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Fluorinated Hg-1223 under pressure: the ultimate T_c of the cuprates?

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Abstract

High pressure experiments have revealed that T_c is affected by two main pressure-dependent parameters: the doping level of the CuO_2 planes and by an intrinsic factor. The origin of the intrinsic factor is still unclear as, depending on the experiment, it is associated with the reduction of the c or the a lattice parameters. F incorporation into the Hg-1223 structure yields an enhancement of T_c up to a susceptibility onset of 138 K, mainly related to a compression of the a crystallographic axis. We have obtained a new high T_c record (166 K ± 1.5 K) by applying pressure (23 GPa) in the fluorinated Hg-1223 superconductor optimally doped. T_c increases with increasing pressure, reaching different maximum values, depending on the F doping level, and decreases for a further increase of pressure. This saturation of T_c may be the highest T_c that can be obtained for the cuprates, considering the particular structural characteristics of this system.

Keywords: Pressure effects; Cuprates; Fluorinated Hg-1223

Many efforts have been devoted in order to understand which are the relevant structural parameters that determine the superconducting transition temperature (T_c) of cuprates. There is a clear consensus, based on structural studies and their correlation with the appearance of superconductivity, that the highest T_cs can be reached if $n = 3$ flat CuO_2 planes and small Cu–O in plane distances ($d_{Cu-O} = a/2$ for flat planes) can be achieved [1]. Besides, the CuO_2 planes should reach an optimum doping ($n_{op} = 0.16$), as underdoped and overdoped samples present a lower T_c, in agreement with an empirical parabolic law [2] between T_c and the doping level n ($T_c(n) = T_c^M[1 - \beta(n_{op} - n)^2]$, with $\beta > 83$).

The fluorinated Hg-1223 compound (Hg-1223F) present all this advantages as a small d_{Cu-O} distance can be achieved with a small CuO_2 plane buckling [3]. This compound reaches a maximum susceptibility onset (T_c^{zac}) of 138 K for a d_{Cu-O} distance where the oxygenated samples show a reduction of their T_c.

We have learnt from high pressure experiments, particularly in the Hg-based cuprate superconductors, that T_c increases with increasing pressure following a quadratic dependence which depends on the doping level of the sample (n), on the pressure-induced charge transfer (dn/dP) and on an intrinsic factor (dT_c^i/dP). In this phenomenological model, pressure increases linearly the doping level of the CuO_2 planes and also increases T_c^M which is related to the intrinsic term. It can be shown that [1]:

$$T_c(P) = T_c + \{dT_c^i/dP + \beta(n_{op} - n)T_c^M dn/dP\}P - \beta T_c^M (dn/dP)^2 P^2 \quad (1)$$

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Resistance measurements as a function of temperature were performed on samples prepared as in Ref. [3], with different fluorine contents labelled #1 ($a = 3.8496$ Å, T_c^{ac} 138 K) and #2 ($a = 3.8536$ Å, T_c^{ac} 135 K), following a conventional 4 terminal DC technique, applying pressures up to 30 GPa by using sintered diamond anvils in a Bridgman setup and steatite as pressure medium.

The effect of pressure on a normalized resistance can be observed in Fig. 1 for sample #1. Similar results are obtained for samples with other F content. Zero resistance is attained at low temperatures 10–60 K, depending on the quality of the intergrain coupling of these ceramic samples. Using the temperature derivative of the resistance we can define the onset critical temperature (T_{co}), where derivative departs from its normal behavior, and a peak transition temperature (T_{cp}) determined by the peak of the derivative (see the inset of Fig. 1). The former criterion, usually dominated by thermal fluctuations, is the same used in Ref. [4] and corresponds to the formation of small superconducting droplets, while the later is mostly related to the appearance of a bulk superconductivity.

Pressure increases both T_{co} and T_{cp} following a parabolic law, as can be depicted in Fig. 2. The obtained values for the transition temperatures are clearly higher than the ones reported for the non-Fluorinated Hg-1223 samples. In particular, a T_{co} of 166 K, to our knowledge the highest ever reported, is obtained for sample #1 at 23 GPa. As can be observed in Fig. 2 a good fit with Eq. (1) is obtained for $T_{co}(P)$ (solid line). In order to obtain reasonable parameters (both samples shown here are near the optimum doping, large variations of dT_c^i/dP are not expected) it was necessary to consider different T_c^M depending on F content, which reflects the increase of T_c produced by F incorporation. We effectively obtained that both samples are very near the optimum doping ($n = 0.155$), with dT_c^i/dP 1.5 K/GPa, and with $(dn/dP)^{\#1} > 1.7 \times 10^{-3}$ holes/GPa and $(dn/dP)^{\#2} > 1.6 \times 10^{-3}$ holes/GPa, which are nearly the same values than for optimally oxygenated Hg-1223.

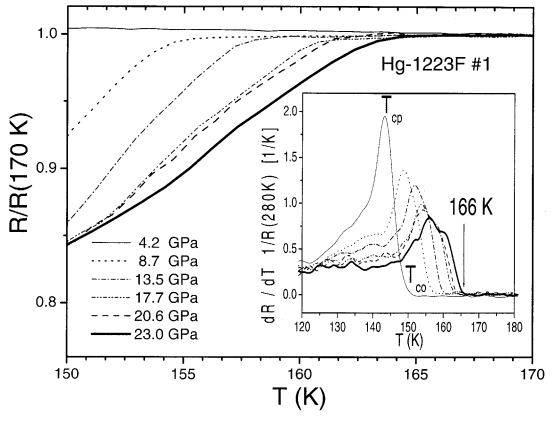

Fig. 1. Normalized resistance of sample #1 as a function of temperature at different pressures. The inset shows the temperature derivative of the resistance which was used to determine T_{co} and T_{cp}.

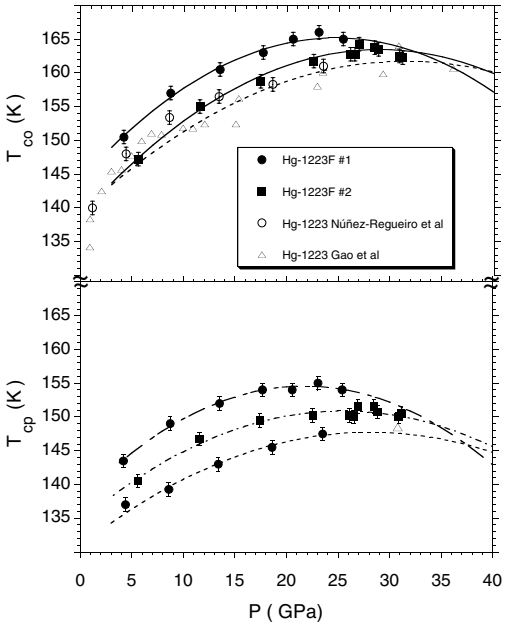

Fig. 2. Pressure dependence of T_{co} and T_{cp}. Solid lines are a fit of T_{co} using the expression (1). Dashed lines are guides to the eye.

This small but easily noticeable increase of dn/dP for sample #1 (as the pressure needed to obtain a maximum T_c decreases) may indicate the proximity of a sudden change of this parameter for further doping, as was observed from optimally to highly oxygenated Hg-1201 samples [5]. Thus, a large overdoping of the CuO_2 planes can be expected for a further chemical compression of the Hg-1223F structure. The overdoping and the chemical difficulties to produce small d_{Cu-O} without increasing the buckling of the CuO_2 planes should be overcome in order to obtain higher T_cs than the obtained for the Hg-1223F compound under pressure.

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