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Uniaxial pressure dependence of the superconducting critical temperature in RBa$_2$Cu$_3$O$_{7-\delta}$ high-T_c oxides

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We have obtained the three uniaxial-pressure derivatives of the critical temperature, T_c, for GdBa$_2$Cu$_3$O$_{7-\delta}$ from the hydrostatic pressure dependence measured on films of different crystalline orientations. The strain derivatives are found to be extremely anisotropic in the a-b plane, inducing a decrease (increase) of T_c when compressing across (along) the CuO chains. A comparison of the results in c-oriented RBa$_2$Cu$_3$O$_{7-\delta}$ films and superlattices for various R and substrates reveals a nonmonotonic relation between the critical temperature and pressure.

One of the issues not yet satisfactorily understood for the high-T_c cuprates is the relation between the crystal structure and superconductivity. The anisotropy of the layered perovskitelike structure is reflected in their properties, superconductivity being no exception. An important clue towards establishing this relation may be provided by the pressure dependence of T_c. In particular, the uniaxial pressure dependence should yield information on the anisotropic coupling of structure and superconductivity along the different crystalline orientations. However, these experiments are complicated by the thin plate shape and extreme fragility of the available high-T_c single crystals. To the best of our knowledge, measurements dealing with the *uniaxial* pressure dependence of superconductivity in high-T_c oxides have been very limited.[1] We present here an alternative approach towards the study of this problem which consists of an investigation of the *hydrostatic* pressure dependence of $T_c(P)$, on highly crystalline, oriented thin films. In this case, the *hydrostatic* pressure applied to the combined system thin-film–substrate is transformed into an *anisotropic* effective stress on the film.

We have studied the hydrostatic pressure dependence of T_c for a variety of RBa$_2$Cu$_3$O$_{7-\delta}$ (RBCO) films (R=Yb, Y, Dy, and Gd) and YBa$_2$Cu$_3$O$_{7-\delta}$/GdBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO/GdBCO) superlattices on single-crystalline SrTiO$_3$ (STO), MgO, and yttria-stabilized zirconia (YSZ) substrates. From $T_c(P)$ measurements in a-, b-, and c-oriented GdBCO films on STO, the three uniaxial strain derivatives for GdBCO, $dT_c/d\varepsilon$, along a, b, and c, can be extracted. In addition, the R dependence of dT_c/dP for c-oriented films exhibits a crossover from negative to positive which is also found in the pressure dependence of c-oriented DyBCO films on MgO.

The films (~2000 Å thick) were deposited by dc magnetron sputtering from ceramic targets in a 0° off-axis geometry[2] (substrate parallel to the target and outside the plasma region) in order to avoid resputtering effects.[3] The a- and c-oriented GdBCO films were grown on (100) STO by depositing at different substrate temperatures while the b-oriented GdBCO film was obtained by deposition on a STO substrate cut at 18° from the (100) plane. The resulting film has the b axis parallel to the [100] STO direction, at 18° from the surface normal.

Measurements from 77 K to room temperature in the 0–2.5 GPa pressure range were performed in a piston-cylinder hydrostatic pressure cell similar to the one used in Ref. 4 with a 40:60 mineral oil:pentane mixture as the pressure-transmitting medium. The pressure was measured at room temperature by means of a Manganin manometer. The change in pressure upon cooling of the cell was corrected according to a previous calibration.[5] Briefly, the low-temperature pressure was calibrated against the room-temperature pressure by means of the pressure dependence of T_c for In and Sn. The superconducting critical temperature was determined from standard four-probe ac resistivity measurements using a current density of ≈ 10 A/cm^2. The temperature was measured with a SiO$_2$ diode in thermal contact with the exterior of the cell. At the typical temperature sweep rate (0.1 K/min) used in the experiments, there was no detectable thermal lag between sample and thermometer and the resistance data taken while cooling and warming were identical. The extrapolation of $T_c(P)$ to $P=0$ coincides with independent dc resistive measurements. Typically the 10%–90% transition width of the samples was 1.5 K and remained constant under applied pressure. The critical temperature was defined at 50% of the resistive transition although other definitions do not change the conclusions presented here.

Figure 1 shows the experimental results for the hydrostatic pressure dependence of T_c for the a-, b-, and c-oriented GdBCO films on STO. The striking feature in this graph is the qualitatively different behavior for the a-oriented film. While the b- and c-oriented films show an *increasing* linear $T_c(P)$, the a-oriented film shows a smaller *decreasing* trend. It is worth noting at this point that dT_c/dP is always found to be positive for bulk 1:2:3 materials under hydrostatic pressure,[6] although there is some spread in the values that depend upon oxygen content.

In order to obtain the strain derivatives of T_c from these measurements the following issues must be taken into consideration: (1) Given the experimental linearity of $T_c(P)$, the pressure derivatives will be related to the strains through

$$T_c(P)-T_c(0)=\frac{\partial T_c}{\partial \varepsilon_x}\varepsilon_x+\frac{\partial T_c}{\partial \varepsilon_y}\varepsilon_y+\frac{\partial T_c}{\partial \varepsilon_z}\varepsilon_z\ , \qquad (1)$$

where ε_x, ε_y (ε_z) are the strains induced in the directions

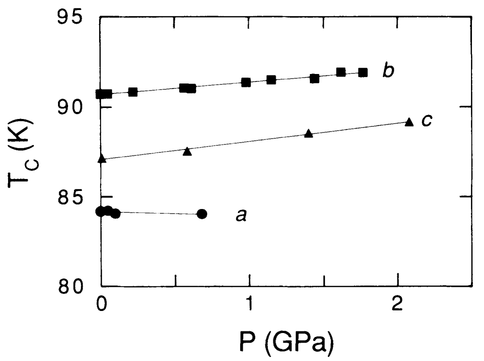

FIG. 1. Hydrostatic pressure, P, dependence of the superconducting critical temperature, T_c, for $GdBa_2Cu_3O_{7-\delta}$ films grown on STO. Letters indicate the crystalline axis normal to the substrate. The "b-oriented" film is actually oriented 18° out of the substrate normal. The slopes of the linear fits to the data (solid lines) are -0.2 ± 0.1 K/GPa for the a-oriented film, 0.69 ± 0.03 K/GPa for the b-oriented film, and 1.01 ± 0.07 K/GPa for the c-oriented film.

parallel (perpendicular) to the substrate surface. We define the strains as positive on compression. (2) Given the cross-sectional area difference between the film and the substrate, we assume that the lateral strain of the film is determined by that of the substrate; i.e., the strain components parallel to the surface are determined only by the substrate elastic constants. Formally, $C^s_{11} = 317.6$ GPa and $C^s_{12} = 102.5$ GPa for STO (Ref. 7) with C_{ij} being the stiffness constants. Notice that since STO is an isotropic material this implies that the possible in-plane polycrystalline character of the film is irrelevant for these estimates. (3) An epitaxial 1:2:3 film on an STO substrate will be subject to anisotropic stresses even without any applied pressure due to the differential thermal contraction and possibly different lattice constant. We assume that the elastic response of the material is not affected by its initial strained state; i.e., the strains are linearly additive. (4) To the best of our knowledge, no complete set of stiffness constants for GdBCO has been reported. However, since YBCO and GdBCO have similar structures,[8] it is expected that their elastic moduli will be the same to within ~5%.[9] Therefore, for the qualitative conclusions presented here it is reasonable to use the C_{ij} estimated for YBCO in Ref. 10; i.e., $C_{aa} = 223$ GPa, $C_{bb} = 244$ GPa, $C_{cc} = 138$ GPa, $C_{ab} = 37$ GPA, $C_{ac} = 89$ GPa, and $C_{bc} = 93$ GPa.[11] (5) The 18° tilt of the b axis with respect to the surface normal for the b-oriented film induces a small shear strain, 1 order of magnitude smaller than the compressive strains. In addition, two different in-plane orientations are possible. This calculation proves to be tedious and does not modify the results in a significant way. Consequently, this tilt is neglected. Under these assumptions Eq. (1) expressed for the three different film orientations defines a system of equations from which the strain derivatives can be obtained. Table I shows the results of this calculation. The surprising feature is the ab-plane anisotropy with a negative value of $\partial T_c/\partial\varepsilon$ along a compared to a positive one for $\partial T_c/\partial\varepsilon$ along b. Given these results and the stiffness constants C, the pressure derivatives of T_c for bulk GdBCO can also be calculated. These estimates are shown in Table I together with the experimental value for the hydrostatic pressure dependence of T_c for bulk samples.[12] Again, there are striking features. Note the much smaller pressure derivative in the b direction and the very good agreement between the experimental and predicted values for the hydrostatic pressure case.

TABLE I. Estimated strain derivatives, $\partial T_c/\partial\varepsilon$, and predicted uniaxial and hydrostatic pressure derivatives, dT_c/dP, for $GdBa_2Cu_3O_{7-\delta}$. Strains and pressures are defined as positive on compression. Errors represent the error propagation from the experimental pressure derivatives in Fig. 1. Errors in the elastic constants are not considered (Ref. 11). The experimental value from Ref. 12 is included for comparison.

Applied strain or pressure		$\frac{\partial T_c}{\partial\varepsilon}$ (K)	$\frac{dT_c}{dP}$ (K/GPa)	$\left.\frac{dT_c}{dP}\right\vert_{\text{expt.}}$ (K/GPa)
Uniaxial	a axis	-362 ± 50	-3.06 ± 0.35	⋯
	b axis	301 ± 30	0.38 ± 0.18	⋯
	c axis	239 ± 24	3.45 ± 0.43	⋯
Hydrostatic		⋯	0.77 ± 0.06	0.83

Figure 2 shows the experimental results for the dependence of dT_c/dP on the trivalent ionic radius, $r(R^{3+})$, for various c-oriented RBCO films and YBCO/GdBCO multilayers on STO, MgO, and YSZ single-crystal substrates. We selected $r(R^{3+})$ as a convenient ordinate because all structural parameters[8] and, possibly, all elastic and thermal expansion coefficients,[9,13] scale with it. For the multilayers $r(R^{3+})$ was defined as the weighted average of the ionic radii. Besides the general trend of the pres-

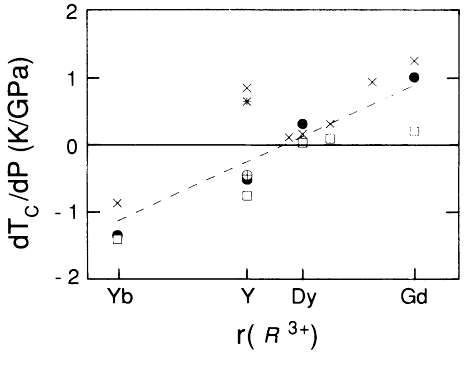

FIG. 2. Hydrostatic pressure derivative of the critical temperature, dT_c/dP, as a function of ionic radius of the trivalent ion, $r(R^{3+})$, for c-oriented $RBa_2Cu_3O_{7-\delta}$ films grown on STO (●), MgO (×), and YSZ (□) substrates. Data for $YBa_2Cu_3O_{7-\delta}/GdBa_2Cu_3O_{7-\delta}$ multilayers with $YBa_2Cu_3O_{7-\delta}$ to $GdBa_2Cu_3O_{7-\delta}$ thickness ratios of 1/3, 3/1, and 1/1 and the data of Voronovskii, Dizhur, and Itskevich (Ref. 15) for $YBa_2Cu_3O_{7-\delta}$ films grown on STO (⊕) and MgO (*), have been included in the graph. For the multilayers $r(R^{3+})$ was defined as the thickness ratio weighted average of $r(Y^{3+})$ and $r(Gd^{3+})$. The dashed line is a guide to the eye.

sure derivative with $r(R^{3+})$ and some minor effect due to the substrate, the most interesting feature is the change in sign of dT_c/dP around the Dy^{3+} ionic radius. Under the assumption that the initial strains due to epitaxy or differential thermal contraction with respect to the substrate do not modify the elastic response, the only other possible explanation for this behavior is a nonmonotonic relation[14] between T_c and pressure. As a consequence, T_c should have a positive slope for small pressures (large ionic radius), go through a maximum, and exhibit a negative slope at large pressures (small ionic radius). This is supported by the experimental pressure dependence of T_c for a c-oriented DyBCO film on MgO shown in Fig. 3. Since the Dy^{3+} ionic radius is close to the sign crossover of dT_c/dP shown in Fig. 2, a nonmonotonic behavior with small slope is expected in the accessible pressure range.

It should be pointed out that the nonlinear dependence of T_c on strain may seem at odds with the estimates of strain derivatives presented above for GdBCO. In particular, the fact that we cannot understand the zero pressure T_c values in terms of only epitaxially induced strains may seem to indicate that the three GdBCO films are in different initial strained states on three different points of the nonlinear T_c vs P relation. However, the agreement with the pressure derivatives measured by Voronovskii, Dizhur, and Itskevich[15] in YBCO films, in spite of the difference in the zero pressure T_c values (0.4 K for MgO and 2.2 K for STO substrate) indicate that this is not the case. Another mechanism has to be included to explain the zero pressure T_c such as small differences in the interdiffusion between the Gd and Ba sites due to different growth temperatures for the films. Also, the linear behavior of T_c with pressure shown in Fig. 1 and the fact that GdBCO on STO in Fig. 2 is far away from the crossover point in the positive slope region, like the bulk materials, and the agreement between the estimated and experimental hydrostatic pressure derivative in Table I suggest that our analysis in GdBCO is not affected by the nonlinear T_c vs P relation.

The substrate dependence for YBCO and GdBCO remains an open question since there is no qualitative systematic difference between the substrates and films, neither in their thermal differential stresses[16] nor in their elastic properties.[7,10,17] The good agreement with the data of Voronovskii, Dizhur, and Itskevich[15] (see Fig. 2) excludes the possibility of an experimental error. Another intriguing fact is that the superlattices follow the same trend with $r(R^{3+})$ as the films although their structure shows clear composition modulation.[18]

Table II shows a comparison between determinations of the uniaxial pressure derivatives made by different groups. Meingast *et al.*[19] estimated the strain derivatives of T_c for YBCO based on high-resolution thermal expansion measurements of untwinned single crystals. Their results show the same ab-plane anisotropy as in this paper. However, the absolute value for the strain derivatives is different. In particular the b-direction derivative is larger than here and the result for the c-direction derivative is $dT_c/dP_c \sim 0$, in contrast with our results. The results of Crommie *et al.*[1] for the c-direction derivative, although smaller than here, are restricted to a very narrow range of pressures (1 kbar) and the data show nonlinear behavior.

A number of studies[12,20-23] have claimed that the CuO_2 plane to apical oxygen interatomic distance controls the pressure-induced T_c changes in RBCO compounds. The difference in sign for the ab-plane strain derivatives can be qualitatively understood in terms of this distance; as the a axis is compressed (across the CuO chains) the Ba tends to separate the apical oxygen from the CuO_2 planes, while in compressing along the b axis (along the CuO chains) there is a competition between the Ba pushing the apical oxygen away from the CuO_2 planes and the oxygens on the chains repelling it towards the planes.

In conclusion, the strain derivatives of T_c in GdBCO are highly anisotropic in the ab plane, inducing a decrease (increase) of T_c when compressing across (along) the CuO chains. A study for a number of RBCO epitaxial films on different substrates implies that the pressure dependence of T_c is nonmonotonic, in agreement with the pressure dependence of T_c for DyBCO on MgO.

Note added. The experiments of G. L. Belenky *et al.*, Phys. Rev. B **44**, 10117 (1991), recently came to our attention. The numerical result presented there for the in-plane strain dependence of T_c is within factors of 2 of our estimates assuming that their film is epitaxially oriented with the b axis parallel to the bending direction of the substrate.

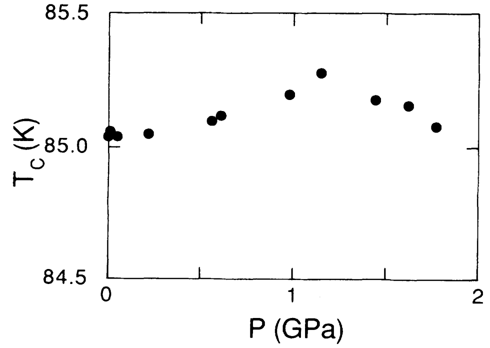

FIG. 3. Hydrostatic pressure, P, dependence of the superconducting critical temperature, T_c, for a c-oriented $DyBa_2Cu_3O_{7-\delta}$ film grown on MgO.

TABLE II. Comparison between experimental determinations of the uniaxial pressure derivatives by different groups. All results are on $YBa_2Cu_3O_{7-\delta}$ except this work which is in $GdBa_2Cu_3O_{7-\delta}$.

	$\frac{dT_c}{dP_a}$ (K/GPa)	$\frac{dT_c}{dP_b}$ (K/GPa)	$\frac{dT_c}{dP_c}$ (K/GPa)
This work	-3.06 ± 0.35	0.38 ± 0.18	3.45 ± 0.43
Meingast *et al.* (Ref. 19)	-1.9	2.2	~ 0
Crommie *et al.* (Ref. 1)	. . .	. . .	0.03–0.1

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