# Pressure dependence of $T_c$ and $H_{c2}$ of $YBa_2Cu_4O_8$

J. J. Scholtz, E. N. van Eenige, R. J. Wijngaarden, and R. Griessen
*Faculty of Physics and Astronomy, Free University, Amsterdam, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands*
(Received 21 June 1991)

We measured resistively the critical temperature $T_c$ and the upper critical field $H_{c2}$ of $YBa_2Cu_4O_8$ up to a pressure of 18 GPa in magnetic fields up to 10 T, using a cryogenic diamond anvil cell. The onset critical temperature is found to increase from 82 K at ambient pressure to 104 K at 8 GPa at the very large rate of 5.5 K/GPa. At higher pressures, $T_c$ first saturates and then decreases with increasing pressure. The upper critical field $H_{c2}(T=0\text{ K})$ is found to strongly decrease up to a pressure of $\sim 8$ GPa; at higher pressures, the rate of decrease as a function of pressure is diminished. The large $\partial T_c/\partial p$ at low pressure in $YBa_2Cu_4O_8$ can be explained by means of a large change of the number of charge carriers $\delta$ in the $CuO_2$ planes as a function of pressure. The volume derivative $\partial \ln\delta/\partial \ln V$ is calculated from our data on $H_{c2}(p)$ and $T_c(p)$. When the pressure decreases, we find a smaller $\partial T_c/\partial p$ and $\partial H_{c2}(T=0\text{ K})/\partial p$ due to an irreversible change of the sample above 20 GPa. When the pressure is increased for the second time, we retain the behavior observed at decreasing pressure. The irreversibility in $T_c$ versus pressure is attributed to an irreversible change in the number of charge carriers.

## INTRODUCTION

In the beginning of research in the field of high-$T_c$ superconductivity it was found by Chu *et al.*,[1] that pressure can be used to increase the critical temperature. They managed to increase the $T_c$ of La-Ba-Cu-O from 32 to 40 K by applying a pressure of 1.3 GPa. This strong pressure dependence of 6.1 K/GPa, one of the largest found for any superconductor so far, led to the discovery of $YBa_2Cu_3O_{7-x}$.[2] The main idea, leading to this important discovery, was that replacement of La by the smaller Y would cause an increase in internal pressure and hence an increase in the ambient pressure $T_c$. Indeed the $T_c$ of $YBa_2Cu_3O_{7-x}$ ($x<0.65$) was high and found to range from 60 to 90 K, depending on the value of $x$. However, these high-$T_c$ values are not only due to a chemical pressure effect, but also to a change in structure. The $T_c$ mentioned for La-Ba-Cu-O is due to the phase $La_{2-y}Ba_yCuO_{4-x}$, which has a single $CuO_2$ plane per unit cell, whereas $YBa_2Cu_3O_{7-x}$ has two such planes. Unfortunately, the replacement of Y by even smaller atoms did not cause an additional increase in $T_c$.

Not only can high-pressure experiments be used to find new materials, they can also be used to test theories: the observed pressure dependences of $T_c$ and $H_{c2}$ must be predicted correctly by any valid theory.

$YBa_2Cu_4O_8$ was originally found as a contaminant in $YBa_2Cu_3O_{7-x}$.[3] This contamination was not a coincidence, because orthorhombic $YBa_2Cu_3O_x$ is thermodynamically unstable with respect to decomposition to $YBa_2Cu_4O_8$ at all temperatures.[4] The crystal structure of $YBa_2Cu_4O_8$ resembles that of $YBa_2Cu_3O_{7-x}$, the difference being a doubling of the so called chains with respect to $YBa_2Cu_3O_{7-x}$, causing the $c$ axis to more than double (see Fig. 1). Due to this double chain all the oxygen atoms have a strong bond with a copper atom, resulting in a fixed and stable oxygen content: the material is stoichiometric. The doubling of the chains also causes the crystal structure to become less orthorhombic, the orthorhombicity of $YBa_2Cu_4O_8$ being half that of $YBa_2Cu_3O_{7-x}$. The decrease of the orthorhombicity

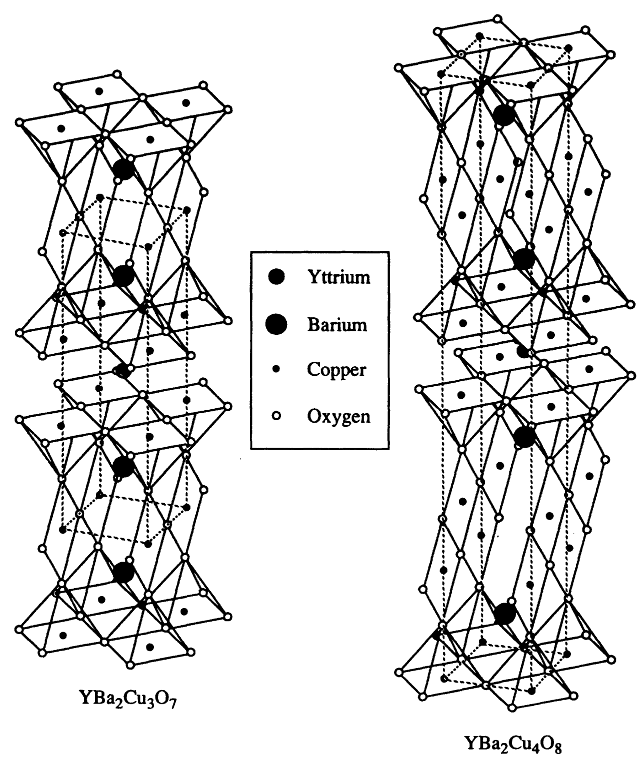


FIG. 1. Crystal structure of both $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$. Note that the chains present in $YBa_2Cu_3O_7$ are doubled in $YBa_2Cu_4O_8$, causing the length of the $c$ axis to more than double.

 

makes the appearance of twin crystals less frequent.

Measurements of the $T_c$ of $YBa_2Cu_4O_8$ as a function of pressure were carried out by Bucher *et al.*,[5] Diederichs *et al.*,[6] and Van Eenige *et al.*[7] They measurements of Van Eenige *et al.* were carried out up to a pressure of 12 GPa. They found a linear increase of $T_c$ with pressure up to $\sim 8$ GPa, with $\partial T_c/\partial p = 5.5$ K/GPa or using $B = (142 \pm 8)$ GPa (Ref. 8): $\partial \ln T_c / \partial \ln V = -9.8$. This large $\partial \ln T_c / \partial \ln V$ is remarkable, considering the large $T_c$ of 80 K, because empirically[9,10] a large $T_c$ nearly always implies a small $\partial \ln T_c / \partial \ln V$. At higher pressures $T_c$ started to saturate as a function of pressure, reaching $T_c = 108$ K at 12 GPa, the highest $T_c$ known for any Y-Ba-Cu-O compound, although very close to the maximum measured by McElfresh *et al.*[11] of 107 K in $YBa_2Cu_3O_7$ at 15 GPa.

So far, high-pressure experiments on $YBa_2Cu_4O_8$ did not include measurements of the critical field $H_{c2}$ as a function of pressure. As we have shown,[12] measurements of $H_{c2}$ as a function of pressure can be used to determine the pressure dependence of the number of charge carriers, which is of major importance for the pressure dependence of $T_c$. Therefore it was of considerable interest to measure $H_{c2}$ for this compound and compare it to similar measurements done on $CaLaBaCu_3O_7$, which has a much smaller $\partial T_c / \partial p$.

In this paper we report measurements on $YBa_2Cu_4O_8$ in magnetic fields up to 10 T and pressures to 18 GPa.

## EXPERIMENTAL TECHNIQUE

The sample studied in this work was synthesized under high oxygen pressure in a high-temperature two-chamber autoclave.[13] A stoichiometric mixture of fine grained $YBa_2Cu_3O_7$ and CuO resulting from the decomposition of oxalates was heated to 980 °C under 21 MPa oxygen for 30 h and then slowly cooled down at a rate of 5 °C/min. From high sensitivity volumetric oxygen analysis[14] it is concluded that the material is nearly stoichiometric with the composition $YBa_2Cu_4O_{7.986 \pm 0.001}$. The $T_c$ of the bulk ceramic is 80 K and thus similar to that of the two phased thin film investigated earlier by Kapitulnik.[15] The low degree of twinning in $YBa_2Cu_4O_8$ makes it possible to accurately determine the *a*-*b* anisotropy by means of x-ray scattering. The orthorhombic lattice parameters are $a = 0.384\,13$ nm, $b = 0.387\,08$ nm, and $c = 2.7240$ nm, with the double (square planar) chains in the *bc* plane.[16]

Magnetic fields for the high-pressure experiments are generated by means of a 12-T Thor-Cryogenics superconducting magnet with a 53-mm room temperature bore and a field homogeneity of $1{:}10^5$ in a 1-cm-diameter sphere.

Pressure is generated by means of a diamond anvil cell[17] made of nonmagnetic stainless steel. This cell can be inserted in the bore of the magnet together with a specially designed optical cryostat enabling optical access to the sample space and allowing experiments in the temperature range 10–300 K. The force mechanism of the cell is situated outside the magnet, thus avoiding large components in the bore.

The diamonds used in this experiment are 16-sided and single beveled, with a bevel angle of 5°, and a culet diameter of 800 $\mu$m. A nonprepressurized stainless-steel gasket is used with an initial thickness of 100 $\mu$m. The gasket hole has a diameter of 300 $\mu$m.

To measure pressure by ruby fluorescence method is used. The pressure is calculated from the shift of the ruby *R*1 line, using the pressure scale of Mao *et al.*,[18] corrected for the temperature dependence of the frequency of the ruby *R*1 line.[19] Ruby chips are homogeneously distributed over the culet, enabling us to measure the pressure gradient over the sample. The pressure which we attribute to a measurement is the maximum pressure measured anywhere on the sample. This maximum pressure is measured in the center of the culet, as anticipated.

The temperature of the sample can be changed continuously down to 10 K by means of a heat exchanger made of sintered copper, which is attached to the bottom of the diamond anvil cell. The estimated uncertainty in temperature determination of the sample (due to small thermal gradients, which are inevitable in such a large cell) imply an uncertainty in $T_c$ of $\pm 0.3$ K. Temperatures are generally above 30 K, thus allowing the use of a platinum resistor, calibrated for magnetic fields up to 10 T.

The resistance of the sample is measured by means of a four-point lock-in technique, which reverses the current every 0.25 sec thus also correcting for thermovoltages. Electrical connections to the sample are made by six gold wires pressed onto the powdered sample, a procedure analogous to that of van Eenige *et al.*[7] We use six gold wires, although only four are needed, to have some redundancy.

In the present experimental set-up no pressure medium is used. We assume however that the sample acts as its own pressure-transmitting medium. In this case this is apparent, because despite the very large $\partial T_c / \partial p$ of 5.5 K/GPa we have a very narrow transition width.

## RESULTS

In this experiment we take as the critical temperature $T_c$ the critical *onset* temperature, because we believe this to be close to the thermodynamic $T_c$. The onset critical temperature is obtained from the intersection of the tangents to the resistivity curve $R(T)$ in the normal state and halfway the transition, respectively (see inset in Fig. 3).

As an example of our measurements of resistance versus temperature we show the data at $p = 10.7$ GPa for several magnetic fields in Fig. 2. The width of the transition increases when the field is increased, mainly as a result of flux flow and flux creep.

In Fig. 3 we show $T_c$ as a function of pressure in zero magnetic field. We find a strong increase of $T_c$ with pressure up to $\sim 8$ GPa after which $T_c$ first saturates and then even decreases. Above 20 GPa the transition suddenly disappeared, possibly indicating a change in the

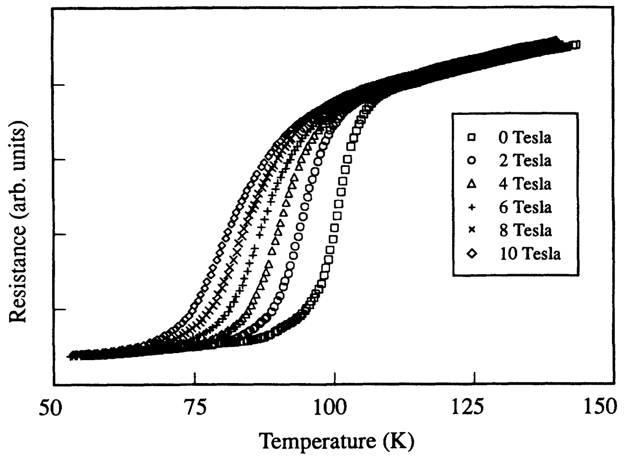


FIG. 2. Resistance as a function of temperature of $YBa_2Cu_4O_8$ at $p = 10.7$ GPa for several magnetic fields.

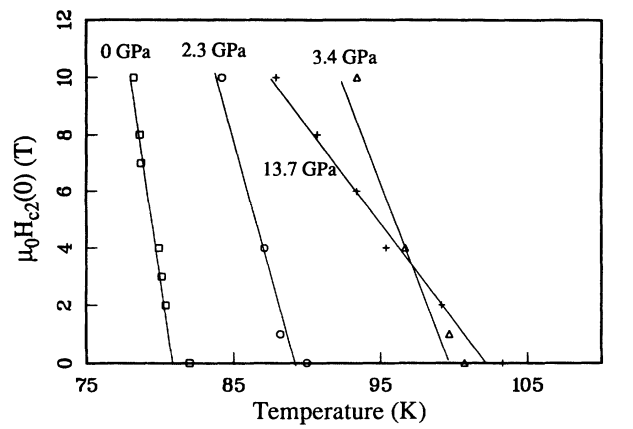


FIG. 4. $H_{c2}$ has a function of temperature for four different pressures.

sample. In our opinion the decreasing pressure measurement should be considered to have taken place on a different sample, due to this transformation at our highest pressure. Interestingly, this "pressure-treated" sample follows upon pressure decrease qualitatively the same curve as the "original" sample upon pressure increase. However, the gradients $\partial T_c/\partial p$ tend to be somewhat smaller. When pressure is increased for the second time we retain the values for $T_c$ at decreasing pressure. Our data during the first increase of pressure for $p < 12$ GPa are fully consistent with those found by Bucher *et al.*[5] and Van Eenige *et al.*[7] It is worthwhile to mention that measurements of $T_c(p)$ with decreasing pressure are not often reported in the literature, because most high-pressure experiments are terminated because of technical breakdown of the resistance measurement.

The upper critical field as a function of temperature, $H_{c2}(T)$, is obtained by measuring the resistive transition as a function of temperature at several magnetic fields at a fixed pressure. As an example we show $H_{c2}(T)$ for four different pressures in Fig. 4. From $H_{c2}$ as a function of temperature, which can only be determined in a small temperature interval near $T_c$ because, compared to the very high $H_{c2}(T=0\text{ K})$ in these materials, the magnetic field at our disposal is relatively limited, we extrapolated our measurements of $H_{c2}$ versus temperature to $T=0$ K. We show the results for $H_{c2}(T=0\text{ K})$ as a function of pressure obtained using the theory by Werthamer, Helfand, and Hohenberg[20] (WHH theory) in Fig. 5. In this theory $H_{c2}(T)$ is completely determined by $T_c$, $\partial H_{c2}/\partial T$ at $T_c$ and a spin-orbit coupling parameter $\lambda_{so}$. Because $\lambda_{so}$ is not known for this compound, we take $\lambda_{so}=2$, a value which is argued by Van Bentum *et al.*[21] to apply to $La_{1.85}Sr_{0.15}CuO_4$.

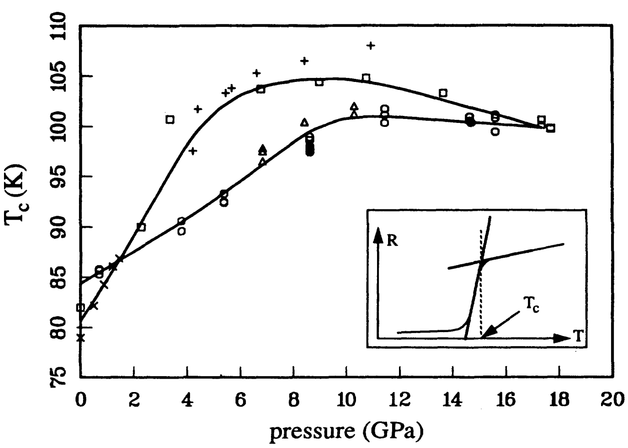


FIG. 3. The onset critical temperature of $YBa_2Cu_4O_8$ as a function of pressure at zero magnetic field. + = Van Eenige *et al.* (Ref. 7), × = Bucher *et al.* (Ref. 5), □ = this work, increasing pressure, ○ = this work, decreasing pressure, △ = this work, increasing pressure for the second time. The inset shows how $T_c$ is defined.

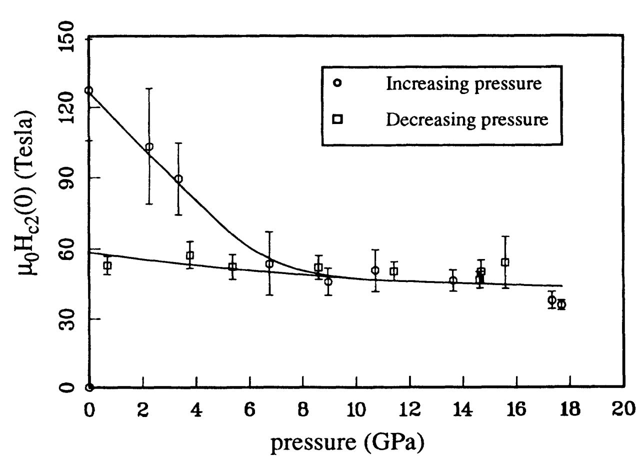


FIG. 5. $H_{c2}(T=0\text{ K})$ as a function of pressure, calculated using the WHH theory with $\lambda_{so}=2$.

TABLE I. Experimental results. The last column is an extrapolation of our measurements using WHH theory with $\lambda_{so}=2$. To determine the values shown for increasing pressure we took the data points at $p<5$ GPa, for decreasing pressure we took the data points at $p<12$ GPa.

| | $\frac{\partial \ln T_c}{\partial \ln V}$ | $\frac{\partial \ln \left[ \frac{-\partial H_{c2}}{\partial T} \Big\|_{T_c} \right]}{\partial \ln V}$ | $\frac{\partial \ln H_{c2}(0)}{\partial \ln V}$ |
|---|---|---|---|
| Increasing pressure | $-8.5\pm0.9$ | $27\pm6$ | $12.4\pm0.9$ |
| Decreasing pressure | $-2.4\pm0.2$ | $3.3\pm0.8$ | $1.0\pm0.8$ |

## DISCUSSION

### $H_{c2}$ as a function of pressure

Although upon decreasing pressure $H_{c2}(p)$ nicely follows the curve for increasing pressure between 8 and 18 GPa, we do speculate that the difference observed below 8 GPa is due to the sample transformation above 20 GPa, already mentioned. In the following analysis we will treat the increasing pressure and decreasing pressure data separately, just as if it were two different samples.

We will now show that from our $H_{c2}(p)$ measurement the variation with pressure in the Fermi velocity and in the charge carrier density can be calculated, pursuing the line of argument outlined in a previous article on $CaLaBaCu_3O_7$.[12] In that paper we have shown that $\partial \ln\delta/\partial \ln V$, which is an important parameter as explained above, can be derived from the volume dependence of the Fermi velocity $\partial \ln v_F/\partial \ln V$. $\partial \ln v_F/\partial \ln V$ can be calculated from our measurements of $T_c$ and $H_{c2}$ as a function of pressure using four different methods. Two of these methods rely on an extrapolation of $H_{c2}$ to 0 K, making use of the empirical relation

$$H_{c2}(T)=H_{c2}(0)\left[1-\left[\frac{T}{T_c}\right]^2\right], \tag{1}$$

or, alternatively, the WHH theory with $\lambda_{so}=2$ to extrapolate our $H_{c2}(T)$ measurements to $T=0$ K and so obtain $\partial \ln H_{c2}(0)/\partial \ln V$, from which we can derive $\partial \ln v_F/\partial \ln V$ using

$$\frac{\partial \ln v_F}{\partial \ln V}=\frac{\partial \ln T_c}{\partial \ln V}-\frac{1}{2}\frac{\partial \ln H_{c2}(0)}{\partial \ln V} \tag{2}$$

while Eq. (1) yields

$$\frac{\partial \ln v_F}{\partial \ln V}=-\frac{1}{2}\frac{\partial \ln \left[ \frac{-1}{T_c}\frac{\partial H_{c2}}{\partial T}\Big|_{T_c}\right]}{\partial \ln V}. \tag{3}$$

The other two methods use relations from both Ginzburg-Landau and BCS theory. For pure superconductors we have

$$\xi(t)=0.74\frac{\xi_0}{(1-t)^{1/2}}, \tag{4}$$

where $\xi$ is the coherence length, $t=T/T_c$, and

$$\xi_0=\frac{hv_F}{2\pi^2\Delta(0)}, \tag{5}$$

where $h$ is Planck's constant, $v_F$ is the Fermi velocity, and $\Delta(0)$ the superconducting gap at $T=0$ K. From Eq. (4) we find

$$\frac{\partial \ln v_F}{\partial \ln V}=-\frac{1}{2}\frac{\partial \ln \left[ \frac{-1}{T_c}\frac{\partial H_{c2}}{\partial T}\Big|_{T_c}\right]}{\partial \ln V}. \tag{6}$$

Using the expression for dirty superconductors

$$\xi(t)=0.855\frac{(\xi_0 l)^{1/2}}{(1-t)^{1/2}}, \tag{7}$$

where $l$ is the mean free path we find

$$\frac{\partial \ln v_F}{\partial \ln V}=-\frac{\partial \ln \left[ -\frac{\partial H_{c2}}{\partial T}\Big|_{T_c}\right]}{\partial \ln V}-\frac{1}{3}. \tag{8}$$

From $\partial \ln v_F/\partial \ln V$ we can calculate the volume dependence of the number of charge carriers using a free electron model, which assumes a cylindrical Fermi surface

$$\frac{\partial \ln\delta}{\partial \ln V}=0.17+\frac{2}{3}\frac{\partial \ln v_F}{\partial \ln V}. \tag{9}$$

The results of our measurements are shown in Table I. All volume derivatives are taken at zero pressure. From these results we have calculated $\partial \ln v_F/\partial \ln V$ and $\partial \ln\delta/\partial \ln V$ as shown in Table II for $p=0$ GPa. From a comparison of the values for $\partial \ln\delta/\partial \ln V$ we see that these values are much more negative than the corresponding values found for $CaLaBaCu_3O_7$.[12] This means

TABLE II. Volume dependence of both the Fermi velocity and the number of charge carriers for four different models.

| Model | Increasing pressure $\frac{\partial \ln v_F}{\partial \ln V}$ | Increasing pressure $\frac{\partial \ln\delta}{\partial \ln V}$ | Decreasing pressure $\frac{\partial \ln v_F}{\partial \ln V}$ | Decreasing pressure $\frac{\partial \ln\delta}{\partial \ln V}$ |
|---|---|---|---|---|
| WHH, $\lambda_{so}=2$ | $-15\pm1$ | $-9.8\pm0.7$ | $-2.9\pm0.4$ | $-1.8\pm0.3$ |
| parabolic | $-18\pm3$ | $-12\pm2$ | $-2.9\pm0.4$ | $-1.8\pm0.3$ |
| GL-BCS clean | $-18\pm3$ | $-12\pm2$ | $-2.9\pm0.4$ | $-1.8\pm0.3$ |
| GL-BCS dirty | $-27\pm6$ | $-18\pm4$ | $-3.6\pm0.8$ | $-2.2\pm0.5$ |

that the number of charge carriers changes much faster with pressure in $YBa_2Cu_4O_8$ than in $CaLaBaCu_3O_7$, thus explaining the larger $\partial T_c/\partial p$ ($p=0$) found in $YBa_2Cu_4O_8$ as compared to $CaLaBaCu_3O_7$ as we will now show.

### $T_c$ as a function of pressure

The behavior of $T_c$ as a function of pressure can be understood in a simple picture, where pressure increases the charge carrier concentration $\delta$ and where $T_c$ is parabolically dependent upon $\delta$. In their review paper Shafer and Penney[22] have demonstrated that for high-temperature superconductors with two $CuO_2$ layers per unit cell, approximately the following relation holds: $T_c \sim T_0 (1-60\delta)$, where $\delta$ is the number of charge carriers per $CuO_2$ unit (i.e., per half a unit cell) and $T_0$ is the maximum $T_c$ observed for a particular compound. In the previous section we have demonstrated (from our $H_{c2}$ measurements), that pressure does increase the charge carrier concentration. It turns out, that this increase is roughly linear at a rate of 0.009 hole/GPa. From this and the Shafer and Penney curve the whole pressure dependence of $T_c$ can be understood.

The fact that pressure can indeed change $\delta$ in $YBa_2Cu_4O_8$ is, for instance, shown theoretically by Yamada *et al.*[23] Using x-ray diffraction measurements they calculated the Madelung energy. They find that when pressure is increased the minimum of the Madelung energy shifts to higher $\delta$. Experimental evidence is found by Furrer *et al.*[24] They show, using inelastic neutron scattering techniques on $ErBa_2O_4O_8$, that $\delta$ increases when pressure is increased from 0 to 1 GPa.

The higher $\partial T_c/\partial p$ ($p=0$) found in $YBa_2Cu_4O_8$ with respect to $YBa_2Cu_3O_7$ is probably due to a larger $\partial\delta/\partial p$ in $YBa_2Cu_4O_8$. To address this point we must realize that $YBa_2Cu_4O_8$ and $YBa_2Cu_3O_x$ are similar in crystal structure, the major difference being the double chains in $YBa_2Cu_4O_8$ as explained above. A larger $\partial\delta/\partial p$ in $YBa_2Cu_4O_8$ could come about through an increased charge transfer from the chains (which are thought to be the charge reservoir) to the $CuO_2$ planes as a function of pressure. One could conjecture that this enhanced charge transfer is the result of a larger shift of the apical oxygen toward the $CuO_2$ plane as a function of pressure. The position of the apical oxygen above the $CuO_2$ plane $[d(\text{Cu(2)-O(1)})]$ both for $YBa_2Cu_3O_x$ and $YBa_2Cu_4O_8$ has been measured as a function of pressure by respectively Jorgensen *et al.*[25] and Nelmes *et al.*[8] Nelmes *et al.* find $\partial[d(\text{Cu(2)-O(1)})]/\partial p = (-1.3\pm0.1)\times10^{-3}$ nm/GPa for $YBa_2Cu_4O_8$, while Jorgensen *et al.* find for $YBa_2Cu_3O_{6.61}$ $\partial[d(\text{Cu(2)-O(1)})]/\partial p = (-2.2\pm0.3)\times10^{-3}$ nm/GPa and for $YBa_2Cu_3O_{6.93}$ $\partial[d(\text{Cu(2)-O(1)})]/\partial p = (-1.1\pm0.4)\times10^{-3}$ nm/GPa. So for $YBa_2Cu_4O_8$ $\partial[d(\text{Cu(2)-O(1)})]/\partial p$ is not much larger than for $YBa_2Cu_3O_x$. However, this enhanced charge transfer could also be caused by the fact that it is *energetically* more favorable for the charge carriers to go to the $CuO_2$ planes in the case of $YBa_2Cu_4O_8$. This could be the result of the difference in crystal structure.

## CONCLUSIONS

We measured the critical temperature and upper critical field of $YBa_2Cu_4O_8$ up to a pressure of 18 GPa in magnetic fields up to 10 T.

From our data we obtained the volume dependence of the Fermi velocity using Ginzburg-Landau or BCS theory. From $\partial \ln v_F/\partial \ln V$ we calculated the volume dependence of the number of charge carriers $\partial \ln\delta/\partial \ln V$ using a free electron model and assuming a cylindrical Fermi surface. We find that $\partial\delta/\partial p$ is much larger for $YBa_2Cu_4O_8$ than for $CaLaBaCu_3O_7$, thus explaining the difference in $\partial T_c/\partial p$, and the maximum in $T_c(p)$ for $YBa_2Cu_4O_8$.

## ACKNOWLEDGMENTS

We are extremely grateful to Dr. J. Karpinski, Dr. E. Kaldis, Dr. S. Rusiecki, and Dr. E. Jilek of the Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule, Zürich, Switzerland for providing us with the high-quality samples on which this work is based. Interesting discussions with Dr. L. F. Feiner are gratefully acknowledged. This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (FOM) which is financially supported by NWO.

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