# Electronic States and Superconducting Transition Temperature based on the Tomonaga-Luttinger liquid in $Pr_2Ba_4Cu_7O_{15-\delta}$

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An NQR experiment revealed superconductivity of $Pr_2Ba_4Cu_7O_{15-\delta}$ (Pr247) to be realized on CuO double chain layers and suggests possibility of novel one-dimensional(1D) superconductivity. To clarify the nature of the 1D superconductivity, we calculate the band dispersions of Pr247 by using the generalized gradient approximation(GGA). It indicates that Fermi surface of CuO double chains is well described to the electronic structure of a quasi-1D system. Assuming the zigzag Hubbard chain model to be an effective model of the system, we derive tight binding parameters of the model from a fit to the result of GGA. Based on the Tomonaga-Luttinger liquid theory, we estimate transition temperature ($T_c$) of the quasi-1D zigzag Hubbard model from the calculated value of the Luttinger liquid parameter $K_\rho$. The result of $T_c$ is consistent with that of experiments in Pr247 and it suggests that the mechanism of the superconductivity is well understood within the concept of the Tomonaga-Luttinger liquid.

KEYWORDS: $Pr_2Ba_4Cu_7O_{15-\delta}$, first-principles calculations, CuO double chain, superconductivity, Tomonaga-Luttinger liquid

## 1. Introduction

The newly discovered superconductor $Pr_2Ba_4Cu_7O_{15-\delta}$ (Pr247) consists of semiconducting single-chain and metallic CuO double-chain besides the Mott insulating $CuO_2$ plane.[1–3] It shows the transition temperature ($T_c$) of superconductivity up to $T_c \sim 20$K in a moderate oxygen defect concentration range of $\delta = 0.2 \sim 0.6$, where $\delta$ can be varied by controlling oxygens in the CuO single chains, and electrons are doped to CuO double chain. It is found that the main carrier is electron from the measurement of the Hall coefficient at low temperature($T < 120$K). An NQR (nuclear quadrupole resonance) experiment has confirmed the superconductivity in the CuO double chain layers.[4] A spin-gap-like behavior was also observed in the recent NMR experiment,[5] although it is not clear yet whether the spin gap is enough large. These experiments suggest that Pr247 shows the possibility of novel one-dimensional(1D) superconductivity.

In the previous papers, we address the 1D double chain models to clarify the superconductivity of Pr247 from the theoretical point of view.[6,7] Based on the Tomonaga-Luttinger liquid theory, we analyze 1D superconducting mechanism proposed generally by Fabrizio,[8] in which the spin gap plays an important role. In fact, the spin gap of the double chain model is found to become up to $\sim 100$K for a typical case and the 1D superconductivity seems to be realistic in Pr247.[7] On the other hand, it is shown that the spin gap is very small at some parameter region by using the density matrix renormalization group (DMRG).[9] Then, Nakano *et al.* proposed a spin fluctuation mechanism of the superconductivity for Pr247 by applying the fluctuation exchange (FLEX) method to a quasi (1D) extended Hubbard model.[10] Berg *et al.* claimed that "$C_1S_{3/2}$" phase, in which the total spin mode is gapless but the half of the relative spin mode is gaped, is crucial to the superconductivity by using the bosonization theory.[11] However, these theoretical works employed the result of energy band calculation[12] for not Pr247 but $YBa_2Cu_4O_8$(Y124) in which CuO double chains are included with the same lattice structure, because the band calculation results of Pr247 has not been available. Although energy dispersions corresponding to the CuO double chain for the both compounds are expected to be almost equivalent, it is very difficult to avoid ambiguity of the model to be solved.

In this article, we calculate the band structure of Pr247 by the density functional calculation with the generalized gradient approximation of Perdew, Burke, and Ernzerhof by using the WIEN2k package.[13] We set the 1D zigzag chain Hubbard model to be an effective model of CuO double-chain and obtain the tight-binding parameters of the model by fitting the energy dispersion. Using these parameters, we estimate $T_c$ of the model based on the Tomonaga-Luttinger liquid theory. It would give an accurate analysis of the electronic state of Pr247 and shed light on the mechanism of the novel quasi-1D superconductivity.

## 2. Band Calculation

The calculations is done using the WIEN code,[13] which uses a full potential-linearized augmented plane wave (FLAPW) calculation. The exchange correlation potential is treated by the generalized gradient approximation of Perdew *et al.*[14] The energy threshold to separate core and valence states is -6 Ry. For the number of plane waves the criterion $R_{\rm MT}$ (muffin tin radius) $\times$ $K_{\rm max}$ (for the plane waves) = 7. Self-consistency of the calculation is yield by using a set of 216 k points in the irreducible wedge of the Brillouin zone and the spin-orbit coupling is also considered in our calculations. The crystal structure is referred by the experiment val-

ues obtained by Yamada *et al.*[2] To check the validity of our calculation, we also obtain the result of GGA for Pr123(data not shown) and confirm that it agrees with previous study well.[15]

Figure 1 shows the band structure of Pr247, where diameter of circles represents weight of contribution from $d_{x^2-y^2}$ orbitals of Cu in the double chain, where the double chain is set to be along the $b$-axis in our calculation. We find that seven bands cross the Fermi energy $E_F$ on $\Sigma-\Gamma$ line among them, two bands which have significant dispersion on $\Gamma-Y$ line, correspond to the CuO double chain. It surely shows that CuO double chains are metallic and the dispersion along the $c$-axis perpendicular to $CuO_2$ layer is very small as well as that of Pr123. Almost flat bands near and above $E_F$ consists mainly of Pr's $4f$ orbital.

Fermi-surfaces of Pr247 are shown in Fig.2 on the $k_x$-$k_y$ plane with $k_z = 0$. Six vertical lines in both sides present the Fermi-surfaces of single-chain and double-chain, where that of single-chain is denoted by the dashed lines, and the solid lines are corresponding to that of double-chain. Four closed Fermi surfaces are corresponding to $CuO_2$ plane. The hybridization between double-chain and $CuO_2$ plane is very small, because the Fermi surfaces of the double-chain are almost straight, though that of the single chain is warped. The result suggests that the band structure of the double chain is well described as a quasi-1D system. Although the existence of the Fermi surfaces of $CuO_2$ plane seems to indicate $CuO_2$ plane to be metallic, it becomes insulator because the electronic conduction in the plane is suppressed due to the so called Fehrenbacher-Rice state formed by the strong hybridization between Pr4$f$ and O2$p$ orbitals.[16] We note that it is not able to take into account this effect within the GGA calculation. Therefore, we should not consider the position of the Fermi level obtained by GGA too serious.

Next, we consider quasi-1D double chain Hubbard model as a minimum model reflecting the band structure of Pr247. It contains intra-chain hopping integrals between Cu sites and inter-chain hoppings as shown in Fig. 3 . The Hamiltonian is given by

$$
\begin{aligned}
H &= t_1 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+1,m,\sigma} + h.c.) \\
&+ t_2 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+2,m,\sigma} + h.c.) \\
&+ t_3 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+3,m,\sigma} + h.c.) \\
&+ t_4 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+4,m,\sigma} + h.c.) + U \sum_{i,m} n_{i,m,\uparrow} n_{i,m,\downarrow} \\
&+ V_1 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i,m+1,\sigma} + h.c.) \\
&+ V_2 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+1,m+1,\sigma} + h.c.) + (c^\dagger_{i,m,\sigma} c_{i-1,m+1,\sigma} + h.c.) \\
&+ V_3 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+2,m+1,\sigma} + h.c.) + (c^\dagger_{i,m,\sigma} c_{i-2,m+1,\sigma} + h.c.) \\
&+ V_4 \sum_{i,m,\sigma} (c^\dagger_{i,m,\sigma} c_{i+3,m+1,\sigma} + h.c.) + (c^\dagger_{i,m,\sigma} c_{i-3,m+1,\sigma} + h.c.) \quad (1)
\end{aligned}
$$

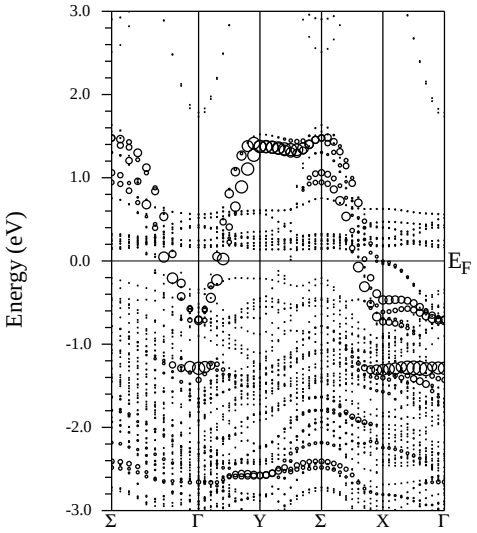


Fig. 1. A band structure of Pr247 in $k_z = 0$ from non-spin polarized calculation. The Γ point is at the corners, the Σ point in center, the $X$ point at the midpoint of the horizontal edge, and the $Y$ point at the midpoint of the vertical edge,

where $c^\dagger_{i,m,\sigma}$ stands for the creation operator of an electron with spin $\sigma$ at site $i$ on $m-$th chain and $n_{i,m,\sigma} = c^\dagger_{i,m,\sigma} c_{i,m,\sigma}$. Here, $t_{n=1,2,3,4}$ is the hopping energy between intra chain sites and $V_{n=1,2,3,4}$ is that between the nearest-neighbor inter-chain sites as shown in Fig.3. Here, $U$ is the on-site Coulomb interaction parameter.

In a noninteracting case ($U = 0$), the Hamiltonian eq. (1) yields the band energies as functions of wave numbers of $k_x$ and $k_y$,

$$
\begin{aligned}
E(k_x, k_y) &= \varepsilon_a + 2t_1 \cos(k_x) + 2t_2 \cos(2k_x) + 2t_3 \cos(3k_x) \\
&+ 2t_4 \cos(4k_x) + 2V_1 \cos(k_y) + 4V_2 \cos(k_x)\cos(k_y) \\
&+ 4V_3 \cos(2k_x)\cos(k_y) + 4V_4 \cos(3k_x)\cos(k_y). \quad (2)
\end{aligned}
$$

We estimate the tight-binding parameters $t_n$ and $V_n$

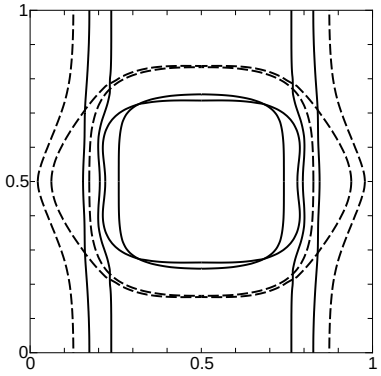


Fig. 2. A fermi surface in $k_z = 0$ for Pr247 from non-spin polarized calculation. The Γ point is at the corners, the Σ point in center, the $X$ point at the midpoint of the horizontal edge, and the $Y$ point at the midpoint of the vertical edge,

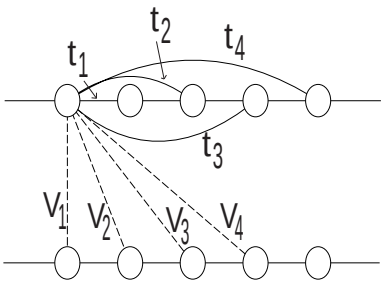


Fig. 3. The definition of hopping parameters in the quasi-1D double chain Hubbard model between intra-chain sites and between inter-chain sites, respectively.

| i | $t_i$ | $V_i$ |
|---|---|---|
| 1 | −0.1227 eV | −0.0203 eV |
| 2 | −0.4908 eV | 0.0060 eV |
| 3 | 0.0876 eV | −0.0097 eV |
| 4 | −0.0652 eV | 0.0026 eV |

Table I. The values of intra-chain transfer $t_i$, and inter-chain transfer $V_i$ for i-th neighbor sight.

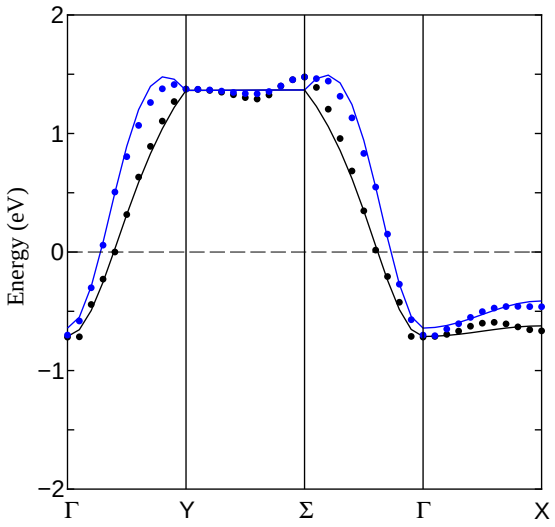


Fig. 4. The dispersion of the tight-binding model with the parameters determined so as to fit the calculated band structure of Pr24, where the closed circles present GGA result.

of the model eq. (2) so as to fit $E(k_x, k_y)$ to the energy dispersion obtained by the GGA. The obtained values of parameters are shown in Table I and the fitted dispersion is shown in Fig.4 . The figure indicates that the obtained dispersion is in good agreement with the GGA result and our quasi-1D model is expected to simulate the electronic state of Pr247 very well. We also performed same calculation for Pr124(data not shown) and found that $t_1/t_2$ of Pr124 is smaller than that of Pr247.[17)]

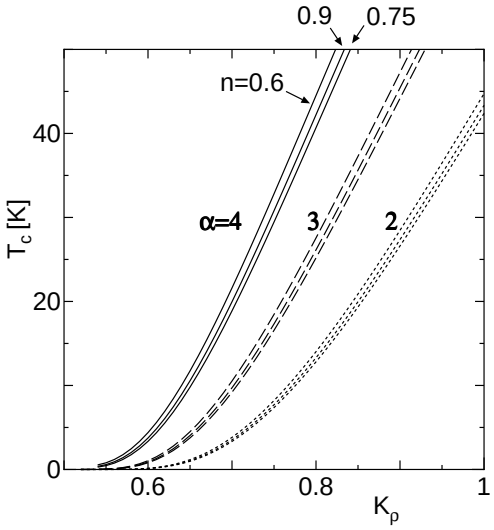


Fig. 5. $T_c$ as a function of $K_\rho$ at $n$ =0.5, 0.75 and 1.0 for $\alpha$=2(dotted line),3(dashed line) and 4(solid line), respectively.

## 3. Tomonaga-Luttinger liquid approach

Next, assuming the spin gap to be large enough to exceed the transition temperature ($T_c$) of the superconductivity,[7,18)] we consider $T_c$ by using the formulation of the Tomonaga-Luttinger liquid in cooperation with the mean-field analysis for quasi-1D systems.[19)] It contains the Luttinger liquid parameter $K_\rho$ of chain part and coupling parameter $\lambda$ between chains, as shown below

$$T_c = \frac{v_F}{2\pi\alpha}\left\{\frac{\lambda}{4\pi}B^2\left(\frac{\gamma}{2},\frac{\gamma}{2}\right)\tan\frac{\pi\gamma}{2}\right\}^{1/(2-2\gamma)}, \qquad (3)$$

where $\lambda = \frac{4\alpha^2}{{v_F}^2}(2V_1^2 + 4V_2^2 + 4V_3^2 + 4V_4^2)$, $B(x,y) = \frac{\Gamma(x)\Gamma(y)}{\Gamma(x+y)}$, and $\gamma = \frac{1}{2K_\rho}$. Here, $v_F$ is the Fermi velocity. We approximate $v_F \simeq {v_F}^*$, where ${v_F}^* = \frac{v_{F_1}v_{F_2}}{v_{F_1}+v_{F_2}}$ is an effective Fermi velocity constructed by two Fermi velocities; $v_{F_1}$ and $v_{F_2}$ in the double chain model.[18,20)] Further, $\alpha$ is a short-wavelength cutoff parameter.[19,21)] This value related to the spatial extension of the quasiparticle, however, it is difficult to determine it without ambiguity. We should chose $\alpha \gtrsim 2$, since the dimer state on the nearest neighbor sites is considered to composite main part of the quasiparticle.[22–24)]

In the double chain model with two different Fermi points, low-energy excitations are given by a single gapless charge mode with a gapped spin mode.[8,25–29)] In this case, the SC and CDW correlations of the charge mode decay as $\sim r^{-\frac{1}{2K_\rho}}$ and $\sim \cos[2(k_{F_2}-k_{F_1})r]r^{-2K_\rho}$, respectively, while the SDW correlation decays exponentially. Hence, the SC correlation is dominant for $K_\rho > 0.5$, while the CDW correlation is dominant for $K_\rho < 0.5$. Since the role of inter-chain couplings is considered to stabilize the SC state at finite temperature, the electronic state in chain part is crucial to the creation of the SC. If $K_\rho > 0.5$, the SC correlation is dominant in the chain and the total system becomes superconductor.

In Fig. 5, we show $T_c$ as a function of $K_\rho$ for severl values of electron number per site $n$ by substituting the

fitting parameters of Pr247 into eq.(3). At first glance, the $n-$dependence of $T_c$ seems to be small, but we stress that $n-$dependence of $K_\rho$ is not small. For example, near $n = 1$, $K_\rho$ becomes very small due to the umklapp effect. The $n-$dependence of $K_\rho$ for similar models has been already discussed in several works.[6,7,18] They indicate that $K_\rho$ has a maximum value almost around $n \sim 0.7$. It suggests that $T_c$ of Pr247 has also a peak near the same point.

We roughly estimate $K_\rho$ of our model by the Hartree-Fock approximation; $K_\rho = \sqrt{\frac{1}{1+\frac{U}{\pi v_F{}^*}}}$,[18,20] where this approximation is valid only in the weak coupling limit and neglects the umklapp process. When $U = 2$eV and $n = 0.75$, we obtain $K_\rho \sim 0.79$ and then $T_c \sim 12$K for $\alpha$=2 and $T_c \sim 23$K for $\alpha$=3. These results are reasonable and consistent with the experiment results.[2] It also suggests that the novel mechanism of the quasi-1D superconductivity is realized in Pr247.

## 4. Summary and Discussion

In summary, we calculate the band dispersions of Pr247 by using the GGA. It shows that the electronic state of Pr247 is well described by the quasi-1D system. We also obtain the tight binding parameters by fitting the band dispersions to the quasi-1D zigzag Hubbard chain model. It would give an accurate analysis of the electronic state and a concrete basis to address the superconductivity of Pr247. On the assumption that the spin gap is large enough to exceed $T_c$, we estimate $T_c$ of the superconductivity in Pr247 by using the formula of $T_c$ for quasi-1D superconductor. The result is consistent with that of experiments and suggests that the mechanism of the superconductivity in Pr247 may be understood within the concept of the Tomonaga-Luttinger liquid.

Finally, we would point out the possibility of the quasi-1D superconductivity in Y124, since the double chains are also contained in Y124. We estimate $T_c$ of the quasi-1D superconductivity in Y124 as $\sim$ 40K for $U = 2$eV and $\alpha$=2 by adopting the tight-binding parameters of Y124 obtained by ref.10, where inter-chain hoppings is larger than those in Pr247 resulting higher $T_c$.

At this stage, the experimental evidence of quasi-1D superconductivity has not been explicitly observed, since the quasi-two-dimensional(2D) superconductivity($T_c \sim$ 80K) in $CuO_2$ plane masks the quasi-1D superconductivity even if it exists. If we can suppress only the quasi-2D superconductivity, we expect the quasi-1D superconductivity to be found in Y124 near 40K. In fact, the experiment of Zn-doped Y124 shows that $T_c$ have a plateau at $T_c \sim 25$K in the case of doping rate being larger than 2%.[30] If doped impurities destroy the superconductivity of $CuO_2$ planes completely, we can expect that the plateau may be corresponding to the indication of quasi-1D superconductivity. Further both of theoretical and experimental studies in this regard is highly desired.


1) M. Matsukawa, Yuh Yamada, M. Chiba, H. Ogasawara, T. Shibata, A. Matsushita and Y. Takano: Physica C **411** (2004) 101.
2) Yuh Yamada and A. Matsushita: Physica C**426-431** (2005) 213.
3) M. Hagiwara, S. Tanaka, T. Shima, K. Gotoh, S. Kanda, T. Saito, K. Koyama: Physica C **468** (2008) 1217.
4) S. Watanabe, Y. Yamada, S. Sasaki: Physica C 426-431 (2005) 473.
5) S. Sasaki: private communication.
6) K. Sano, Y. Ōno and Y. Yamada: J. Phys. Soc. Jpn. **74** (2005) 2885.
7) K. Sano and Y. Ōno: J. Phys. Soc. Jpn. **76** (2007) 113701.
8) M. Fabrizio: Phys. Rev. B **54** (1996) 10054.
9) K. Okunishi : Phys. Rev. B **75** (2007) 174514.
10) T. Nakano, K. Kuroki, and S. Onari: Phys. Rev. B **76** (2007) 014515.
11) E. Berg, T. H. Geballe, and S. A. Kivelson: Phys. Rev. B **76** (2007) 214505.
12) C. Ambrosch-Draxl, P. Blaha, and K. Schwarz: Phys. Rev. B **44** (1991) 5141.
13) P. Blaha, K. Schwarz, G. Madsen, D. Kvasnicka, and J. Luitz: WIEN2k, An Argumented Plane Wave + Local Orbitals Program for Calculating Crystal Properties (Technische Universiat Wien, Vienna, 2002) [http://www.wien2k.at].
14) J. P. Perdew, K. Burke, and Y. Wang: Phys. Rev. B **54** (1996) 16533.
15) D. J. Singh: Phys. Rev. B**50** (1994) 4106.
16) R. Fehrenbacher and T. M. Rice: Phys. Rev. Lett. **70** (1993) 3471.
17) When we consider the metallic state of double chain in $PrBa_2Cu_4O_8$, we should probably add the Coulomb interactions between the nearest-neighbor and the next nearest-neighbor sites; H. Seo and M. Ogata: Phys. Rev. B **64** (2001) 113103.
18) S. Nishimoto, K. Sano, and Y. Ohta: Phys. Rev. B **77** (2008) 085119.
19) H. J. Schulz and C. Bourbonnais: Phys. Rev. B**27** (1983) 5856. It is noted that the equation of $T_c$ has been derived for the quasi-1D *single* chain model without spin gap in the above reference. In order to apply the equation to the spin gap system such as our quasi-1D *double* chain model, we should substitute $\frac{1}{2K_\rho}$ for the scaling dimension $\gamma$ in eq.(3). See also S. T. Carr and A. M. Tsvelik: Phys. Rev. B**65** (2002) 195121.
20) K. Sano: J. Phys. Soc. Jpn. **69** (2000) 1000.
21) J. Voit: Rep. Prog. Phys. **58**, (1995) 977.
22) I. Doi, K. Sano, and K. Takano: Phys. Rev. B**45** (1992) 274.
23) K. Sano, and K. Takano: J. Phys. Soc. Jpn. **61** (1992) 4661.
24) K. Takano, and K. Sano : Phys. Rev. B**48** (1993) 9831.
25) L. Balentz and M. P. A. Fisher: Phys. Rev. B **53** (1996) 12133.
26) V. J. Emery, S. A. Kivelson and O. Zachar: Phys. Rev. B **59** (1999) 15641.
27) K. Kuroki, R. Arita, and H. Aoki: J. Phys. Soc. Jpn. **66** (1997) 3371.
28) S. Daul and R. M. Noack: Phys. Rev. B **58** (1998) 2635.
29) S. Daul and R. M. Noack: Phys. Rev. B **61** (2000) 1646.
30) I. Felner and B. Brosh: Phys. Rev. B**43** (1991) 10364.