The Transition from the d- to s-State Due to Thermal Fluctuation for High-T_c Superconductors as an Evidence from the Microwave Penetration-Depth Measurement

Li Jui Chen and Juh Tzeng Lue

***Abstract*— A temperature dependence of the penetration depth $\lambda(T)$ measurement for the high-T_c superconductors $YBa_2Cu_3O_{7-\delta}$ and $Tl_2Ba_2CaCu_2O_7$ thin films elucidates a T^2 dependence at low temperatures and an exponential dependence at high temperatures. The transition temperature for the shift from T^2 to exponential dependence decreases as the duration for the samples exposed to air increases. An impurity scattered mechanism to fluctuate a pure d-wave to the s-wave by thermal fluctuaion is proposed for the pairing states of these high-T_c superconducting films.**

I. Introduction

One of the important issue in high-T_c superconductivity is the controversy about the order parameters in superconductors. In order to determine the superconducting pairing states, the radio frequency (RF) properties detected by the temperature dependence of microwave surface-impedance measurements are used to illustrate the surface resistance R_s and the change of magnetic penetration depth $\Delta\lambda$. Conceptually, the classical Drude model is used to interpret the frequency dependence on the complex conductivity and scattering mechanism for conduction electrons in metals. Although a number of theoretical investigations have proposed that d-wave symmetry of the pairing state occurs in high-T_c superconductors [1], [2], there is currently no undeniable consensus on this concern [3]. A second intriguing possibility is the proposal of the s and $d_{x^2-y^2}$ mixed waves [4]–[7].

In conventional Bardeen–Kooper–Schriefer (BCS) frameworks, the wavefunction of the superconducting electrons is isotropic as the so-called s-wave pairing. If the pairing electrons are the ideal s-wave, the energy gap will have the same value for all momentum directions on the Fermi surface. However, if the electron pairs have the form of d-wave pairing called $d_{x^2-y^2}$, the shape of both energy gaps in k space and wavefunction in real space exhibits a four-leaf shape and the gap function can be described by $\Delta(k) = \Delta_0(\cos k_x a - \cos k_y a)$. By the special gap function there are zeros, namely "nodes," at some momentum values by which less energy is needed to excite the superconducting electrons. For probing the symmetry of the high-T_c superconducting states, a large variety of experiments were designed, which are characterized into three categories by: 1) spectroscopy; 2) magnetization (or transport phenomena); and 3) Josephson tunneling measurements. The first two methods are intend to measure the excitation of superconducting electrons, the last one is to investigate the shape of wavefunction.

In the aspect of microwave absorption measurement, following the delicate split-ring resonator experiments done by Hardy's group [8], [9], the high-T_c superconductors behave nearly and evidently as a d-wave. However, recent studies [4] focusing on nonperfect single crystals show that the effect of impurities has the implication of exciting the s-wave. As this is a fuller perspective from manipulating the magnetic-field penetration depth based on Ginsberg–Landau theory and Gorkov's equations, an infinitesimal concentration of impurities can excite an amount of the s-wave density of states at the Fermi surface even for a d-wave superconductor. The theory proposed by Ichioka *et al.* [10] suggests that the s-wave (or d_{xy}-wave) component of the order parameter can be induced around a vortex in $d_{x^2-y^2}$-wave superconductors with four-lobe clovers. Josephson tunneling experiments seem to provide the similar results for highly twinned crystals or on the film surfaces [7], [11]. A model composing a small admixture of the s-component to the $d_{x^2-y^2}$-wave without increasing the number of nodes, but merely displacing the node away from the original 45° angular position is also proposed [12]. Recent important works by Jacobs' group [13]–[15] suggest that the temperature-dependent $R_s(T)$ and $\lambda(T)$ can be explained by a weakly coupled $d_{x^2-y^2}$-order paramater in the form of $\Delta(T,\varphi) = \Delta_d(T)\cos(2\varphi)$ and they can satisfactorily fit the data by the modified energy gap $\Delta_d(0) = 2.16kT_c$ or larger temperature range with respect to the crossover from d- to s-behavior. There seems no incontrovertible evidence to determine the symmetry of the order parameter without ambiguity.

To explore this fact, various experimental techniques are designed such as field exclusion and the split-ring resonator method [8], [9] for single crystals, cavity wall replacement, the transmission method, parallel-plane resonator method, confocal cavity method, microstrip-resonator method, and dielectric-

Manuscript received September 2, 1997; revised March 22, 1998. This work was supported by the National Science Council, Taiwan, R.O.C., under Contract NSC86-2112-M007-002.

The authors are with the Department of Physics, National Tsing Hua University, Hsinchu, Taiwan 30043, R.O.C.

Publisher Item Identifier S 0018-9480(98)06144-4.

resonator method [16]–[18] for thin films. Among these methods, the dielectric-resonator method has the advantage of simplicity and accuracy. If the loss of the dielectric is small, this method, in principle, gives a very direct measurement of the surface impedance of unpatterned films. It is unsuitable for measurement of absolute penetration depth λ by a conventional dielectric resonator [19], while only the $\Delta\lambda(T)$ might be extracted if thermal expansion and other background effects are taken into account. We will present details of calculations and experiments in this paper.

In this paper, we attempt to probe the pairing symmetry by measuring $\lambda(T)$ of high-T_c $YBa_2Cu_3O_{6.95}$ (YBCO) and $Tl_2Ba_2CaCu_2O_7$ (Tl-2212) thin films. This experiment clearly reveals that $\lambda(T)$ follows the d-wave symmetry at low-temperature regions and becomes more prominent to be fitted by the BCS symmetry at high temperatures near the critical temperature T_c. It seems plausible that the pairing state is fluctuated from a pure d-wave to mixed s- and d-waves, and then a pure s-wave as temperature increases. For samples stored in air, the transition temperatures from d- to s-waves decreases as the stored time increases, implying that the s-states are more easily excited from the d-states as the impurity phase increase for these high-T_c superconductors.

II. Electrodynamics of Surface Resistance

In the case of local limit, where the superconducting coherence length ξ is much smaller than the London penetration depth λ, i.e., $\xi \ll \lambda$, the surface impedance can be expressed by $Z_s = R_s + iX_s$, where X_s is the surface reactance. The complex surface impedance is written as

$$Z_s = \left(\frac{i\mu_0\omega}{\sigma}\right)^{1/2} \tag{1}$$

where σ is a complex conductivity, which is written as [16]

$$\sigma(\omega, T) = \sigma_1(\omega, T) + i\frac{1}{\mu_0\omega\lambda^2(T)}. \tag{2}$$

In the superconducting state, the real and imaginary parts of Z_s at the microwave frequency are derived as

$$\begin{aligned} R_s &= \frac{\mu_0^2}{2}\omega^2\lambda^3(T)\sigma_1(\omega, T) \\ X_s &= \mu_0\omega\lambda(T) \end{aligned} \tag{3}$$

where $R_s(T)$ and $\lambda(T)$ are related by $\sigma_1(\omega, T)$, which is manipulated by the scattering of normal electrons.

By using a cavity perturbation technique, the deviation of the penetration depth $\lambda(T)$ from its low-temperature value $\lambda(0)$, defined as $\Delta\lambda(T) \equiv \lambda(T) - \lambda(0)$, is measured. For an s-wave (BCS) pairing states excited by finite energy, the $\Delta\lambda(T)$ calculated by Mattis–Bardeen (MB) [20], [21] is given by

$$\frac{\Delta\lambda(T)}{\lambda(0)} \sim 3.33\left(\frac{T_c}{T}\right)^{1/2}\exp(-\Delta/T) \tag{4}$$

where $\Delta = \alpha T_c$ is the energy gap at 0 K, which is equal to $1.76T_c$, as derived by Mühlschlegel [22]. The MB equation is appropriate for most pure elemental and A15 superconductors.

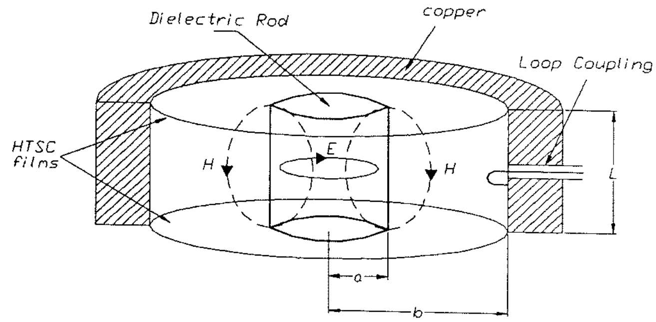

Fig. 1. Schematic structure of the dielectric resonator composed of HTSC films and dielectric material (sapphire). The TE_{011} field pattern is also plotted.

However, in an MB fitting of $\Delta\lambda(T)$ for high-temperature superconductors (HTSC's), α might span a range from 0.4 to 3 and does not reveal any definite value in some experiments [23]–[25]. Another possible mechanism is the Gorter–Casimir (GC) relation [26], derived from the two-fluid model as given by

$$\frac{\lambda(T)}{\lambda(0)} = \left[1 - \left(\frac{T}{T_c}\right)^4\right]^{-1/2}. \tag{5}$$

However, the GC relation is closely related to the BCS theory [21], [27]. If there are line nodes on the Fermi surface, $\Delta\lambda(T)$ is calculated to be proportional to T^p [28], in which $p = 1$ for the simplest form of a gap with the d-wave. An impurity or defect scattering can change the T dependence to T^2 [29]–[31]. In general, powers of T, T^2, T^3, and T^4 are possible depending on the type of nodes and the orientation of the applied fields with respect to the crystal axis [28], [31].

The studies of the temperature dependence of R_s have been performed by many researchers with two main themes. One is, what is the lowest limit of the surface impedance at zero temperature for high-T_c superconductors? To solve this problem, a criterion for the d-wave limit connecting the superconducting pairing states is proposed by Lee [32], [33]. The second point is the temperature-dependent behavior of $R_s(T)$. From the BCS theory, the R_s for either isotropic or anisotropic gap superconductors yields an $\exp(-\Delta/T)$ relation [21], since the density of the excited quasi-particles roughly depends on temperatures exponentially. For the d-wave superconductors, the temperature dependence of $R_s(T)$ is generally recognized to be linear at low temperatures [34]. Furthermore, Hardy *et al.* show that nonmonotonic decrease of $R_s(T)$ with temperature can be observed in the clean limit.

III. Experimental Setup

A dielectric microwave resonator consisting of a small sapphire rod sandwiched by two high-T_c superconducting films is used for the surface-impedance measurement, as shown in Fig. 1. The cavity is preferentially excited by the fundamental TE_{011} mode by which the electromagnetic (EM) fields are mostly confined in the sapphire rod due to its high dielectric constant, while with low-current loss on the outside copper wall, resulting in a high Q-value. The input antenna is a current loop connected to an HP 8510 (some data are performed by an HP8722D) network analyzer by a coaxial transmission line and 2.4-mm connectors for detecting the s_{11} reflection parameter. The copper cavity is intimately mounted on a cryogenic cooling stage, with the achieved lowest temperature to be 17 K. As the temperature decreases, the resonant frequency shifts toward a higher value of above 28.75 GHz, predominantly due to the reduction of penetration depth of thin films and the copper wall.

IV. Theoretical Calculations

Resonant Frequency ω_0

The EM fields solved from Maxwell wave equations in a cylindrical coordinate system (ρ, θ, z) are expressed as follows [17]:

for $\rho \le a$:

$$\begin{aligned} H_{\rho 1} &= -\frac{\beta}{\xi_1} A J_1(\xi_1 \rho)\cos\beta z \\ E_{\theta 1} &= i\frac{2\pi\mu_0 f}{\xi_1} A J_1(\xi_1\rho)\sin\beta z \\ H_{z1} &= A J_0(\xi_1\rho)\sin\beta z. \end{aligned} \tag{6}$$

for $a \le \rho \le b$:

$$\begin{aligned} H_{\rho 2} &= \frac{\beta}{\xi_2}[CK_1(\xi_2\rho) - DI_1(\xi_2\rho)]\cos\beta z \\ E_{\theta 2} &= -i\frac{2\pi\mu_0 f}{\xi_2}[CK_1(\xi_2\rho) - DI_1(\xi_2\rho)]\sin\beta z \\ H_{z2} &= [CK_0(\xi_2\rho) + DI_0(\xi_2\rho)]\sin\beta z \end{aligned} \tag{7}$$

where a is the radius of the sapphire rod, b is the inner radius of the copper cavity, L is the length of the sapphire rod

$$\begin{aligned} \beta &= \frac{\pi}{L} \\ \beta^2 &= k^2\epsilon_r^2 - \xi_1^2 = k^2 + \xi_2^2 \end{aligned} \tag{8}$$

and A, C, D are undetermined amplitude coefficients, μ_0 is the magnetic permeability of free space, and f is the frequency. The $J_n(x)$ is the nth-order Bessel function of the first kind, $K_n(x)$ and $I_n(x)$ are the modified Bessel functions of nth order. ξ_1 and ξ_2 are wavenumbers along the ρ-direction inside and outside the rod, respectively, and β is the propagation constant in the z-axis.

To expedite the experimental measurement, the dimension of the dielectric resonator should be carefully designed. The continuity equations on boundary at $\rho = a$ and b imply

$$\begin{aligned} \frac{J_1(\xi_1 a)}{\xi_1 a}A + \frac{K_1(\xi_2 a)}{\xi_2 a}C - \frac{I_1(\xi_2 a)}{\xi_2 a}D &= 0 \\ J_0(\xi_1 a)A - K_0(\xi_2 a)C - I_0(\xi_2 a)D &= 0 \\ K_1(\xi_2 b)C - I_1(\xi_2 b)D &= 0. \end{aligned} \tag{9}$$

To obtain a nonzero solution for the coefficients A, C, D, the secular determinant must be zero or

$$\begin{vmatrix} \xi_2 J_1(\xi_1 a) & \xi_1 K_1(\xi_2 a) & -\xi_1 I_1(\xi_2 a) \\ J_0(\xi_1 a) & -K_0(\xi_2 a) & -I_0(\xi_2 a) \\ 0 & K_1(\xi_2 b) & -I_1(\xi_2 b) \end{vmatrix} = 0 \tag{10}$$

the above equation yields the resonant frequency ω_0, and the coefficients C and D, which are given by

$$\begin{aligned} C &= \frac{J_0(\xi_1 a)I_1(\xi_2 b)A}{K_0(\xi_2 a)I_1(\xi_2 b) + K_1(\xi_2 b)I_0(\xi_2 a)} \\ D &= \frac{K_1(\xi_2 b)J_0(\xi_1 a)A}{K_0(\xi_2 a)I_1(\xi_2 b) + K_1(\xi_2 b)I_0(\xi_2 a)}. \end{aligned} \tag{11}$$

Using similar approaches, the resonant frequency for higher order TE_{0ml} modes can be calculated.

B. Surface Resistance R_s

In this experiment, the surface resistance R_s can be transformed from the quality (Q) factor of the resonator, which is defined as

$$Q \equiv \omega_0 \frac{W}{P} = 2\pi f_0 \frac{W_1 + W_2}{P_s + P_c + P_d} \tag{12}$$

where W is the total energy of the cavity, including the field energy stored inside and outside the rod, as denoted by W_1 and W_2. The power loss P is composed of P_s, P_c, and P_d, representing, respectively, with the power dissipation of superconducting films, copper wall, and dielectric rod. The power loss expressed as $P_d = 2\pi f W_1 \tan\delta$ is much smaller than P_s and P_c since the loss tangent $\tan\delta \approx 10^{-7}$ for sapphire at 30 GHz.

The components of stored energies and power losses are listed as follows:

$$\begin{aligned} W_1 &= \frac{\epsilon_0\epsilon_r}{2}\int_{V1} E_{\theta 1}^* E_{\theta 1}\, dv \\ &= \frac{\epsilon_0\epsilon_r \pi L}{2}\left(\frac{2\pi\mu_0 f}{\xi_1}\right)^2 \int_0^a J_1^2(\xi_1\rho)\rho\, d\rho \\ W_2 &= \frac{\epsilon_0}{2}\int_{V2} E_{\theta 2}^* E_{\theta 2}\, dv \\ &= \frac{\epsilon_0 \pi L}{2}\left(\frac{2\pi\mu_0 f}{\xi_2}\right)^2 \int_a^b [CK_1(\xi_2\rho) - DI_1(\xi_2\rho)]^2 \rho\, d\rho \\ P_s &= \frac{R_s}{2}\left(\int_{S1} H_{\rho 1}^* H_{\rho 1}\, ds + \int_{S2} H_{\rho 2}^* H_{\rho 2}\, ds\right) \times 2 \\ &= R_s \cdot 2\pi \cdot \left\{ \left(\frac{\pi}{\xi_1 L}\right)^2 \int_0^a J_1^2(\xi_1\rho)\rho\, d\rho \left(\frac{\pi}{\xi_2 L}\right)^2 \right. \\ &\qquad \left. + \int_a^b [CK_1(\xi_2\rho) - DI_1(\xi_2\rho)]^2 \rho\, d\rho \right\} \\ &\equiv R_s \cdot gs, P_c \\ &= \frac{R_c}{2}\int_{\mathrm{Wall}} H_{z2}^* H_{z2}\, ds \\ &= \frac{R_c \pi b L}{2}[CK_0(\xi_2 b) + DI_0(\xi_2 b)]^2 \\ &\equiv R_c \cdot gw. \end{aligned} \tag{13}$$

To check the correct measurement of $R_s(T)$, we first examine the $R_c(T)$ by substituting oxygen-free copper plates for HTSC films. The unloaded Q reveals the information of $R_c(T)$ by $Q_0 = 2\pi f_0 \cdot (W_1 + W_2)/R_c(gs + gw)$. The measured R_c can fit quite well with the conductivity of copper derived from the anomalous skin-effect region, which is valid at high frequency and low temperatures.

C. Extraction of $\Delta\lambda(T)$ from $\Delta f(T)$

The change of penetration depth $\Delta\lambda$ from the frequency shift Δf is derived in this paper. Slater [35] first formulated the resonant-frequency shift of a cavity from the change of the stored energy by

$$\frac{\omega-\omega_0}{\omega_0}=\frac{\int_{\delta V}(\mu H_0\cdot H_0^*-\epsilon E_0\cdot E_0^*)\,dv}{\int_{V_0}(\mu H_0\cdot H_0^*+\epsilon E_0\cdot E_0^*)\,dv} \tag{14}$$

where ω is the perturbed frequency and ω_0 is the unperturbed value, $\delta V\equiv V-V'$ is the change of volume, which is defined to be positive for a contracted cavity. The volume change can be replaced by the energy, which implies

$$\frac{\delta\omega}{\omega_0}=\frac{\delta W_m-\delta W_e}{W} \tag{15}$$

where δW_m and δW_e are the change of magnetic and electric energies due to the volume change δV, respectively. Presumably, the size change of the copper cavity with temperatures is small, also due to the weak field on the copper wall, and it is plausible to use the perturbation theory to calculate (14).

Assuming the volume change of the sapphire rod with temperatures is negligible, while those for the copper cavity in the ρ- and z-directions are $\Delta\rho(T)$ and $\Delta z(T)$, the change of resonant frequency depends only on $\Delta\rho$ and Δz. As the temperature decreases below T_c, we have

$$\begin{aligned} d\rho &= \Delta\rho \\ dz &= \Delta\lambda+\Delta z/2 \end{aligned} \tag{16}$$

where $d\rho$ and dz are the total length reductions along the radial and azimuthal directions of the cavity, respectively. For a magnetic-field coupled TE_{011} cavity mode, $\delta W_e\approx 0$ and δW_m at wall and plates are

$$\begin{aligned} \delta W_{m,\text{wall}} &= \mu\int_{\delta V}[CK_0(\xi_2 b)+DI_0(\xi_2 b)]^2 \\ &\quad\cdot\sin^2\beta z\cdot 2\pi b\cdot d\rho\,dz \\ &= \mu\pi bL[CK_0(\xi_2 b)+DI_0(\xi_2 b)]^2\,d\rho \\ &= 2\mu\cdot gw\cdot d\rho \\ \delta W_{m,\text{plates}} &= 2\mu\left\{\left(\frac{\pi}{\xi_1 L}\right)^2\int_0^a J_1^2(\xi_1\rho)2\pi\rho\,d\rho\,dz+\left(\frac{\pi}{\xi_2 L}\right)^2\right. \\ &\quad\left.\cdot\int_a^b[CK_1(\xi_2\rho)-DI_1(\xi_2\rho)]^2 2\pi\rho\,d\rho\,dz\right\} \\ &= 2\mu\cdot gs\cdot dz. \end{aligned} \tag{17}$$

Substituting (16) into (14), we obtain

$$\begin{aligned} \frac{\delta\omega}{\omega_0} &= \frac{2\mu\cdot gw\cdot d\rho+2\mu\cdot gs\cdot dz}{W_1+W_2} \\ \frac{\delta\omega}{\omega_0} &= \frac{2\mu\cdot[gw\cdot\Delta\rho+gs\cdot(\Delta\lambda+\Delta z/2)]}{W_1+W_2}. \end{aligned} \tag{18}$$

The coefficient of thermal expansion $\alpha(T)$ is conceptually relative to the phonon energy dispersion [36] and can be estimated to yield

$$\alpha(T)=9.35\times 10^{-12}T+6.49\times 10^{-13}T^3,\quad \text{for pure copper.} \tag{19}$$

After obtaining the desired $\Delta\rho(T)$ and $\Delta z(T)$, we can readily calculate $\Delta\lambda(T)$ by using (18) and (19).

V. Results and Conclusion

We first transform the values of frequency shift into the temperature dependence of the change of magnetic penetration depth $\Delta\lambda(T)\equiv\lambda(T)-\lambda(20K)$ for $Tl_2Ba_2CaCu_2O_7$ thin films, as depicted in Fig. 2. The thickness of the superconducting film prepared by laser ablation on $LaAlO_3$ substrates is approximately 700 nm, thus, we can subsequently estimate $\lambda(20K)\approx 190$ nm by the value of the highest point. Above 65 K, the penetration depth is too large to confine the EM waves for a cavity; the Q values measured at this temperature cannot correctly inform us of the true characteristics of the thin film.

Below 60 K, the measured data can be satisfactorily fitted by the T^2 law. However, only the MB or GC equations can yield better agreement above 60 K. The inserted magnetization curve showing a slow change of shoulder clearly addresses that the films contain some defects or impurities. For comparison, the samples stored in a dry box for two months were measured, as shown in the same plot, by which we observed that the transition temperature from d- to s-wave is lowered down from 60 to 32 K. This suggests that the superconducting films with impurity phases are much instable to be readily fluctuated from their original d- to s-states. The muon–spin–depolarization measurement of the magnetic-field penetration depth of superconducting $GdBa_2Cu_3O_x$ performed by Cooke *et al*. [37], [38], as depicted in Fig. 2, indicates that for pure phase samples, the $\Delta\lambda(T)$ plot is well described by BCS theory.

Another $\Delta\lambda(T)$ plot for a YBCO film with an onset critical temperature near 90 K (see Fig. 3) also shows the same trends, but yields a transition temperature near 36 K. In Fig. 3, the treatment of the sample is arranged: 1) by being stored in a dry box for one month; 2) after being stored in a dry box, we exposed them in air with a relative humidity of 70% for 12 h; and 3) by being exposed to air for 52 h. The critical temperature T_c degrades as the duration of the films exposed in air increases. In this measurement, we first observed that the temperature dependence on $\Delta\lambda(T)$ is different from those measured for high-quality YBCO single crystals [8], [9]. A close scrutiny of these experiments exhibits the extrinsic effects on the pairing states. Fig. 3 clearly indicates that increasing the amounts of impurities or defects, the $\Delta\lambda(T)$ for the d-wave pairing states can be fluctuated from T to T^2. The crossover temperatures which separate the T^2 (d-wave) and MB (s-wave) fittings increase with the same trend as the result for the T1-2212 sample. At higher temperatures, the thermal fluctuation will violate the pure d-state to become the mixed $d_{x^2-y^2}$ and s-wave, and then to the s-wave as the temperature increases further.

Compared to previous studies [8], [9], [29], we conclude that for a high-quality high-T_c single crystal, the temperature dependence on $\Delta\lambda(T)$ is linear from 3 to 25 K. For a

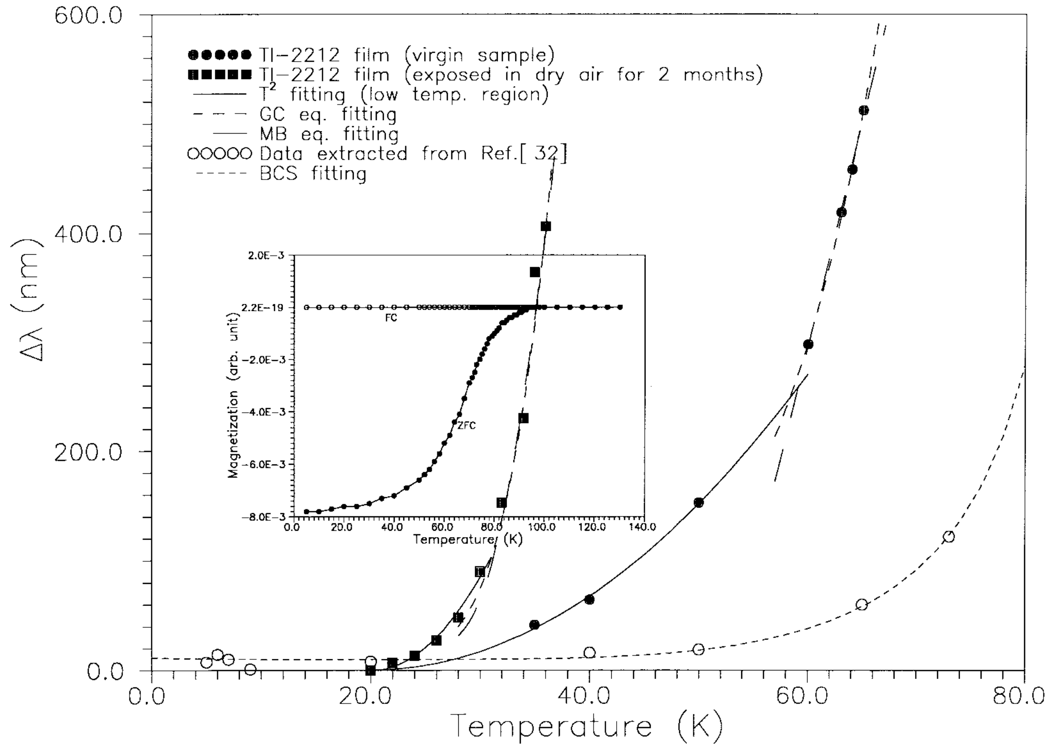

Fig. 2. The measured $\Delta\lambda(T)$ and theoretical fitting for the Tl-2212 superconducting film. The inset is the SQUID data.

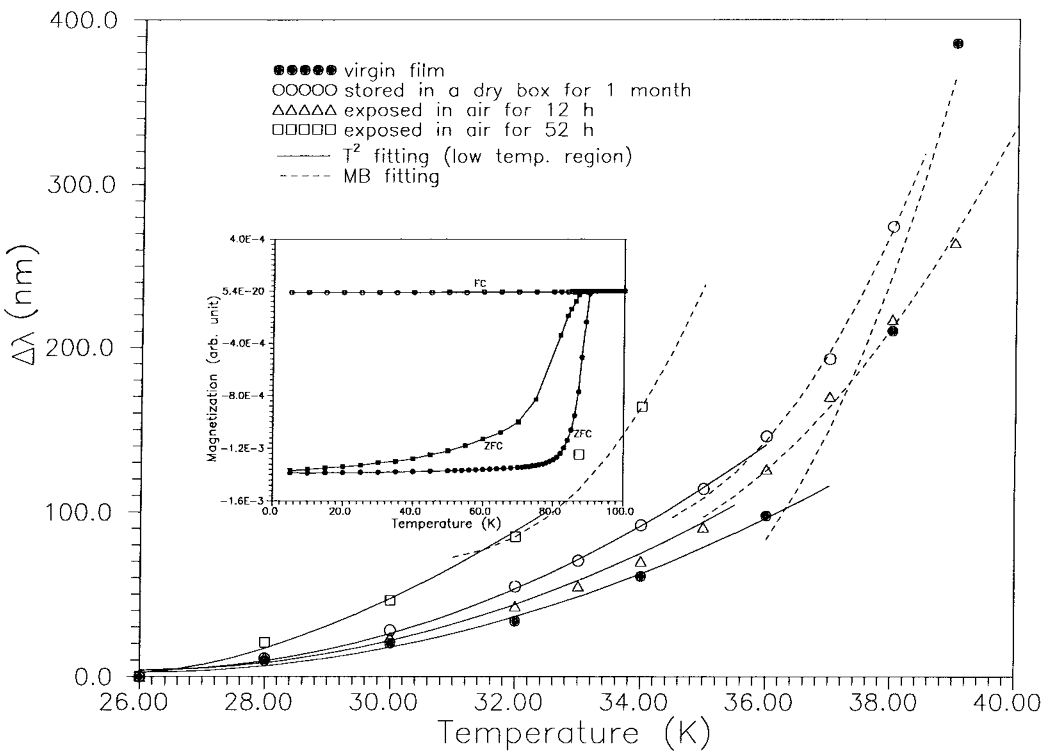

Fig. 3. The $\Delta\lambda(T)$ of the YBCO thin film of various storing conditions. The transition temperatures from d- to s-states decrease as the sample stored in air is longer.

perfect thin film or common-quality crystal, this temperature dependence will follow T^2. Presumably, for films containing impurity phases or defects, $\Delta\lambda(T)$ is proportional to T^2 in the low-temperature region, and then exhibit exponential tendency at high temperatures. The MB and GC fittings exhibit the same trend in the high-temperature region. However, there is no way to fit the data by the MB equation at low temperature, irrespective to whatever adjusting of the gap parameter α in (4). Our results are debatable with those of Klein *et al.* [23], [24].

Our results are compared to the works of Jacobs *et al.* [13]–[15]. In order to understand the deviations from the clean-limit calculation, they suggest that three effects can be taken into consideration. These are: 1) the strong coupling to incorporate larger gaps; 2) the temperature dependent inelastic scattering; and 3) the fluctuation effect. In our fittings, however, the use of a larger modified gap function does not exhibit better agreements, showing that the strong-coupled d-wave model is not profitable to explain our data, and we recommend that the fluctuations be dominant. Also, as seen in the calculations of Xu *et al.* [5], [6] [considering both the Born limit (weak scattering) as well as the unitary limit (strong scattering case)], an s-wave can be regenerated from a pure d-wave in bulky superconductors. Accompanied with d-wave components, both states are weakened by the impurity scattering. This experimental work demonstrate a fact that the generating temperature and the magnitude of s-wave pairing states are not accompanied with the same strategy. It needs further investigation by other experimental methods for observing the four-folded symmetric structure of a $d_{x^2-y^2}$-wave vortex. However, one recent t–U–V (hopping, repulsive on-site, and attractive at distance) [39] Hubbard calculation, leading to the result that the binding energy for the d-pairing state is lower than that for the s-state, strongly provides a correct prospect to this presumption.

ACKNOWLEDGMENT

The authors are indebted to Dr. H. C. Lai, Industrial Technology Research Institute, Hsinchu, Taiwan, R.O.C., and Prof. K. H. Wu, National Chiao Tung University, Hsinchu, Taiwan, R.O.C., for providing the films.

REFERENCES

[1] H. J. Schulz, *Europhys. Lett.*, vol. 4, pp. 609–615, 1987.
[2] C. Gross, R. Joynt, and T. M. Rice, *Z. Phys. B, Condens. Matter*, vol. 68, pp. 425–430, 1989.
[3] W. A. Little, *Science*, vol. 242, pp. 390–395, 1988.
[4] R. Joynt, *Phys. Rev. B, Condens. Matter*, vol. B41, pp. 4271–4277, 1990.
[5] W. Xu, Y. Ren, and C. S. Ting, *Phys. Rev. B, Condens. Matter*, vol. B53, pp. 12 481–12 495, 1996.
[6] ——, *J. Phys. Chem. Solids*, vol. 56, p. 1749–1756, 1995.
[7] M. B. Walker, *Phys. Rev. B, Condens. Matter*, vol. B53, pp. 5835–5838, 1996.
[8] W. N. Hardy and D. A. Bonn *et al.*, *Phys. Rev. Lett.*, vol. 70, pp. 3999–4002, 1993.
[9] ——, *Phys. Rev. B, Condens. Matter*, vol. B47, pp. 11 314–11 328 1993.
[10] M. Ichioka, N. Enomoto, N. Hayashi, and K. Machida, *Phys. Rev. B, Condens. Matter*, vol. B53, pp. 2233–2236, 1996.
[11] K. A. Müller, *J. Phys. Soc. Jpn.*, vol. 65, pp. 3090–3091, 1996.
[12] B. Chattopadhyay, D. M. Gaitonde, and A. Taraphder, *Europhys. Lett.*, vol. 34, pp. 705–710, 1996.
[13] T. Jacobs, S. Sridhar, C. T. Rieck, K. Sharnberg, T. Wolf, and J. Halbritter, *J. Phys. Chem. Solids*, vol. 56, p. 1945–1949, 1995.
[14] ——, *Phys. Rev. Lett.*, vol. 75, pp. 4516–4519, 1995.
[15] ——, *J. Low. Temp. Phys.*, vol. 105, pp. 503–508, 1996.
[16] O. Llopis and J. Graffeuil, *J. Less-Common Met.*, vols. 164 and 165, pp. 1248–1251, 1990.
[17] Z. Y. Shen, C. Wilker, P. Pang, W. L. Holstein, D. Face, and D. J. Kountz, *IEEE Trans. Microwave Theory Tech.*, vol. 40, pp. 2424–2427, 1992.
[18] A. P. Gusev, Z. F. Kaplun, V. P. Kiryushin, V. V. Malov, and A. S. Tager, *Instrum. Exp. Tech.* vol. 37, pp. 314–316, 1994.
[19] D. A. Bonn and W. N. Hardy, "Microwave surface impedance of high temperature superconductors," in *Physical Properties of High Temperature Superconductors V*, D. M. Ginsberg, Ed. Singapore: World Scientific, 1996, p. 7.
[20] D. C. Mattis and J. Bardeen, *Phys. Rev.*, vol. 111, pp. 412–421, 1958.
[21] J. P. Turneaure, J. Halbritter, and H. A. Schwettman, *J. Supercond.*, vol. 4, pp. 341–355, 1991.
[22] B. Mühlschlegel, *Z. Phys.*, vol. 155, pp. 313–327, 1959.
[23] N. Klein, N. Tellmann, H. Schulz, K. Urban, S. A. Wolf, and V. Z. Kresin, *Phys. Rev. Lett.*, vol. 71, pp. 3355–3358, 1993.
[24] ——, *J. Supercond.*, vol. 5, pp. 195–200, 1992.
[25] S. M. Anlage, B. W. Langley, G. Deutscher, J. Halbritter, and M. R. Beasley, *Phys. Rev. B, Condens. Matter*, vol. 44, pp. 9764–9767, 1991.
[26] C. J. Gorter and H. B. G. Casimir, *Phys. Z*, vol. 35, pp. 963–969, 1934.
[27] A. J. Berlinky, C. Kallin, G. Rose, and A. C. Shi, *Phys. Rev. B, Condens. Matter*, vol. B48, pp. 4074–4079, 1993.
[28] J. F. Annett, N. Goldenfeld, and S. R. Renn, *Phys. Rev. B, Condens. Matter*, vol. 43, pp. 2778–2782, 1991.
[29] Z. Ma, R. C. Taber, L. W. Lombardo, and A. Kapitulnik *et al.*, *Phys. Rev. Lett.*, vol. 71, pp. 781–784, 1993.
[30] F. Gross, B. S. Chandrasekhar, D. Einzel, K. Andres, P. J. Hirschfeld, H. R. Ott, J. Beuers, Z. Fisk, and J. L. Smith, *Z. Phys. B, Condens. Matter*, vol. 64, pp. 175–188, 1988.
[31] M. Prohammer and J. P. Carbotte, *Phys. Rev. B, Condens. Matter*, vol. 43, pp. 5370–5374, 1991.
[32] P. A. Lee, *Phys. Rev. Lett.*, vol. 71, pp. 1887–1890, 1993.
[33] Y. Hatsugai and P. A. Lee, *Phys. Rev. B, Condens. Matter*, vol. 48, pp. 4204–4207, 1993.
[34] M. R. Beasley, *IEEE Trans. Appl. Superconductivity*, vol. 5, pp. 141–146, 1995.
[35] J. C. Slater, *Microwave Electronics*. New York: Van Nostrand, 1950, p. 289.
[36] H. M. Rosenberg, *Low Temperature Solid-State Physics*. London, U.K.: Oxford Univ. Press, 1988, p. 36.
[37] D. W. Cooke *et al.*, *Phys. Rev. B, Condens. Matter*, vol. B37, pp. 9401–9404, 1988.
[38] ——, *Phys. Rev. B, Condens. Matter*, vol. B39, pp. 2748–2751, 1989.
[39] A. Nazarenko, A. Moreo, E. Dagotto, and J. Riera, *Phys. Rev. B, Condens. Matter*, vol. B54, pp. R768–R771, 1996.

Li Jui Chen, photograph and biography not available at the time of publication.

Juh Tzeng Lue, photograph and biography not available at the time of publication.