

c -axis thermal expansion of the $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ superconductor

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The low-temperature c -axis thermal expansion of a thin-film sample of the superconducting material $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ was measured to 10 K. Based on standard ac resistance measurements, the material exhibits a detectable drop at 110 K and zero resistance at 78 K. Careful x-ray measurements between 10 and 300 K reveal a uniform, monotonic c -axis expansion of about 0.4%, similar to what has been previously found for $\text{La}_{1.9}\text{Ba}_{0.1}\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$. Although anomalous c -axis thermal expansion has been recently reported in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, a careful search in the vicinity of T_c failed to reveal anything unusual in this material.

I. INTRODUCTION

In the absence of a clear theoretical explanation of superconductivity in the new class of high- T_c , ceramic superconductors, a wealth of research is underway to provide extensive experimental data. Recently, Srinivasan *et al.*¹ reported an anomalous variation in the length of the c axis of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($\delta=0.01-0.15$) through the superconducting transition. Although the origin of this anomaly is not understood, Srinivasan *et al.*¹ argue that it is consistent with other anomalous behavior found in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, viz., in the nuclear quadrupole resonance frequencies as reported by Riesemeier *et al.*,² in positron-annihilation lifetimes as reported by Ming-Kang Teng *et al.*,³ and in sound velocity measurements as reported by Srinivasan, Ramachandran, Seshadri, and Ananda Ramdass.⁴ Given the similarity in the structures of all the newer higher- T_c materials, i.e., they are all distorted perovskites with alternating metal and copper oxide layers, the experiments described in this work were undertaken to search for a possible similar anomaly in $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$.

II. EXPERIMENTAL PROCEDURES

Thin films of $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ were prepared by evaporating a bulk sample of same composition in vacuum with an electron beam. The reactants were deposited on a polished (100) face of an MgO single crystal maintained at 300°C. The films were then annealed in air at 840°C. Complete details of the preparation procedures are given elsewhere.⁵

The presence of superconductivity was confirmed by standard low-frequency, four-terminal ac techniques with pressure contacts forming the electrodes and a current of approximately 5 μA . A small, but detectable drop in the resistance was observed at 110 K and a zero resistance at 78 K. The structure of the material is consistent with that reported by Zandbergen *et al.*⁶ for $\text{Bi}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$.

Due to the layered nature of these structures, upon deposition, the metal oxide planes tend to align themselves with the polished (100) surface of the single-crystal MgO substrate. This was confirmed by an x-ray-diffractometer scan made with the film normal coincident with the

diffraction vector; only (00 l) reflections were observed in the spectrum. Independently centering the MgO (200) and (400) and the superconductor (00 l) reflections on an automated diffractometer showed the two principal reciprocal-lattice vectors to be coincident to within 0.06°. Because of this configuration, it was not possible to make precision measurements of ($hk0$) reflections from the thin film. Thus, the thermal expansion of the other axes could not be determined. However, the anomalous behavior found in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ was restricted to the c axis.

For purposes of measuring the c -axis thermal expansion, the sample was mounted with thermally conducting grease on the cold stage of a Helitran cryogenic refrigerator. This refrigerator system is coupled to a computer controlled x-ray diffractometer in a manner similar to that previously described.⁷ A calibrated Si-diode thermometer is also bonded to the low-temperature block, and coarse temperature control is achieved by regulating the flow rate of He fluid from a remote storage Dewar. Fine control is accomplished with an electrical heater and feed-

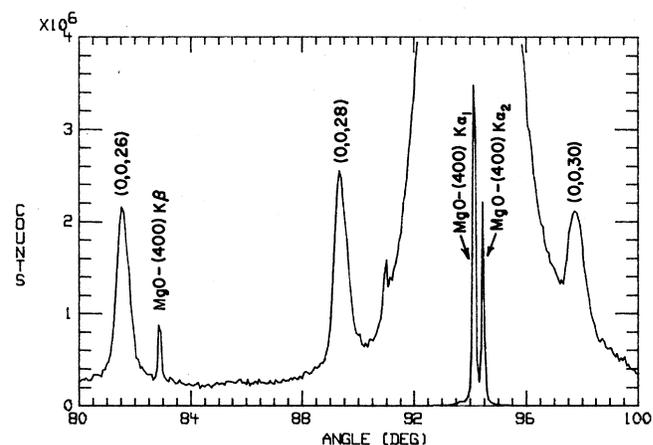


FIG. 1. X-ray-diffraction spectrum showing the three (00 l) peaks ($l=26, 28,$ and 30) of $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ and the MgO (400) peaks as excited by the Cu $K\beta$ radiation at 82.89°, by the Cu $K\alpha_1$ radiation at 94.14°, and by the Cu $K\alpha_2$ radiation at 94.53°. The ordinate scale is for the MgO (400) $K\alpha$ peaks; the ordinate scale is reduced by 10^3 for the second plotting of the same spectrum.

back circuit using either a Pt or Ge sensor, depending on the temperature range. The measured temperatures are estimated to be accurate and stable to within ± 0.5 K.

The sample was illuminated with radiation from a Cu x-ray tube and the scattered radiation measured with a NaI(Tl) detector through a graphite monochromator. Data were collected at fixed temperatures in a step-scanning mode over the range from 80° to 100° in 2θ . In this region, the three (00 l) peaks ($l = 26, 28,$ and 30) are recorded. Also recorded within this angular range is the MgO (400) peak as excited by the Cu $K\beta$ radiation at 82.89° , the Cu $K\alpha_1$ radiation at 94.14° , and the Cu $K\alpha_2$ radiation at 94.53° . As may be seen in Fig. 1, the intensity of the MgO $K\alpha$ peaks is 3 orders-of-magnitude greater than that of the other peaks. The MgO $K\beta$ peak is discriminated against by the graphite monochromator, but nevertheless, it is observable. The presence of these MgO peaks provide a convenient calibrant. A correction for small displacements in the sample, which may occur during thermal cycling, was made by using the known thermal expansion of MgO.⁸

III. RESULTS AND DISCUSSION

Although the c -axis temperature dependence can be determined from any of the three measured $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ (00 l) reflections, only the (0,0,28) data were used because they exhibited the least scatter. The fractional change in the c axis is plotted in Fig. 2. For purposes of comparison, similar data obtained earlier from a bulk sample of superconducting $\text{La}_{1.9}\text{Ba}_{0.1}\text{CuO}_4$ are plotted on the same figure.⁹ The solid curve in the figure is based on measurements on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ taken from the work of Horn *et al.*¹⁰ Clearly, within the combined experimental error, as reflected by the scatter in the data, the c -axis thermal expansivities of each of these superconductors are equivalent.

The data fail to reveal any unusual behavior in the c -axis thermal expansion. It is further noted that the size of

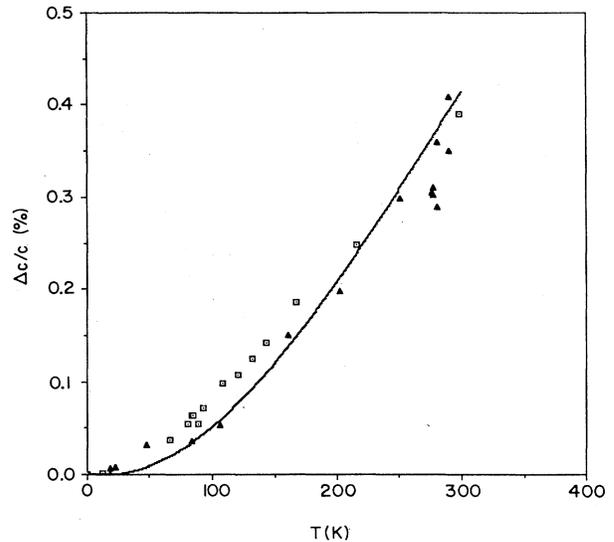


FIG. 2. Fractional change in the c -axis length as a function of temperature for $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ (squares), for $\text{La}_{1.9}\text{Ba}_{0.1}\text{CuO}_4$ (triangles) from Ref. 9, and for $\text{YBa}_2\text{Cu}_3\text{O}_7$ (solid curve) from Ref. 10.

the c -axis expansion anomaly seen by Srinivasan *et al.*¹ is well within the detection limits of these measurements. Between 82 and 102 K, Srinivasan *et al.*¹ find a discontinuous change in the c -axis length in excess of 3.5%, almost ten times greater than the thermal expansion seen here. Srinivasan *et al.*¹ have noted that others have not observed this anomalous behavior and suggest that different annealing treatments may be a cause for the contradictory results. In conclusion, we find no evidence of anomalous behavior in the c -axis thermal expansion of $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ and the linear thermal expansivity in this direction amounts to about 0.4% for each of the high- T_c superconductors, $\text{La}_{1.9}\text{Ba}_{0.1}\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_7$, and $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$.

¹R. Srinivasan, K. S. Girirajan, V. Ganesan, V. Radhakrishnan, and G. V. Subba Rao, Phys. Rev. B **38**, 889 (1988).

²H. Rieseheimer, Ch. Grabow, E. W. Scheidt, V. Muller, K. Luders, and D. Riegel, Solid State Commun. **64**, 309 (1987).

³Ming-Kang Teng, D. Shen, L. Chen, C. Yi, and G. Wang, Phys. Lett. A **124**, 363 (1987).

⁴R. Srinivasan, V. Ramachandran, A. T. Seshadri, and G. Ananda Ramdass, Pramana **29**, L603 (1987).

⁵M. S. Osofsky, P. Lubitz, M. Z. Harford, A. K. Singh, S. B. Qadri, E. F. Skelton, W. T. Elam, R. J. Soulen, Jr., W. L. Lechter, and S. A. Wolf, Appl. Phys. Lett. (to be published).

⁶H. W. Zandbergen, P. Groen, G. Van Tendeloo, J. Van Lan-

duyt, and S. Amelinckx, Solid State Commun. **66**, 397 (1988).

⁷E. F. Skelton, I. L. Spain, S. C. Yu, C. Y. Liu, and E. R. Carpenter, Jr., Rev. Sci. Instrum. **55**, 849 (1984).

⁸D. K. Smith and H. R. L der, J. Appl. Crystallogr. **1**, 246 (1968).

⁹E. F. Skelton, W. T. Elam, D. U. Gubser, S. H. Lawrence, M. S. Osofsky, L. E. Toth, and S. A. Wolf, Phys. Rev. B **35**, 7140 (1987).

¹⁰P. M. Horn, D. T. Keane, G. A. Held, J. L. Jordan-Sweet, D. L. Kaiser, F. Holtzberg, and T. M. Rice, Phys. Rev. Lett. **59**, 2772 (1987).