

Electric Charge Transfer and Scattering of Its Carriers in Cuprates of the 1–2–3 System

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Abstract We show that the temperature dependences of the basal-plane electrical resistance in cuprates of the 1–2–3 system can be described as a consequence of scattering of charge carriers on phonons and defects in conjunction with the fluctuation conductivity. The electron–phonon parameters values deduced from fitting the experimental data to recognized models are close to those for metallic alloys of complex composition. It is revealed that at a large oxygen deficit (low superconducting transition temperatures T_c), the superconducting behavior of the studied cuprates has similarities with that of complex superconducting alloys. At the optimum oxygen deficit (maximal T_c s), superconductivity in the investigated cuprates is likely governed by some other mechanisms.

Keywords High-temperature superconductors · Cuprates · Electric charge transfer · Fluctuation conductivity

1 Introduction

Despite a nearly thirty-year long history of high-temperature superconductivity (HTSC), its microscopic theory has not been elaborated so far. This is why the interest to study the properties of HTSC compounds in the superconducting as well as in the normal state is not diminishing. As is known [1], the charge transfer mechanisms in HTSCs have peculiarities stipulated by the manifestations of a series of specific phenomena observed therein in the normal (non-superconducting) state. To

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these phenomena, one attributes the fluctuation [2–4] and pseudogap [5–7] anomalies, transitions of the metal-to-insulator type [8,9], the incoherent electronic transport [10,11], and others. According to the contemporary views [1,12], it is that these phenomena which may be the key to understand the nature of HTSC. However, despite of considerable efforts of theorists and experimentalists, many aspects of these phenomena remain unclear so far. A certain role in this is played by a pronounced anisotropy of the crystal structure of HTSC compounds [13], the presence in these of a non-trivial disorder ensemble containing defects of different types [14,15], the presence of a labile component [16–18] in the system, and a series of other peculiarities leading to difficulties in studying the charge transfer mechanisms. This is why understanding the charge transfer mechanisms [3] and the peculiarities of their transfer in the presence of structural and kinematic anisotropies [19,20] is of great importance.

For investigations in this aspect, the most asked-for compounds are ones from the 1–2–3 system $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($\text{Re}=\text{Y}$ or lanthanides) that is stipulated by several reasons. First, these compounds have a rather high critical temperature $T_c \approx 90$ K above the nitrogen liquefaction temperature [21,22]. Second, the electric transport characteristics of these compounds can rather easily be varied by doping of the compound with substituting elements [23,24] or varying the oxygen content [25,26]. Third, there are routine technologies for the preparation of single-crystal samples with a given defect structure [27,28] that is crucial for doing fundamental research.

In particular, it is worth noticing that the behavior of the conductivity in HTSCs in the normal state is similar to the behavior of the conductivity in such typical systems with a pseudogap as amorphous alloys, quasicrystals, and approximants. For these systems, the electronic transport mechanisms were divided into two regimes [29–31]. One of these is determined by the electron mean-free path of the charge carriers, while the other one by the effects of the electronic structure specific for systems with a pseudogap.

A transition from one regime to the other takes place upon attaining a certain conductivity value equal to the minimal metallic conductivity. This value can be estimated using the conventional formula for the electrical conductivity obtained on the basis of the transport Boltzmann equation [29]

$$\sigma = \rho^{-1} = \frac{e^2 S_F \Lambda}{12\pi^3 \hbar}, \quad (1)$$

where S_F is the area of the Fermi surface and Λ is the mean-free path of the charge carriers. At $S_F = 4\pi k^2$ and $k = \pi/a$ (a is the interatomic distance) for the half-filled band, one obtains the Ioffe–Regel limit $\sigma_{\min} = e^2/(3\hbar a)$. If one sets $a \approx 3 \text{ \AA}$, then $\rho_{\max} = (\sigma_{\min})^{-1} \approx 370 \mu\Omega\text{cm}$.

For the regime which is determined by the mean-free path of the charge carriers is typical a temperature dependence of the metallic type. This dependence can be described by the conventional Bloch–Grüneisen formula accounting for scattering of the charge carriers on phonons, i. e., effects of the electron–phonon interaction [32], and defects, viz.,

$$\rho_n(T) = \rho_0 + C_n \left(\frac{T}{\theta}\right)^n \int_0^{\theta/T} \frac{x^n e^x dx}{(e^x - 1)^2}. \quad (2)$$

Here ρ_0 is the residual resistivity stipulated by the defects, while the second term is the ideal phonon resistivity with C_n being the material constant of the metal. In Eq. (2), θ is the kinetic Debye temperature which can somewhat differ from the thermodynamic value and $n = 3, 5$ [33]. For $n = 3$, C_3 is the coefficient of the ideal phonon resistivity. In accordance with Eq. (2), $\rho(T) \sim T^n$ at $T \ll \theta$, while $\rho(T)$ tends to a linear dependence with increasing temperature.

Here one should emphasize two circumstances. First, for HTSCs at least from the 1–2–3 system, the derivatives $d\rho(T)/dT$ calculated from the experimental curves $\rho(T)$ tend to constant values in the region $T \geq \theta$ only, see e.g., [3,34–37]. Second, the dependence (1) has a saddle point; hence, $d\rho(T)/dT$ has a maximum at $T/\theta \approx 0.33$ ($n = 3$) and $T/\theta \approx 0.36$ ($n = 5$). This maximum of the derivative is observed in the experimental dependences $d\rho(T)/dT$ for underdoped samples with $T_c < \theta/3$ [35,36]. For optimally doped samples from the 1–2–3 system, a significantly stronger maximum in $d\rho(T)/dT$ is located in the region $T/\theta \approx 0.3$. This maximum is related to the transition to the superconducting state and in its background the maximum described by Eq. (1) is not seen.

Naturally, Eq. (1) is an approximation. At high temperatures, one often notices deviations from linearity (see, e.g., Ref. [38] and references therein). These deviations are attributed in [39] to the change in the location of the Fermi level and can be accounted for by multiplying Eq. (1) by the factor $1 + BT^2$, where B depends on the shape of the density of electronic states curve, the effective mass of the charge carriers, and the Fermi energy.

For the regime which is determined by the effects of the electronic structure, one approximates $\rho(T)$ by dependences of the semiconductor type. Near T_c a fluctuation paraconductivity appears (see, e.g., [40]). This is why the temperature dependence of the resistivity above T_c can be written as

$$\rho(T) = [\rho_n^{-1}(T) + \Delta\sigma_{\text{fluct}}]^{-1}. \tag{3}$$

Near T_c one has to account for the fluctuation conductivity which within the framework of the 3D Aslamazov–Larkin model [40] reads

$$\Delta\sigma_{\text{fluct}} = \Delta\sigma_{\text{AL}}^{3D} = \frac{e^2}{16\hbar\xi_c(0)\sqrt{2\varepsilon_0 \sinh(2\varepsilon/\varepsilon_0)}}. \tag{4}$$

In Eq. (4), $\varepsilon = \ln[(T - T_c)/T_c]$ is the reduced temperature, T_c is the critical temperature, $T > T_c$, ξ_c is the coherence length along the c -axis, ε_0 determines the temperature interval of superconducting fluctuations $\varepsilon_0 = \ln(T^* - T_c)/T_c$, and T^* is the characteristic temperature determining the collapse of superconducting fluctuations [41].

Here, we comparatively analyze the electrical resistance data for several cuprates [3,24,37,37,42–44] of the 1–2–3 system and study the electric charge transfer and scattering of its carriers therein.

2 Results and Discussion

In Refs. [3,37,42] for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, [24,37,43] for $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$, and [44] for $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$, it was shown that in the temperature interval from T_c to 300 K the resistance in the ab plane can be described by Eqs. (1)–(4). This means that the cumulative experimental curves $\rho^{ab}(T)$ can be approximated as a result of scattering of electrons on phonons and defects, as well as the fluctuation conductivity.

The approximation parameters were determined by their variation to achieve a minimal average error of 1–4 % in the entire range of temperatures from T_c to 300 K. An analysis of these parameters in dependence of the conditions, namely the composition, the pressure, and the aging time allows one to reveal their effect on the mechanisms of the conductivity and the charge carriers scattering. We note that approximations using Eqs. (1)–(4) require 5 or even more fitting parameters. However, the electrical resistance in HTSCs is a result of simultaneous contributions of different mechanisms to the conductivity and the charge carriers scattering. This is why one generally does not expect to fit experiment data to simple relations with a small number of fitting parameters.

Figure 1 displays the change of the residual resistivity ρ_0 as a function of the transition temperature T_c which, in turn, is determined by the oxygen index δ in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [39] and $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [44] and the Pr content y in $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ [42] (here δ has an optimal value). Despite of the data scattering caused by the fact that in Eq. (1) $\rho_0 = \rho_n(T \rightarrow 0)$, one clearly sees a correlation between ρ_0 and T_c . Namely, a decrease of T_c , caused by the increase of the oxygen deficit or the increase of the Pr concentration, is accompanied by a rise of the residual resistivity. In this way, vacancies in the oxygen and yttrium subsystems as well as the praseodymium in this case are defects, scattering on which increases the residual resistivity.

In the temperature region $T_c \geq 60$ K, the changes in ρ_0 caused by the changes in the oxygen concentration in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [39] and $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [44] are

Fig. 1 Residual resistivity ρ_0 versus the superconducting transition temperature T_c in *open circle*— $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [39], *open square*—optimally doped $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ [42] and *open triangle*— $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals [44] (Color figure online)

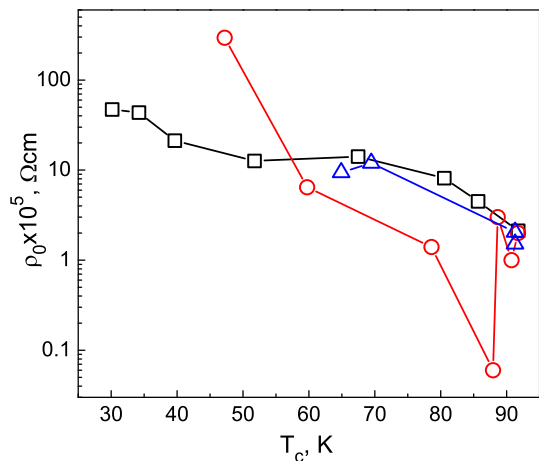
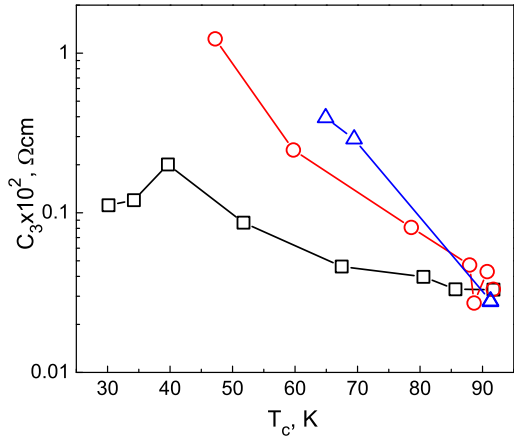


Fig. 2 Coefficient of the ideal phonon resistivity versus T_c . The designations are the same as in Fig. 1 (Color figure online)



rather close to those caused by the changes in the Pr concentration in the system $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ [42]. In Ref. [45], it was observed that in the system $YBa_2Cu_3O_{7-\delta}$, Pr substitutes not only Y but also Ba that also leads to the appearance of vacancies at the positions of Cu. This is why we believe that at high transition temperatures close to the maximal T_c , the changes in ρ_0 are largely stipulated by defects in the CuO_2 layers.

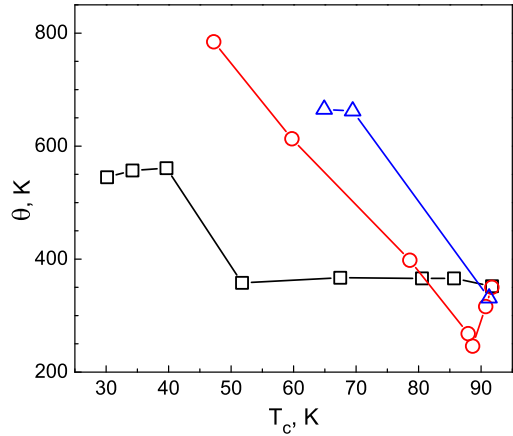
It should be noted that with the decrease of T_c from 90 to 30 K (in dependence on the oxygen deficit and the Pr concentration) the residual resistivity ratio $RRR = \rho(300\text{ K})/\rho_0$ varies from about 10 to ≈ 1.6 . Such RRR values are typical for metallic alloys of complex composition and this means that the contribution of the residual resistivity in the total resistivity is rather large in the entire temperature range from T_c to 300 K.

Figure 2 depicts the coefficient of the ideal phonon resistivity C_3 as a function of the superconducting transition temperature T_c . One sees that for $YBa_2Cu_3O_{7-\delta}$ [39] and $HoBa_2Cu_3O_{7-\delta}$ [44], the values of C_3 are rather similar and only at $T_c \leq 80$ K ($\delta \geq 0.15$ [27]) this parameter for $HoBa_2Cu_3O_{7-\delta}$ becomes larger than for $YBa_2Cu_3O_{7-\delta}$. At the same time for $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$, the coefficient of the ideal phonon resistivity C_3 is smaller than for $HoBa_2Cu_3O_{7-\delta}$ and $YBa_2Cu_3O_{7-\delta}$ at already rather small Pr concentrations ($y \geq 0.05$ [27]). In this way, the oxygen deficit influences scattering of electrons on phonons more strongly than praseodymium.

Figure 3 presents the Debye temperatures θ as a function of T_c which, in turn, is determined by the oxygen deficit [39,44] and the Pr concentration [42] at the optimal oxygen content. The values of θ correspond to the literature data, see [46,47].

Since $\Delta\theta(y)/\theta \approx -\alpha\Delta V/V + \beta\Delta f/f$ (ΔV is the change of the elementary cell, Δf is the change of the force constants), the growth of θ with increasing T_c for $YBa_2Cu_3O_{7-\delta}$ and $HoBa_2Cu_3O_{7-\delta}$ is connected with the increase of the force constants owing to the increase of the oxygen deficit. The behavior of θ for $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ is caused by the concurrence of both terms, and in the broad interval of the T_c values (50–90 K), the increase of the lattice parameter [48] is compensated by the increase of the force constants.

Fig. 3 The Debye temperature versus T_c . The designations correspond to those in Fig. 1 (Color figure online)



The correlation between T_c and θ for the case of strong coupling is described by the McMillan formula

$$T_c = \frac{\theta}{1.45} \exp \left[\frac{-1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right], \quad (5)$$

where λ is the electron–phonon interaction constant and μ^* is the Coulomb pseudopotential ($\lambda, \mu > 0$) [49]. The McMillan formula is correct at $\lambda \leq 1.5$ only [49]. In [49], are presented the data for correlation between T_c and λ (λ is calculated using the McMillan formula for $\mu^* = 0.13$) for 12 transition metals. In Fig. 4, these data are presented along with the cumulative data for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$, and $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$. One sees that only at $T_c \leq 70$ K, the data for transition metals agree with the data for HTSCs from the 1–2–3 system that attests to a strong electron–phonon interaction in these compounds. For $T_c \geq 70$ K, λ rapidly grows (in [49] $\lambda \geq 10$ corresponds to the superstrong coupling limit) and then transits to the negative area. In this way, HTSCs from the 1–2–3 system can be termed superconductors with a strong coupling only for $T_c \leq 70$ K; at higher T_c s, the McMillan formula is evidently not reasonable.

Formally, the applicability of the McMillan formula to HTSCs from the 1–2–3 system at lower T_c values is connected with the fact that upon change of the composition (increase of the disorder degree) of HTSCs, the change (namely, reduction) of T_c is more strong than the change of the Debye temperature. In other words, if superconductivity in the cuprates with a high degree of disorder (low T_c s) can be explained similar to superconductivity in alloys, then higher T_c s stipulated by the optimal oxygen deficit and the minimal degree of disorder are related to some other mechanisms which appear from the conventional ones upon reduction of the disorder degree and optimization of the oxygen content.

Figure 5 displays the dependence of T_c on the transverse coherence length $\xi_c(0)$. One sees that in $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$, where the reduction of T_c is caused by the increase of the Pr concentration at the optimum oxygen content, $\xi_c(0)$ increases with reduction of T_c that is typical for the conventional BSC theory. The reduction of $\xi_c(0)$ at $T_c \leq 35$ K

Fig. 4 Correlation between T_c and λ calculated by the McMillan formula: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (open circle), $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (open square) $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (open triangle), and transition metals (open diamond) [49] (Color figure online)

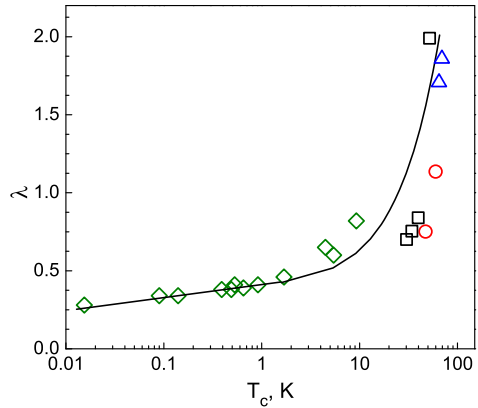
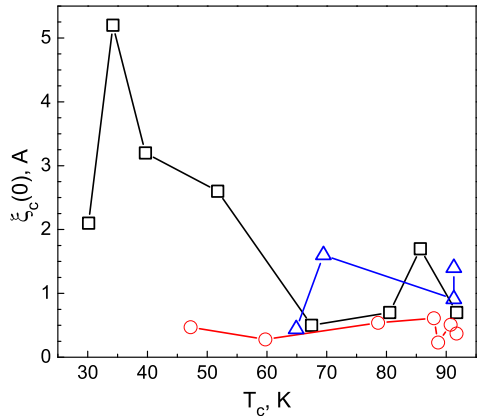


Fig. 5 Dependence of the transverse coherence length $\xi_c(0)$ on the superconducting transition temperature T_c . The designations correspond to those in Fig. 1 (Color figure online)

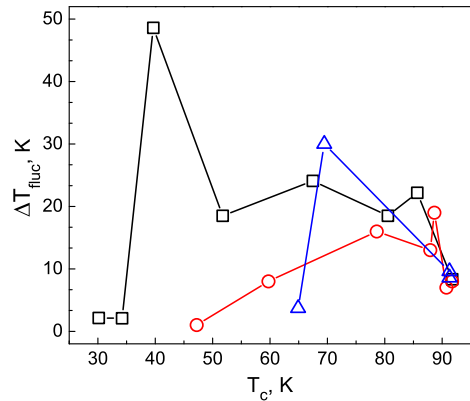


is related in this case to a structural reordering which also leads to the appearance of a semiconductor-like contribution in the resistance of $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ [24]. This attests to a transition from the regime in which the resistance is determined by the electron mean-free path to the regime where the resistance is determined by the changes in the electronic structure.

For $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$, where the reduction of T_c is caused by the increase of the oxygen deficit, which in turn gives rise to the oxygen mobility, the correlation between T_c and $\xi_c(0)$ is not seen. We note that the values of the transverse coherence length $\xi_c(0)$ obtained from the approximation of the experimental data to Eqs. (2)–(4) are substantially smaller than the interlayer distance (11.7 \AA [48]) that attests to the 3D character of superconducting fluctuations, as assumed in Eq. (4).

In this way, the correlation between $\xi_c(0)$ and T_c^{-1} is observed for the system $\text{Y}_{1-y}\text{Pr}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ only; for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the reduction of T_c owing to the increase of the oxygen deficit (increase of the concentration of the labile oxygen) weakly influences the behavior of the transverse coherence length, at least in the investigated range of concentrations of the labile oxygen.

Fig. 6 Dependence of the interval of superconducting fluctuations $\Delta T_{\text{fluct}} = T^* - T_c$ on the superconducting transition temperature T_c . The designations are the same as in Fig. 1 (Color figure online)



In Fig. 6, is presented the dependence of the interval of superconducting fluctuations $\Delta T_{\text{fluct}} = T^* - T_c$ on the critical temperature T_c . We note that according to [40], an important consequence of the presence of fluctuational Cooper pairs above T_c is a reduction of the one-electron density of states at the Fermi level. In this sense, one can speak of the opening of the fluctuation gap at the Fermi level, which begins at $T = T^*$. With reduction of T down to T_c , the fluctuation pseudogap turns into the conventional superconducting gap. In this way, the temperature T^* introduced in [41] can be considered as a temperature of the pseudogap opening. In Fig. 6, one sees that the reduction of T_c caused by the change of the defect concentration and the composition leads, in the very end, to the vanishing of the fluctuation conductivity. The range for the latter to exist tends to zero as in conventional superconductors. This correlates with the behavior of the electron–phonon interaction constant (Fig. 4), which at low T_c is close to that for transition metals. From Fig. 6, it follows that the most broad range of existence of superconducting fluctuations and, hence, the pseudogap regime corresponds to the intermediate values of T_c but not to the maximal ones. A certain role in this can be played by specific quasiparticles scattering mechanisms [50–53] appearing in consequence of the presence of structural and kinematic anisotropies in the studied compounds.

3 Conclusion

To summarize, the temperature dependences of the basal-plane resistance above T_c in various high-temperature superconductors from the 1–2–3 system can be described by scattering of the charge carriers on phonons and defects, in conjunction with the effect of the fluctuation conductivity. The parameters of the electron–phonon interaction, primarily the residual resistivity ratio and the Debye temperature have values typical for metallic alloys of complex composition. The application of the McMillan formula reveals that at strong disorder degrees (low T_c s), superconductivity in the investigated cuprates is similar to conventional superconductivity in alloys. To the same attests, the behavior of the range of existence of superconducting fluctuations as a function of T_c , which rapidly shrinks at low T_c . At the optimal oxygen deficit and low disorder

degrees (maximal T_{cS}), superconductivity in the investigated cuprates is likely to be related to some other mechanisms. The increase of the oxygen deficit (increase of the concentration of the labile oxygen) affects $\xi_c(0)$ slightly. At the same time, substitution of Pr instead of Y leads to the appearance of a correlation of $\xi_c(0)$ with T_c^{-1} that is also similar to the behavior of the transverse coherence length in classical superconducting alloys.

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