

Charge-fluctuation mechanism of high- T_c superconductivity and the isotope effect in oxide superconductors

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In connection with high- T_c superconductivity, superconductivity in a system of Fermi particles interacting via a pairing interaction mediated by charge fluctuation and LO phonons is theoretically studied. The anomaly observed in the LO phonon dispersion of the oxides suggests that the electronic dielectric constants with certain wave numbers are negative due to the strong Coulomb correlation. The superconducting transition temperature T_c of the system calculated by an Eliashberg-type equation is unusually high and the isotope effect for T_c is very weak.

I. INTRODUCTION

Extensive experimental and theoretical studies to elucidate the mechanism of high- T_c superconductivity in the oxides have been made.¹⁻³ The measurements of the flux quantization^{4,5} and the ac Josephson effect^{6,7} show that the unit of the superconducting current in the oxides is twice the electronic charge. This fact indicates that even in the oxide superconductors, superconductivity is generated by electron pairs or hole pairs. The coherence lengths estimated from the upper-critical field are 30 to 40 Å in the basal planes of $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$,^{8,9} and Bi-Sr-Ca-Cu-O .^{10,11} These values are shorter than those in usual superconductors, but much longer than the distance between Cu and O ions which is approximately 2 Å. The specific heat of $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ shows a sharp jump at the superconducting transition temperature.^{12,13} Some of the tunneling experiments using $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ show a clear energy gap.¹⁴ These experimental results suggest that the nature of the pairing in the oxide superconductors is of the Bardeen-Cooper-Schrieffer (BCS) -type pairing.

The only experimental result which appears inconsistent with the BCS-type pairing is the nuclear relaxation of Cu in the two-dimensional layers in the crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$: The hump of the nuclear relaxation rate $1/T_1$ which is usually seen just below T_c in BCS superconductors is not observed and $1/T_1$ sharply decreases below T_c .¹⁵⁻¹⁸ To explain the anomalous temperature dependence of the nuclear relaxation, superconducting states other than the BCS superconducting state have been proposed. However, recently the anomalous temperature dependence was found to be explained on the basis of the BCS pairing, if the correlation effect of electrons is taken into account.¹⁹ Very recently, Kitaoka *et al.* measured the nuclear relaxation rate of ^{17}O 's which replace ^{16}O 's in the layers of CuO_2 and observed the BCS-type temperature dependence of $1/T_1$ with a hump.²⁰ This observation confirms that the superconductivity in the oxides is generated by the pairing with s -wave symmetry. Therefore, the remaining problem is what is the origin of the strong binding energy of the pairing accountable for the high- T_c superconductivity in the oxides.

As inferred from the photoemission experiments of the

copper oxides, the copper d -electron state at 3-4 eV below the Fermi level is almost localized at the lattice sites.²¹⁻²³ Due to the strong on-site Coulomb interaction, each divalent copper ion has a spin of $\frac{1}{2}$.²⁴ In metallic samples holes are mainly on oxygen ions octahedrally surrounding copper ions.^{25,26} Since the copper d -electron levels are relatively close to the oxygen p -electron levels in the copper oxides, electrons can transfer from the copper ions to the oxygen ions, and a small number of holes are created on the copper ions. These holes will form a Fermi-liquid state near the Fermi level. Recently, the existence of the Fermi-liquid state has been confirmed by angular-resolved photoemission experiments by Takahashi *et al.*²⁶ The Fermi liquid is responsible for normal conductivity and superconductivity of the copper oxides. The Fermi liquid is accompanied by large spin and charge fluctuations. Due to the large charge fluctuations, the electronic distribution in the metallic sample is almost unstable and a small perturbation could cause the formation of charge-density waves. In this case, the dielectric constant of the Fermi liquid can be negative as discussed in the previous paper²⁷ (hereafter referred to as paper I). It has also been shown that the theoretical dielectric constant in an electron gas system becomes negative when the dimensionless mean radius per electron r_s increases above a critical value, since the correlation effect becomes important for large r_s .²⁸

In the copper oxide system the charge fluctuations strongly couple with the longitudinal-optical (LO) phonons, especially of the breathing modes. Evidence of the strong coupling has been provided by the intensity enhancement of the LO phonons observed by electron-loss-energy spectroscopy using $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$.²⁹ The interaction mediated by the charge fluctuations and the LO phonons yields the effective attractive potential between electrons. In Sec. II, using this effective potential, we formulate the equation for calculating the superconducting transition temperature T_c . In Sec. IV, using this equation and assuming some form for the electronic dielectric constant of the copper oxide, we estimate the value of T_c . When the electronic dielectric constants are negative for some wave numbers, T_c is extremely enhanced. We also calculate the decrease of T_c when ^{16}O 's are replaced by ^{18}O 's in the oxide, and show that the isotope effect is

much weakened as T_c increases.

When the electronic dielectric constants are negative for some range of wave numbers, the LO phonons with the wave numbers are strongly renormalized by the charge fluctuations and their frequencies shift lower than the frequencies of the corresponding TO phonons. In Sec. III, the calculated frequency shift is compared with the frequency shift observed in some oxides.

The value of T_c predicted from the present mechanism is much higher than T_c estimated from the pure phonon mechanism. The predicted systematics of the isotope effect and the frequency shift of LO phonons are in agreement with those observed in the experiments. Therefore, we suggest that the interaction mediated by the charge fluctuations and the LO phonons is responsible for the high- T_c superconductivity in the copper oxides.

II. FORMULATION OF THE EQUATION FOR CALCULATING T_c

In this section we derive the equation which is used for calculating the superconducting transition temperature T_c . The effective interaction $V_{\text{eff}}(\mathbf{q}, \omega)$ between two electrons is defined by Fig. 1, where \mathbf{k} , \mathbf{k}' , and \mathbf{q} are wave numbers and ω is frequency. The effective interaction is expressed as^{27,30,31}

$$V_{\text{eff}}(\mathbf{q}, \omega) = \frac{v(\mathbf{q})}{\epsilon(\mathbf{q}, \omega)}, \quad (2.1)$$

where $v(\mathbf{q})$ is the Fourier component of the Coulomb interaction, and $\epsilon(\mathbf{q}, \omega)$ is the total dielectric constant given by

$$\epsilon(\mathbf{q}, \omega) = \epsilon_{\text{ion}}(\mathbf{q}, \omega) + \epsilon_{\text{el}}(\mathbf{q}, \omega) - \epsilon_{\infty}, \quad (2.2)$$

where $\epsilon_{\text{ion}}(\mathbf{q}, \omega)$ is the ionic dielectric constant, and $\epsilon_{\text{el}}(\mathbf{q}, \omega)$ is the electronic dielectric constant, and ϵ_{∞} is the high-frequency part of the dielectric constant.²⁷ If we use the spectral representation of $1/\epsilon(\mathbf{q}, \omega)$, $V_{\text{eff}}(\mathbf{q}, \omega)$ is written as

$$V_{\text{eff}}(\mathbf{q}, \omega) = v(\mathbf{q}) \left[1 - 2 \int_0^{\infty} d\Omega \frac{\Omega \rho(\mathbf{q}, \Omega)}{\Omega^2 - (\omega + i\delta)^2} \right], \quad (2.3)$$

where $\rho(\mathbf{q}, \Omega)$ is the spectral intensity defined by

$$\rho(\mathbf{q}, \omega) = -\frac{1}{\pi} \text{Im} \left[\frac{\epsilon_{\infty}}{\epsilon(\mathbf{q}, \omega)} \right]. \quad (2.4)$$

Using the effective interaction, we set up the Eliashberg equation linearized with respect to the gap function $\Delta(\mathbf{k}, i\omega_n)$ as

$$\Delta(\mathbf{k}, i\omega_n) = -T \sum_l \sum_{\mathbf{k}'} V_{\text{eff}}(\mathbf{k} - \mathbf{k}', i\omega_n - i\omega_l) \frac{\Delta(\mathbf{k}', \omega_l)}{\xi_{\mathbf{k}}^2 + \omega_l^2}, \quad (2.5)$$

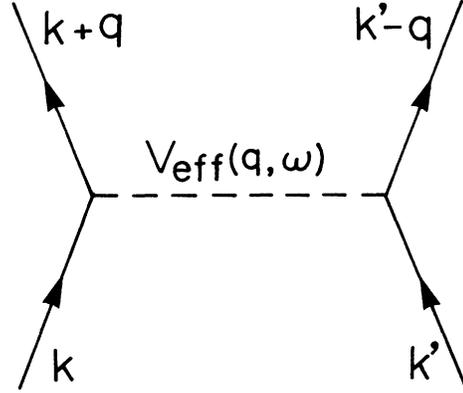


FIG. 1. Effective interaction $V_{\text{eff}}(\mathbf{q}, \omega)$ between two electrons. The wave numbers \mathbf{k} and \mathbf{k}' denote the electron wave numbers, and \mathbf{q} and ω the differences in wave number and frequency between the two electrons, respectively.

where $\omega_n = (2n + 1)\pi T$ with n being integer, and $\xi_{\mathbf{k}}$ is the quasiparticle energy of carriers. If we introduce the pair function defined by

$$F(\mathbf{k}, i\omega_n) = \Delta(\mathbf{k}, i\omega_n) / (\omega_n^2 + \xi_{\mathbf{k}}^2), \quad (2.6)$$

Eq. (2.5) is written as

$$F(\mathbf{k}, i\omega_n) = -\frac{1}{(\omega_n^2 + \xi_{\mathbf{k}}^2)} T \sum_l \sum_{\mathbf{k}'} V_{\text{eff}}(\mathbf{k} - \mathbf{k}', i\omega_n - i\omega_l) F(\mathbf{k}', i\omega_l). \quad (2.7)$$

To reduce the computation load for numerical calculation of T_c , Krizhznits, Maksimov, and Khomskii have obtained an approximate but simpler Eq. (2.13) for (2.7).³² We briefly reproduce the procedure of deriving the equation to make clear the approximation which is made in the course of the derivation. In order to make a summation of the imaginary frequencies in (2.7), we use the spectral representation for $F(\mathbf{k}, i\omega_n)$

$$F(\mathbf{k}, i\omega_n) = 2 \int_0^{\infty} d\nu \frac{\nu f(\mathbf{k}, \nu)}{\omega_n^2 + \nu^2}. \quad (2.8)$$

Inserting (2.8) into (2.7), we have the equation for $f(\mathbf{k}, \nu)$ in the form

$$f(\mathbf{k}, \omega) = A(\mathbf{k}) \delta(\omega^2 - \xi_{\mathbf{k}}^2) + B(\mathbf{k}, \omega) P \frac{1}{\omega^2 - \xi_{\mathbf{k}}^2}, \quad (2.9)$$

where

$$A(\mathbf{k}) = -\frac{1}{2} \sum_{\mathbf{k}'} v(\mathbf{k} - \mathbf{k}') \int_{-\infty}^{\infty} d\nu \left\{ \tanh \frac{\nu}{2T} + \int_{-\infty}^{\infty} d\Omega \frac{\rho(\mathbf{k} - \mathbf{k}', \Omega)}{|\xi_{\mathbf{k}}| - \nu - \Omega} \left[\tanh \left[\frac{\nu}{2T} \right] + \coth \left[\frac{\Omega}{2T} \right] \right] \right\} f(\mathbf{k}', \nu), \quad (2.10)$$

$$B(\mathbf{k}, \omega) = -\frac{1}{2} \sum_{\mathbf{k}'} v(\mathbf{k} - \mathbf{k}') \int_{-\infty}^{\infty} d\nu \rho(\mathbf{k} - \mathbf{k}', \omega - \nu) \left[\tanh \left(\frac{\nu}{2T} \right) + \coth \left(\frac{\omega - \nu}{2T} \right) \right] f(\mathbf{k}', \nu), \quad (2.11)$$

From (2.6), (2.8), and (2.9) we can show that $A(\mathbf{k})$ is the real part of $\Delta(\mathbf{k}, \omega)$ at $\omega = \xi_{\mathbf{k}}$, i.e., $A(\mathbf{k}) = \text{Re}[\Delta(\mathbf{k}, \xi_{\mathbf{k}})]$. Let us introduce the following function:

$$\Phi(\mathbf{k}) = 2|\xi_{\mathbf{k}}| \int_0^{\infty} d\nu f(\mathbf{k}, \nu). \quad (2.12)$$

In the energy region near the Fermi level where the quasi-particle is well defined, the first term in (2.9) mainly contributes to $f(\mathbf{k}, \omega)$. In this case, Eq. (2.12) gives $\Phi(\mathbf{k}) \approx A(\mathbf{k}) = \text{Re}[\Delta(\mathbf{k}, \xi_{\mathbf{k}})]$. If we approximate $f(\mathbf{k}, \omega)$ in (2.10), (2.11), and (2.12) by the first term in (2.9), we have the equation for $\Phi(\mathbf{k})$

$$\Phi(\mathbf{k}) = -\sum_{\mathbf{k}'} K(\mathbf{k}, \mathbf{k}') \frac{\tanh(\xi_{\mathbf{k}'}/2T)}{2\xi_{\mathbf{k}'}} \Phi(\mathbf{k}'), \quad (2.13)$$

where the kernel $K(\mathbf{k}, \mathbf{k}')$ is given by

$$K(\mathbf{k}, \mathbf{k}') = v(\mathbf{k} - \mathbf{k}') \left[1 - 2 \int_0^{\infty} d\Omega \frac{\rho(\mathbf{k} - \mathbf{k}', \Omega)}{\Omega + |\xi_{\mathbf{k}}| + |\xi_{\mathbf{k}'}|} \right]. \quad (2.14)$$

We use the equation (2.13) for the calculation of T_c of the present system. The ionic dielectric constant in (2.2)

$$K(\mathbf{k}, \mathbf{k}') = v(\mathbf{q}) \left[\frac{1}{\epsilon_{\text{el}}^{\infty}(\mathbf{q})} + 2 \int_0^{\infty} d\Omega \frac{(|\xi_{\mathbf{k}}| + |\xi_{\mathbf{k}'}|)}{\Omega(\Omega + |\xi_{\mathbf{k}}| + |\xi_{\mathbf{k}'}|)} \rho_{\text{el}}(\mathbf{q}, \Omega) - \frac{1}{\epsilon_{\text{el}}^{\infty}[\mathbf{q}, \omega^*(\mathbf{q})]} \frac{\omega^*(\mathbf{q}) - \omega_{\text{TO}}^2}{\omega^*(\mathbf{q})[\omega^*(\mathbf{q}) + |\xi_{\mathbf{k}}| + |\xi_{\mathbf{k}'}|]} \right], \quad (2.18)$$

where $\mathbf{q} = \mathbf{k} - \mathbf{k}'$, $\epsilon_{\text{el}}^{\infty}(\mathbf{q})$ is the static electronic dielectric constant, and $\rho_{\text{el}}(\mathbf{q}, \omega)$ is the spectral intensity of $1/\epsilon_{\text{el}}^{\infty}(\mathbf{q}, \omega)$

$$\rho_{\text{el}}(\mathbf{q}, \omega) = -\frac{1}{\pi} \text{Im} \left[\frac{1}{\epsilon_{\text{el}}^{\infty}(\mathbf{q}, \omega)} \right]. \quad (2.19)$$

In deriving (2.18), the Kramers-Kronig relation for $1/\epsilon_{\text{el}}^{\infty}(\mathbf{q}, \omega)$ was used. In (2.18) the first two terms are purely electronic contributions and the third term is a phonon contribution from the renormalized LO phonons. Roughly speaking, the first two terms correspond to μ and the second term to λ in conventional notations. The negative $1/\epsilon_{\text{el}}^{\infty}(\mathbf{q})$, therefore, corresponds to the negative value of μ . However, as seen from (2.18), the electron and phonon contributions cannot completely be separated in the highly correlated electron system.

III. NEGATIVE ELECTRIC DIELECTRIC CONSTANT AND FREQUENCY SHIFT OF LO PHONON

As discussed in paper I, $1/\epsilon_{\text{el}}^{\infty}(\mathbf{q}, \omega)$ should be less than 1 from the causality.^{27,30,31} This region includes the nega-

is expressed in a conventional form as²⁷

$$\epsilon_{\text{ion}}(\mathbf{q}, \omega) = \epsilon_{\infty} \frac{\omega^2 - \omega_{\text{LO}}^2}{\omega^2 - \omega_{\text{TO}}^2}, \quad (2.15)$$

where ω_{LO} and ω_{TO} are, respectively, the frequencies of the longitudinal- and transverse-optical phonons in the insulating state. For simplicity, we consider a single optical-phonon mode whose vibrational frequencies are independent of wave number. If (2.15) is used in (2.2), the spectral function (2.4) is written as

$$\rho(\mathbf{q}, \omega) = -\frac{1}{\pi} \text{Im} \frac{1}{\epsilon_{\text{el}}^{\infty}(\mathbf{q}, \omega)} + \frac{\omega_{\text{LO}}^2 - \omega_{\text{TO}}^2}{\{\epsilon_{\text{el}}^{\infty}[\mathbf{q}, \omega^*(\mathbf{q})]\}^2} \delta[\omega^2 - \omega^{*2}(\mathbf{q})], \quad (2.16)$$

where $\epsilon_{\text{el}}^{\infty}(\mathbf{q}, \omega) = \epsilon_{\text{el}}(\mathbf{q}, \omega)/\epsilon_{\infty}$ and the $\omega^*(\mathbf{q})$ is the renormalized frequency of the longitudinal phonon and written as

$$\omega^{*2}(\mathbf{q}) = \omega_{\text{TO}}^2(\mathbf{q}) + \frac{(\omega_{\text{LO}}^2 - \omega_{\text{TO}}^2)}{\epsilon_{\text{el}}^{\infty}[\mathbf{q}, \omega^*(\mathbf{q})]}, \quad (2.17)$$

In the derivation of (2.16), we neglected the damping of the phonon. Using (2.16), we rewrite the kernel (2.14) as

tive $\epsilon_{\text{el}}^{\infty}(\mathbf{q})$ which corresponds to the case of a test charge being overscreened. We expect that the state of the negative $\epsilon_{\text{el}}^{\infty}(\mathbf{q})$ occurs in the superconducting oxides, since the electronic distribution in the metallic state of the oxides is almost unstable due to the strong Coulomb correlation and small perturbation could cause the formation of charge-density waves with certain wave numbers. We are now trying to calculate the dielectric constant using a model appropriate for the oxides. However, this calculation has not yet been completed. Therefore, in this paper, according to the reason mentioned below, we assume the form $1/\epsilon_{\text{el}}^{\infty}(\mathbf{q})$

$$1/\epsilon_{\text{el}}^{\infty}(\mathbf{q}) = \left[\frac{q^2}{1+q^2} \Theta(1-q) - \eta \Theta(q-1) \right] \times \tanh(3|1-q|), \quad (3.1)$$

where $\Theta(x)$ is the usual step function and η is a parameter. In (3.1), q is measured in units of the Fermi momentum. If the form (3.1) is used, the anomalous dispersion curve of the LO phonon observed in some oxides is fairly well reproduced as shown later. The choice of the form (3.1) is also consistent with the fact that $1/\epsilon_{\text{el}}^{\infty}(\mathbf{q})$ should be proportional to q^2 for small \mathbf{q} in a three-dimensional

system. The q dependence of $1/\epsilon_{el}^{\infty}(\mathbf{q})$ is shown for several values of η in Fig. 2. Using these dielectric constants, we calculate the renormalized frequency of the LO phonon from (2.17). Since $\omega^*(\mathbf{q})$ is much smaller than the characteristic frequency of the electronic charge fluctuation, the value of $\epsilon_{el}^{\infty}(\mathbf{q})$ is used for $\epsilon_{el}^{\infty}[\mathbf{q}, \omega^*(\mathbf{q})]$. The dispersion curves of $\omega^*(\mathbf{q})$ when $\omega_{LO}=0.3$, and $\omega_{TO}=0.26$ and 0.28 , in units of the Fermi energy, are shown in Fig. 3. We see in Fig. 3 that the renormalized frequencies $\omega^*(\mathbf{q})$ shift lower than the frequencies of the corresponding TO phonons in the range of the wave numbers where the electronic dielectric constants are negative.

Unfortunately, the measurement of the phonon dispersion curve in the high- T_c oxide superconductors has not yet been done. However, the phonon dispersion curves of La_2NiO_4 , which is isostructural to La_2CuO_4 , have been observed by inelastic neutron scattering by Pintschovius *et al.*³³ They found a pronounced anomaly in the LO phonon branch Σ_1 which corresponds to in-plane oxygen-nickel stretching vibrations. In particular, the frequencies of the vibrations of breathing modes around $\mathbf{q}=(0.5, 0.5, 0)$ shift lower than the frequencies of the vibrations of the TO phonon branch Σ_3 which corresponds to the LO phonon branch Σ_1 . The behavior of the dispersion curve is qualitatively reproduced by the dispersion curves shown in Fig. 3. The fact that the anomalous dispersion of LO phonon mentioned above occurs in spite of La_2NiO_4 being a semiconductor may be understood in the following way. La_2NiO_4 has a very narrow energy gap of 50 meV which is inferred from the temperature dependence of the dc conductivity. The frequencies ω_{LO} and ω_{TO} are around 80 meV which is much larger than the energy gap. The electronic dielectric constants with the frequencies behave metallic and may be negative for the wave numbers around $(0.5, 0.5, 0)$.

The phonon densities of states in polycrystalline samples of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ and La_2NiO_4 observed by inelastic neutron scattering measurements show the striking similarity, in particular, in the frequency range above 50 meV.^{34,35} Therefore, the phonon anomaly mentioned above is also expected in $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. The similar anomaly in the LO phonon dispersion has been observed in a superconducting oxide $\text{BaPb}_{0.75}\text{Bi}_{0.25}\text{O}_3$ by Reichardt and Weber.³⁶

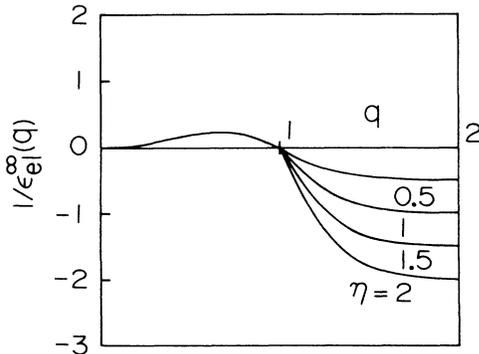


FIG. 2. Inverse of electronic dielectric constants $1/\epsilon_{el}^{\infty}(\mathbf{q})$ as a function of wave number q for several values of η .

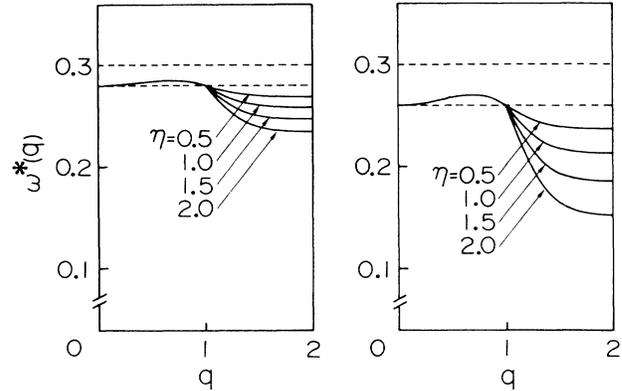


FIG. 3. Dispersion curves of renormalized LO phonon frequencies $\omega^*(\mathbf{q})$ calculated using the inverse of electron dielectric constant $1/\epsilon_{el}^{\infty}(\mathbf{q})$ shown in Fig. 2. (a) $\omega^*(\mathbf{q})$ for the parameters of the bare phonon frequencies $\omega_{LO}=0.3$ and $\omega_{TO}=0.28$, and (b) $\omega^*(\mathbf{q})$ for $\omega_{LO}=0.3$ and $\omega_{TO}=0.26$.

IV. SUPERCONDUCTING TRANSITION TEMPERATURE AND ISOTOPE EFFECT

Using $\epsilon_{el}^{\infty}(\mathbf{q})$ given in the previous section, we calculate the superconducting transition temperature T_c from (2.13) in the following way. We introduce an eigenvalue equation with an eigenvalue $\lambda(T)$,

$$\lambda(T)\Phi(\mathbf{k}) = -\sum_{\mathbf{k}'} K(\mathbf{k}, \mathbf{k}') \frac{\tanh(\xi_{\mathbf{k}'}/2T)}{2\xi_{\mathbf{k}'}} \Phi(\mathbf{k}'). \quad (4.1)$$

We numerically solve the equation and calculate $\lambda(T)$ as a function of temperature T . The temperature corresponding to $\lambda(T_c)=1$ is the superconducting transition temperature. The kernel $K(\mathbf{k}, \mathbf{k}')$ (2.18) is inserted in (4.1) and a form $k^2/2m$ is used for $\xi_{\mathbf{k}}$. Since the expression for the spectral function $\rho_{el}(\mathbf{q}, \omega)$ is not available at present, we use the expression of PRA for $\rho_{el}(\mathbf{q}, \omega)$ in the second term of (2.18) as an approximation. The calculated T_c is shown as a function of η in Fig. 4 where the measure of units of T_c is the Fermi energy and η is the parameter characterizing $\epsilon_{el}^{\infty}(\mathbf{q})$ (see Fig. 2). The solid curves show the values of T_c when the charge fluctuation and the LO phonon contribute to the interaction. The dashed curve shows the values of T_c for the case of no phonon contribution. When η is small, T_c is very small due to the Coulomb repulsion acting between electrons. As η increases, the negative electronic dielectric constants for large wave numbers decrease more as seen in Fig. 2. In this case, the Coulomb interaction works as an attractive interaction and also works as enhancing the intensity of the LO phonon. These two effects combined yield the drastic increase of T_c when η increases, as seen in Fig. 4. Equation (2.13) does not include the wave-function renormalization constant. If the constant is included, T_c shown in Fig. 4 will be smaller for large η .³⁷ However, the qualitative features of the dependence of T_c on η is not changed.

If the LO phonon modes responsible for superconductivity are the vibrational modes of oxygen ions, the fre-

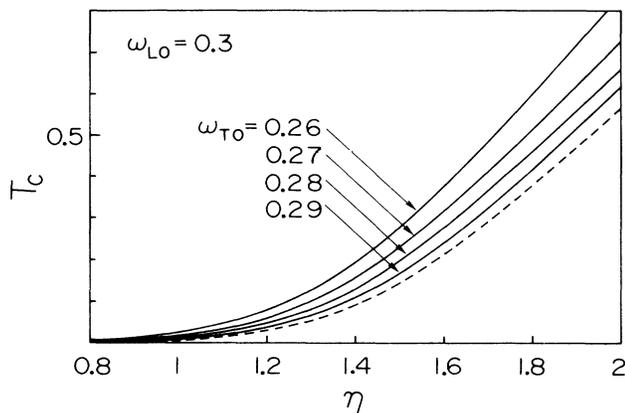


FIG. 4. Superconducting transition temperature T_c vs η . The solid curves show T_c when the charge fluctuations and the LO phonons combining contribute to the interaction, and the dashed curve shows T_c when only the charge fluctuations contribute to the interaction.

frequencies ω_{LO} and ω_{TO} are proportional to $M^{-1/2}$, M being the oxygen mass. The transition temperature for the sample in which ^{16}O 's are replaced by ^{18}O 's is calculated in the same way. The calculated result shows that T_c for the sample of ^{18}O 's decreases from T_c for the sample of ^{16}O 's. The calculated value of $\delta T_c/T_c$, δT_c being the decrease of T_c , is plotted by the solid curves in Fig. 5. The dashed lines indicate the values of $\delta T_c/T_c$ when only the phonon contribution, the third term in (2.18), is included in the calculation. This value is equal to $[1 - (M_{16}/M_{18})^{1/2}] = 0.0572$, M_{16} and M_{18} being the masses of ^{16}O and ^{18}O , respectively. This value is called the BCS value of the isotope effect. As seen in Fig. 5, $\delta T_c/T_c$ is smaller than the BCS value since the contributions from the first and second terms in (2.18), the nonphonon contribution, to the kernel is large compared with the third-term which is related to the phonons in a highly correlated system like the copper oxide superconductors. When the values of ω_{LO} and ω_{TO} are fixed, $\delta T_c/T_c$ increases as η increases, attains a maximum, and then decreases as η increases fur-

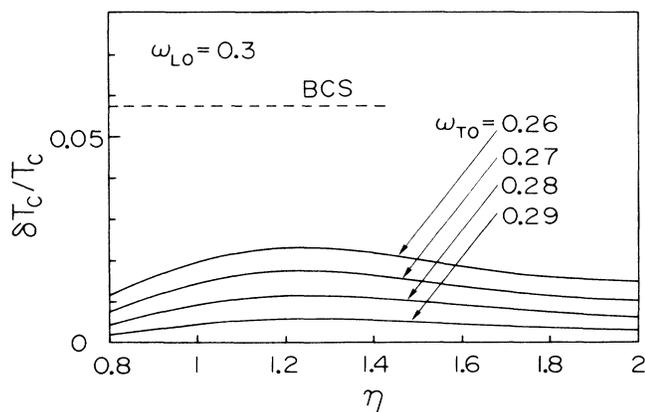


FIG. 5. Isotope shift $\delta T_c/T_c$ vs η . The dashed line shows the BCS value for the isotope shift.

ther. This dependence can be understood in the following way. Since the Coulomb repulsion dominates when η is small, $\delta T_c/T_c$ becomes small due to the same reason as in conventional superconductors. When η is large, $\delta T_c/T_c$ also becomes small, since the attractive interaction mediated by the charge fluctuations predominates.

As seen in Fig. 5, when ω_{TO} is close to ω_{LO} , $\delta T_c/T_c$ becomes smaller for the following reason. The third term in (2.18) is proportional to $\omega_{LO}^2 - \omega_{TO}^2$ and thus the phonon contribution to the kernel becomes small when ω_{TO} is close to ω_{LO} .

The theoretical result of the isotope effect mentioned above is consistent with the experimental results that $\delta T_c/T_c$'s observed in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($T_c \approx 35$ K), $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ ($T_c \approx 90$ K), and Bi-Sr-Ca-Cu-O ($T_c \approx 110$ K) are, respectively, 0.013,^{38,39} 0–0.005,^{40–43} and 0.003,⁴⁴ which are much smaller than the BCS value 0.0572. The band theoretical calculation shows that the electron-phonon interaction in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ is stronger than that in $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$.^{45,46} This fact may indicate that $\omega_{LO}^2 - \omega_{TO}^2$ is larger in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ than in $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$. The value of η in $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ may be larger than that in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, since T_c of $\text{YBa}_2\text{Cu}_3\text{O}_7$ is higher than T_c of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. From these two facts we may explain the difference between the values of $\delta T_c/T_c$ of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$.

V. SUMMARY AND DISCUSSION

A recent experiment of angular resolved photoemission using single crystals of Bi-Sr-Ca-Cu-O shows a clear Fermi edge and dispersion in the excitation spectra.²⁶ This experiment suggests that the Fermi-liquid state appears near the Fermi level in the metallic samples of the copper oxides. It seems natural to consider that the normal and superconducting currents are carried in this Fermi liquid. The experimental fact that the carrier unit of the superconducting current in the oxides is twice the electronic charge^{4–7} implies that the superconductivity is generated by the pairs of the particles constituting the Fermi liquid. The nature of the pairing is of the BCS type.

The problem is the origin of the large binding energy for the pairing. The phonon-mediated interaction has been shown to be too weak to account for the high- T_c superconductivity in the oxides.⁴⁵ The observed isotope effect is much weaker than that predicted from the BCS theory.^{38–44} We, therefore, should seek a nonphonon mechanism accountable for the high- T_c superconductivity in the oxides. Since large spin and charge fluctuations are in the copper oxides, the interaction mediated by these fluctuations are candidates for the pairing interaction. The recent experiments of the nuclear spin relaxation of Cu and ^{17}O confirm that the pairing is of the s -wave symmetry.²⁰ The spin fluctuations work destructively rather than constructively on the pairs.

When the charge fluctuations are large in a highly correlated electron system such as the oxide superconductors, the electronic dielectric constant can be negative. In this system, the charge fluctuations strongly couple with LO phonons. We studied the superconductivity in the sys-

tem and found that T_c is very much enhanced. We found also that the isotope effect, that is, the decrease of T_c when ^{16}O 's in the sample are replaced by ^{18}O 's, is weakened as T_c increases, consistent with the experiments.³⁸⁻⁴⁴ In the above, we assume that the electronic dielectric constants are negative for some wave numbers in the oxides. This assumption will turn out to be reasonable from the following experimental results. As discussed in Sec. III, the frequencies of the LO phonons with some wave numbers for which the electronic dielectric constants are negative shift to the frequencies lower than the frequencies of the corresponding TO phonons. This kind of shift has been observed in single crystals of $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ and La_2NiO_4 .^{33,36} Measurements of the phonon dispersion using single crystals of other superconducting oxides are anticipated. The fact that the spectral intensity of the phonon measured by electron energy loss spectroscopy is enhanced in the metallic sample of $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ (Ref. 29) may provide further evidence for the negative electronic dielectric constant, as discussed in paper I.

Recently, Cava *et al.* discovered a new high- T_c superconducting oxide, $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$.⁴⁷⁻⁴⁹ Since this oxide in-

cludes no Cu ion, we may not expect a large spin fluctuation and thus the strong interaction mediated by the spin fluctuation. The photoemission experiments in the copper oxides show that the d -electron levels of Cu are close to the p -electron levels of O in the crystals.²¹⁻²³ This electronic level configuration causes large charge fluctuations due to the Cu-O charge transfer.^{50,51} If the $6s$ electron level of Bi is close to the p -electron level of O in $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$, we expect a large charge fluctuation in the crystal too. In this case the mechanism for high- T_c superconductivity presented in this paper may also be applied to the superconductivity in $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$.

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