiometry is below 6.6, the YBCO will have tetragonal structure. For the orthorhombic to tetragonal transition, the length of *c* axis changed. The x-ray diffraction data for the two phases are shown in Figure 2.

Before the orthorhombic to tetragonal transition, the film contained some microcracks indicated in the SEM examination, whick cracks might reduce electrical current density (J_c) [3]. After the

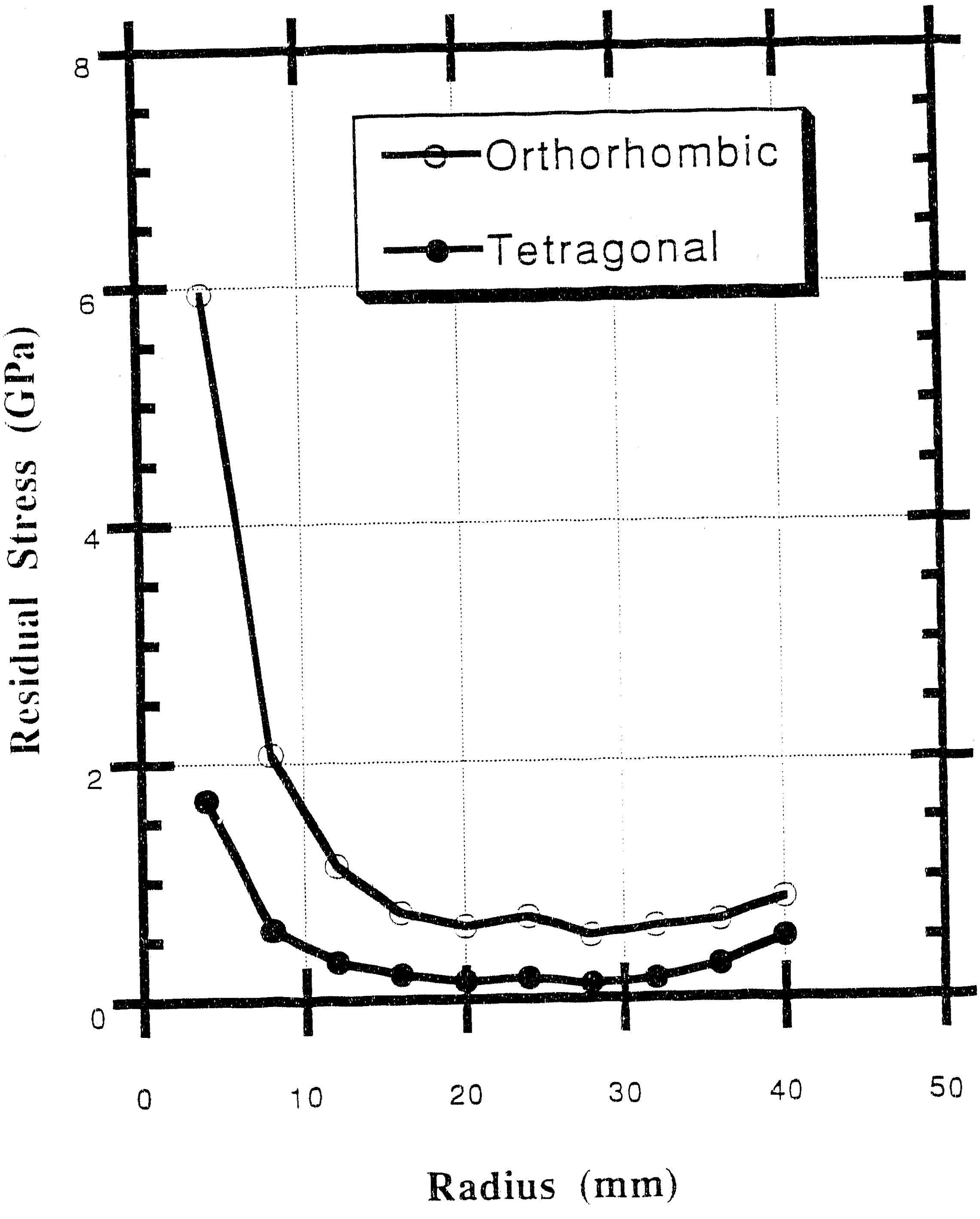

Figure 1. Residual stresses change during the phase transformation of YBCO.

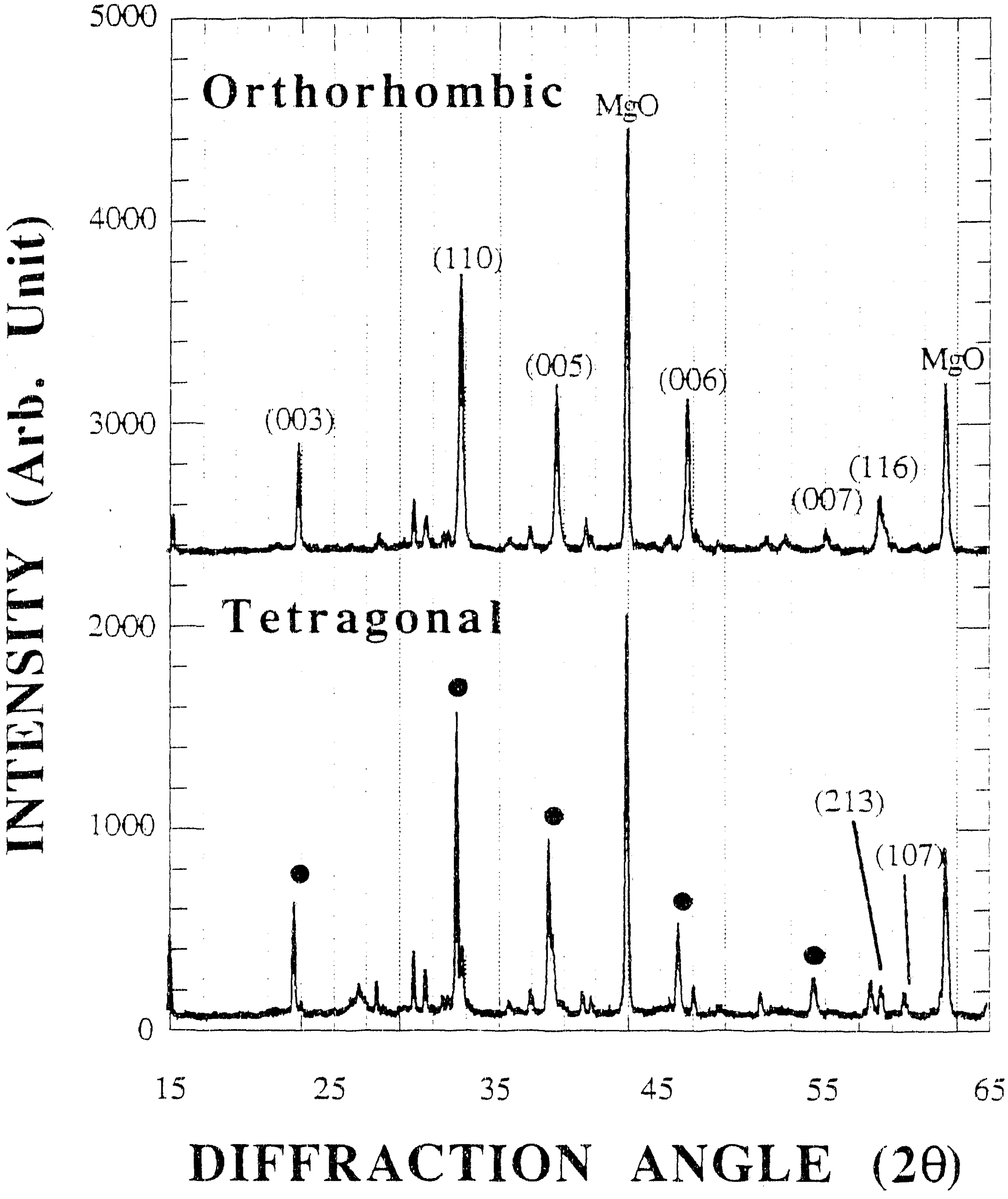

Figure 2. X-ray diffraction patterns of orthorhombic-tetragonal phase transformation.

transition, the microcracks were difficult to find. This may indicate the internal stress was change from the large tensile stress to the very small tensile stress and push the cracks closed. There may be some reasons for this tetragonal film to show low tensile stress replacing the compressive stresses. The first reason is even though the film had a tetragonal structure its oxygen stoichiometry was still higher than 6; therefore, lattice expansion maybe little higher than MgO. The second reason is the internal stresses of the film may not be just caused by lattice expansion mismatch like as the growth stress. The total residual stress can be comprised of misfit and intrinsic components, that is

$$\sigma_{rs} = \sigma_{misfit} + \sigma_{intrinsic} \qquad (2)$$

Dauksher et al. [5] had gat high percentage intrinsic stress in SiO_2 films. The third reason is the surface roughness. The rough surface can improve the adhesion between film and substrate because it increases the interface area and mixing the chemical and mechanical bonding together.

Conclusion

The in-plane residual stress measurement of YBCO films during the orthorhombic to tetragonal phase transition on polycrystalline MgO has been studied. The highest in-plane residual stresses were always found to be close wafer center, and the stress decreased toward the edge. The orthorhombic-to-tetragonal phase transformation resulted in a relaxation of residual stress from 0.84 to 0.34 GPa.

Acknowledgments

The authors would like to thank Dr. Marc L. Kullberg and Mr. Biao Wang for technique discussion.

This work was supported by the U.S. Department of Energy, Conservation and Renewable Energy, as part of a program to develop electric power technology, under Contract W-31-109-Eng-38.

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