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## Hg,Re-1223 system: $T_c$ dependence on hydrostatic pressure and thermopower measurements

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### Abstract

Samples of  $\text{Hg}_{1-x}\text{Re}_x\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  superconductor (Hg,Re-1223) were prepared with varying rhenium (Re) nominal content ( $0.15 \leq x \leq 0.20$ ) in order to study the changes on the critical temperature under external hydrostatic pressure. At low pressures the  $\partial T_c / \partial P$  depends strongly on the Re doping and the sample with  $x = 0.18$  shows the largest measured value. However, thermopower measurements reveal that there is no significant change in the numbers of carriers for the different Re content ( $0.15 \leq x \leq 0.20$ ) meaning that  $\partial T_c / \partial n$  is the same for all samples which, in principle, have an optimal oxygen content. Taking into account that  $dT_c/dP = \partial T_c^i / \partial P + [\partial T_c / \partial n][\partial n / \partial P]$  and  $\partial T_c / \partial n$  does not depend on the Re content we can argue that the intrinsic term ( $\partial T_c^i / \partial P$ ) determines the influence of external hydrostatic pressure on  $T_c$ . © 2001 Elsevier Science B.V. All rights reserved.

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### 1. Introduction

After the discovery of high temperature superconductivity [1], the highest  $T_c$  of 134 K (ambient pressure) was recorded in the new family of mercury-based superconductors for  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  [2] compounds. The rhenium (Re) substitution at the Hg site have improved the preparation of (Hg,Re)- $\text{Ba}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_y$  in a quartz tube method [3].

Gao et al. [4] have found that  $T_c$  increases under pressure ( $P$ ), in the range of 0.0–2.0 GPa with a rate of 1.7 K/GPa, a behavior which was assumed to be typical of an underdoped sample. Other measurements with optimally doped samples confirmed that  $T_c$  increases with a lower rate, reaching 153 K at 15 GPa [5] and the maximum  $T_c$  as function of pressure ( $T_c(P)$ ) rises to 164 K at 30 GPa [6]. A linear  $\partial T_c / \partial P$  dependence close to 4 K/GPa for the optimally doped samples was found in the low pressure range 0.0–1.0 GPa [7].

The Re substitution provides a reduction of cell volume (chemical pressure), but the effect on  $T_c$  is distinct of an external hydrostatic pressure [8]. It is

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now generally accepted that an applied external pressure  $P$  induces a charge transfer (PICT) to the  $\text{CuO}_2$  planes, which leads to a change in  $T_c$  [9]. The derivative  $\partial T_c/\partial n$  ( $n$  being the charge density or hole concentration) may be estimated phenomenologically through the  $T_c$ - $x$ - $n$  diagram for  $P = 0$ , which satisfies an inverse *universal* parabolic behavior given by:

$$T_c(n) = T_c^{\max}[1 - \eta(n - n_{\text{op}})^2] \quad (1)$$

where  $T_c^{\max}$  is the maximum  $T_c(n)$  at the optimal doping  $n_{\text{op}}$ . However, since  $T_c$  can increase above the optimal doping  $T_c^{\max}$ , it is clear that another pressure effect, probably due to structural changes, is present and it is known as *intrinsic effect*. Thus one assumes, for a given compound with a charge density or hole concentration  $n$ , an expansion of  $T_c(n, P)$  in powers of  $P$  [10]. The linear coefficient (on the pressure) is given by:

$$dT_c/dP = \partial T_c^i/\partial P + [\partial T_c/\partial n][\partial n/\partial P] \quad (2)$$

where  $\partial T_c^i/\partial P$  is the intrinsic term and  $[\partial T_c/\partial n] \times [\partial n/\partial P]$  is attributed to PICT. Both  $\partial T_c^i/\partial P$  and  $\partial n/\partial P$  depend on the given compound. There is no theory about the origin of the intrinsic term, despite some attempts have been made to relate it with changes in the phenomenological attractive potential of an extended Hubbard model [10].

## 2. Experimental procedures

In our previous paper [8], the synthesis of the  $\text{Hg}_{1-x}\text{Re}_x\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  samples with ( $0.15 < x \leq 0.20$ ) was optimized. The X-ray powder diffraction and magnetic characterization of the samples reveals a unique phase with  $n = 3$  [8]. The measurement of resistance under pressure was performed in a BeCu piston-cylinder cell [8]. The superconducting transition temperature  $T_{\text{cd}}$  was determined from the variation of resistance  $R$  with temperature for all the pressure values.

The thermopower measurements were carried out at constant temperatures [11]. The temperature gradients  $\nabla_x T$  of the order of 0.5 K/mm were applied using a small heating resistance mounted on

the top of the samples, and the  $\nabla_x T$  were measured by a calibrated AuFe-chromel thermocouples.

## 3. Results and discussion

The samples  $\text{Hg}_{1-x}\text{Re}_x\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ ,  $x = 0.15, 0.18$ , and  $0.20$ , have nearly the same  $T_c \approx 133$  K as the pure optimal doped Hg-1223 [8], which it is in agreement with the thermopower measurements (see Fig. 1). Fig. 1 seems to indicate that the addition of Re does not produce a detectable charge variation (or hole content). The thermopower data have indicated to be all Re content samples optimal doped ( $S(290 \text{ K}) \approx 3.0 \mu\text{V/K}$ ) [12]. Therefore, it can be assumed that the Re doped compounds have the same  $P = 0$  parabolic  $T_c(n)$  of the pure Hg-1223 and consequently the same small  $\partial T_c/\partial n$ .

On the other hand, Fig. 2 shows an unusual high  $dT_c/dP = 4.8(2) \text{ K/GPa}$ , at low pressures for our Re 18% ( $x = 0.18$ ) sample (starting at 133 K), which is about three times higher than the value measured for optimal doped Hg-1223 (Gao et al. [6]). Since our  $\text{Hg}_{1-x}\text{Re}_x$ -1223 compound is optimal doped, which is assured by its high  $T_c \approx 133$  K [8] and the thermopower measurements (Fig. 1), the second term in the right hand side of Eq. (2) is zero or very small. This fact implies that the high  $dT_c/dP \approx 4.8(2) \text{ K/GPa}$  is due almost entirely to the intrinsic term.

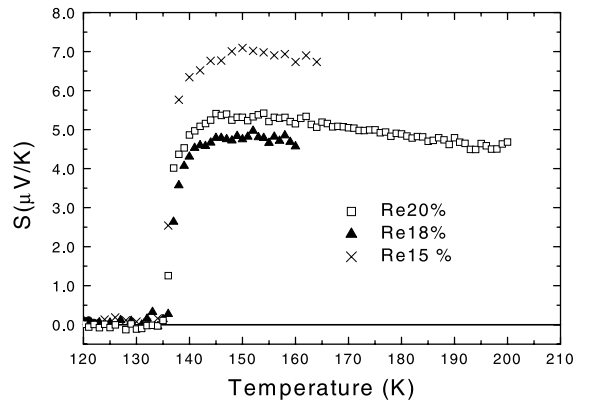


Fig. 1.

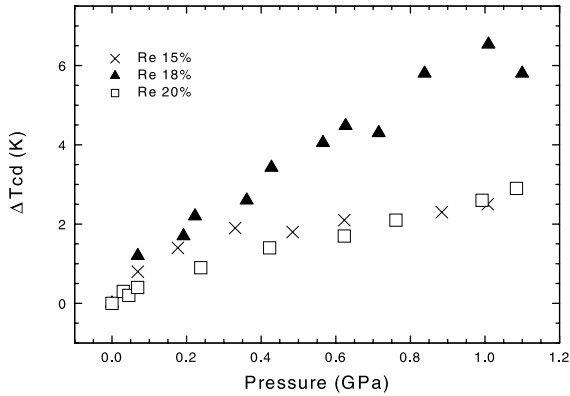


Fig. 2.

#### 4. Conclusions

Based on the analysis above, it can be concluded that the chemical pressure made by the Re doping produces modification that, under hydrostatic pressure, gives a very large intrinsic term. The sample with Re 18% ( $x = 0.18$ ) content has the maximum variation of  $T_c$  (4.8(2) K/GPa) with external hydrostatic pressure, determined from AC resistance. The thermopower measurements have shown that all samples  $\text{Hg}_{1-x}\text{Re}_x\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  ( $x = 0.15, 0.18, \text{ and } 0.20$ ) have the same carrier number (optimal doped). Since  $\partial T_c / \partial n$  is very small for all the samples, the non-equivalent behavior under external hydrostatic pressure, among

samples with different Re content, can be related to the *intrinsic* term and the PICT term of Eq. (2).

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