利用環型及線型共振器研究摻鈣釔系高溫超導薄膜之微波物理性質

研究生：賴良星　指導教授：郭義雄 教授

國立交通大學
電子物理系

中文摘要

經由脈衝雷射蒸鍍技術，我們成功地在鋁酸鑭(LAO)(100)基板的上、下二面蒸鍍釔鋇銅氧（$YBa_2Cu_3O_{7-\delta}$）及摻鈣釔鋇銅氧($Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$)之高溫超導薄膜以研製成環型或線型的超導微條共振器，並且藉由控氧技術，控制電洞濃度p從過摻雜到低摻雜區，致使我們可以在相同樣品上完成微波量測工作。經由量測該微波元件的微波表面阻抗，$Z_s(T) = R_s(T) + jX_s(T)$，可獲知在ab平面上的複數導電率$\sigma(T)$。利用上述之結果，輔以二流體模型，我們可獲知同一樣品在不同電洞濃度時之物理特性，如倫敦穿透深度、超流體密度和複數導電率等，及它們對溫度的關係，並且將該實驗結果用來檢測現有的高溫超導理論模型，特別是李(Lee)的模型、溫-李(Wen and Lee)的模型、艾莫利-肯文生(Emery and Kivelson)的模型及米其氏瓦拉-費雪(Vishveshwara and Fisher)的模型，本論文的實驗結果如下：

I. 從量測環型及線型共振器之共振頻率的變化，並利用張的公式及利用 THz 等實驗的輔助，我們獲得釔鋇銅氧及摻鈣釔鋇銅氧在各種不同電洞濃度時，在 ab 平面上其 5K 的穿透深度之絕對值，例如在最適宜(optimum)摻雜時，ab 平面上的穿透深度$\lambda(5K) = 150nm \sim 200nm$，該結果和目前所有單晶量測$\lambda(5K) = 150nm$或薄膜量測$\lambda(5K) = 200nm$的實驗結果都頗具一致性。

II. 在低摻雜區(電洞濃度改變)，量測到的超流體密度$1/\lambda^2(5K)$和臨界溫度T_c成正比，這和Uemura關係式是一致的。但對所有摻雜區而言(從過摻雜到低

摻雜)，量測到的超流體密度$1/\lambda^2(5K)$和在T_c的直流電導率σ_{dc}跟T_c乘積值成正比，這個普遍性的關係式(universal relation)可以用Ferrell-Glover-Tinkham sum rule (FGT總和定理)解釋，並且利用FGT總和定理，得到超導能隙的大小對電洞濃度的相依性，和掃描式電子顯微鏡所量測到d-wave能隙的結果相同，這個間接證據也證實了超導能隙的對稱性是d-wave並且指出在此系統中電荷是守恆者。

III. 經由實驗和理論搭配間接得到複數電導率$\sigma = \sigma_1 - j\sigma_2$，發覺到實部電導率$\sigma_1$跟虛部電導率$\sigma_2$都具有一熱活化能隙(thermal activation gap)的存在，我們認為這個結果不僅與李(Lee)的理論模型一致並且指出古典熱擾動(thermal fluctuations)在高溫態的超導物理行為上扮演很重要的角色。並且，利用$\sigma_2(5K)(1-e^{-E_{g2}(T)/k_BT})$來擬合$\sigma_2$，發覺到$\sigma_2 = 0$時有一高於臨界溫度之能量尺度$T_{\sigma 2}^*$存在，此能量尺度$T_{\sigma 2}^*$對摻雜電洞濃度之相圖，若與Emery和Kivelson之理論預言的相圖相較，頗為一致，因此，我們推論在低摻雜區時，$T_{\sigma 2}^*$即可能代表相位有序溫度之上限$T_\theta^{\max}$，而在過摻雜區時，$T_{\sigma 2}^*$即可能代表平均場轉變溫度T^{MF}。

IV. 實部電導率$\sigma_1(T)$對溫度的關係有兩種情況，第一種情況，$\sigma_1(T)$在低溫時($T<0.1T_c$)不再明顯隨溫度變化而趨平，第二種情況，$\sigma_1(T)$在低溫時隨溫度降低而降低，然而此兩種情況實部電導率$\sigma_1(T)$皆具有一熱活化能隙$E_{g1}(T)$存在，此熱活化能隙乃是被侷限著(localized)的準粒子在此弱連(weak-link)結構中藉由穿隧所需克服的能量障壁。並且，根據米其氏瓦拉-費雪(Vishveshwara and Fisher)的理論模型，對第一種情況，$\sigma_1(T)$在超導態時是處於熱的絕緣(thermal insulator)相;但對第二種情況，$\sigma_1(T)$需再加上考慮對溫度有線性關係，這與哈地(Hardy)等人領導之研究群所做之高純度的單晶在接近潔淨極限(clean limit)下的實驗結果頗為一致。我們認為此線性溫度的貢獻可視為代表準粒子之去侷限化(delocalized)而未受弱連效應所操控。根據米其氏瓦拉-費雪(Vishveshwara and Fisher)的理論模型，此種去侷限化(delocalized)的準粒子是處於熱的金屬(thermal metal)相。因此，對第二種情況而言，此高純度之樣品，$\sigma_1(T)$在超導態時

應包含熱的絕緣(thermal insulator)與熱的金屬(thermal metal)兩相共存。

V. 實部電導率$\sigma_1(T)$在溫度趨於 0K時，皆具有一殘存的電導率,實驗上我們取此殘存電導率為$\sigma_1(5K)$，根據李的理論，在二維d-波超導体內若是存在少量雜質，則這些雜質散射子產生之準粒子可能在Unitary極限下散射。在我們樣品中，由於這些準粒子之平均自由徑l(~75nm)小於侷限長度ξ_L(~202nm)，致使這些準粒子被侷限住(localized)，此時其電導率趨近於一個常數$e^2/2\pi\hbar\times\xi_0/a$($\xi_0$是相干常度，$a$是晶格常數)。之後，德斯特-李(Durst and Lee)的理論模型提出此常數尚要加入費米液體修正因子α^2和頂點修正因子(vertex correction factor)β才是正確的殘存電導率。因此，我們根據實驗上量得的殘存電導率$\sigma_1(5K)$而求出β值約為 1.5±1.0，此結果與德斯特跟李的理論預期頗具一致性。事實上，我們也觀測到穿透深度在低溫區時($T<0.1T_c$)幾乎趨平而不隨溫度改變，此結果和李的理論預期一致，顯示出在有雜質或無序(disorder)的二維d-波超導体內，準粒子很容易因為雜質存在而被侷限化。

VI.根據溫-李(Wen and Lee)的理論，估算在最適宜(optimum)摻雜時高溫超導體之費米液體修正因子α^2約為 0.5 附近($\alpha\approx0.7$)，這跟溫與李氏理論預測是一致的($\alpha\le1.0$)，這表示在超導態時的準粒子可用正常費米液體來描述。此外，在低摻雜跟過摻雜區時，費米液體修正常數α也都小於 1.0 並且近乎和摻雜電洞濃度無關，這顯示出在所有摻雜區中超導態時的準粒子的特性皆可用費米液體來描述。

VII.虛部電導率$\sigma_2(T)$具有一熱活化能隙$E_{g2}(T)$，此熱活化能隙代表超流體(古柏對)藉由約瑟芬穿隧(Josephson tunneling)形成長距離有序(long range order)時所需克服的能量障壁。

VIII.經由σ_1與σ_2隨溫度變化($\sigma_1=\sigma_1(5K)(1-e^{-E_{g1}(T)/k_BT})$和$\sigma_2=\sigma_2(5K)(1-e^{-E_{g2}(T)/k_BT})$)中可估算得到不同之能量尺度與摻雜電洞濃度之間關係，而此關係之相圖有類同於艾莫利-肯文生(Emery and Kivelson)以古典相位漲落說明之超導古柏對形成之理論預言的相圖，有某

程度之良好吻合。經由此吻合，我們推敲在高溫超導薄膜中超導態形成之機制由此而來，如下所述:

根據我們的實驗結果顯示，在具有少許無序(disorder)的薄膜樣品內，控制電洞濃度從過摻雜到低摻雜區，在超導態時的準粒子其基本特性皆可用費米液體來描述。當溫度高於臨界溫度T_c時，這些準粒子早已形成短距離有序(short range order)的古柏對，此時古典相位漲落扮演著一個很重要的角色。在溫度等於臨界溫度T_c時，超流體(古柏對)藉由約瑟芬穿隧(Josephson tunneling)形成長距離有序(long range order)的超導體。至於在超導態時($T<T_c$)，在我們樣品中，虛部電導率$\sigma_2(T)$皆起因於古柏對在弱連結構裡的穿隧效應，而實部電導率$\sigma_1(T)$包含兩種可能的情況，第一種，$\sigma_1(T)$可完全歸諸於準粒子在弱連結構裡的穿隧效應，亦即準粒子完全被侷限化(localized)，此時在超導態時的$\sigma_1(T)$為熱的絕緣(thermal insulator)相。第二種，$\sigma_1(T)$包含準粒子經由弱連結構的穿隧效應(準粒子被侷限化(localized))與未經由弱連效應者(準粒子去侷限化(delocalized))，亦即在超導態時的$\sigma_1(T)$包含熱的絕緣(thermal insulator)與熱的金屬(thermal metal)兩相。

Study of Physical Properties of YBCO and Ca-YBCO Thin Films Using Microstrip Ring and Line Resonators

Student : Liang-Shing Lai Advisor : Prof. Yih-Shun Gou

Institute of Electrophysics
National Chiao-Tung University

ABSTRACT

Superconducting microstrip ring and line resonators were successfully fabricated using double-side $YBa_2Cu_3O_{7-\delta}$ (YBCO) and $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ (Ca-YBCO) thin films deposited on $LaAlO_3$ (LAO) substrates by pulsed laser deposition. By controlling the oxygen contents of the same resonator, the hole concentration p determined from the empirical relation, $T_c/T_{c,max}=1-82.6(p-0.16)^2$, was controlled from the over- to the underdoped regime. The microwave surface impedance measurements, $Z_s(T) = R_s(T) + jX_s(T)$, allow us to determine the complex conductivity, $\sigma(T)$, in the ab-plane of YBCO and Ca-YBCO thin films. Using results of $Z_s(T)$ and $\sigma(T)$ together with the modified two-fluid model, the doping and temperature dependences of London penetration depth, superfluid density and complex conductivity can be systematically studied. In particular, these experimental results can be used to test the current theoretical models of high-Tc superconductivity, especially for Lee's model, Wen and Lee's model, Emery and Kivelson's model and Vishveshwara and Fisher's model. Some salient results found in the dissertation are listed below:

I. From the measurements of the temperature dependence of resonance frequency in the microstrip ring and line resonators, the absolute values of the ab-plane London penetration depth $\lambda(T)$ at 5K were obtained for YBCO and Ca-YBCO thin films with various hole concentrations by using Chang's formula together with the help of THz measurements. For example, $\lambda(5K)$ was obtained to be $150nm \sim 200nm$ at optimum doping, and this result was consistent with the one obtained from single crystal measurement ($\lambda(5K) = 150nm$) or thin film measurement ($\lambda(5K) = 200nm$).

II. In the underdoped regime, the measured superfluid density $1/\lambda^2(5K)$ is proportional to critical temperature T_c. This relation has been revealed by the Uemura relation. However, for all doping levels (from over- to underdoped regime), the measured superfluid density $1/\lambda^2(5K)$ is proportional to the product of critical temperature (T_c) and the d.c. conductivity σ_{dc} at T_c. This universal relation can be explained by using Ferrell-Glover-Tinkham (FGT) sum rule. Furthermore, the doping dependence of the normalized superconducting energy gap $2\Delta_0 / k_B T_c$ can be obtained from the FGT sum rule. Compare to the doping dependence of the superconducting d-wave gap $2\Delta_d / k_B T_c$ directly measured by scanning tunneling spectroscopy (STS), the doping dependence of $2\Delta_0 / k_B T_c$ is consistent with that measured by STS. This indirect evidence not only leads further support that the superconducting energy gap is of d-wave symmetry but also point out that the charge is conserved in the ab-plane of the cuprates.

III. The complex conductivity $\sigma = \sigma_1 - j\sigma_2$ extracted from experiments together with the help of some theoretical model gives the evidence that the real part of complex conductivity, σ_1, and the imaginary part of it, σ_2, both have thermal activation gap. This result is consistent with the prediction of Lee's model. It thus points out that the classical thermal fluctuations play an important role on physical properties of the high temperature state of high-T_c superconducting cuprates. Also, by fitting σ_2 with $\sigma_2(5K)(1-e^{-E_{g2}(T)/k_BT})$, it was found that a characteristic energy scale $T^*_{\sigma 2}$ emerges. $T^*_{\sigma 2}$ is higher than T_c, when $\sigma_2 = 0$. The phase diagram of $T^*_{\sigma 2}$ versus hole concentration p is similar to that predicted by Emery and Kivelson's model. Hence, we propose that $T^*_{\sigma 2}$ would be the upper bound on the phase ordering temperature $T_\theta^{\max}$ in the underdoped regime, and in the overdoped regime, $T^*_{\sigma 2}$ is corresponding to the mean field transition temperature T^{MF}.

IV. There are two cases for the temperature dependence of σ_1 in the low temperature regime ($T < 0.1\ T_c$). Case 1, $\sigma_1(T)$ reaches a plateau in the low temperature limit. Case 2, $\sigma_1(T)$ decreases with temperature in the low temperature limit. However, in both cases, $\sigma_1(T)$ contains a thermal activation gap $E_{g1}(T)$. The meaning of the thermal activation gap $E_{g1}(T)$ in $\sigma_1(T)$ can be attributed to the energy barrier for quasiparticle tunneling in the weak-link structures. And also, in case 1, the cuprate is the thermal insulator in the superconducting state according to the Vishveshwara and Fisher's model. But for case 2, the real part conductivity $\sigma_1(T)$ contains an extra linear-T contribution, which has been also observed for pure crystals, as reported by Hardy et al. We propose that the linear-T contribution is due to the delocalized quasiparticles free from the effects of weak links. According to Vishveshwara and Fisher, the delocalized quasiparticles would further result in the thermal metal phase in the superconducting state. Thus, for case 2, the σ_1(T) results imply that there are two phases, the thermal-metal and the thermal-insulator, in the superconducting state of some samples displaying the clean limit behaviors similar to those observed in high purity single crystal. The fact reveals that the feature of $\sigma_1(T)$ is very sensitive to the degree of disorder in the cuprate samples.

V. As the temperature approaching 0 K, σ_1(T) has a finite residual value, which we denoted as σ_1(5K). According to Lee's model, impurity scattering, particularly in the unitary limit, produces low energy quasiparticles in a two-dimensional d-wave superconductor even with small impurity concentrations. In our thin film samples, these quasiparticles are localized because the mean free path of quasiparticles ($l \sim 75nm$) is smaller than the localization length ($\xi_L \sim 202nm$). In this condition, the conductivity σ_1(T) approaches to a universal value $e^2/2\pi\hbar \times \xi_0/a$ at $T \to 0$, where ξ_0 is the coherence length and a is the lattice constant. Moreover, Durst and Lee proposed a theoretical model based on Lee's model that the universal value $e^2/2\pi\hbar \times \xi_0/a$ should be corrected by the Fermi liquid correction factor α^2 and the vertex correction factor β, which accounts for the charge current renormalization and anisotropic impurity potential, respectively. Based on the residual conductivity σ_1(5K) obtained in our experiments, we found that the value of β is about 1.5±1.0, which is consistent with that predicted by the Durst and Lee's model. In fact, we have observed that a flattening of the temperature dependence of $\lambda(T)$ in the low temperature limit, which is also consistent with that predicted by Lee's model. All of these results revealed that the quasiparticles have the tendency of being localized in a two-dimensional superconductor with small impurity concentrations or disorder.

VI. The Fermi-liquid model, proposed by Wen and Lee in 1998 to describe the basic nature of low-lying excitations, was analyzed in a quantitative manner. The obtained Fermi-liquid correction factor, α, was formed to be always smaller than one ($\alpha < 1.0$) over the entire doping range (from underdoped to overdoped) and is almost independent of hole concentration p, which is consistent with that predicted by Wen and Lee's model ($\alpha \leq 1.0$). The results revealed that the basic nature of quasiparticles in the superconducting state can be attributed to the normal Fermi-liquid in all doping levels.

VII. The imaginary part of complex conductivity $\sigma_2(T)$ has a thermal activation gap $E_{g2}(T)$, which shows the thermal-averaged Josephson coupling energy of superfluid (Cooper pairs) in forming the long range order across the barriers macroscopically.

VIII. A closed form of the empirical formula of the real part $\sigma_1 = \sigma_1(5K)(1-e^{-E_{g1}(T)/k_BT})$ and the imaginary part $\sigma_2 = \sigma_2(5K)(1-e^{-E_{g2}(T)/k_BT})$ is obtained respectively. From that, the phase diagram of several energy scales versus hole concentration p was presented. The phase diagram is very similar to the one predicted by Emery and Kivelson's model based on the classical phase fluctuations on explaining the Cooper pair formation for high-T_c superconductors. We conjecture that the formation of superconducting state in the high-T_c superconducting thin films is owing to the phase fluctuations mechanism.

The experimental results have revealed that a number of the basic natures of quasiparticles in the superconducting state can be attributed to the normal Fermi liquid in the all levels of doping. When $T > T_c$, the classical phase fluctuations play a key role in forming the Cooper pairs of short range order. When $T \cong T_c$, the superconductivity with a long range order is formed by the Josephson tunneling of superfluid (Cooper pairs). In the superconducting state ($T < T_c$), the $\sigma_2(T)$ was attributed to the effect of Josephson tunneling in the weak links in our disordered samples. However, for all of the experiments in the microwave measurements till now there are two cases for $\sigma_1(T)$. One of them is that $\sigma_1(T)$ can be completely attributed to the effect of quasiparticle tunneling in the weak links. It means that the quasiparticles are localized, and $\sigma_1(T)$ is in thermal insulator phase in the superconducting state. Another is that $\sigma_1(T)$ can be partly attributed to the effect of quasiparticle tunneling through the weak links, i.e. quasiparticles are localized, and partly attributed to the effect of delocalized quasiparticles. In this case, $\sigma_1(T)$ indicates that the system contains both the thermal insulator and the thermal metal phases. All of the above results have led us to the conjecture that in the HTSC, there are two different energy scales which correspond to two temperatures: T_c below which coherence is established, and some higher temperature $T^*_{\sigma 2}$ where the pairs are formed. These two scales are close to each other, in the all levels of doping. The conjecture is also to endow the two associated superconducting phases - the thermal metal and the thermal insulator – and the critical point between them.