

# High pressure studies of high $T_c$ superconductors in A–Cu–O system: The role of apical oxygen

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## Abstract

We address the role of apical oxygen that plays the bridges linking charge reservoir and  $\text{CuO}_2$  conducting layers for high  $T_c$  superconductors (HTS). The presentation will introduce our recent work on high pressure synthesis of novel superconductors in the A–Cu–O simple system by tuning apical oxygen content and arrangement (A represents alkaline earth element). The topic will focus on 214 type compounds highlighting apical oxygen as a novel doping mechanism in oxychloride HTS, or dopant at a partially occupied apex, especially the unusual enhancement of  $T_c$  up to 95 K in a monolayered HTS by improving ordering state.

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## 1. Introduction

Since the historical discovery of high  $T_c$  superconductors (HTS) of cuprate oxide in 1986 by Bednorz and Muller [1], a lot of HTS compounds have been synthesized [2–18]. Crystallographically HTS consists of periodic stacking of charge reservoir and  $[\text{CuO}_2]$  conducting layers, respectively. They are usually bridged via apical oxygen as shown in Fig. 1 [1–6]. Charge reservoir is responsible for generating carriers to the  $[\text{CuO}_2]$  superconducting plane through an appropriate doping. According to the geometric relationship with the  $[\text{CuO}_2]$  plane, the charge reservoir block can be further classified into two sub blocks: the nearest neighbor to  $[\text{CuO}_2]$  plane; and the next nearest neighbor to  $[\text{CuO}_2]$  plane as shown in Fig. 2. The oxygen which connects the nearest neighbor charge reservoir layer in turn is named “apical oxygen” since it is located at the apical position relative to the  $[\text{CuO}_2]$  plane (this is validate to p type HTS). Tuning the oxygen content at the charge reservoir layer is the most fundamental chemical doping mechanism of HTSs [2].

It is noted that most of oxygen doping are taking place at the next nearest neighbor charge reservoir block. One can expect that  $T_c$  will be even more sensitive to the oxygen evolution at the nearest charge reservoir block via the apical bonding nature [3,6,11]. It was found that the distance of apical oxygen with the  $[\text{CuO}_2]$  plane has an essential effect on the hopping integral  $t'$  between the second nearest Cu–Cu within the  $[\text{CuO}_2]$  plane [6], and consequently it is crucial to modify  $T_c$  value of p-type HTS. However almost all typical HTSs have a fully occupied oxygen site, leaving the apical oxygen amount stoichiometrically unchangeable. Searching compounds with tunable apical oxygen content is thus of fundamental physical interests in terms of deeply understanding the unknown doping effects of oxygen at the nearest charge reservoir block.

Since HTS is of perovskite crystal nature [2,3], high pressure is thus thermodynamically favorable to stabilize novel HTSs [11]. High oxygen pressure can effectively modulate the copper valence. The obvious merit of high pressure is to build up novel HTS, which otherwise will be hard to approach in ambient conditions. On the other hand, the positive pressure effect on  $T_c$ , such as the record 160 K  $T_c$  of Hg-1223 above 20 GPa will shed lights on pursuing new HTS with higher  $T_c$  as well as solving mechanism puzzle. Here we will briefly introduce our recent

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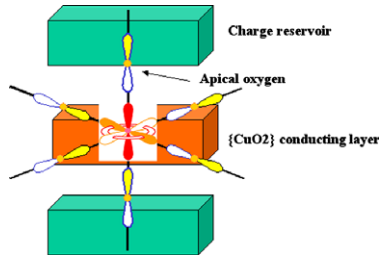


Fig. 1. A schematic view of the crystal structure of HTS, being composed of the charge reservoir and the [CuO<sub>2</sub>] conducting block, which are usually connected via apical oxygen.

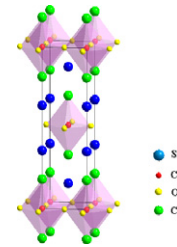


Fig. 3. The crystal structure of Sr<sub>2</sub>CuO<sub>2</sub>Cl<sub>2</sub>.

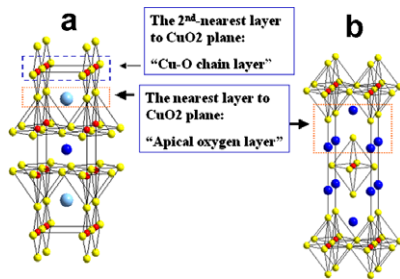


Fig. 2. The charge reservoir layer can be further viewed into the nearest layer and the 2nd nearest layer with respect to [CuO<sub>2</sub>] plane as shown for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (a). The oxygen at the nearest layer is named “apical oxygen” since it is located at the apex site of the [CuO<sub>2</sub>] plane. Crystallographically, 214 (i.e., 0201) type only has the nearest charge reservoir layer as indicated for La<sub>2</sub>CuO<sub>4</sub> (b).

research progress on the high pressure studies of high  $T_c$  superconductors in A–Cu–O system (A is alkaline earth element) highlighting the role apical oxygen as a dopant mechanism in 0201 type Sr<sub>2</sub>CuO<sub>2+δ</sub>Cl<sub>2-x</sub> series, the ordering effects to enhance  $T_c$  up to 95 K in Sr<sub>2</sub>CuO<sub>3+δ</sub>.

## 2. The “apical oxygen doping” to introduce p type superconductivity

So far there are two major types of hole carrier doping mechanisms for a pure oxide HTS. One is to apply hetero-valence metallic ions substitution as very well established in (La<sub>2-x</sub>Sr<sub>x</sub>)CuO<sub>4</sub>. The other is to tune oxygen nonstoichiometry as classically performed for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> or other 12( $n-1$ ) $n$  or 22( $n-1$ ) $n$  homologous series. But those oxygen modifications all happen in the 2nd nearest charge reservoir layer. The stoichiometric oxychloride A<sub>2</sub>CuO<sub>2</sub>X<sub>2</sub> (A = Ca, Sr) is an antiferromagnetic insulator similar to that of La<sub>2</sub>CuO<sub>4</sub> as shown in Fig. 3. This affinity makes A<sub>2</sub>CuO<sub>2</sub>X<sub>2</sub> (A = Ca, Sr) favorable prototype candidates for new HTS. Considering the crystal structural characteristics, we established a new hole doping mechanism to induce carriers in oxyhalide cuprates. The doping is realized through a heterovalent anion substitution, i.e., a partial replacement of monovalent halide with divalent oxygen at the apical site. Since all Cl ions reside at the apex anion position, it is called “apical oxygen doping” [9], to

address the doping nature that introduces apical oxygen otherwise absent into the pristine A<sub>2</sub>CuO<sub>2</sub>X<sub>2</sub>.

The apical oxygen doping was first realized in the double-[CuO<sub>2</sub>]-layer compound (Sr, Ca)<sub>3</sub>Cu<sub>2</sub>O<sub>4+δ</sub>Cl<sub>y</sub> [9,10] which is isostructural to (La, Sr)<sub>2</sub>CaCu<sub>2</sub>O<sub>6</sub>.

Using high pressure synthesis, we also found a new homologous series (Sr,Ca)<sub>n+1</sub>Cu<sub>n</sub>O<sub>2n+δ</sub>Cl<sub>2-x</sub>, or termed as Cl- $''02(n-1)n''$ . Cl-02( $n-1$ ) $n$  crystallizes into tetragonal phase with space group I4/mmm. The unique crystal structure of Cl-02( $n-1$ ) $n$  oxyhalide cuprates enables more to modify the apical oxygen amount, studying its effects on superconductivity.

## 3. The Sr<sub>2</sub>CuO<sub>2+δ</sub>Cl<sub>2-x</sub> superconductor induced by “apical oxygen doping”

Using apical oxygen doping, we recently have been able to get the new Sr<sub>2</sub>CuO<sub>2+δ</sub>Cl<sub>2-x</sub> superconductors using high pressure synthesis [13]. Bulk superconductivity was realized with  $T_c$  up to 30 K.

The sample was synthesized under high pressure high temperature at 5 GPa and 1000 °C. Fig. 4 shows the temperature dependence of the DC magnetic susceptibility of

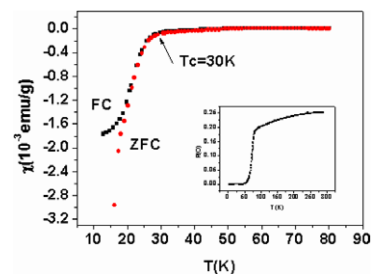


Fig. 4. The superconducting transition of Sr<sub>2</sub>CuO<sub>2+δ</sub>Cl<sub>1.2</sub>.

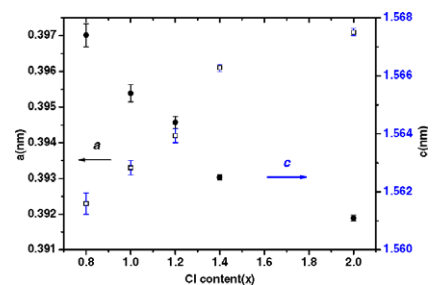


Fig. 5. The change of lattice parameters as a function of apical oxygen doping in Sr<sub>2</sub>CuO<sub>2+δ</sub>Cl<sub>2-x</sub>.

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