Anisotropic pressure dependence of T_c in single-crystal $YBa_2Cu_3O_7$ via thermal expansion

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High-resolution anisotropic-thermal-expansion measurements of single-crystalline and oriented-grained $YBa_2Cu_3O_7$ at the superconducting transition are presented for the first time. Discontinuities in the thermal-expansion coefficient $\alpha_{ab}[\Delta\alpha_{ab}=(15-23)\times10^{-8}\ K^{-1}]$, measured with a capacitance dilatometer, are found to occur in both samples. No discontinuity in $\alpha_c(|\Delta\alpha_c|<1\times10^{-8}\ K^{-1})$ is observed in either sample, although $\alpha_c(T)$ shows a distinct change of slope at T_c. The specific-heat discontinuity ΔC_p of both samples was also measured and is used, along with the $\Delta\alpha$'s, to calculate the dependence of T_c on uniaxial pressure and uniaxial strain to first order. T_c is predicted to increase with pressure applied perpendicular to the c axis ($dT_c/dp_{ab}=0.04-0.09$ K/kbar) and to be insensitive to pressure parallel to the c axis. Uniaxial strain, on the other hand, is found to increase T_c about equally in both directions.

INTRODUCTION

The large positive hydrostatic pressure dependence of the critical temperature ($dT_c/dp=0.1-0.8$ K/kbar is a unique and, therefore, important feature of oxide superconductors.[1] $YBa_2Cu_3O_7$ has a relatively small pressure dependence of T_c ($dT_c/dp=0.03-0.1$ K/kbar), although doping this system with only 2.5% Fe can increase dT_c/dp to 0.6 K/kbar.[2,3] $YBa_2Cu_4O_8$ also has a large positive pressure dependence of 0.5 K/kbar at a relatively high critical temperature of 81 K.[4] A successful microscopic theory of superconductivity in these materials will have to not only correctly predict the high critical temperatures, but also these exceptionally strong pressure dependences. So far dT_c/dp has been measured almost exclusively using hydrostatic pressure,[1] however due to the highly anisotropic nature of these materials, measurements of T_c under uniaxial pressure conditions are of utmost importance. For example, determining dT_c/dp for uniaxial pressure along the c axis provides a measure for the importance of coupling between the superconducting Cu-O layers. To our knowledge, only two such experiments have been reported to date. Crommie *et al.*[5] found that T_c increases at a rate of 0.03–0.1 K/kbar for uniaxial pressure (0–1 kbar) along the c axis in $YBa_2Cu_3O_7$ single crystals. However, only the pressure dependence of T_c onset (measured resistively) is presented and it is, therefore, not clear whether this is a true bulk effect. The effect of pressure perpendicular to the c axis was not examined. Koch *et al.*[6] examined $YBa_2Cu_3O_7$ single crystals in a high-pressure cell (0–100 kbar) which has both hydrostatic and uniaxial pressure components. Their results show a clear anisotropy with a decrease in T_c for pressure applied parallel to the c axis, and an initial increase in T_c for pressure perpendicular to the c axis. These latter measurements also illustrate the difficulties associated with making direct pressure measurements on these brittle materials. Applying pressure considerably broadened the superconducting transitions and resulted in irreversible damage to the crystals.

Measurements of the discontinuity in the thermal-expansion coefficient $\alpha(T)$ at T_c along different crystallographic directions, along with specific-heat data, can provide an alternative method for determining the first-order uniaxial pressure derivatives of T_c.[7,8] The advantages of this approach are that it is a true bulk method, the difficulties in applying uniform uniaxial pressure are eliminated, and the derivatives are determined at zero pressure which can only be obtained by extrapolation from the direct pressure measurements. This thermodynamic derivation assumes that the jumps in C_p and α are solely due to superconductivity, and not, for example, due to a structural transformation at T_c. That these conditions are met in $YBa_2Cu_3O_7$ is indicated by the thermodynamic consistency between the temperature dependence of the thermodynamic critical field and the jump in specific heat,[9] and by the good correlation between the measured jumps in α and C_p, and dT_c/dp obtained in polycrystalline samples of $YBa_2Cu_3O_7$ and $YBa_2(Cu_{1-x}Fe_x)_3O_7$ with use of the Ehrenfest relationship.[3,10,11]

In this paper we present, to our knowledge for the first time, high-resolution anisotropic-thermal-expansion measurements as well as specific-heat data on single-crystalline and oriented-grained $YBa_2Cu_3O_7$ samples. Well-defined second-order jumps in $\alpha(T)$ are observed perpendicular to the c axis (α_{ab}) at the same temperatures where the measured specific-heat jumps are found to occur. No jump in α was observed parallel to the c axis (α_c). The uniaxial pressure dependence of T_c, calculated using these data, is found to be very anisotropic, with

$dT_c/dp_{ab} \gg dT_c/dp_c$. The uniaxial strain dependence of T_c does not exhibit this anisotropy; decreasing only the c axis is predicted to increase T_c at the same rate as decreasing the a,b axis. This finding indicates that the coupling between superconducting Cu-O layers is not of primary importance in determining T_c.

EXPERIMENTAL DETAILS

Two $YBa_2Cu_3O_7$ samples were investigated: a single-crystalline (SC) and an oriented-grained sample (OG) of approximate dimensions $2\times2\times1$ mm^3 (1 mm in c axis) and $4\times2\times4$ mm^3 (4 mm in c axis), respectively. The SC was grown using the CuO-BaO self-flux method in an Al_2O_3 crucible. A detailed description of the growth parameters and further characterization of these crystals are presented elsewhere.[12] Inductive T_c measurements gave $T_c=90$ K; $\Delta T_c=2$ K. The OG sample was fabricated using a liquid phase processing method[13] which produces many thin platelike grains stacked on top of each other. Typical grain dimensions are 10–20 μm (in c axis) times several mm^2 (in ab plane). Misorientation between grains is estimated to be about 1–3°. T_c, measured resistively, is unusually high ($T_c=93.4$ K) and also quite sharp with $\Delta T_c=1$ K. Both samples are heavily twinned as observed with optical methods. It is, therefore, impossible to differentiate between the a and b axes from bulk measurements, and both samples are assumed to have tetragonal symmetry for the purpose of these investigations. This was confirmed by measurements of $\Delta\alpha$ along two orthogonal directions in the a-b plane for the SC.

The thermal expansion was measured with a high-resolution capacitance dilatometer similar, but with several important differences, to previous designs.[14] Rather than stabilizing the temperature for each length measurement, the temperature is slowly raised (5–7 mK/sec) at a very "smooth" and reproducible rate over the whole temperature range by a computer controlled heater. To ensure good thermal equilibrium of the sample with the dilatometer, the fairly massive dilatometer was designed so that the much less massive sample is totally surrounded by and in good thermal contact with the dilatometer through He exchange gas. Length, temperature, and heating rate are measured every 2 sec, then averaged over 0.1 K intervals, and are finally stored in a computer. The relative errors between consecutive temperature readings, taken with a platinum thermometer, are estimated to be on the order of ±200 μK from the measured heating-rate fluctuations. The length changes are measured with an analog capacitance bridge[15] with $\pm5\times10^{-12}$ m resolution. The coefficient of thermal expansion $\alpha(T)$ is defined by

$$\alpha(T)=(1/LT)(\Delta L/\Delta T)\ , \qquad (1)$$

where ΔL and ΔT are the differences in length and temperature between consecutive data points and $L(T)$ is the length of the sample. A resolution in $\alpha(T)$ of about 10^{-8} K^{-1} can be obtained in our experimental setup, even for samples only 1mm long, if ΔL and ΔT in Eq. (1) are the differences between averaged sets of 10 data points (i.e., $\Delta T\approx1$ K). All α's presented in this paper were calculated in this manner. However, this resolution was only achieved by cycling back and forth in a restricted temperature range of $T\pm30$ K. Failure to do this resulted in many "glitches" (non-reversible length changes of several angstroms) which are also observed by other groups,[16] which worsened the relative resolution considerably. The uncertainty in α is estimated to be $\pm3\%$ and $\pm10\%$ for the OG and SC samples, respectively.

THERMAL EXPANSION AND SPECIFIC-HEAT RESULTS

The anisotropy in the thermal expansion is shown in Fig. 1 for both samples. The thermal-expansion coefficient is considerably larger along the c axis (α_c) than along the a,b axis (α_{ab}) in agreement with x ray[17] and neutron diffraction measurements.[18] Both samples exhibit the same anisotropy within the errors of measurements. The OG sample shows a pronounced dip in both α_{ab} and α_c (for increasing T) between 200 and 300 K. This behavior was reproducible, almost an order of magnitude larger than the experimental uncertainty, and temperature hysteretic. Upon cooling, this anomaly disappeared as indicated by the dashed lines in Fig. 1. The origin of this anomaly is unclear at this point. Expanded views of the thermal-expansion coefficients near T_c (for runs where T was cycled between 60 and 110 K) are presented in Figs. 2–4 (upon heating). The specific-heat anomalies at T_c, measured using an adiabatic continuous heating calorimeter,[19] of both samples are also shown in Figs. 2 and 3. Unmistakable second-order transitions in both α_{ab} and C_p are seen for both samples. Both transitions ($\Delta\alpha$ and ΔC_p) occur at exactly the same temperature and also have approximately the same width, illustrating their common origin. In fact one can hardly distinguish between α and C_p from the shape of these curves. It is interesting to note that while the specific-

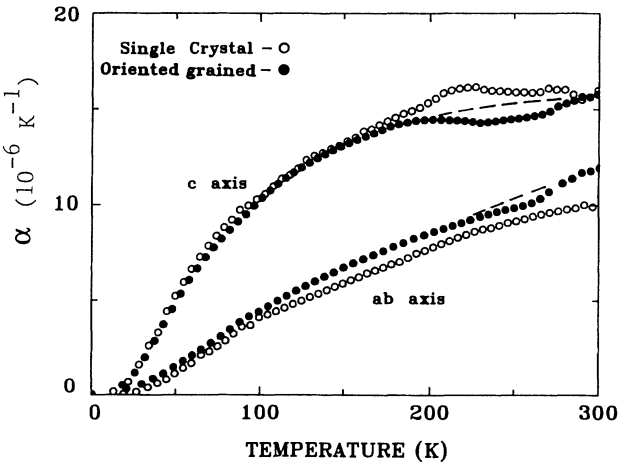

FIG. 1. Linear thermal-expansion coefficients vs temperature for the single-crystal (open circles) and the oriented-grained (solid circles) $YBa_2Cu_3O_7$ samples along and perpendicular to the c axis. The dashed lines indicate the behavior of the oriented grained sample upon cooling.

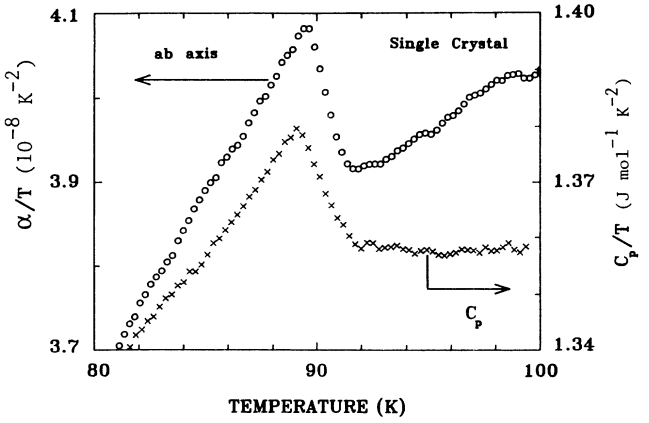

FIG. 2. Expanded view of the thermal-expansion coefficient α_{ab}/T and specific heat C_p/T (crosses) near the superconducting transition for the single-crystalline sample. Both quantities exhibit clear second-order transitions.

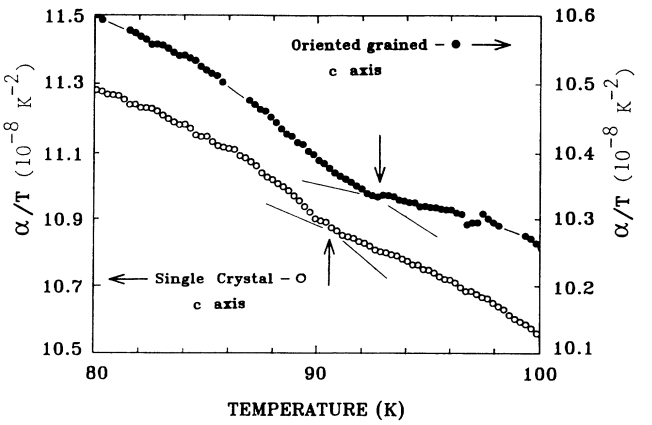

FIG. 4. Expanded view of the thermal-expansion coefficients α_c/T for both samples near the superconducting transition. No second-order transition (jump) is observed. However, a distinct change of slope occurs at T_c (marked by vertical arrows) for both samples.

heat jump is larger in OG than in SC (43 versus 27 mJ/mol K^2), the jump in α_{ab} is smaller in OG than in SC (15×10^{-8} versus 23×10^{-8} K^{-1}). In α_c, no jumps ($|\Delta\alpha_c|<1\times10^{-8}$ K^{-1}) are observed in either sample. However, a well defined change of slope of $\alpha_c(T)$ occurs at T_c as shown in Fig. 4. The approximate magnitudes for these changes in slope $\Delta(d\alpha/dT)$ are 2×10^{-8} K^{-2} (SC) and 1.2×10^{-8} K^{-2} (OG). Closer inspection of α_{ab} shows that, along with the jump, there is also a change of slope at T_c with magnitudes -3.5×10^{-8} K^{-2} (SC) and -2.5×10^{-8} K^{-2} (OG). The jumps and slope changes of $\alpha(T)$ observed at T_c were found to be reproducible during several runs, and also showed the same effects upon cooling.

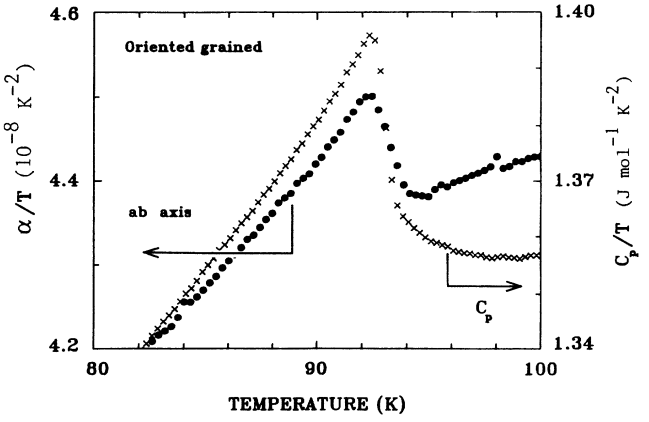

FIG. 3. Expanded view of the thermal-expansion coefficient α_{ab}/T and specific heat C_p/T (crosses) near the superconducting transition for the oriented-grained sample. Both quantities exhibit clear second-order transitions.

CALCULATED UNIAXIAL PRESSURE DEPENDENCE OF T_c

The uniaxial pressure (stress) dependence of T_c can be calculated from the measured jumps in α and C_p.[7,8] The jump in α is generally given by[20]

$$\Delta\alpha_i=\alpha_{iS}-\alpha_{iN}=-\left[\frac{1}{4\pi}\right](dH_c/dT)(dH_c/dp_i)\ , \quad (2)$$

where H_c and p_i are the thermodynamic critical field and the pressure (stress) along the ith direction, respectively. $H_c(p_i,T)$ can be written, close to T_c and for a parabolic temperature dependence of H_c, as

$$H_c(p_i,T)=A(p_i)[T_c(p_i)-T]\ , \quad (3)$$

where A is related to the specific-heat discontinuity at T_c as $\Delta C_p/T_c=(C_{pS}-C_{pN})/T_c=A^2/4\pi$. Expanding $T_c(p)$ in first- and second-order derivatives of p_i we obtain

$$T_c(p)=T_c(0)+\Sigma a_i p_i+\Sigma b_{ij}p_i p_j\ , \quad (4)$$

where $a_i=dT_c/dp_i$ and $b_{ij}=d^2T_c/dp_i dp_j$. For tetragonal symmetry there are two independent a_i's and six b_{ij}'s. Using Eqs. (3) and (4), and evaluating the derivatives in Eq. (2) at $T=T_c$, we arrive at the following expression for the jump in α_i at zero applied pressure:

$$\Delta\alpha_i=(A^2/4\pi)a_i=(\Delta C_p/T_c)a_i\ . \quad (5)$$

From Eq. 5 it is seen that the jumps in α_i are directly related to the first-order pressure derivatives of T_c and that the second-order b_{ij} terms do not contribute to the jump in the thermal-expansion coefficient at zero stress. Evaluating the coefficients a_1 and a_3 for our samples, using Eq. (5), we find the following: $a_1=dT_c/dp_{ab}=0.089$ K/kbar (SC); 0.036 K/kbar (OG), $|a_3|=|dT_c/dp_c|<0.004$ K/kbar (SC and OG). The values of the measured thermal-expansion and specific-heat discontinuities, and the calculated pressure derivatives are summarized in Table I.

TABLE I. Measured values of the jumps in thermal-expansion coefficients and specifit heat, and calculated uniaxial and hydrostatic pressure dependencies of the critical temperature. The calculted uniaxial pressure dependence is highly anisotropic.

	$\Delta\alpha_{ab}$ (10^{-8} K)	$\lvert\Delta\alpha_c\rvert$ (10^{-8} K)	$\Delta C_p/T_c$ ($mJ/\text{mol K}^2$)	dT_c/dp_{ab} (K/kbar)	$\lvert dT_c/dp_c\rvert$ (K/kbar)	dT_c/dp (K/kbar)
Single crystalline	23	<1	27	0.089	<0.004	0.177
Oriented grained	15	<1	43	0.036	<0.003	0.073

DISCUSSION

From Table I one can see that the behavior found is highly anisotropic [$a_1/a_3>22$ (SC) and 12 (OG)]: pressure applied perpendicular to the c axis increases T_c, while pressure applied parallel to the c axis has little effect upon T_c. The dependence of T_c on hydrostatic pressure is simply obtained by summing over the a_i's ($dT_c/dp=2a_1+a_3$, for tetragonal symmetry). The values thus obtained [$dT_c/dp=0.177$ K/kbar (SC) and 0.073 K/kbar (OG)] fall well within the scatter of hydrostatic pressure measurements reported in the literature.[1] dT_c/dp is significantly larger for the SC than for OG, which may have its origin in the strong dependence of dT_c/dp on even small amount of doping.[2,3] The SC contains up to 2% Al due to its processing method.[12] Al doping has been shown to decrease the orthorhombicity, which has been empirically correlated to an increase in dT_c/dp.[21] The fact that the specific-heat jump and T_c are reduced from the values for "ideal" $YBa_2Cu_3O_7$ is also consistent with Al doping.[22] An oxygen content below 7.0 can also increase dT_c/dp.[3] (Unfortunately, exact values for dT_c/dp are not known to us for Al doping, and, therefore, no definite explanation can be given for this effect at this time.)

The uniaxial stress dependence of T_c measured directly by Koch *et al.*[6] are in rough agreement with our calculated values. They found an initial increase of T_c for stress perpendicular to the c axis ($dT_c/dp_{ab}\sim 0.1-0.2$ K/kbar), and a decrease for stress parallel to the c axis ($dT_c/dp_c\sim -0.1$ K/kbar). It must, however, be stressed again that these measurements can only provide very approximate results due to the non-ideal nature of their experiment. A more direct comparison can be made with the results of Crommie *et al.*[5] since the stresses were applied uniaxially (in the c direction) and were of much smaller magnitude, which makes the extraction of a linear term possible. A closer inspection of their data reveals that their results are quite nonlinear with possibly a very small linear term, which is in agreement with our calculated result. However, since only the onset was measured, it is not clear whether this represents a real bulk effect. The positive pressure dependence found at $p>0$ is also in disagreement with the results of Koch *et al.* More precise uniaxial pressure measurements on well characterized samples are needed for a more meaningful comparison, and for determining the importance of second-order effects. The magnitude of dT_c/dp_{ab} has also been determined by the change in sound velocity at T_c in single crystals,[23] and was found to equal ± 0.18 K/kbar, which is also in fair agreement with our results.

The large anisotropic nature of the T_c pressure dependence derived from our experiment is greatly reduced if one considers the physically more meaningful *uniaxial strain,* instead of the *uniaxial pressure (stress),* dependence of T_c. These two quantities are different because the application of uniaxial stress along one direction also produces strain along orthogonal directions. The T_c strain dependence coefficients ($dT_c/d\epsilon_i$) can be calculated directly from the pressure dependence coefficients (a_i's) using the elastic modulus tensor C_{ij}.[7] For tetragonal symmetry, the first-order coefficients are: $dT_c/d\epsilon_{ab}=(C_{11}+C_{12})a_{ab}+C_{13}a_c$ and $dT_c/d\epsilon_c=2C_{13}a_{ab}+C_{33}a_c$. Since a_c is approximately zero, we see that both strain coefficients are positive and the strain anisotropy depends only on a ratio of elastic constants as $(dT_c/d\epsilon_{ab})/(dT_c/d\epsilon_c)=(C_{11}+C_{12})/2C_{13}$. The constants C_{11} and C_{33} have been determined from sound-velocity measurements on a single crystal (see Table II).[24] The other elastic constants can be easily calculated using measured values of the bulk modulus (C_{bulk}) and the result that under hydrostatic pressure the ratio of strain parallel and perpendicular to the c axis is given by $\epsilon_{ab}/\epsilon_c=0.74$.[25] Unfortunately, the bulk modulus determined using sound velocity measurements[26] ($C_b\approx 1000$ kbar) is much smaller than C_b measured by x-ray investigations under hydrostatic pressure ($C_b\approx 1800$ kbar),[25] and accordingly we will use both values as upper and lower bounds. We find $C_{12}=916$ kbar, $C_{13}=2080$ kbar and $C_{12}=213$ kbar, $C_{13}=834$ kbar for $C_b=1800$ kbar and $C_b=1080$ kbar, respectively. Evaluating the strain derivatives yields $dT_c/d\epsilon_{ab}=226$-288 K (SC), $dT_c/d\epsilon_{ab}=93$-118 K (OG), and $dT_c/d\epsilon_c=148$-369 K (SC), $dT_c/d\epsilon_c=61$-151 K (OG). A compilation of the elastic constants and strain derivatives are given in Table II, where one can see that both strain derivatives are positive and of approximately equal magnitude. The exact ratio of strain coefficients $(dT_c/d\epsilon_{ab})/(dT_c/d\epsilon_c)$, varies between 0.8 and 1.5 depending on the choice of C_b. Measurements of the change in sound velocity at T_c can provide a direct method for determining the magnitude of the strain derivatives.[8] Such measurements have recently been made on $YBa_2Cu_3O_7$ single crystals[24] and it was found that $\lvert dT_c/d\epsilon_{ab}\rvert=160$ K and $\lvert dT_c/d\epsilon_c\rvert=125$ K, which is in very good agreement with our results. Sound-velocity measurements can, however, only determine the magnitude of the strain coefficients and not the

TABLE II. Calculated elastic constants and uniaxial strain dependence of T_c for upper and lower bounds of the bulk modulus (see text for details). The strain dependence of T_c is found to be approximately isotropic.

C_{bulk} (kbar)	C_{11}[a] (kbar)	C_{33}[a] (kbar)	C_{12} (kbar)	C_{13} (kbar)	$dT_c/d\epsilon_{ab}$ (K)	$dT_c/d\epsilon_c$ (K)
1080	2340	1450	213	834	226 (SC)	148 (SC)
					93 (OG)	61 (OG)
1800	2340	1450	916	2080	288 (SC)	369 (SC)
					118 (OG)	151 (OG)

[a]Values from sound velocity measurements on a single crystal (Ref. 24).

sign. Our calculation also provide the sign of the strain coefficients, and from Table I, it is seen that reducing the c-axis length is predicted to increase T_c at about the same rate as decreasing the dimensions of the a,b axes. Therefore, the coupling between the Cu-O layers is not judged to be of primary importance in determining T_c in $YBa_2Cu_3O_7$, since, if it were, a much larger anisotropy of the T_c strain dependence would be expected.

The stress coefficient a_c can also be written as $a_c = 2S_{13}dT_c/d\epsilon_{ab} + S_{33}dT_c/d\epsilon_c$, where S_{ij} are components of the elastic compliance tensor, which can have negative values. The large anistropy of the a_i's ($a_{ab} \gg a_c \approx 0$) can then be considered as an accidental cancellation of the two strain coefficients. However, it is surprising that this "accidental" cancellation occurs for both SC and OG, which have quite different values for a_{ab}. This implies that either the strain coefficients always have the same anisotropy, irrespective of magnitude, if the elastic constants do not change, or that the elastic constants are strongly correlated with the strain coefficients, so that always $2S_{13}dT_c/d\epsilon_{ab} = -S_{33}dT_c/d\epsilon_c$.

Another interesting aspect of these measurements is the, relatively, large change of slope in the $\alpha(T)$ curves observed at T_c for both samples. For α_c, $\Delta(d\alpha_c/dT) = (d\alpha_c/dT)_S - (d\alpha_c dT)_N$ is positive, while for α_{ab}, $\Delta(d\alpha_{ab}/dT)$ is negative and almost twice as large in magnitude. Both $\Delta(d\alpha/dT)$'s are about 1.5 times larger in the SC than in the OG, and, therefore, are also proportional to the jump in α_{ab}, which is about 1.5 times larger in the SC. Sound-velocity measurements show a similar effect where the sound velocity $v(T)$, increases upon cooling at a greater rate below than above T_c, which is attributed to a faster increase of the elastic constants below T_c.[23] Our measurements indicate that this hardening upon cooling occurs in the a-b plane, since $\Delta(d\alpha_{ab}/dT)$ is negative and α correlates with the magnitude of the elastic constants. Since $\Delta(d\alpha_c/dT)$ is positive, one could argue that softening occurs in the c direction, however, the effect in the c direction may also be a reflection of the a-b hardening, coupled to the c axis via the elastic constants. The change of slope in both samples are about twice as large in the a-b direction than in the c direction, which supports the latter view.

CONCLUSIONS

In conclusion, the jumps in the thermal expansion coefficient at T_c were measured along and perpendicular to the c axis in single-crystalline and oriented-grained $YBa_2Cu_3O_7$ and are found to be highly anisotropic in both cases, with $\Delta\alpha_{ab} \gg \Delta\alpha_c \approx 0$. The specific-heat jump at T_c was also measured for both samples and is found to occur at exactly the same temperature, and also has nearly the same shape, as the jump in thermal expansion coefficient in the a-b plane. The calculated first-order uniaxial pressure derivatives of T_c exhibit the same anisotropy as the α's and are in fair agreement with direct uniaxial pressure measurements. Using measured and calculated values of the elastic moduli (C_{ij}), the uniaxial strain dependence of T_c is calculated and is shown to be positive in both directions, and approximately isotropic. Coupling between Cu-O layers is, therefore, not of primary importance in determining T_c. The temperature derivatives of α_{ab} and α_c are also discontinuous at T_c, and this is most likely due to increased hardening of the elastic constants along the a-b direction (C_{11}) below T_c.

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