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Effect of oxygen deficiency (δ) on normal state resistivity of $YBa_2Cu_3O_{7-\delta}$ superconductors

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Abstract. **We have investigated theoretically the effect of oxygen deficiency (δ) on normal state resistivity (ρ) as well as its temperature dependence in $YBa_2Cu_3O_{7-\delta}$ superconductors. This has been based on a potential which incorporates the structure factors and various interactions for double two-dimensional (2-D) conducting CuO_2 plane. Using the Coulomb and electron–phonon terms of the interaction potential, we have then worked out the coupling strength (γ) for neighbouring electrons linked via 2-D acoustic phonons ($\hbar\omega_-$). Furthermore, the scattering time ($\tau_{e\text{-}ph}$) due to electron–phonon interaction is deduced. The variations in $\tau_{e\text{-}ph}$ and $\rho_{e\text{-}ph}$ are studied with oxygen deficiency (δ) which is in the range of $0{\cdot}0 \le \delta \le 1{\cdot}0$, and the results thus obtained are found to be consistent with the earlier reported data. The residual resistivity ρ_0 obtained by extrapolation from experimental data together with $\rho_{e\text{-}ph}$ will predict the nearly-linear behaviour of normal state resistivity at temperatures (T) [$90 \le T \le 300$ K] in $YBa_2Cu_3O_{7-\delta}$ superconductors.**

Keywords. **Oxygen deficiency; electron–phonon coupling strength; inelastic scattering rate; normal state resistivity.**

1. Introduction

High temperature copper oxide superconductors of the type $YBa_2Cu_3O_{7-\delta}$ (Wu *et al* 1987) have been the focus of theoretical and experimental studies. Intensive experimental investigations have yielded the existence of two-dimensional CuO_2 planes responsible for contributing to the anomalous transport properties. The study of $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconductors is of particular interest because its crystal structure contains double two-dimensional (2-D) CuO_2 planes as well as single one-dimensional (1-D) CuO chains. The relative importance of chains and planes is still not clear. Efforts have been made in the recent past to look for the pairing mechanism as well as the anomalous physical properties (Ginsberg 1990). As regards to the transition temperature, YBCO possess a transition temperature (T_c) of 90 K higher than that of LaSrCuO superconductor and has a single 2-D CuO_2 plane in a unit cell. The study of normal state transport properties as resistivity, is of specific interest for it offers clues to the function of their counterparts—the inelastic scattering rate mechanisms. The behaviour of normal state resistivity (ρ) as functions of the oxygen deficiency (δ) and temperature (T) in the polycrystalline samples of YBCO superconductors have earlier been investigated by several experimental groups (Cava *et al* 1987; Tarascan *et al* 1987; Hariharan *et al* 1988).

Tarascan *et al* (1987) have reported the normal state resistivity (ρ) behaviour of polycrystalline $RBa_2Cu_3O_{7-\delta}$ (R = Nd, Sm, Eu, . . . , Lu and Y), and obtained a roughly linear temperature dependence at high temperatures. Subsequently, Hariharan *et al* (1988) measured the normal state behaviour of resistivity for oxygen and air-annealed samples (in single phase) of YBaCuO superconductors and reported the behaviour of ρ as a function of oxygen deficiency (δ). They found a correlation between the transition temperature (T_c) and the resistivity, and showed that for lower values of δ where the T_c is maximum, the resistivity is minimum; and with the increase in δ values, the T_c decreases while ρ increases. The Bloch–Boltzmann theory (Ziman 1960) explains the normal state transport properties of the conventional superconductors. The linear temperature dependence of the normal state resistivity of high T_c cuprates has always been a puzzle for theorists, and one of the checks for the theory to be correct was this dependence. The linear temperature dependence of the normal state resistivity can be explained on the basis of different assumptions such as RVB (Anderson and Zou 1988) and spin fluctuations (Morita *et al* 1990). Recently, Varshney and Singh (1995) have explained the behaviour

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of normal state resistivity as a function of doping concentration and temperature of mono CuO_2 layer LaSrCuO superconductors, based on Bloch–Boltzmann theory within the framework of a free electron layered electron gas (FELEG) approach.

Recently, Singh *et al* (1996) and Varshney *et al* (1996) have made efforts to develop an interaction potential for YBaCuO superconductors with double 2-D conducting CuO_2 plane in a unit cell. The approach incorporates the structure factors and various interactions involved, and predicts the oxygen deficiency dependence of transition temperature. Motivated from the earlier success of superconducting state property on YBCO systems and the normal state resistivity behaviour in LSCO superconductors, we thought it pertinent to understand the reported behaviour of ρ as a function of oxygen deficiency (δ) and temperature (T) in the YBCO systems. We have used the electron–phonon interaction part of the interaction potential to obtain the expressions for the coupling strength between the neighbouring electrons and the inelastic scattering rate, to deduce the normal state resistivity.

The plan of the present investigations are as follows. In § 2 we briefly introduce the earlier developed interaction potential for YBCO systems with double 2-D planes in a unit cell. In the analysis for normal state resistivity, we have separated the electron–phonon and electron–plasmon interactions and used the former following the Bloch–Boltzmann theory to interpret the reported behaviour. The expressions for coupling strength linking between two electrons and the inelastic scattering rate are then worked out to evaluate the resistivity due to electron–phonon interaction. The expressions obtained are computed numerically and the results thus obtained are analysed and discussed in § 3.

2. The model

The single crystal of Y-based cuprate superconductors is modelled to be an array of planes of Y, CuO_2, BaO, oxygen-deficient CuO (chain), CuO_2, BaO, CuO_2, and Y. In a unit cell of the crystal, two conducting planes are considered, and the non-conducting planes between CuO_2 planes are regarded as an uniform dielectric medium with a background dielectric constant, ε_0. The unit cell dimension along c axis is d.

The effective interaction potential for a single conducting CuO_2 plane is represented (Varshney and Singh 1995) as:

$$V(q, q_z, \omega) = \frac{2\pi e^2}{q\varepsilon_0 \varepsilon(q, q_z, \omega)} S(q, q_z), \tag{1}$$

where, $\varepsilon(q, q_z, \omega)$ is the dielectric response function, and for a single band of charge carriers

$$\varepsilon(q, q_z, \omega) = 1 + 2P(q, \omega)\, S(q, q_z) + P^2(q, \omega) S(q, q_z)\, S'(q), \tag{2}$$

with

$$P(q, \omega) = \frac{-2\pi e^2}{q\varepsilon_0} \prod (q, \omega), \tag{3}$$

$$S(q, q_z) = \frac{\sinh(qd)}{\cosh(qd) - \cos(q_z d)}, \tag{4}$$

and

$$S'(q) = \frac{\cosh(qd) - \cosh(qd')}{\sinh(qd)}. \tag{5}$$

Here, $d' = 2d_1 - d$ and $d' = d/3$. The in-plane wave vector, denoted as q and q_z, is in the z-direction. Keeping in mind the anisotropy in physical properties, we wish to restrict ourselves to the a–b plane conduction and hence averaging (1) over q_z as

$$V(q, \omega) = \frac{\mathrm{d}}{2\pi} \int_{-\pi/\mathrm{d}}^{+\pi/\mathrm{d}} V(q, q_z, \omega)\, \mathrm{d}q_z. \tag{6}$$

The integral can thus be evaluated by making use of (1) and (6) to get

$$V(q, \omega) = \frac{4\pi e^2}{q\varepsilon_0} \frac{[1 + P(q, \omega) S'(q) + R(q, \omega)] \sinh(qd)}{[\,|D^2(q; \omega) - 1|\,]^{1/2}}, \tag{7}$$

with

$$R(q, \omega) = \frac{\sinh[q(d - d_1)] + D(q, \omega) \sinh(qd_1)}{\sinh(qd)}, \tag{8}$$

and

$$D(q, \omega) = \cosh(qd) + 2P(q, \omega) \sinh(qd) + P^2(q, \omega) \sinh(qd) S'(q). \tag{9}$$

The polarization function $P(q, \omega)$ is due to charge carriers and ions as

$$P(q, \omega) = P_c(q, \omega) + P_i(q, \omega), \tag{10}$$

$$= \frac{\Omega_1^2}{A^2 - \omega^2} - \frac{\Omega_2^2}{\omega^2}, \tag{11}$$

where, Ω_1 and Ω_2 are the 2-D electron and ion plasmon frequency and are essentially expressed as:

$$\Omega_1^2 = \frac{2\pi n_c e^2 q}{m^* \varepsilon_0}, \tag{12}$$

$$\Omega_2^2 = \frac{2\pi n_i Z^2 e^2 q}{M\varepsilon_0}, \tag{13}$$

$$A^2 = q^2 V_F^2/2 . \tag{14}$$

Here, n_c [cm^{-2}], n_i [cm^{-2}], m^* [m_e], M [amu], and Ze are the 2-D charge carrier density, ionic areal density, effective mass, mass of the unit cell and the sum of ionic as well as electron bound charge lying on the conducting CuO_2 plane, respectively. We represent V_F as the Fermi velocity.

Zeros of the longitudinal dielectric function will yield the frequencies of coupled 2-D acoustic plasmon and phonon modes. The resonant frequencies of the coupled modes in the long wavelength limit ($q_z \to 0$) are described as:

$$2\omega_\pm^2 = [(\Omega_1^2 + \Omega_2^2)\alpha + A^2] \pm [\{[\Omega_1^2 + \Omega_2^2]\alpha + A^2\}^2 - 4\alpha A^2 \Omega_2^2]^{1/2}, \tag{15}$$

with $\alpha = qd$. Further simplification will yield the two coupled modes, i.e. $\hbar\omega_+(q)$ and $\hbar\omega_-(q)$ as

$$\omega_-^2(q) \cong \alpha\Omega_2^2 [1 + \alpha\Omega_1^2 A^{-2}]^{-1}. \tag{16}$$

Here, we have used an approximation that $\Omega_1 \gg \Omega_2$. Also, the higher frequency mode $\omega_+(q)$ is

$$\omega_+^2(q) \cong \alpha\Omega_1^2 + A^2. \tag{17}$$

These modes follow the dispersion relations for 2-D acoustic phonon and plasmon modes as:

$$\omega_-(q) = cq , \tag{18}$$

and

$$\omega_+(q) = aq^{1/2} + bq , \tag{19}$$

with a, b and c as constants.

The normal state resistivity is well explained from the linearized Bloch–Boltzmann transport equation where the electron–phonon interaction is dominant (Ziman 1960). The evaluation of electron–phonon scattering rate and hence the resistivity essentially requires the coupling strength $\gamma_{e\text{-}ph}(q)$ linking electrons at the frequencies $\omega_-(q)$. Considering only the Coulomb and electron–phonon interaction part of the interaction potential, we obtained the coupling strength from the residue of $V(q, \omega)$ at the frequency ω_- (Varshney and Singh 1995) as:

$$|\gamma_{e\text{-}ph}(q)|^2 = \frac{\pi e^2 \hbar^2}{q\varepsilon_0} \frac{\omega_-^2(\omega_-^2 - A^2)}{(\omega_-^2 + \omega_+^2)} , \tag{20}$$

which on simplification becomes,

$$\frac{\gamma_{e\text{-}ph}}{\omega_-} = \frac{\pi e^2 \hbar^2}{q\varepsilon_0} \frac{(D' - A^2)}{(D'' - A^2)} , \tag{21}$$

with

$$D' = \alpha[\Omega_2^2 - \Omega_1^2] , \tag{22}$$

and

$$D'' = \alpha[\Omega_2^2 - 2\Omega_1^2] . \tag{23}$$

A feature of the copper oxide superconductors is that, at T_c, the inelastic scattering rate is very large. We have deduced the mean time it takes to absorb or emit a phonon of energy $\hbar\omega_-(q)$, which is included in the resistivity expression. Thus, the mean time is expressed as:

$$\frac{1}{\tau_{e\text{-}ph}} = \frac{2\pi}{\hbar} \int_0^{2k_F} \frac{d^2q}{(2\pi)^2} |\gamma_{e\text{-}ph}(q)^2| (1 - \cos\theta) \times [n_q \delta(\varepsilon_{k+q} - \varepsilon_k - \hbar\omega_-) + (n_q + 1)\delta(\varepsilon_{k+q} - \varepsilon_k + \hbar\omega_-)] . \tag{24}$$

Here, the scattering of the charge carriers at the Fermi surface is considered for all possible values of scattering angle θ. The scattering angle between k and k + q is denoted as $\cos\theta$. The phonon occupancy factor is represented as n_q and in the high temperature limit it is reduced to $k_B T/\hbar\omega_-$ as the Debye temperature YBaCuO superconductors is of the order of 400 K (Sankar *et al* 1988). Using the exact form of (24) and substituting in (21),

$$\frac{1}{\tau_{e\text{-}ph}} = \frac{m^* k_B T}{\pi\hbar^4 k_F^3} \int_0^{2k_F} \frac{|\gamma_{e\text{-}ph}|^2}{\omega_-} \frac{q^2 dq}{[1 - (q/2k_F)^2]^{1/2}} . \tag{25}$$

Integrating over the two-dimensional wave vector (q), the final expression can be obtained as:

$$\frac{1}{\tau_{e\text{-}ph}} = \frac{2\pi e^2 m^* k_B T}{\hbar^3 k_F \varepsilon_0} \frac{[V' - V_F^2]}{[V'' - V_F^2]} , \tag{26}$$

with

$$V' = (\Omega_2^2 - \Omega_1^2) dq^{-1} , \tag{27}$$

and

$$V'' = (\Omega_2^2 - 2\Omega_1^2) dq^{-1} . \tag{28}$$

Usually, the δ function fixes the angle between the energies in 2-D case and hence a nearly linear temperature dependence is obtained of the inelastic scattering rate. The normal state resistivity due to electron–phonon interaction in terms of scattering time is expressed as:

$$\rho_{\text{e-ph}} = [m^*/ne^2]\,[1/\tau_{\text{e-ph}}]\,, \tag{29}$$

where n is the carrier density in cm^{-3}. Using (26) and (29), the temperature dependent part of the normal state resistivity due to the scattering of 2-D acoustic phonons is obtained as:

$$\rho_{\text{e-ph}} = \frac{2\pi m^{*2} k_B T}{\hbar^3 k_F \varepsilon_0 n}\,\frac{[V' - V_F^2]}{[V'' - V_F^2]}\,. \tag{30}$$

In cuprate superconductors, scattering of charge carriers via phonons as well as scattering via defects and impurities exist. Moreover, the scattering of charge carriers due to impurities is temperature independent. The temperature dependent part of the resistivity is attributed to electron–phonon scattering process.

The two-component model for resistivity can thus be expressed as:

$$\rho_{\text{total}}(T) = \rho(0) + \rho_{\text{e-ph}}(T), \tag{31}$$

with $\rho(0)$ as the residual resistivity developed due to impurity scattering.

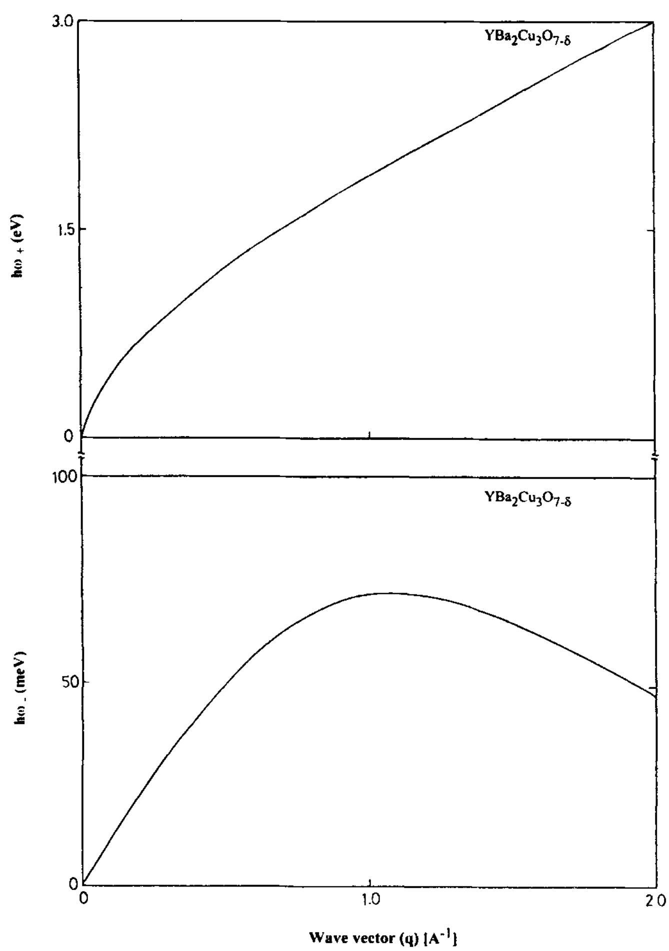

Figure 1. Variations of 2-D acoustic plasmon (phonon) energy $\hbar\omega_+(\hbar\omega_-)$ as a function of wave vector (q) [$Å^{-1}$].

Following the Bloch–Boltzmann theory of normal state resistivity, we have obtained the temperature and oxygen deficiency dependence of resistivity in YBaCuO superconductors. The analysis is presented and discussed in the following section.

3. Results and discussion

To compute the normal state resistivity (ρ) as a function of oxygen deficiency (δ) and temperature (T) in YBaCuO superconductors, we have used the value of effective mass $m^* = 1{\cdot}5\ m_e$ which is in accordance with the specific heat measurement data (Inderhees *et al* 1987). The background dielectric constant, ε_0 is taken as 4·5 (Bozovic 1990) from the electron energy loss spectroscopy. Using $a = 3{\cdot}8136$ Å, $b = 3{\cdot}8845$ Å and $c = 11{\cdot}6603$ Å (Cava *et al* 1990), we obtained n_c as $2{\cdot}38 \times 10^{14}\,cm^{-2}$ and $n_i = 6{\cdot}74 \times 10^{14}\,cm^{-2}$ for the condition of optimized pairing in cuprate superconductors. We make them oxygen deficiency dependent as:

$$n_{c(i)} = n_{c(i)}\{\delta = 0{\cdot}0\}\,[\exp - (\delta/\delta_c)^2]\,, \tag{32}$$

with $\delta_c = 0{\cdot}4$. The sum of ionic and bound electronic charge $Ze = -2e$. M denotes the reduced mass of Cu and O in CuO_2 plane and is 12·77 amu indicating that the vibration considered is the CuO vibration.

The 2-D acoustic plasmon frequency, ω_+ and phonon frequency, ω_- were plotted from (17) and (16) as a function of wave vector ($q = 2k_F \sin\theta$) in $Å^{-1}$ along the a–b plane for YBaCuO superconductors in figure 1. The higher frequency mode $\hbar\omega_+(q)$ displays a characteristic

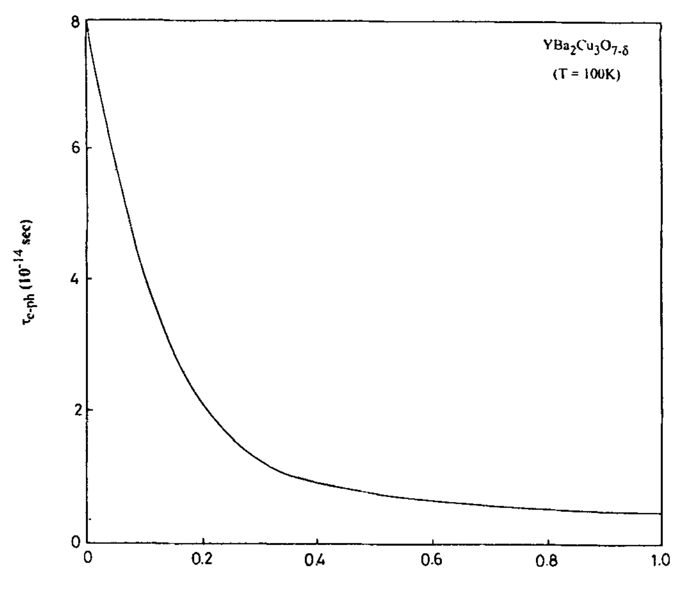

Figure 2. Variations of electron–phonon scattering time ($\tau_{\text{e-ph}}$) as a function of oxygen deficiency (δ).

square root behaviour for small wave vector and becomes linear for higher values of q. This is consistent with the 2-D acoustic plasmon dispersion relation. The plot of $\hbar\omega(q)$ as a function of q is characterized by the low acoustic phonon frequencies. Using the relation $V_s = ak_B\omega_-/\hbar\pi$, the long wavelength sound velocity was estimated. Using $a = 3{\cdot}8136$ Å and $\omega_- = 46$ meV, $V_s = 6{\cdot}2 \times 10^5$ cm sec^{-1} was deduced, which is higher than the reported data on single crystal of YBaCuO superconductors with the ultrasound velocity measurements as $5{\cdot}0 \times 10^5$ cm sec^{-1} (Saint Paul *et al* 1989).

It is evident from (26) that the scattering rate between electrons and phonons is inversely proportional to the temperature (T). The calculated value of τ_{e-ph} is plotted for different oxygen deficiency values in figure 2 at $T = 100$ K for the YBaCuO systems. The figure shows that the electron–phonon scattering time τ_{e-ph} is high for the lower values of δ and decreases with the increased value of oxygen deficiency. Timusk and Tanner (1989) had pointed out that the inelastic scattering rate is very large and of the order of T_c in cuprate superconductors. We deduced $\tau_{e-ph} = 8{\cdot}0 \times 10^{-14}$ sec at $\delta = 0{\cdot}0$. The obtained variation of τ_{e-ph} with δ could not be compared due to paucity of data. Furthermore, the scattering time due to electron–phonon interaction is rather insensitive for higher oxygen deficiency as the system approaches the insulating phase. Besides this, the presence of multiple scattering mechanism lowers the scattering time value and oxygen deficiency independently. Earlier, Kim *et al* (1989, 1991) had evaluated the electron–phonon life time by solving the Bloch–Boltzmann transport equation variationally and the Eliashberg function in LaSrCuO superconductors. Their analysis predicts that τ_{e-ph} is rather insensitive in the metallic region in La based cuprates. In YBaCuO superconductors, the result on τ_{e-ph} dependence with δ is consistent with those obtained by Kim *et al* (1989, 1991).

The normal state resistivity due to electron–phonon interaction at room temperature is plotted with oxygen deficiency (δ) in YBaCuO superconductors along with the reported data (Hariharan *et al* 1988) on pellets in figure 3. The ρ_{e-ph} includes the extrapolated value of residual resistivity ρ_0. It is noticed from the graph that at low oxygen deficiency level, ρ_{e-ph} is lower and is almost δ independent. For $\delta \geq 0{\cdot}4$, ρ_{e-ph} increases and it increases further monotonously for $\delta \geq 0{\cdot}7$. The lower value of ρ_{e-ph} in the range $\delta \leq 0{\cdot}4$ signified the metallic origin. The deduced results on the variation of ρ with δ are consistent with those reported earlier by Hariharan *et al* (1988) for oxygen-annealed YBaCuO superconductors.

In figure 4, we have plotted the behaviour of normal state resistivity as a function of temperature for YBaCuO superconductors with $\delta = 0{\cdot}0$. For this purpose, we have used the developed expression (30) and compared them with the experimental data (Tarascan *et al* 1987). It is found that the temperature dependence of ρ is almost linear at high temperatures in the range $90 \leq T \leq 300$ K and is consistent with the experimental data. The higher values of resistivity is indeed due to the large inelastic scattering rate. It may be pointed out that for a inelastic scattering rate of $\approx 10^{14}$ sec^{-1} one obtained ρ of the order

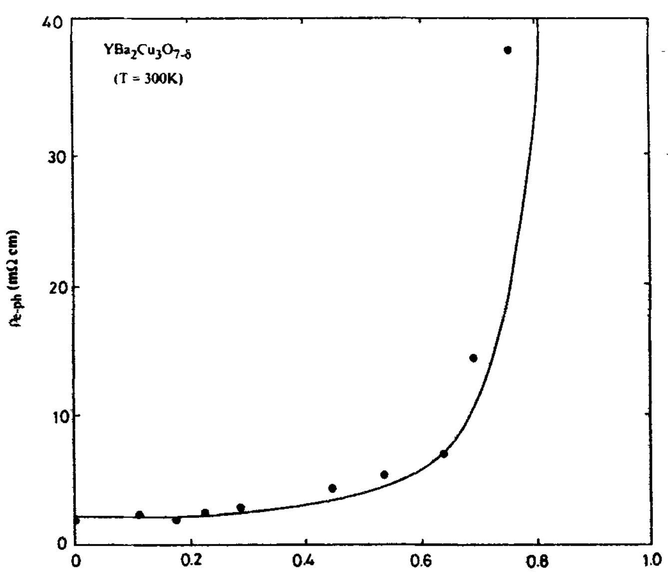

Figure 3. Variations of normal state resistivity ρ_{e-ph} with oxygen deficiency (δ) along with the experimental data (•) from Hariharan *et al* (1988).

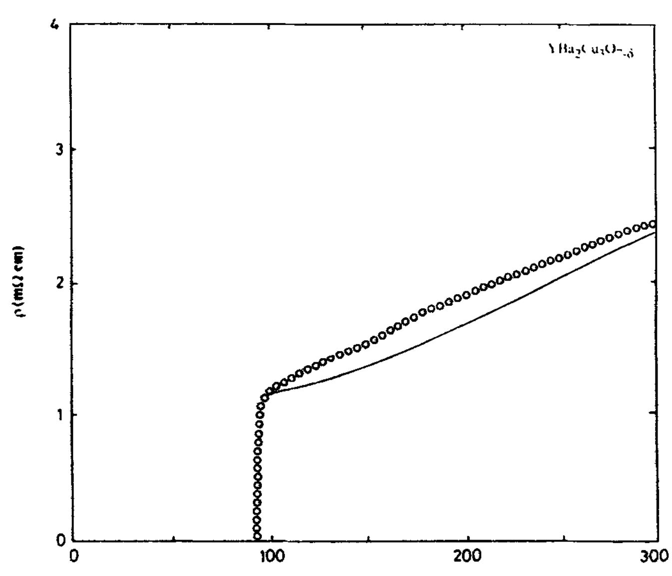

Figure 4. Variations of normal state resistivity (ρ) of YBaCuO superconductors as a function of temperature. Experimental data (•) refer to Tarascan *et al* (1987).

of mΩ cm. Furthermore, the above analysis yields a finite intercept at a temperature when ρ is extrapolated to zero. The intercept is interpreted in terms of residual resistivity that is temperature independent, and arises from the impurity scattering mechanism. We have estimated $\rho(0) = 1.14$ mΩ cm from the extrapolation of experimental data. We believe that in YBaCuO superconductors the nearly linear behaviour of normal state resistivity with temperature is definitely due to the inelastic scattering of electrons via phonons.

4. Conclusions

In the present investigation, we have made an effort to understand the observed behaviour of normal state resistivity with oxygen deficiency as well as temperature in YBaCuO superconductors. We have used the earlier developed potential which properly incorporates the structure factors and various interactions for double 2-D conducting CuO_2 planes. For the purpose of normal state resistivity we have used the Coulomb and electron–phonon part of the interaction potential and Bloch–Boltzmann theory. We have succeeded in understanding the reported behaviour of ρ with δ and T in YBaCuO systems within a two-component model. The appropriateness of the above approach is attributed to the proper care of layered structure and the estimation of the model parameters as $\hbar\omega_-$, $\gamma_{e\text{-}ph}$ and $\tau_{e\text{-}ph}$ based on the realistic physical parameters. The consistency of the normal state resistivity data with the experimental data strongly supports the earlier proposed approach for pairing mechanism and transition temperature in YBaCuO superconductors. The estimated value of long wavelength sound velocity and inelastic scattering rate are consistent with the earlier reported data and favours the strong participation of phonons. In conclusion, the two-component model based on Bloch–Boltzmann theory for normal state resistivity in YBaCuO superconductors can explain fairly well the reported data on polycrystalline samples.

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