Effect of stress along the *ab* plane on the J_c and T_c of $YBa_2Cu_3O_7$ thin films

G. L. Belenky,* S. M. Green, A. Roytburd,† C. J. Lobb, S. J. Hagen, and R. L. Greene‡
Center for Superconductivity Research, Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742-4111

M. G. Forrester and J. Talvacchio
Westinghouse Science and Technology Center, 1310 Beulah Road, Pittsburgh, Pennsylvania 15235

(Received 24 May 1991)

We studied the influence of elastic stress along the *ab* plane (σ_{ab}) on the critical current density (J_c) and transition temperature (T_c) of epitaxial *c*-axis-oriented $YBa_2Cu_3O_7$ thin films. We observed reversible and reproducible increases (with compression) and decreases (with extension) of J_c and T_c from their equilibrium values. By comparing $dT_c/d\sigma_{ab}=0.045-0.050$ K/kbar with the results of hydrostatic-pressure and thermodynamic experiments reported in the literature, we conclude that pressure-induced changes in the *ab* plane completely account for the changes in T_c (within our experimental uncertainty). We also show that the mismatch strain between film and substrate does not substantially affect the change in T_c caused by the external stress.

INTRODUCTION

Continued study of the effect of pressure (P) on the transition temperature (T_c) of high-temperature superconductors (HTSC) is important to our understanding of the nature of superconductivity in these materials. In addition, external strain (ε) can affect another crucial characteristic of superconductors—the critical current density (J_c). Thus, deformation effects in HTSC is an important area of superconductivity research. The basic measured parameter is, as a rule, the pressure derivative dT_c/dP. Most data were obtained by studying the effect of hydrostatic compression (P_h) on HTSC ceramics and crystals. The results vary, but most fall within the range $dT_c/dP_h=0.05-0.12$ K/kbar.[1-4] Less data is available pertaining to *c*-axis uniaxial compression (P_c), and even the sign of dT_c/dP_c is not consistent.[5,6]

In this paper, we present and discuss observed changes in T_c and J_c with compression and extension along the *ab* plane (parallel to the Cu-O planes) in the absence of external stress in the *c* direction. These conditions are realized by bending substrates on which sputtered epitaxial thin films have been deposited and patterned. Our preliminary results were presented previously.[7] The effect of stress along the substrate plane was reported earlier by Park *et al.*[8] for polycrystalline $YBa_2Cu_3O_7$ films on Al_2O_3 substrates. They attributed the interesting and complex behavior of their films to the effects of granularity and inhomogeneity. The consistency of our results, among several films on different substrates, strongly suggests that our observations are intrinsic to $YBa_2Cu_3O_7$.

EXPERIMENT

Epitaxial *c*-axis-oriented thin films of $YBa_2Cu_3O_7$ were deposited by dc magnetron sputtering from a stoichiometric target onto rotating (100) $LaAlO_3$ and MgO substrates. A layer of Au sputtered onto the films *in situ* provided low-resistance contacts. The films had midpoint T_c's of 87–89 K, and resistivities at $T=100$ K, $\rho(100\text{ K})$, of 150–200 or 250–300 $\mu\Omega$ cm for $LaAlO_3$ or MgO substrates, respectively. $\rho(T)$ curves and *I-V* characteristics (at $T=77$ K) were measured on 50-μm-wide patterned strips. J_c was determined based on a threshold criterion of 2 μV/mm. Conventional four-terminal configurations were used for both the *I-V* and $\rho(T)$ measurements.

Strain along the *ab* plane was created by bending the substrate in a cantilevered arrangement (Fig. 1). When a force (F) is applied to the end of the substrate, the resulting strain$[\varepsilon_{xx}(x)]$ is given by[9]

$$\varepsilon_{xx}(x)=F\frac{6(l-x)}{E^s bh^2} . \quad (1)$$

Here, l is the length of the substrate between the clamp and the point of force application, x is the distance between the clamp and the film, b is the width of the substrate, and E^s is the flexural modulus (or corresponding combination of the elastic constants) of the substrate.

For an epitaxial film, the film's atoms have strong bonds with the substrate, so that substrate and film strains are identical. In a cantilevered experiment, the stress tensor component in the z direction, σ_{zz}, vanishes. In our experiment, $l-x\cong l$ allows a simplification of Eq. (1), and also implies $\varepsilon_{yy}\cong 0$. The stress along the *ab* plane, $\sigma_{ab}\equiv\sigma_{xx}+\sigma_{yy}$, is then given by

$$\sigma_{ab}=F\frac{6l}{E^s bh^2}\frac{(c_{11}+c_{12})c_{33}-2c_{13}^2}{c_{33}} . \quad (2)$$

Equation (2) is obtained from Hooke's law ($\sigma_{ik}=C_{iklm}\varepsilon_{lm}$ using the compact notation $xx\rightarrow 1$, $yy\rightarrow 2$, $zz\rightarrow 3$ for components of the elasticity tensor) and Eq. (1). The components of the elasticity tensor (C_{iklm}) of an

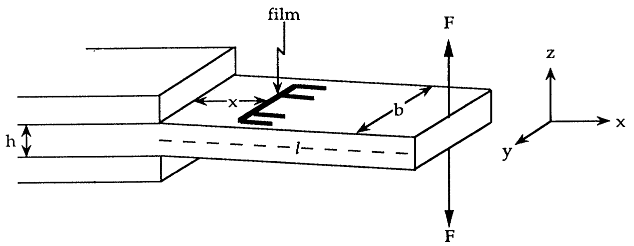

FIG. 1. The arrangement of the film on the substrate in the bending experiment. *l*, *b*, and *h* are the length, width, and thickness of the substrate, respectively.

$YBa_2Cu_3O_7$ crystal as determined by neutron scattering are[10] $c_{11}=274$ GPa, $c_{12}=36.7$ GPa, $c_{33}=183$ GPa, and $c_{13}=61.3$ GPa. We measured the values of E^s which control the flexural modes of the single crystal substrates directly using a flexural resonance technique.[11,12] For $LaAlO_3$ and MgO, $E^s=250$ and 300 GPa, respectively. The typical dimensions for our substrates were $(l,b,h)=(10$ mm, 6 mm, 0.5 mm). The strain ε_{xx} did not exceed 6×10^{-4}.

Figure 2 shows the shift of $\rho(T)$ with extension and compression. The values of the derivative as determined from $\Delta T_C^{\text{midpoint}}/\sigma_{ab}$ for different samples are

$$\frac{dT_c}{d\sigma_{ab}}=0.045\text{–}0.050\ \text{K/kbar}\ .$$

The error of the derivative is dominated by the uncertainties in the components of the elasticity tensor (15–20 %),[10] and is estimated to be 30%.

Figures 3 and 4 show the change in J_c with extension and compression for films on $LaAlO_3$ and MgO, respectively. The insets to Figs. 3 and 4 present the stress dependence of J_c. The critical current density increases with compression along the *ab* plane, and decreases with extension. The shifts in both T_c and J_c are reversible and reproducible.

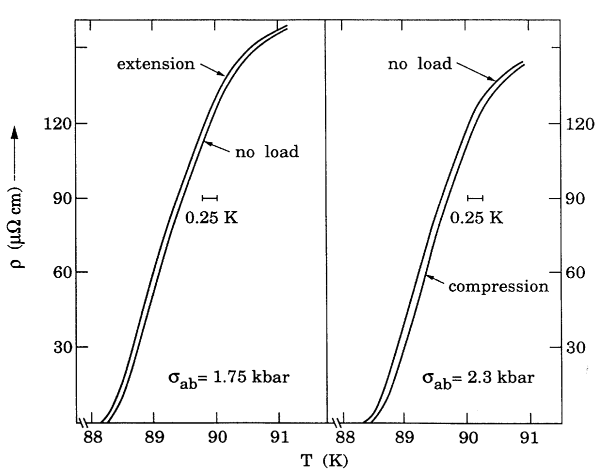

FIG. 2. Resistivity vs temperature of $YBa_2Cu_3O_7$ film on $LaAlO_3$ substrate, shown with extension and with compression. Film thickness = 1000 Å.

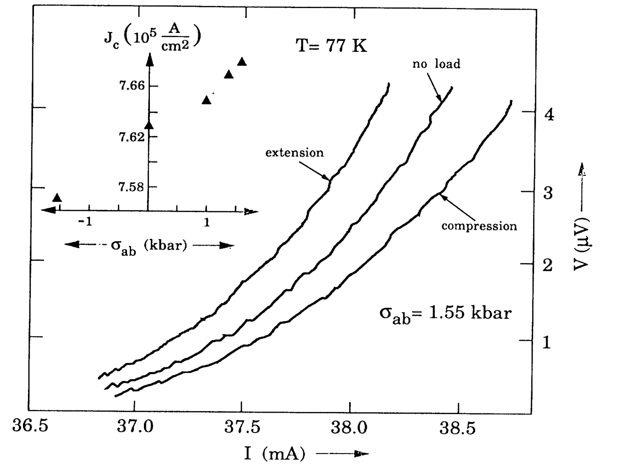

FIG. 3. The effect of stress along the *ab* plane on the *I-V* characteristics of $YBa_2Cu_3O_7$ film on $LaAlO_3$. Film thickness = 1000 Å; $\rho(100\ \text{K})=200\ \mu\Omega$ cm. Inset: the dependence of J_c on stress ($\sigma_{ab}>0$ is compression; $\sigma_{ab}<0$ is extension).

DISCUSSION

The experimental results described above show that compression along the *ab* plane leads to an increase in T_c, while extension along this plane decreases T_c in $YBa_2Cu_3O_7$ films. The results clearly indicate a direct correlation between changes in T_c and changes in interatomic spacing in the *ab* plane. A correlation has been noted, in previous work,[13] between the *c* parameter and T_c. This correlation could be taken to suggest that

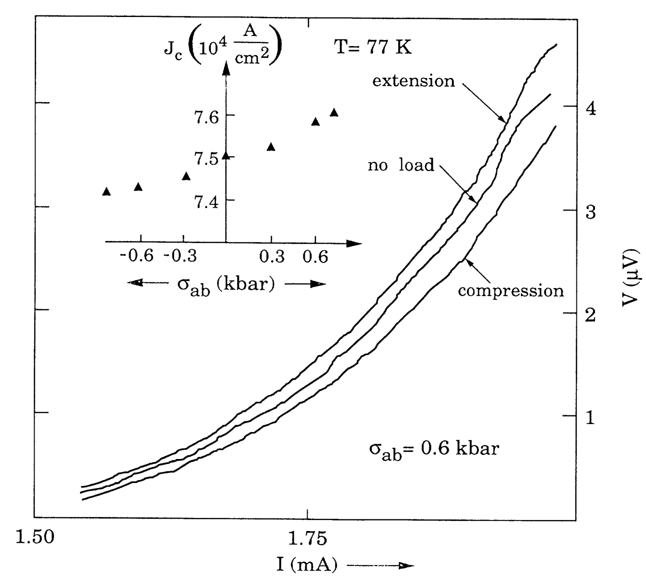

FIG. 4. The effect of stress along the *ab* plane on the *I-V* characteristics of $YBa_2Cu_3O_7$ film on MgO. Film thickness = 500 Å; $\rho(100\ \text{K})=380\ \mu\Omega$ cm. Inset: the dependence of J_c on stress ($\sigma_{ab}>0$ is compression; $\sigma_{ab}<0$ is extension).

changes in c parameter *cause* changes in T_c. Analysis of our data, combined with thermodynamic data from the literature, indicates that this is not the case, as discussed below.

Another result is the increase of J_c under compression and the decrease of J_c with extension (the increase of J_c in thin $YBa_2Cu_3O_7$ films under hydrostatic pressure was reported in Ref. 14). The value $\Delta J_c/J_c^0$ (J_c^0 is the critical current density without deformation) did not exceed several percent. The behavior of $J_c(\sigma_{ab})$ turned out to be similar in experiments with several films on both $LaAlO_3$ and MgO. These films had different normal-state resistivities, and critical current densities which differed by two orders of magnitude (see, for example, Figs. 3 and 4). The similarity in their behavior cannot be readily explained within the framework of "weak link" models. It is our point of view that a direct connection between the changes in T_c and J_c cannot be neglected since compression along the *ab* plane causes both to increase just as extension leads to their decrease. It is also necessary to consider that external stress tends to concentrate near defects which may influence the pinning force. Thus, the nature of the reversible change in J_c remains unclear.

We can compare our value of $dT_c/d\sigma_{ab}$ with the results of hydrostatic pressure experiments using the fact that the derivative dT_c/dP_h is obtained by summing two contributions:

$$\frac{dT_c}{dP_h}=2\frac{dT_c}{d\sigma_{ab}}+\frac{dT_c}{d\sigma_c} ,$$

where σ_c is stress (or pressure) along the c axis. The value calculated from our experiments, $2dT_c/d\sigma_{ab} \approx 0.09-0.10$ K/kbar, falls well within the range reported from hydrostatic experiments in the literature. This fact supports the conclusion that pressure-induced changes in interplanar spacing along the c axis do not contribute significantly to the change in T_c for $YBa_2Cu_3O_7$.

Confirmation of this statement follows by considering both our results and the experimental results of thermodynamic investigations.[15,16] From the generalization of Ehrenfest's relation for second-order phase transitions, it is possible to write the following equation:

$$\frac{\Delta\alpha_{xx}\sigma_{xx}+\Delta\alpha_{yy}\sigma_{yy}+\Delta\alpha_{zz}\sigma_{zz}}{\Delta C_p/T_c}=\Delta T_c . \quad (3)$$

Here, $\Delta\alpha_{ij}$ are the thermal expansion jumps of the lattice at T_c, ΔC_p is the jump in heat capacity at T_c, σ_{ij} are the stress tensor components, and ΔT_c is the stress-induced change in T_c. For $YBa_2Cu_3O_7$ single crystals and oriented grained samples, $\Delta\alpha_{xx}+\Delta\alpha_{yy}$ is equal to 2.3×10^{-7} K^{-1} and 1.5×10^{-7} K^{-1}, and $\Delta C_p/T_c$ is 27 mJ/mol K^2 and 43 mj/mol K^2, respectively.[15] Assuming that $\sigma_{ab}=\sigma_{xx}+\sigma_{yy}$ and $\sigma_{zz}=0$, we can calculate $dT_c/d\sigma_{ab}=0.089$ K/kbar for the single crystal and 0.038 K/kbar for the oriented grained sample. Taking into account that the value 0.089 K/kbar is too large due to the presence of Al doping impurities,[16] we believe that our experimental result of 0.045–0.050 K/kbar is in good agreement with Eq. (3).

Our deduction that pressure-induced changes in interplanar spacing do not contribute significantly to the change of T_c is strongly supported by the fact that the jump in the component of thermal expansion along the c axis at T_c is very small: $\Delta\alpha_{ab}/\Delta\alpha_{zz}>20$.[15] Our deduction agrees with the theoretical conclusion that a large change in Cu-O interplanar distance affects T_c only slightly,[17] as well as data that establish a correlation between the T_c and the in-plane Cu-O bond length in cuprate superconductors.[18]

Finally, we discuss the role of thermal contraction mismatch between the film and substrate. [A discussion of this internal strain ($\hat{\varepsilon}_m$) is presented in the literature.[19]] It is possible to describe the free energy of a film strained due to interaction with its substrate (strain tensor $\hat{\varepsilon}_m$) and an external stress $\hat{\sigma}$ (corresponding strain $\hat{\varepsilon}_\sigma$) by the following equation (using for simplicity the notation $\hat{\varepsilon}$ and $\tilde{C}$ instead of ε_{ik} and C_{iklm}):

$$F(\varphi,\varepsilon)=F_0(T)+\tfrac{1}{2}a(T-T_c^0)\varphi^2+\tfrac{1}{4}B\varphi^4 + \tfrac{1}{2}[\hat{\varepsilon}-\hat{\varepsilon}_0(\varphi)]\tilde{C}[\hat{\varepsilon}-\hat{\varepsilon}_0(\varphi)] . \quad (4)$$

The first three terms characterize the free energy of the unstrained state. Here φ is the order parameter, T_c^0 is the critical temperature in the unstrained state, and $a/(2B)=\Delta C_p/T_c$.[20] The last term in (4) corresponds to the elastic energy of the superconductor at a given φ.[21] The elastic modulus $\tilde{C}=\tilde{C}_0+\Delta\tilde{C}\varphi^2$ near the transition point, $\tilde{C}_0$, corresponds to the normal state and ΔC characterizes the superconducting state, ε is the total strain tensor $\hat{\varepsilon}=\hat{\varepsilon}_m+\hat{\varepsilon}_\sigma$, and $\hat{\varepsilon}_0(\varphi)=\hat{\varepsilon}_0(T)+\hat{\omega}\varphi^2$ is the self-strain directly connected with $\hat{\alpha}$ for equilibrium φ: $d\hat{\varepsilon}/dT=\hat{\alpha}$, $\Delta\hat{\alpha}=\hat{\omega}a/B$. It is possible to estimate the critical temperature T_c of a film with internal (mismatch) strain [$T_c\neq T_c^0$ in (4)] as well as the shift of this temperature under external stress ΔT_c, by equating to zero the sum of all terms containing φ^2 in (4). Taking into account that only terms containing $\hat{\varepsilon}_\sigma$ need to be kept, and neglecting the quadratic term $\hat{\varepsilon}_\sigma\Delta\tilde{C}\hat{\varepsilon}_\sigma$, we can write the following equation for ΔT_c, which was measured in our experiment:

$$\Delta T_c=\frac{\Delta\hat{\alpha}\hat{\sigma}-(a/B)\hat{\varepsilon}_m\Delta\tilde{C}\hat{\varepsilon}_\sigma}{\Delta C_p/T_c} . \quad (5)$$

The first term of Eq. (5) corresponds to the generalization of Ehrenfest's relation, Eq. (3), which has been shown to describe the results of our experiments satisfactorily. For all practical purposes, then, substrate mismatch does not contribute significantly to the change in T_c caused by external stress. The consistency in our experimental results for films on different substrates strongly supports the conclusion that the J_c and T_c changes observed are intrinsic to $YBa_2Cu_3O_7$ and are not mismatch induced.

CONCLUSION

We observed reversible and reproducible changes in J_c(77 K) and T_c of c-axis-oriented epitaxial $YBa_2Cu_3O_7$ thin films due to compression and extension along the *ab* plane. Compression increases and extension decreases both J_c and T_c from their unstressed values. We have determined that $dT_c/d\sigma_{ab}=0.045-0.050$ K/kbar. Furthermore, it was shown that pressure-induced changes within the *ab* plane dominate the changes in T_c. Comparison of our result with thermodynamic considerations shows that mismatch strain between the film and substrate does not affect the change in T_c caused by external stress.

ACKNOWLEDGMENTS

We are grateful for helpful discussions with E. Suhir, T. Venkatesan, and M. Wuttig. The authors also appreciate the assistance provided by M. Wuttig and Harsh Deep Chopra during the measurements of the substrates' flexural moduli. Partial support for this work was provided by the Electric Power Research Institute.

[*]Present address: AT&T Bell Laboratories, Murray Hill, NJ 07974.

[†]Also at Department of Material and Nuclear Engineering.

[‡]Also at IBM Research Division, Yorktown Heights, NY 10538.

[1]C. W. Chu, Z. J. Huang, R. L. Meng, L. Gao, and P. H. Hor, Phys. Rev. B **37**, 9730 (1988).

[2]R. Wingaarden and R. Griessen, in *Studies of High Temperature Superconductors,* edited by A. Narlikar (NOVA Science, New York, 1989).

[3]R. Wingaarden, E. N. van Eenige, J. I. Scholtz, and R. Griessen (unpublished).

[4]S. Klotz, W. Reith, and J. C. Shilling, Physica C **172**, 423 (1991); J. E. Schirber, B. Morosin, and D. S. Ginley, *ibid.* **157**, 237 (1989).

[5]U. Koch, J. Wittig, and B. Gegenheimer, Physica C **162-164**, 739 (1989).

[6]M. F. Crommie, A. Y. Liu, A Zettl, M. L. Cohen, P. Parillo, M. F. Hundley, W. N. Creager, S. Hoen, and M. S. Sherwin, Phys. Rev. B **39**, 4231 (1989).

[7]G. L. Belenky, S. M. Green, S. J. Hagen, A. Roytburd, R. L. Greene, M. G. Forrester, and J. Talvacchio (unpublished); G. L. Belenky, S. M. Green, S. J. Hagen, R. L. Greene, M. G. Forrester, and J. Talvacchio (unpublished).

[8]S. I. Park, M. R. Schenermann, C. C. Chi, and C. C. Tsuei, in *High Temperature Superconductors, Boston, 1987,* edited by M. B. Brodsky, R. C. Dynes, K. Kitazawa, and H. Tuller, MRS Symposia Proceedings No. 99 (Materials Research Society, Pittsburgh, 1988), p. 685.

[9]R. P. Feynman, R. Leighton, and M. Sands, *The Feynman Lectures on Physics* (Addison-Wesley, Reading, MA, 1964), Vol. 2, p. 38.

[10]S. K. Chan, B. W. Veal, and C. K. Loonge (private communication).

[11]A. S. Nowic and B. S. Berry, in *Anelastic Relaxation in Crystalline Solids* (Academic, New York, 1972), p. 360.

[12]M. Wuttig and C. H. Lin, Acta Metall. **31**, 1117 (1983).

[13]See, for example, C. B. Eom, J. Z. Sun, B. M. Lairson, S. K. Streiffer, A. F. Marshall, K. Yamamoto, S. M. Anlage, J. C. Bravman, T. H. Geballe, S. S. Laderman, R. C. Taber, and R. D. Jacowitz, Physica C **171**, 354 (1990).

[14]Q. Xiong, M. F. Davis, S. Dechmuch, Y. Q. Wang, Y. Y. Xue, J. C. Wolfe, D. Economou, P. H. Hor, and C. W. Chu (unpublished).

[15]C. Meingast, B. Blank, H. Bürkle, B. Obst, T. Wolf, and H. Wüne, Phys. Rev. B **41**, 11 299 (1990).

[16]C. Meingast, R. Ahrens, B. Blank, H. Bürkle, B. Rudolf, and H. Wüne, Physica C **173**, 309 (1991).

[17]D. Yoshioka, J. Phys. Soc. Jpn. **59**, 2627 (1990).

[18]M. H. Whangbo, D. B. Kang, and C. C. Torardi, Physica C **158**, 371 (1989).

[19]E. Suhir, J. Electron. Packag. **111**, 281 (1989); E. Suhir, in Proceedings of the International Society for Optical Engineering [SPIE J. **1187**, 227 (1989)].

[20]*Landau and Lifshitz Course on Theoretical Physics* (Pergamon, Elmsford, NY, 1980), Vol. 9, pt. 1.

[21]A. Roytburd, Fiz. Tverd. Tela (Leningrad) **26**, 2025 (1984) [Sov. Phys. Solid State **26**, 1229 (1984)].