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Composition dependence of transition temperature in some ceramic superconductors

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Abstract. The composition dependence of transition temperature in some ceramic superconductors ($La_{2-x}(Ba, Sr)_xCuO_4$) was studied by modifying our earlier approach and developing a Fourier-transformed effective potential which involves the effect of two-dimensional (2D) acoustic plasmons. This potential was used to obtain the pairing (electron–electron attraction) parameter (λ), the averaged Coulomb repulsive parameter (μ^*) and the cut-off 2D acoustic plasmon frequency (ω_c) required to compute the superconducting transition temperature (T_c) from the strong coupling theory. The variations of T_c with compositions (x) obtained for $La_{2-x}(Ba, Sr)_xCuO_4$ show reasonably good agreement with experimental data.

Keywords. Plasmons; phonons; transition temperature; coupling parameter; superconductors.

1. Introduction

The discovery of high temperature superconductivity (HTSC) in rare-earth (RE) cuprates (Bednorz and Muller 1986) has led to unprecedented research activity. The doping of transition and RE elements into the copper oxide ceramics plays a crucial role in determining their normal and superconducting properties. For instance, the phase-transition from normal to superconducting state, the stabilization of superconducting phase, and the maximum value of the critical temperature (T_c) strongly depend on the doping concentration (x) of these elements. Various superconducting properties of $La_{2-x}(Ba/Sr)_xCuO_4$ and those of similar materials have earlier been investigated as a function of x (Bednorz *et al* 1987; Dover *et al* 1987; Jha 1987; Shafer *et al* 1987; Weber 1987; Markert *et al* 1988; Moodenbaugh *et al* 1988; Torrance *et al* 1988; Seaman *et al* 1989; Tewari and Gumber 1990; Singh *et al* 1991).

Recently, Dover *et al* (1987) and Shafer *et al* (1987) measured T_c as a function of x in $La_{2-x}Sr_xCuO_4$ system and found that T_c first increased with x and then decreased after reaching its maximum value at $x = 0{\cdot}15$. Subsequently, Torrance *et al* (1988) observed that the variation of T_c was below 5 K for $0{\cdot}06 \leqslant x \geqslant 0{\cdot}30$ in $La_{2-x}Sr_xCuO_4$ with its maximum ($T_c = 36$ K) at $x = 0{\cdot}15$. However, it behaved like a normal metal at $x > 0{\cdot}30$. Moodenbaugh *et al* (1988) also observed the variation of T_c with x in $La_{2-x}Ba_xCuO_4$ system and found that T_c had two maximum values, one at $x = 0{\cdot}09$ and another at $x = 0{\cdot}15$. Further, Bednorz *et al* (1987) and Markert *et al* (1988) reported the variation of T_c as a function of x in $La_{2-x}Ca_xCuO_4$ and $La_{2-x}Na_xCuO_4$ superconductors, respectively. Motivated from these experimental observations, we thought it pertinent to perform an extensive theoretical analysis of the composition dependence of T_c in $La_{2-x}(Ba/Sr)_xCuO_4$.

Singh *et al* (1991) have recently computed the variation of T_c with x in $La_{2-x}(Ba/Sr)_xCuO_4$ superconductors using the weak coupling theory of Kresin (1987).

Although their results are in good agreement with experimental data for the intermediate range of x ($0{\cdot}06 \leqslant x \leqslant 0{\cdot}31$), the agreement is not so good for the lower and higher concentrations (x). These deviations between experimental and earlier values of T_c calculated by Singh *et al* (1991) might be due to the following limitations present in their interaction potential: (a) it does not take proper account of the involvement of electronic excitations (2D acoustic plasmons) in the pairing mechanism and (b) their formalism to calculate the cut-off frequency and the coupling parameter for plasmons does not properly include the effect of structural changes in $La_{2-x}(Ba/Sr)_xCuO_4$ superconductors.

In this paper, we have attempted to eliminate the drawbacks of our earlier results (Singh *et al* 1991). The main features of our modified approach are (i) the transition temperature has been computed from the formalism based on the strong coupling theory of Eliashberg (1960) and (ii) the model parameters (coupling parameter (λ), modified averaged Coulomb repulsion (μ^*), and upper cut-off 2D acoustic plasmon frequency (ω_c)) are calculated by using the effective interaction potential (Jha 1987; Sharma 1989). The essentials of the potential formulation are presented in § 2 and the computed results in § 3.

2. Theoretical formalism

The change in concentration (x) in $La_{2-x}(Ba/Sr)_xCuO_4$ superconductors introduces two types of changes in the system. The first is in carrier states and free-carrier density (n_c) and the other in crystal structure. The usual bulk properties such as phonon, plasmons, etc. are not expected to change significantly for small variations in x. Therefore, one cannot expect that BCS type theory, based on the pairing of charge carriers by the exchange of only phonons, is capable of explaining the drastic change in T_c as a function of x. This, in turn, suggests the involvement of the collective electronic excitations (plasmons, Bozovic 1990) along with phonons in the pairing of the current carrying charge carriers.

The present paper thus includes the effects of electronic excitations (2D acoustic plasmons) in V_{eff} used for the evaluation of T_c and potential parameters (λ, μ^* and ω_c) in $La_{2-x}(Ba/Sr)_xCuO_4$. The model interaction potential has been formulated for $La_{2-x}(Ba/Sr)_xCuO_4$ and allied superconductors, based on the assumptions that (i) there is only one conducting Cu–O plane per unit cell, (ii) the Cu–O planes form an infinite array of planes along c-axis, (iii) the non-conducting planes between Cu–O planes are considered as an uniform dielectric medium with a background dielectric constant (ε_∞), (iv) there are two types of acoustic plasmons corresponding to the electronic motion along and perpendicular to a–b plane, (v) only long wave acoustic plasmons along a–b motion of electrons (Bozovic 1990) take part in the pairing mechanism, and (vi) in the long wavelength limit, the Coulomb interactions between the charges on nearby CuO planes have a significant influence (Fetter 1974) on the interaction potential. In order to discuss the possible changes in the dielectric screening of these nearby planes, we have considered the screened Coulomb potential for a series of identical planes (see figure 1).

The Fourier-transformed effective interaction potential has been expressed as (Jha 1987; Sharma 1989)

$$V_{\text{eff}}(q, q_z, \omega) = \frac{2\pi e^2 S(q, q_z)}{\varepsilon_\infty q \varepsilon(q, q_z, \omega)} \tag{1}$$

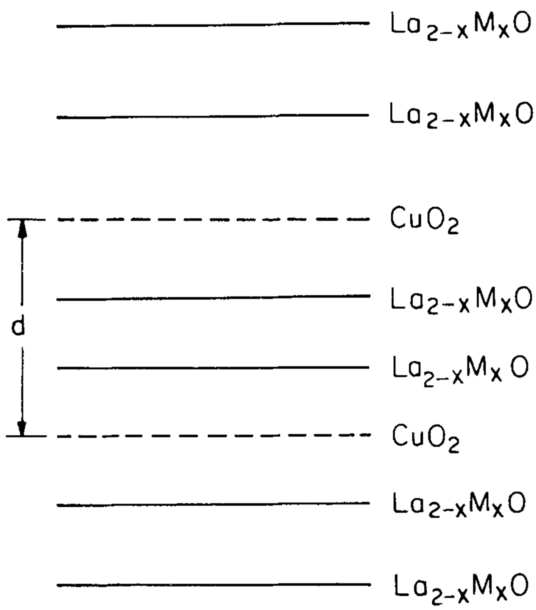

Figure 1. Schematic diagram of layered structure of cuprate superconductors.

in terms of the structure factor (Fetter 1974)

$$S(q,q_z)=\frac{\sinh(qd)}{\cosh(qd)-\cos(q_z d)}\simeq\frac{qd}{\phi} \tag{2}$$

for small values of the wave vector (q) and $\phi=1-\cos\theta$ with θ as an angle between q_z and d. Here, d is the distance between the consecutive Cu–O planes, q and q_z, the components of the momentum along and perpendicular to the Cu–O plane, and ε_∞, the high frequency background dielectric constant of the surrounding material.

In the limit $d\to\infty$, V_{eff} reduces to a single plane result and $d\to 0$ to an ordinary three-dimensional electron potential. Thus, we can rewrite (1) as

$$V_{\text{eff}}(q,q_z,\omega)=\frac{2\pi e^2 d}{\varepsilon_\infty}R(q,q_z,\omega) \tag{3}$$

with

$$R(q,q_z,\omega)=\frac{1}{\phi\,\varepsilon(q,q_z,\omega)} \tag{4}$$

and

$$\varepsilon(q,q_z,\omega)=1+P(q,\omega)S(q,q_z) \tag{5}$$

as expressed by Sharma (1989). Here, $P(q,\omega)$ is the polarization function due to the charge carriers (c) and the ions (i) expressed as

$$P(q,\omega)=P_c(q,\omega)+P_i(q,\omega) \tag{6}$$

$$=\frac{2\pi e^2 n_c q}{m^*\varepsilon_\infty[(q^2v_F^2/2)-\omega^2]}-\frac{\Omega_i^2 qd}{2\varepsilon_\infty\omega^2}, \tag{7}$$

where n_c and v_F, being the 2D carrier density and Fermi velocity of charge carriers, respectively.

Using (3) to (7) we can obtain the value of the function $R(q,q_z,\omega)$ as

$$R(q,q_z,\omega)=\frac{\omega^2}{\phi\omega(\omega+i\delta)-Bq^2} \tag{8}$$

with

$$B=2d\left[\frac{v_F^2}{2a^*}+\frac{\Omega_i^2 d}{4\varepsilon_\infty}\right] \tag{9}$$

where ω is the frequency, Ω_i, the usual three-dimensional ionic plasma frequency and a^*, the effective Bohr radius.

The effective interaction between the charge carriers in a given conducting Cu–O layer can now be expressed as

$$V_{\text{eff}}(q,\omega)=\frac{d}{2\pi}\int_{-\pi/d}^{+\pi/d}V_{\text{eff}}(q,q_z,\omega)\mathrm{d}q_z. \tag{10}$$

This integral can be evaluated by making use of (1) and (5) to get

$$V_{\text{eff}}(q,\omega)=\frac{2\pi e^2 D(q,\omega)\sinh(qd)}{q\varepsilon_\infty|D(q,\omega)|\{|D^2(q,\omega)-1|\}^{1/2}} \tag{11}$$

with

$$D(q,\omega)=P(q,\omega)\sinh(qd)-\cosh(qd) \tag{12}$$

The interaction potential, $V_{\text{eff}}(q,\omega)$, includes the effects of direct Coulomb repulsion as well as those of 2D acoustic plasmon and phonon modes. This interaction energy is attractive, if

$$D(q,\omega)<0 \tag{13}$$

and is responsible for the pairing mechanism. For small q values, the attractive interaction from (11) is given by

$$V_{\text{eff}}(q,\omega)=\frac{\sqrt{2}\pi e^2 d}{\{|qd\,P(q,\omega)R(q,\omega)|\}^{1/2}} \tag{14}$$

in view of (5), (11) and (12). Here,

$$R(q,\omega)=1+P(q,\omega)\tfrac{1}{2}qd. \tag{15}$$

In the limit $d\to\infty$, the effective interaction potential given by (11) reduces to 2D effective potential. Using the condition for attractive interaction (13), we may express

$$V'_{\text{eff}}(q,\omega)=\frac{2\pi e^2}{q\varepsilon_\infty}R'(q,\omega) \tag{16}$$

with

$$R'(q,\omega)=1+P(q,\omega). \tag{17}$$

An attractive potential energy for $R'(q,\omega)<0$ from (13) to (17) is obtained. In terms of frequencies of the collective excitations of charge carriers and those of ions obtained from (14), we get the condition for attractive interaction energy as

$$1+\frac{2(k_1-1)}{(2k_1-1)[1-k_1(\omega/\omega_{\text{pl}})^2]}-\frac{1}{(2k_1-1)(\omega/\omega_{\text{ph}})^2}<0 \tag{18}$$

with

$$K_1 = 1 + \frac{m^* e^2 d}{\hbar^2 \varepsilon_\infty}, \tag{19}$$

$$\omega_{ph} = \Omega_i q d/\sqrt{2} \tag{20}$$

and

$$\omega_{pl} = q v_F((k_1\sqrt{2})) \tag{21}$$

with ω_{ph} and ω_{pl} as the 2D acoustic phonon and plasmon frequencies, respectively. The second and third terms in (18) represent the contributions from the charge carriers and ions, respectively.

If we consider only phonons, (18) reduces to the following condition

$$0 < \omega < \omega_{ph}/(\sqrt{2}k_1 - 1) \tag{22}$$

and if one includes only 2D acoustic plasmons, (18) becomes

$$[\omega_{pl}/\sqrt{k_1}] < \omega < [\omega_{pl}(\sqrt{4}k_1 - 3)/k_1]. \tag{23}$$

Now, it is obvious that the frequency range of an attractive potential obtained from (22) is too low to account for the high transition temperatures. While the frequency range obtained from (23) is adequate to reveal the high transition temperature, this also indicates the inclusion of 2D acoustic plasmon in $La_{2-x}(Ba/Sr)_xCuO_4$ superconductors.

From (19) and (21), the plasma frequency dispersion relation can be written as

$$\omega_{pl}^2 = \frac{q^2 v_F^2}{2} + \frac{2\pi n_c e^2 q}{m^* \varepsilon_\infty}, \tag{24}$$

which is the same as that obtained by Fetter (1974) for free plasmon oscillations in the plane. It is seen from this equation that ω_{pl} approaches a nondispersive wave with propagation speed, $u = v_F/\sqrt{2}$, for short wavelengths and ω_{pl} varies like $q^{1/2}$ at long wavelengths ($q \to 0$). Thus, it is found from these descriptions that 2D acoustic plasmon effects included in our potential are responsible for the high temperature superconductivity in the layered structure cuprates.

In the strong coupling theory of Eliashberg (1960), the superconducting transition temperature requires knowledge of the modified Coulomb repulsive parameter (μ^*) and coupling parameter (λ) between attractive electrons. To obtain these, one needs to evaluate the following real and imaginary parts of the effective potential given by (3) as

$$\text{Real } R(q, q_z, \omega) = \frac{\omega^2}{(\phi\omega^2 - Bq^2)}, \tag{25}$$

$$\text{Imag. } R(q, q_z, \omega) = \frac{\pi B q^2}{\phi}\delta(\phi\omega^2 - Bq^2). \tag{26}$$

Here, $\delta(\phi\omega^2 - Bq^2)$ is the well-known Dirac delta function. In view of the above description, the averaged Coulomb repulsive parameter (μ) can be expressed as

$$\mu = N(0)\int_0^{2q_F} [\text{Real } V(q, q_z, \omega)/q_F^2]\, q \cdot dq$$

$$= \frac{2\pi e^2 d}{\varepsilon_\infty} N(0) \ln\left(\frac{2+D}{D}\right) \tag{27}$$

with $D = 2d/a^*$ and $N(0)$ as the density of states per unit cell.

Finally, the modified Coulomb repulsive parameter (μ^*) representing the Coulomb electron–electron interaction is expressed as

$$\mu^* = \frac{\mu}{1+\mu \ln(E_F/\omega_c)} = \frac{\mu}{1+\mu \ln(\pi \hbar^2 n_c/m^* \omega_c)} \tag{28}$$

with $E_F(=\pi\hbar^2 n_c/m^*)$ being the Fermi energy and ω_c, the cut-off frequency of 2D acoustic plasmons.

Using the Eliashberg theory for strongly coupled superconductors, the expression for the electron–electron pairing parameter (λ) has been derived from the relation (Eliashberg 1960)

$$\lambda = 2\int_0^{\omega_c} \alpha^2(\omega) \frac{F(\omega)\mathrm{d}\omega}{\omega} \tag{29}$$

with $F(\omega)$ as the Boson density of states and $\alpha^2(\omega)$ as the coupling strength between fermions and bosons. In view of the present potential given in (3), we have written the function $\alpha^2(\omega)\ F(\omega)$ as

$$\alpha^2(\omega)F(\omega) = \frac{\pi^2 e^2 d N(0)\omega^2}{\varepsilon_\infty B K_F^2 (1-\cos\theta_c)(1-\bar{\omega}_c)^{1/2}} \tag{30}$$

to obtain the value of λ from (29) and (30) we get

$$\lambda = \frac{\pi[1-((1+\bar{\omega}_c)/2)(1-\bar{\omega}_c)^{1/2}]}{[3+(\Omega_i^2 a^* m^* d/12\varepsilon_\infty E_F)](1-\cos\theta_c)}, \tag{31}$$

where $q_c(=2K_F=(8\pi n_c)^{1/2})$ is the cut-off wave vector and the critical angle (θ_c) between q_z and d is 16°. The Fermi energy (E_F) and the average cut-off 2D acoustic plasmon frequency ($\bar{\omega}_c$) are given by

$$E_F = \hbar^2 K_F^2/2m^* = n_c(\pi\hbar^2/m^*), \tag{32}$$

$$\bar{\omega}_c = \omega_c/E_F = m^*\omega_c/\pi\hbar^2 n_c. \tag{33}$$

The poles of (25) yield the frequency of 2D acoustic plasmon modes. The frequency of these modes can be divided into two parts, i.e. $\omega_p^{\parallel}$ and $\omega_p^{\perp}$ corresponding to the motion of electron along and perpendicular to a–b plane. We have, however, assumed that $\omega_p^{\perp}$ does not take part in pairing mechanism but only screens the electron motion along a–b plane. In order to obtain the cut-off 2D acoustic plasmon frequency (ω_c) for which the effective potential remains negative, we have averaged (3) over q and q_z and hence obtained poles of the averaged effective potential to get

$$\omega_c = 0{\cdot}556 K_F B^{1/2}[1-(\hbar K_F/E_F)B^{1/2}/2{\cdot}83], \tag{34a}$$

$$= 1{\cdot}4[(n_c B)^{1/2} - 0{\cdot}28 m^* B]. \tag{34b}$$

Using the above expressions for λ, μ^* and ω_c, the expression for the superconducting transition temperature (T_c) can be written as (Ruvalds 1987)

$$T_c = 0{\cdot}7\omega_c \exp[-(1+\lambda)/(\lambda-\mu^*)] \tag{35}$$

following the strong coupling theory of Eliashberg (1960). We have computed T_c for different concentrations (x) using the values of the potential parameters μ^*, λ and ω_c obtained from (28), (31) and (34) from the knowledge of $n_c(=n_0 \times 10^{14}\text{cm}^{-2})$ determined as a function of composition (x) as the values of n_0 are reported (Hasegawa *et al* 1987; Ong *et al* 1987; Que *et al* 1987) for Ba and Sr-doped superconductors for the range $0 \leqslant x \leqslant 0{\cdot}3$. The results on T_c thus computed by us as a function of x for $La_{2-x}(Ba/Sr)_xCuO_4 (0 \leqslant x \leqslant 0{\cdot}3)$ have been presented and discussed below.

3. Results and conclusion

In order to calculate T_c as discussed above, we have used the values of the effective mass m^* equal to $6m_e$ and $5m_e$ for $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$, respectively. The values of high frequency background dielectric constant (ε_∞) appearing in (28), (31) and (34) have been obtained as 4·5 as reported by Bozovic (1990) and Mahan and Wu (1989) for Ba and Sr doped superconductors. The values of the model parameters, particularly λ and ω_c, obtained as discussed earlier, have been plotted against the composition (x) in figures 2 and 3 for $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$, respectively. It is seen from figure 2 that λ remains almost constant, while ω_c first increases and decreases after reaching the first maximum once a minimum value of $x = 0{\cdot}10$ is attained. It again increases and reaches a second maximum in the range of x from 0·10 to 0·25. We thus find two peaks when ω_c is plotted as a function of x for $La_{2-x}Ba_xCuO_4$. Figure 3 shows that the values of λ are almost constant throughout the range of x, while the values of ω_c first increase up to $x = 0{\cdot}20$

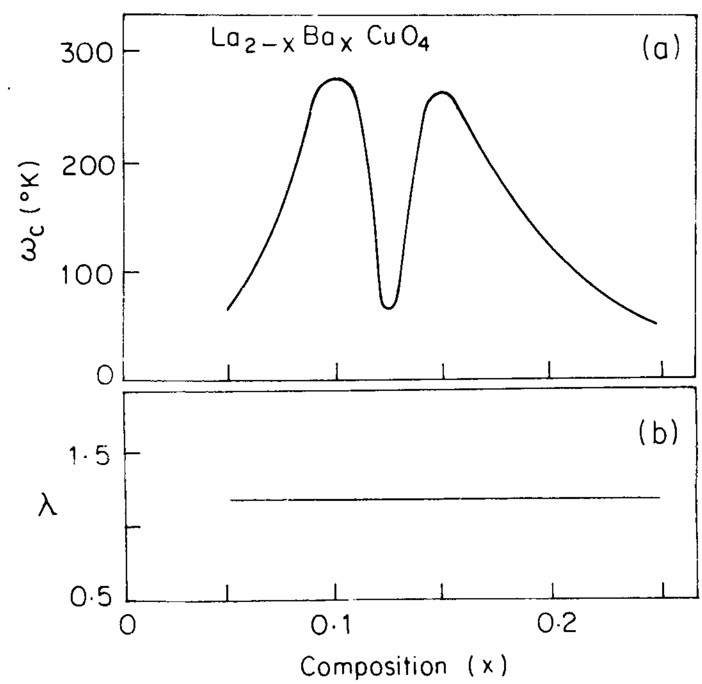

Figure 2. Variation of the parameters ω_c and λ with composition (x) shown in **a** and **b**, respectively.

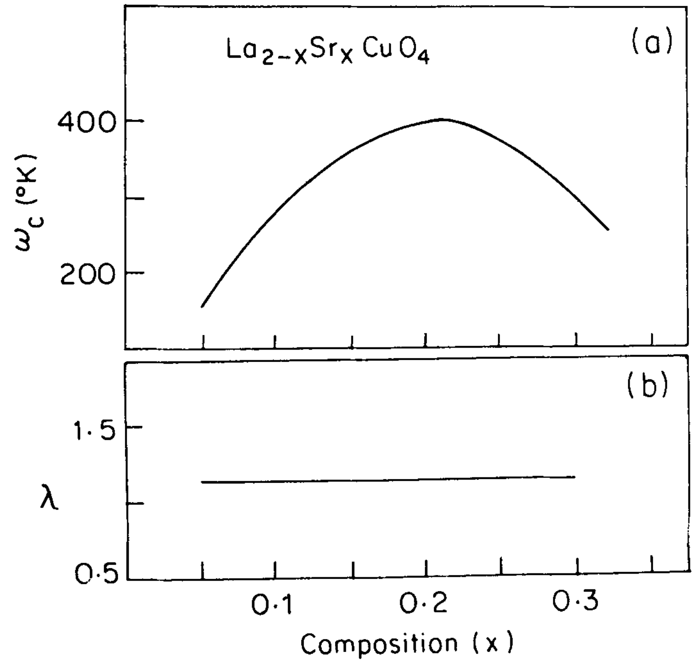

Figure 3. Variation of the parameters ω_c and λ with composition (x) shown in **a** and **b**, respectively.

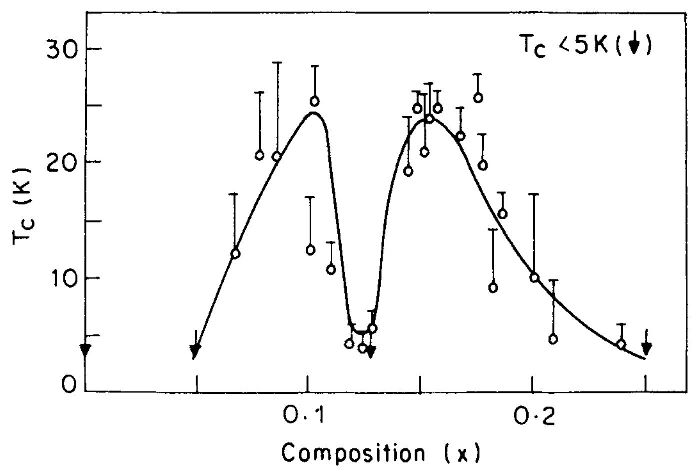

Figure 4. Variation of transition temperature (T_c) with composition (x). The circles (Ⴋ) are the experimental data taken from Moodenbaugh *et al* (1988).

and then gradually decrease with the increase of x. The characteristic of ω_c with x for $La_{2-x}Sr_xCuO_4$ is in keeping with the measured ω_c at different x by Tajima *et al* (1988) and Suzuki (1989). However, such experimental data are not available for $La_{2-x}Ba_xCuO_4$. The values of $\lambda \geqslant 1{\cdot}0$ obtained by us at different concentrations (x) indicate that the coupling due to 2D acoustic plasmons is strong in nature.

The values of T_c, computed by us using the model parameters λ, μ^* and ω_c obtained for different x as discussed earlier, have been plotted as functions of x in figures 4

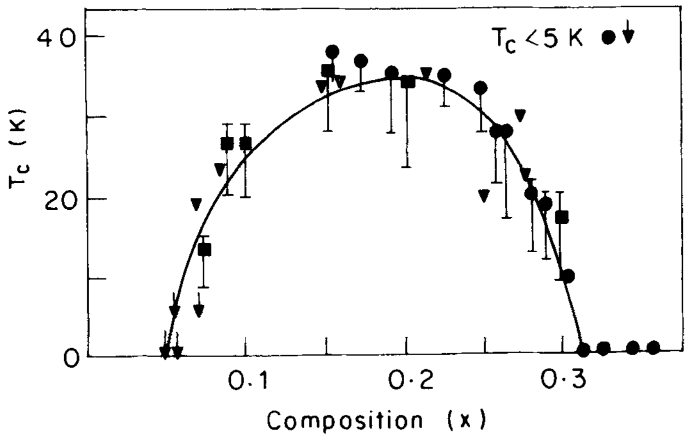

Figure 5. Variation of transition temperature (T_c) with composition (x). The ■, ▼ and ● are taken from Dover *et al* (1987), Shafer *et al* (1987) and Torrance *et al* (1988), respectively. The values of T_c indicated by ▼ and ● are below 5 K.

and 5 along with their experimental values for $La_{2-x}Ba_xCuO_4$ (Moodenbaugh 1988) and $La_{2-x}Sr_xCuO_4$ (Dover *et al* 1987; Shafer *et al* 1987; Torrance *et al* 1988), respectively. It is seen from these figures that our calculated values of T_c are in a reasonably good agreement with their experimental values. Figure 4 shows that there are two maxima: one near composition $x = 0{\cdot}10$ with $T_c = 24$ K and another near $x = 0{\cdot}16$ with $T_c = 26$ K. These maxima of T_c correspond to the recently observed peaks by Moodenbaugh *et al* (1988) at $x = 0{\cdot}10$ and 0·17 with $T_c = 26$ and 26·5 K, respectively. In $La_{2-x}Ba_xCuO_4$ superconductor, we have obtained an anomalous dip around $x = 0{\cdot}12$ besides two maxima. This indicates that above and below the Ba composition $x = 0{\cdot}12$, there are two superconducting phases and hence double transition reflects the intrinsic change of the electronic states around $x = 0{\cdot}12$. It is thus almost clear that at $x = 0{\cdot}12$, a structural transformation occurs from high temperature tetragonal to low temperature orthorhombic phase. The reduction of T_c near $x = 0{\cdot}12$, is related to a significant change in the electronic state. This seems to be an interesting feature in $La_{2-x}Ba_xCuO_4$ superconductor and deserves detailed exploration.

We notice from figure 5 that there is a maximum for T_c in $La_{2-x}Sr_xCuO_4$ near $x = 0{\cdot}15$ with $T_c = 36$ K, which is in good agreement with the experimental results (Dover *et al* 1987; Shafer *et al* 1987; Torrance *et al* 1988). It is also noted from figure 5 that our calculated values of T_c are closer to zero for $x \leqslant 0{\cdot}06$ and $x \geqslant 0{\cdot}30$. This feature is in keeping with the experimental data (Dover *et al* 1987; Shafer *et al* 1987; Torrance *et al* 1988). It can also be seen from this figure that $La_{2-x}Sr_xCuO_4$ is superconducting only for $0{\cdot}06 \leqslant x \leqslant 0{\cdot}30$ according to our calculations, while Torrance *et al* (1988) have reported $T_c < 5$ K near $0{\cdot}06 \leqslant x \geqslant 0{\cdot}3$. We thus find that our calculations, based on the mechanism of pairing of charge carriers by the exchange of 2D acoustic plasmons, satisfactorily explain the observed variations of T_c with composition (x) in both $La_{2-x}(Ba/Sr)_xCuO_4$ superconductors.

We may further add that the theoretical work of Weber (1987) on the calculation of T_c for $La_{2-x}Sr_xCuO_4$, which is based on the pairing of charge carriers by exchange of phonons only, does not exhibit the entire trend of the experimentally observed T_c as a function of x. It is, therefore, obvious that the inclusion of the effects of electronic excitations (2D acoustic plasmons) in the calculation of T_c is essential to describe the composition (x) dependence of T_c in $La_{2-x}(Ba/Sr)_xCuO_4$ ceramic superconductors for entire range ($0 \leqslant x \leqslant 0{\cdot}3$). Another important point which emerges from our calculations and those of Weber (1987) is that for $0{\cdot}06 \leqslant x \leqslant 0{\cdot}15$, the pairing by exchange of electronic excitations (2D acoustic plasmons) seems to dominate over the pairing by exchanged phonons. On the other hand, for $x > 0{\cdot}15$, T_c has decreasing trend and hence the phonon mechanism seems to dominate over the electronic excitation (2D acoustic plasmons) mechanism in charge-carrier pairing.

In conclusion, we have succeeded in predicting the observed effect of Ba and Sr doping on the transition temperature (T_c) in $La_{2-x}(Ba,Sr)_xCuO_4$ superconductors. Our calculations of T_c are based on a model effective interaction potential which includes properly the effects of 2D acoustic plasmons in pairing mechanism. The drawbacks present in our earlier work (Singh *et al* 1991) have almost been eliminated in this paper. Our results on composition dependence of T_c demonstrate that the carrier collective excitations (2D acoustic plasmons) play an important role in the pairing of charge-carriers, especially for lower values of x in cuprate superconductors. Such investigations in the cases of other members of this family are in progress and the results will be reported subsequently.

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