Universal Relationship between T_c and the Hole Content in p-Type Cuprate Superconductors

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A critical analysis of experimental data reveals a universal relationship between $T_c/T_{c,\max}$ and hole content among the p-type high-T_c cuprate superconductors. Each individual compound is characterized by the value of its maximum T_c, $T_{c,\max}$, while the variation of $T_c/T_{c,\max}$ with the hole content is independent of the compound considered. The universal curve is characterized by a plateau, rather than a parabola, with sharp bends at both sides. The T_c versus hole content curve has a close relation to hole contents determined by ordered arrangements of holes in the two-dimensional CuO_2 layer.

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The high-T_c cuprate superconductors constitute a special group of materials that share many common features among one another in crystal structures and properties. One of the characteristic structural features is that they have layered crystal structures with the CuO_2 layer as the most essential structural unit. It has now been well established that charge carriers (holes for p type) are mainly confined to the two-dimensional (2D) CuO_2 layers of the essential structural units [1].

Because of the common features among the high-T_c superconductors, it is suspected that these high-T_c superconductors share common superconducting characteristics. Earlier, Uemura *et al.* [2] pointed out the existence of a universal correlation between T_c and n_s/m^* (charge carrier density over effective mass) in the high-T_c cuprates. Other than this, no universal relations have been known. Here, we report a universal relation between T_c and the hole content (to be defined later) in the CuO_2 layer among high-T_c cuprates if T_c is normalized with respect to the $T_{c,\max}$ of each particular series of cuprates.

The high-T_c cuprate superconductors have been known to have a strong dependence of T_c on the hole content and a certain similar trend between T_c and the hole content exists in these individual series of compounds [3,4]. Beyond this, it has been difficult to draw any general conclusions despite a large number of works on a series of high-T_c cuprate superconductors which range from $La_{2-x}Ba_xCuO_4$ with $T_{c,\max}\simeq 40$ K [5] to Tl-based compounds with $T_{c,\max}\simeq 125$ K [6]. The lack of understanding of the general correlation between T_c and hole content in the high-T_c cuprate superconductors can be partly attributed to the relatively large scatter of experimental data due to the difficulties in dealing with ceramic samples of cuprate superconductors which frequently exhibit a wide range of nonstoichiometry. In addition, because of the complication of phase stability, etc., many high-T_c cuprates have only been studied over a limited range of hole content. In fact, as far as the T_c versus hole content relationship is concerned, the $La_{2-x}Sr_xCuO_4$ (2:1:4) system [7] is the only one that has been comprehensively studied over both the underdoped and overdoped regions of hole content. Such difficulties have been preventing us from reaching any conclusion about the general correlation between T_c and hole content based on the results from studying a single compound.

We have made a critical and extensive analysis of a large amount of existing experimental data of several representative high-T_c cuprates. Based on this study, we selected a number of typical works which we believe to be reliable. The selection has been limited to the p-type cuprate superconductors. The result of the study shows that a universal relationship exists between the normalized T_c and the hole content. Our analysis utilizes the common features of high-T_c cuprates mentioned above. Here, τ_c ($\equiv T_c/T_{c,\max}$) is used to indicate the normalized T_c of a compound. The hole content, p_{sh}, is defined as the number of holes per CuO_2 unit in the 2D CuO_2 layer. When there are more than one CuO_2 layers in a structural unit of a compound, the distribution of holes is assumed to be uniform among the available CuO_2 layers. If the charge reservoir [8] (denoted by Φ) which controls the hole content is defined as a structural unit that is composed of any layer but the CuO_2 layer(s), then each structural unit of the high-T_c cuprate superconductors can be written symbolically as $\Phi(CuO_2)_n$, where n indicates the number of the CuO_2 layers in a structural unit. From the symmetry of the structure, the uniform distribution of holes among CuO_2 layers is guaranteed for $n=1$ and 2, though it is not as clear [9] for $n\geq 3$. The use of the symbol $\Phi(CuO_2)_n$ to indicate high-T_c cuprate superconductors thus makes the relationship between the charge carrier and the charge reservoir much clearer. It also underlines the difference as well as the similarities between cuprate superconductors.

The result of the analysis is shown in Fig. 1. It is clearly seen that a universal relationship exists between τ_c and the hole content p_{sh}. Selected works for four important superconducting systems such as $YBa_2Cu_3O_{7-\delta}$ (1:2:3) [10], $La_{2-x}Sr_xCuO_{4+\delta}$ (2:1:4) [7], and $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4+\delta}$ [11-13], $Tl_mBa_2Ca_{n-1}Cu_n$-$O_{2n+2+m+\delta}$ [14-16] ($m=1$ or 2) are shown. It can be seen that all results are represented by a single curve, including the single ($n=1$), double ($n=2$), or triple ($n=3$) layered compounds in Bi- and Tl-based cuprates.

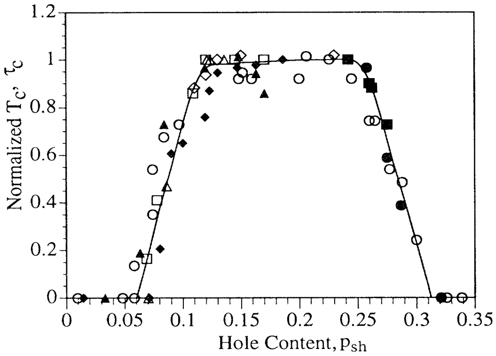

FIG. 1. The universal relationship between $\tau_c = T_c/T_{c,\max}$ and p_{sh} for the cuprate superconductors. The data are from, ◆, Tallon [10] (1:2:3); □, Manthiram and Goodenough [11] (Bi2212); △, Tarascon *et al.* [12] (Bi2212); ◇, Hattori, Nakamura, and Ogawa [13] (Bi2223); ○, Torrance *et al.* [7] (2:1:4); ●, De Leeuw *et al.* [14] (Tl2201); ■, Nakajima *et al.* [15] (Tl1212); and ▲, Gopalakrishnan *et al.* [16] (Tl1212). The solid line is a guide to the eye.

In particular, it should be emphasized that even the 1:2:3 compound, which has been well recognized as having two distinct plateaus of T_c at 90 and 60 K in relation to oxygen content, also fits well into the universal relationship [10] when normalized T_c is plotted against the hole content. In fact, the similarity between the 2:1:4 and 1:2:3 compounds with respect to the relationship of T_c with hole content has been duly pointed out by Tallon [10]. For the Tl-based compounds, it should be pointed out that a reliable correlation between T_c and hole content was not made until the work of Gopalakrishnan *et al.* [16] because of experimental difficulties in determining the hole content. Concerning the results of De Leeuw *et al.* [14] and Nakajima *et al.* [15] shown in Fig. 1, the values of the hole content were derived from other relevant experimental data given in Refs. [14] and [15], respectively. The values of $T_{c,\max}$ for each of the compounds shown in Fig. 1 are listed in Table I.

The universal relation in Fig. 1 is characterized by a sharp increase of τ_c with hole content in the range $0.06 < p_{sh} < 0.12$, a saturation of τ_c in the range $0.12 < p_{sh} < 0.25$, and a rapid decrease of τ_c in the range $0.25 < p_{sh} < 0.31$. The appearance of a plateau is important

TABLE I. Values of $T_{c,\max}$ for some selected high-T_c cuprate superconductors.

Compounds	$T_{c,\max}$ (K)
$La_{2-x}Sr_xCuO_{4+\delta}$	37
$YBa_2Cu_3O_{7-\delta}$	92
$Bi_2Sr_2CaCu_2O_{8+\delta}$	85
$Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$	110
$TiBa_2Ca_{1-x}Y_xCu_2O_7$	85
$Tl_2Ba_2CuO_{6+\delta}$	90

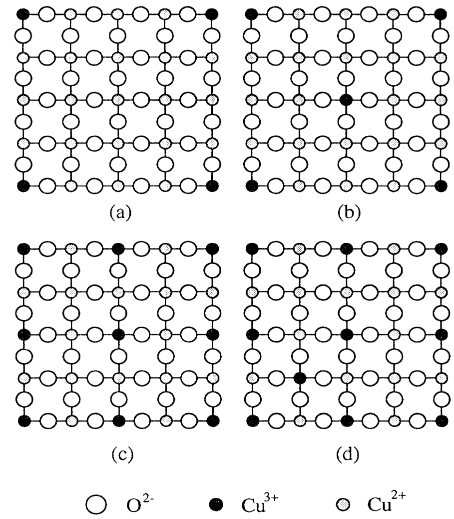

FIG. 2. Atomic arrangement in the CuO_2 layer in the cuprate superconductors. The hole contents illustrated in this figure correspond to (a) $p_{sh}^{cal}=0.0625$, (b) $p_{sh}^{cal}=0.125$, (c) $p_{sh}^{cal}=0.25$, and (d) $p_{sh}^{cal}=0.3125$. For convenience of graphic illustration, holes are put on copper sites.

since it contradicts the previously suggested parabolic relationship between T_c and hole content [4]. The existence of a plateau can also be seen in many other experimental results, in particular, in the universal correlation between T_c and n_s/m^* reported by Uemura *et al.* [2].

The universal relationship in Fig. 1 is also characterized by sharp bends at $p_{sh} \approx 0.06$, 0.12, 0.25, and 0.31. These critical values of hole content correspond to the onset of superconductivity from the antiferromagnetic insulating state, the start and the end of the T_c plateau, and the disappearance of superconductivity to a normal metallic state, respectively. It may be important to point out that these four values of hole content are related to the ordered arrangements of holes in the CuO_2 layer as depicted in Fig. 2. For example, if R_{nn} denotes the distance between two nearest neighboring (nn) holes on the square-planar CuO_2 layer, then a hole content $p_{sh}^{cal}=0.0625$, which is in excellent agreement with $p_{sh} \approx 0.06$ in Fig. 1, can be calculated from the ordered arrangement of holes which forms a square lattice with lattice constant $R_{nn}=4a$, where a is the Cu-O-Cu bond length and $a \approx 4$ Å for the high-T_c cuprates. If more holes are gradually introduced into the CuO_2 layer and arranged in an ordered fashion, hole contents of $p_{sh}^{cal}=0.125$, 0.25, and 0.3125 are obtained with $R_{nn}=2\sqrt{2a}$, $2a$, and $\sqrt{2a}$, respectively. It is seen that the hole contents calculated from ordered arrangements of holes in the CuO_2 layer are all in very good agreement with the critical values shown in the universal relationship in Fig. 1. With $a \approx 4$ Å, it is found that the values of R_{nn} are in the range 5.7 Å $\le R_{nn} \le 16$ Å, which is the range of in-plane coherence length for the high-T_c cuprates. Interestingly, the nn hole distance, $R_{nn}=4a$, at the onset of superconductivity also agrees well with the average polaron radius, $R_{polaron}\approx 4a$, as calculated by Spalek [17] for the high-T_c cuprates. Thus, if the polaron model of superconductivity is considered, the onset of superconductivity at $p_{sh}^{cal}=0.0625$ corresponds to $R_{nn}=R_{polaron}$, which can be interpreted as that a bipolaron forms when the wave functions of two polarons overlap strongly. It should be emphasized that the hole content $p_{sh}^{cal}=0.0625$ also corresponds to the onset of metallicity from the antiferromagnetic insulating state without invoking any pairing mechanism for superconductivity [17].

An important conclusion from the universal relation, i.e., $\tau_c=T_c/T_{c,max}=f(p_{sh})$, is that T_c for any cuprate superconductor is determined by two more or less independent parameters: p_{sh} and $T_{c,max}$. While the parameter p_{sh}, which is regulated by the charge reservoir Φ, is related only to the 2D CuO_2 layer, $T_{c,max}$ is perhaps mainly determined by the interplanar coupling between the CuO_2 layers and the surroundings such as that discussed by Ohta, Tohyama, and Maekawa, [18] who showed that $T_{c,max}$ correlated with the energy level of apical oxygen. The result of this study indicates that $T_{c,max}$ is not affected by the number of charge carriers on the CuO_2 layers since τ_c as well as the universal relationship is obtained by normalizing T_c with respect to a single, constant value of $T_{c,max}$ for each of the high-T_c cuprates. This conclusion is also in accordance with the result of Ohta, Tohyama, and Maekawa [18].

Last, we would like to point out the possible connection between the universal relation reported by Uemura *et al.* [2] and the present one. In Ref. [2], it has been shown that the variation of T_c with n_s/m^* before the saturation of T_c falls into a universal linear line. The tendency of the variation of T_c with n_s/m^* is very similar to the universal curve of our result. A major difference between the two studies is that a complete universal relationship is established in our study but only a portion of the curves falls on the universal linear line in Ref. [2]. However, the two universal relationships may be intimately connected to each other through the maximum T_c, $T_{c,max}$, and the effective mass m^*. From the two universal relationships, a conclusion can be reached that effective mass *actually* decreases with the increase of $T_{c,max}$ and that the increase of $T_{c,max}$ from one system (such as 2:1:4, 37 K) to another (such as 1:2:3, 90 K) is mainly due to the decrease of effective mass from one system to another. This conclusion is not evident from the result of Ref. [2] by only considering the ratio of n_s/m^*. An interesting question from the above conclusion is how the effective mass varies with hole content and what is the relationship between T_c and the effective mass after T_c saturates. The two universal relationships might be able to lead to answers or give clues to these important questions.

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