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Honma, T. ; Hor, P. H.

Intrinsic electronic superconducting phases at 60 K and 90 K in double-layer $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$

T. Honma¹ and P. H. Hor²

¹*Department of Physics, Asahikawa Medical College, Asahikawa, Hokkaido 078-8510, Japan*

²*Department of Physics and Texas Center for Superconductivity, University of Houston, Houston, Texas 77204-5005, USA*

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We study superconducting transition temperature (T_c) of oxygen-doped double-layer high-temperature superconductors $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ ($0 \leq \delta \leq 1$) as a function of the oxygen dopant concentration (δ) and planar hole-doping concentration (P_{pl}). We find that T_c , while clearly influenced by the development of the chain ordering as seen in the T_c vs δ plot, lies on a universal curve originating at the critical hole concentration (P_c)=1/16 in the T_c vs P_{pl} plot. Our analysis suggests that the universal behavior of $T_c(P_{pl})$ can be understood in terms of the competition and collaboration of chemical phases and electronic phases that exist in the system. We conclude that the global superconductivity behavior of $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ as a function of doping is electronically driven and dictated by pristine electronic phases at magic doping numbers that follow the hierarchical order based on P_c , such as $2 \times P_c$, $3 \times P_c$, and $4 \times P_c$. We find that there are at least two intrinsic electronic superconducting phases of $T_c=60$ K at $2 \times P_c=1/8$ and $T_c=90$ K at $3 \times P_c=3/16$.

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It has become clear in recent years that various physical properties of high temperature superconductors (HTS) are manifestations of a complex electronic texture of intrinsic electronic inhomogeneities due to dopants and, more importantly, electronic phase separation (EPS). In this paper we show that the key to understanding the underlying electronic texture and the corresponding superconducting properties are electronic phases that exist at magic planar doping concentrations (hole content per CuO_2 plane, P_{pl}) $P_{pl}=m/n^2$, where both m and n are positive integers with $4m \leq n^2$ and P_{pl} is determined based on a universal hole scale. Studies of the electronic phase diagram under ambient and high-pressure in cation (Sr) and anion (O) co-doped polycrystalline $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4+\delta}$ (CD-La214) revealed that there are two intrinsic electronic superconducting phases with superconducting transition temperature $T_{c1}=15$ K and $T_{c2}=30$ K.¹ The far-infrared charge dynamics studies on the T_{c1} and T_{c2} phases indicated that they are very peculiar electronic phases which have a very small amount (<1% of total doped hole) of free holes moving in otherwise pinned two-dimensional (2D) electronic lattice. The T_{c1} and T_{c2} phases start at the critical “magic” planar hole-doping levels $P_{pl}=1/16 \equiv P_c$ and $P_{pl}=1/8=2 \times P_c$, respectively.² The existence and the clear competition of T_{c1} and T_{c2} phases observed in the pure cation (Sr)-doped polycrystalline $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (SrD-La214) indicated that, independent of the nature of the dopants, these intrinsic T_c 's phases are energetically favored electronic phases that exist in the CuO_2 planes.³ Most recently magnetic studies of SrD-La214 single crystals confirmed the existence of the T_{c1} and T_{c2} transitions and the onset of the superconducting transition temperature were surprisingly robust with little magnetic field dependence up to 5 T.⁴ These “2D square electronic lattices” formed at magic doping concentrations are the most fundamental building blocks of electronic states, the pristine electronic phase(s) [PEP(s)], for the understanding of both normal and superconducting properties of HTS. While three-dimensional ordered PEPs seem to be firmly established in single-layer La214 system,⁴ it is not clear how PEPs will manifest themselves in other HTS. In this paper we show that, indeed, in pure anion (O) doped double-layer $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ (OD-Y123) system there exist at

least two intrinsic T_c 's of $T_c=60$ K and $T_c=90$ K that are based on PEPs at $P_{pl}=2/16=2P_c$ and $3/16=3P_c$, respectively. Furthermore we find that the famous “60 K plateau” in the electronic phase diagram of OD-Y123 can be naturally explained by the EPS of PEPs.

In order to sort out the PEPs in an OD-Y123 system we need to take care of the complications due to sensitive dependences of T_c on both the amount and the arrangement of oxygen dopants.^{5,6} The oxygen dopants tend to form long Cu-O chain ordering along the b axis that results in energetically favored meta-stable superstructures consisting of alternative arrangements of full Cu-O chain (full chain) and O-vacancy chain (empty chain).⁷ It was proposed that there are, starting from complete full-chain-ordering ortho-I phase, ortho-II (-II), -III (-III*), and -IV (-IV*) chemical phases will have single-, double-, and triple-full chains (empty chains) between any two empty chains (full chains), respectively.⁷ It is well known that in the T_c vs P_{pl} phase diagram of OD-Y123 there are two prominent plateaus located at $T_c \sim 60$ K (60 K plateau) and ~ 90 K (90 K plateau). It was generally assumed that 60 K and 90 K phases corresponded to the ortho-II and ortho-I chemical phases, respectively.^{8,9} There were two possible origins of 60 K plateau proposed: one is the chemical phase separation¹⁰ and the other is purely electronic in origin.¹¹ Recent theoretical study indicated that chemical phase arguments based on chain ordering alone cannot account for the observed 60 K plateau. In stead, a chain-ordering induced charge imbalance model was used to account for the 60 K plateau.¹² The electronic scenario attributed 60 K plateau to the well-known 1/8 anomaly identified in the SrD-La214.¹³ However, the analysis was, unfortunately, based on a questionable planar hole scale.¹⁴⁻¹⁶ The problem concerning the origin of the $T_c=60$ K and 90 K phases and the associated plateaus in OD-Y123 are still unresolved. In this paper we show that the electronic phase diagram of OD-Y123 can be understood in terms of the competition and the collaboration among the chemical phases and the PEPs.

A universal hole scale based on the thermoelectric power at 290 K (S^{290}) was constructed and used for comparing vari-

ous physical properties in HTS.¹⁶ It was shown that both normal and superconducting properties can be compared consistently with systematic doping dependences among different HTS.^{16,17} We analyze the reported $T_c(\delta)$ data of OD-Y123 as a function of P_{pl} determined by the universal scale. We find that $T_c(P_{pl})$ lies on one universal curve originating at the critical hole concentration $P_c=1/16$, while $T_c(\delta)$ lies on several curves which strongly depend on the development of the chain ordering. Further, we also find that the robust 60 and 90 K plateaus appear at $P_{pl}\sim 1/8=2P_c$ and $\sim 3/16=3P_c$, respectively, and the superconductivity is always suppressed beyond $P_{pl}=1/4=4P_c$. We extracted the T_c data from the published paper, irrespective of the definition of T_c .^{5,11,13,18–28} A sample's P_{pl} was directly determined from the reported S^{290} data by using the universal scale.^{11,13,14,18,23–29} For samples with either δ or T_c reported but not S^{290} , the P_{pl} of the sample were determined from the P_{pl} vs δ or the P_{pl} vs T_c relation as discussed below.^{19–22,30–32}

In Fig. 1(a) we plot T_c vs δ for OD-Y123. The $T_c(\delta)$ behavior can be roughly separated into two curves with small loop structures for $\delta<0.47$ and $0.7<\delta<0.8$. Curve 1 is $T_c(\delta)$ behavior corresponding to the polycrystals,^{5,13,20,23,24} crystals,^{21,22,26} and c -axis oriented films.²⁵ Curve 2 is $T_c(\delta)$ behavior as observed in the crystals detwinned by a uniaxial pressure of 100 MPa (Refs. 11 and 27) applied along $a(b)$ axis. For all the curve 1 samples, the T_c appears at $\delta\sim 0.35$, reaches the 60 K plateau for $0.47<\delta<0.65$ and reaches the 90 K plateau for $0.82<\delta<0.92$. Finally, the T_c slightly decreases for $\delta>0.92$. For the curve-2 samples, the T_c appears at $\delta\sim 0.4$, exhibits the 60 K plateau at $0.6<\delta<0.75$ and reaches the 90 K plateau at $\delta\sim 0.9$. The $T_c(\delta)$ curve of the detwinned crystals is lower than that of the curve-1 samples. However, the $T_c(\delta)$ -curve of crystals detwinned by the uniaxial pressure of 10 MPa^{22,36,37} follows that of the curve-1 samples. This indicates that the artificially prepared very long chain ordering actually suppresses the superconductivity. All of above are intrinsically consistent with the facts that the PEPs are two-dimensional (2D) square lattice. Furthermore the report that the highest T_c in $\text{La}_{2-x}\text{M}_x\text{CuO}_4$ ($M=\text{Nd, Ca, Sr}$) system is always observed in the trigonal phase with flat and square CuO_2 planes is also consistent with the above picture.³³ Clearly superconductivity of OD-Y123 is greatly affected by the level of the chain ordering and no systematic universal behavior can be inferred from the T_c vs δ phase diagram depicted in Fig. 1(a).

We plot P_{pl} vs δ in Fig. 1(b). Starting from $\delta\sim 0.35$ where $P_{pl}\sim 1/16=P_c$, the $P_{pl}(\delta)$ curve separated into three curves that merge into a common curve for $\delta>\sim 0.85$, where $P_{pl}\sim 2/9$. The upper, middle, and lower curves are determined from the S^{290} of polycrystals,^{13,14,18,23,24,29} the in-plane S^{290} of twinned crystals including c -axis oriented films,^{25,26} and the S^{290} measured along a axis (S_a^{290}) of crystals detwinned by the uniaxial pressure of 100 MPa,¹¹ respectively. P_{pl} increases monotonically with oxygen doping. The continuous increase in P_{pl} is not consistent with the chemical phase separation.³⁵ The common P_{pl} curve for $\delta>0.85$ suggests, in contrast to the charge imbalance model,¹² that chain ordering has no influences on hole concentration in the CuO_2 planes. Accordingly, the S^{290} scale in Ref. 16 can also be

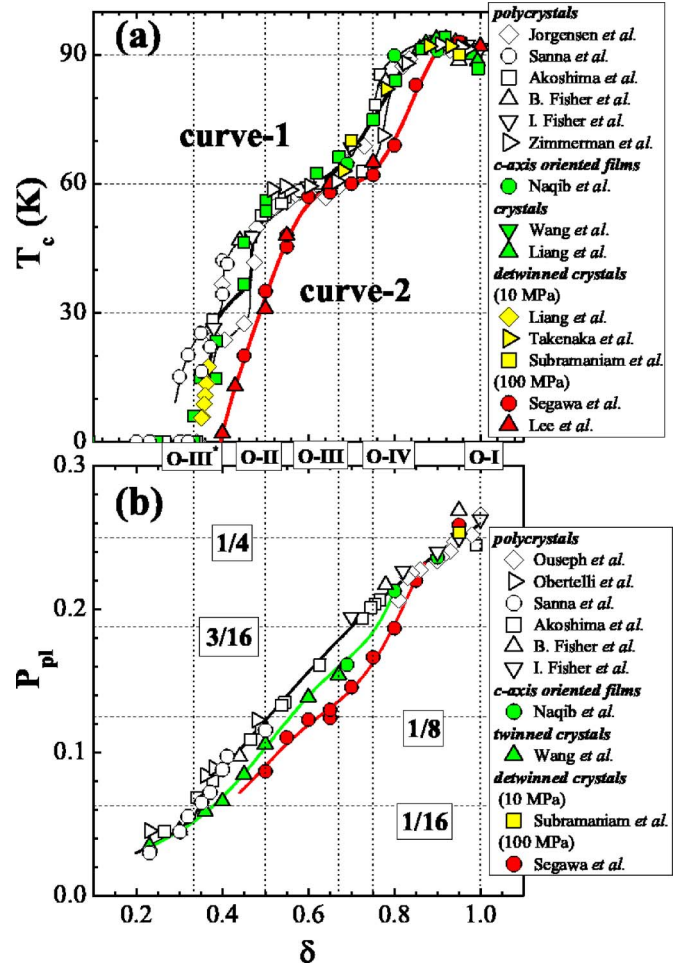


FIG. 1. (Color online) (a) Superconducting transition temperature (T_c) as a function of excess oxygen content (δ) for $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ (Refs. 5, 11, 13, 18, 20–27, 36, and 37). The solid curves are guide to the eyes. (b) Hole concentration (P_{pl}) as a function of δ for $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ (Refs. 11, 13, 14, 18, 23–27, and 29). The dotted horizontal lines show the magic number of the hierarchy of $P_c=1/16$. P_{pl} of all data, except of Akosima *et al.*'s work, are directly determined from the reported S^{290} by using of the universal scale in Ref. 16. Akosima *et al.* reported the δ vs the hole concentration determined from their own S^{290} by using the scale of Ref. 34. We plotted the re-determined P_{pl} by converting their used scale into our scale. In the Figs. 1(a) and 1(b), The dotted vertical lines show the idealized chemical phases of the ortho-I (O-I), -II (O-II), -III (O-III), -IV (O-IV), and ortho-III* (O-III*). The solid curves are guide to the eyes.

used for $\delta>0.85$ in OD-Y123. In the δ range from ~ 0.35 to ~ 0.85 , the planar hole concentration at the same δ values is successively suppressed in the order of the polycrystals, twinned crystals, and detwinned crystals with increasing length of chain ordering. Therefore, in contrast to the common belief, the perfect chemical phase with long chains does not favor the electronic state of HTS. Hereafter, if we only know the value of δ in the OD-Y123, we estimate the P_{pl} for the polycrystals, twinned crystals and artificially detwinned crystals from the corresponding curve of P_{pl} vs δ plot in Fig. 1(b)

In Fig. 2(a) we plot T_c vs P_{pl} for

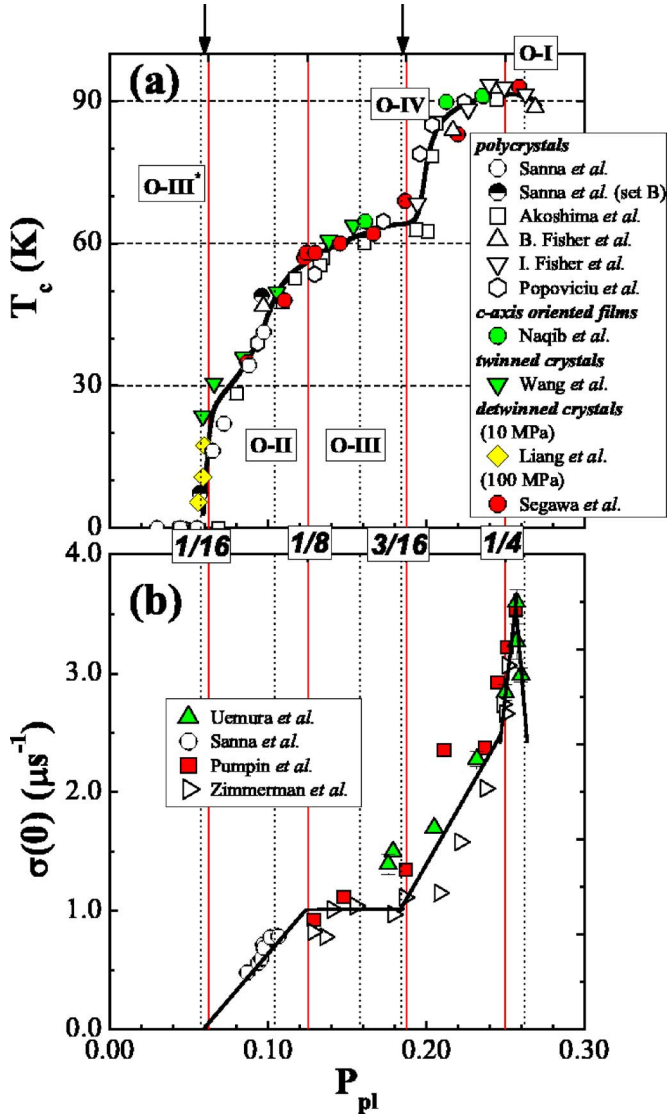


FIG. 2. (Color online) (a) T_c vs P_{pl} for $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ (OD-Y123). (Refs. 11, 13, 18, 19, 21, and 23–28). The P_{pl} for T_c data with δ value alone by Liang *et al.* (Ref. 21) was determined from the P_{pl} vs δ plot of Fig. 1(b). The second data set by Sanna *et al.* (set B) was reported in Ref. 19. According to Ref. 19, we can extract the lower and upper bound of δ and T_c for their OD-Y123 samples. The (δ, T_c) are (0.32, 7 K) and (0.42, 49 K). We plotted these two points by determining P_{pl} from the δ value. All other P_{pl} were directly determined from the reported S^{290} . (b) N_S vs P_{pl} for the OD-Y123 (Refs. 19 and 30–32). P_{pl} for $\sigma(0)$ data with T_c value alone by Sanna *et al.* (Ref. 19) were determined from the P_{pl} vs T_c plot of Fig. 2(a). All other P_{pl} for $\sigma(0)$ with δ and T_c (Refs. 30–32) were determined from the P_{pl} vs δ plot of Fig. 1(b). In the Figs. 2(a) and 2(b), the vertical solid lines show the magic number of the hierarchy of $P_c=1/16$. The vertical dotted lines show the idealized chemical phases of the ortho-I (O-I), -II (O-II), -III (O-III), -IV (O-IV), and ortho-III* (O-III*). The solid curves guide to the eyes.

OD-Y123.^{11,13,18,19,21,23–28} Surprisingly, T_c data of all samples collapsed into a single universal curve in the T_c vs P_{pl} plot. The superconductivity appeared at $P_{pl} \sim 0.06$ ($\sim P_c$) that exhibits an extremely sharp T_c jump to ~ 30 K followed by a broad T_c increase to 60 K at $P_{pl} \sim 0.1$. The 60 K plateau is

observed between 0.12 ($\sim 2P_c$) $< P_{pl} < 0.19$ ($\sim 3P_c$). Further, the T_c suddenly jumps to ~ 90 K over ~ 0.19 ($\sim 3P_c$) and goes into the 90 K plateau for $0.21 < P_{pl} < 0.25$ ($\sim 4P_c$). Finally, the T_c decreases for $P_{pl} > 0.25$ ($\sim 4P_c$). The phase diagram is characterized by fast T_c jumps and much flatter T_c regions in between. Noted that the sharp T_c jumps occurred whenever the P_{pl} of a chemical phase is almost identical to that of a PEP [see two arrows in Fig. 2(a)]. In contrast, the broad T_c increase at $\sim 1/8$ seems to come from the mismatch between the P_{pl} of chemical phases to $2P_c$. The above observations suggest that the global behavior of the electronic phase diagram is dictated by the EPS of PEPs under the influences of chemical phases: the jump in T_c is due to the matching of a PEP and a chemical phase and the flat region is a two PEPs region. There are some fine structures in the T_c vs P_{pl} plot; a “hint” of a 30 K intrinsic phase and a 30 K plateau around $P_{pl} = P_c$ and a small T_c jump to $T_c \sim 45$ K occurred when ortho-II matched with magic doping concentration at $P_{pl} \sim 1/9$. It is also interesting to note that the intrinsic T_c 's increase as integer multiples of 30 K in the double-layer OD-Y123 in contrast to that of 15 K in the single-layer La214 system.

The above EPS picture for the 60 K plateau is further collaborated by the observation of a clear plateau of another intrinsic property, the superfluid density (N_S), of a superconductor in the same two phase region. We used N_S estimated from the low temperature muon-spin relaxation (μSR) rate $\sigma(0)$. In Fig. 2(b), we plotted $\sigma(0) \propto N_S$ vs P_{pl} .^{19,30–32} While there is a slight scattering, in the magnitude among the reported $\sigma(0)$, each reported $\sigma(0)$ data set have similar P_{pl} dependence. The N_S linearly increases with doping for $\sim 1/16$ (P_c) $< P_{pl} < \sim 1/8$ ($2P_c$) followed by a clear plateau for $\sim 1/8$ ($2P_c$) $< P_{pl} < \sim 3/16$ ($3P_c$). It then linearly increases for $\sim 3/16$ ($3P_c$) $< P_{pl} < \sim 1/4$ ($4P_c$) again, and ends with a sharp peak at $\sim 1/4$ ($4P_c$). Finally, the N_S rapidly decreases for $P_{pl} > 1/4$ ($4P_c$). Therefore, in superfluid density vs P_{pl} plot, there is also a flat two-phase region bounded by $P_{pl} = 1/8$ and $3/16$, consistent with the co-existence of two PEPs of $2P_c$ and $3P_c$. It is interesting to point out that the famous linear T_c vs N_S plot, the Uemura plot, failed to reveal the two phase region.³⁰ Therefore in order to have a physically meaningful comparison of various physical properties of HTS, it is of critical importance that the physical properties should always be plotted in terms of P_{pl} first as determined by the universal hole scale.¹⁶

In summary, we have examined both the electronic-phase and chemical-phase diagrams of double-layer high temperature superconductors $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ as a function of the hole content per CuO_2 plane P_{pl} and oxygen doping concentration δ , respectively. The $T_c(P_{pl})$, regardless of the sample quality, is a universal curve originating at $P_c = 1/16$. The 60 K plateau, T_c jump and 90 K plateau occur at a series hierarchical doping concentration based on P_c such as $P_{pl} = 2P_c$, $3P_c$, and $4P_c$. Our analysis suggests that the electronic phase diagram of T_c vs P_{pl} can be understood, although modified by the chemical phases, in term of the existence and the EPS of PEPs. We conclude that there are at least two PEPs with

$T_c=60$ K at $P_{pl}=2P_c=1/8$ and $T_c=90$ K at $P_{pl}=3P_c=3/16$. Beyond $P_{pl}=4P_c=1/4$, the superconductivity, such as T_c and N_S , is always suppressed. The observation of superconducting transitions at magic doping levels in the double-layer OD-Y123 and single-layer La-214 strongly suggest that the PEPs are generic intrinsic properties of all high temperature

superconductors.

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