

VOLUME DEPENDENCE OF HIGH- $T_c$  SUPERCONDUCTORS

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The pressure dependence of  $T_c$  is found to be large for the superconductors (La-Ba-Cu-O, La-Sr-Cu-O) with a relatively low  $T_c$  and small for Y-Ba-Cu-O. The implications of this trend for the theories of high- $T_c$  superconductivity (especially those based on a Hubbard-like model) are discussed.

1. INTRODUCTION

Shortly after the publication of Bednorz and Müller (1) on superconductivity in La-Ba-Cu-O, Chu et al. (2) reported that the  $T_c$  of this compound could be increased from 32 K to 40 K by applying a hydrostatic pressure of 14 kbar. With  $dT_c/dp = 0.64$  K/kbar La-Ba-Cu-O has one of the largest positive pressure dependence measured so far. On the other hand for Y-Ba-Cu-O Hor et al. (3) found a very weak pressure dependence of  $T_c$  up to 19 kbar. This behaviour was confirmed in our laboratory up to 170 kbar (4). For this investigation we used the same diamond-anvil-cell as Hemmes et al. (5) for the high pressure synthesis of superconducting stoichiometric PdH and PdD. The good agreement between the low-pressure and the high pressure data obtained by means of this diamond cell show that the low  $dT_c/dp$  is an intrinsic property of  $Y_1Ba_2Cu_3O_{7-x}$ .

2. TRANSITION

As shown in Fig. 1 both the low and high pressure data exhibit a small increase with pressure for the onset temperature  $T_{co}$  and a small decrease of  $T_{cf}$ , the temperature below which the sample is in the zero-resistivity state. The middle of the transition  $T_m$  is

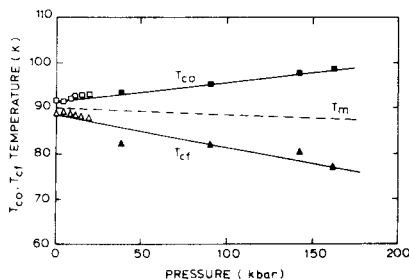


Fig. 1 Pressure dependence of  $T_c$  for  $Y_1Ba_2Cu_3O_{7-x}$

almost pressure independent while the transition width increases with 0.113 K/kbar. This large broadening cannot be explained in terms of pressure gradients present in the diamond anvil cell as  $dT_m/dp \approx 0$ . It is more likely related to the presence of local uniaxial stresses due to the granular nature of the sintered samples used in this work. Using the simple model of Salomons et al. (6) we estimate that for a sample with a density of 80% of the single crystal density, at a pressure of 170 kbar, the c/a-ratio in the uniaxially stressed regions differ by 7.5% from that of the unstressed unit cell. As at 170 kbar the transition width is ~ 19 K we estimate that  $T_c$  increases (or decreases) by 1.3 K for a 1% change in c/a.

3. THEORETICAL MODELS

In Fig. 2 the relative volume dependence of  $T_c$  is shown as a function of  $T_c$ . There is a clear trend which cannot be understood within the classical BCS-theory. Bipolaronic superconductivity could in principle lead to large negative volume derivatives but would also predict large isotope effects in contradiction

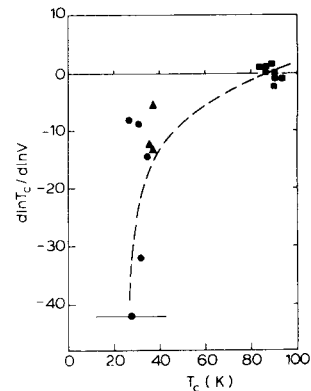


Fig. 2 Volume derivative of  $T_c$  as a function of  $T_c$  (for references see ref. 7)

with available experimental data. The volume derivatives predicted by several theoretical models are discussed in ref. 7.

In the remaining part of this paper we present some implications of the data in Fig. 2 for the theoretical models based on the Hubbard-Hamiltonian,

$$H = \sum_{i,j} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad (1)$$

where  $U$  is the intra-atomic Coulomb integral and  $t_{ij}$  is the hopping integral between sites  $i$  and  $j$ . Depending on the approximations used and the additional terms considered by various authors in the hamiltonian indicated above, different expressions have been proposed so far for  $T_c$ . Some results are clearly incapable of giving large negative  $d\ln T_c/d\ln V$ . To illustrate this point we consider some specific types of expressions for  $T_c$ .

In general,  $T_c$  depends on  $t$ ,  $U$  and the average number  $n$  of electrons (or holes) per site. From dimensional considerations one expects that,

$$T_c = (t^2/U)f(t/U, n) \quad (2)$$

For La-Ba-Cu-O, La-Sr-Cu-O and Y-Ba-Cu-O band structure calculations lead to  $t \approx 0.5$  eV and experiments on copperoxides indicate that  $U \approx 5$  eV. This implies that within the models considered here the variations in  $T_c$  are mainly due to different values of  $n$  in these superconducting materials.

We consider now two special forms of eq. 2:

i) If  $f(t/U, n) = g(t/U)h(n)$ , then neglecting the volume dependence of  $U$  we obtain

$$\frac{d\ln T_c}{d\ln V} = \frac{d\ln t}{d\ln V} \left( 2 + \frac{d\ln g}{d\ln(t/U)} \right) \quad (3)$$

and consequently  $d\ln T_c/d\ln V$  is independent of  $n$  in contradiction with experimental data. This applies, for example, to the two limiting cases of (eqs. 4.21 and 4.22) considered by Robaszkiewicz et al. (8) in their treatment of the generalized periodic Anderson model with strong local attraction. It applies also to models in which  $T_c$  is calculated on the basis of a free Bose gas on a lattice as then  $T_c \propto t n^{-2/3}$ .

ii) If  $f(t/U, n) = g(Uh(n)/t)$  then,

$$\frac{d\ln T_c}{d\ln V} = \frac{d\ln t}{d\ln V} \left( 2 + \frac{d\ln g}{d\ln(Uh(n)/t)} \right) \quad (4)$$

and  $d\ln T_c/d\ln V$  may strongly be dependent on the band filling. One of the model which leads to an expression of the form indicated here is that of Cyrot (9) for which  $h(n) = n$ . It is then possible to express  $d\ln T_c/d\ln V$  in terms of  $T_c$  as shown in Fig. 3. This universal curve

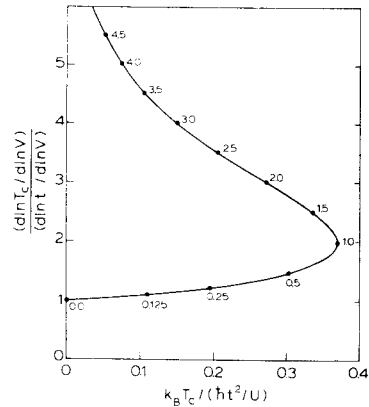


Fig. 3 Prediction of a Hubbard-like model (9).  $Un/t$  is indicated at various points

shows that large (and negative, because of  $d\ln t/d\ln V \approx -2$ )  $d\ln T_c/d\ln V$  values are possible for superconductors with low  $T_c$  when the concentration of carriers is such that  $Un/t \ll 1$ .

#### 4. CONCLUSIONS

We have shown that the general trend observed for  $d\ln T_c/d\ln V$  may be used to test the validity of theoretical models. In this context it is highly desirable to investigate La-Ba-Cu-O and La-Sr-Cu-O single crystals as these systems have very large volume derivatives of  $T_c$

- (1) J.G. Bednorz and K.A. Müller, Z. Phys. **B64** (1986) 189
- (2) C.W. Chu, P.H. Hor, R.L. Meng, L. Gao, Z.I. Huang and Y.Q. Wang, Phys. Rev. Lett. **58** (1987) 405
- (3) P.H. Hor, L. Gao, R.L. Meng, Z.J. Huang, Y.Q. Wang, K. Forster, J. Vassiliou, C.W. Chu, M.K. Wu, J.R. Ashburn and C.J. Torng, Phys. Rev. Lett. **58** (1987) 911
- (4) A. Driessen, R. Griessen, N. Koeman, E. Salomons, R. Brouwer, D.G. de Groot, K. Heeck, H. Hemmes and J. Rector, Phys. Rev. **B36** (1987) 5602
- (5) H. Hemmes, A. Driessen, R. Griessen, R.J. Wijngaarden, J. Caro, S. Radelaar and M. Gupta, this conference
- (6) E. Salomons, H. Hemmes, J.J. Scholtz, N. Koeman, R. Brouwer, A. Driessen, D.G. de Groot and R. Griessen, Physica **145B** (1987) 253
- (7) R. Griessen, Phys. Rev. **B36** (1987) 5284
- (8) S. Robaszkiewicz, R. Micnas and J. Ranninger, Phys. Rev. **B36** (1987) 180
- (9) M. Cyrot, Solid State Commun. **62** (1987) 821