Chapter 1

Introduction

Very currently, high-T_c superconductivity has been a subject of challenging issue [1-5] and the pairing mechanism responsible for superconductivity in the high-T_c superconductors (HTSC) is still unknown yet. Experiments almost to address these fundamental questions have been focused on varying the doping levels of high-T_c superconducting materials [6-9]. It is conventional to differentiate the states of doping levels into 'overdoped', 'optimally doped' and 'underdoped' ones. The optimally doping is generally defined as the carrier concentration (hole) which maximizes the resistively defined superconducting transition temperature T_c. The overdoped materials have more added carriers and the underdoped materials have fewer in comparison with optimally doping. So it is naturally for us to ask how the electronic properties evolves with various hole dopings (Fig. 1.1(a)) [10-13]. And this is believed to be a key issue to elucidate the mechanism of superconductivity in the high-T_c cuprates. So far, actually, the experimentalists have reported some exciting results on the nature of low-lying excitations (quasiparticles) in the high-T_c cuprates, which provide evidences of central importance in helping us understand the physics of the high-T_c cuprates. In particular, angle-resolved photoemission spectroscopy (ARPES) [14, 15] experiments, which probe the spectral density A(k,w) near the Fermi surface, provide a direct test for the existence of well defined quasiparticle state at and near the Fermi level. Results obtained for the $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) samples near optimal doping [16] indicated that the existence of a small but finite discontinuity at the Fermi level. This, in turn, may provide a direct evidence that the basic nature of the quasiparticles near optimal doping is not remote from the Landau Fermi liquid. It immediately raises the following questions: what is the basic nature of the quasiparticles with various dopings? And whether the quasiparticles can be described by the Fermi liquids in the normal and/or the superconducting state irrespective of dopings of all levels. To this end the more experimental data in this respect would be very valuable.

From the theoretical and experimental point of view, the reasons of no consensus for the superconducting mechanism of the cuprates are primarily due to complications incurred by a number of competing orders (or the presence of competing phenomena) in these strongly correlated doped (Mott) insulators [17-19] arising from the basic nature of the above quasiparticles possibly. The competing orders are expected to result in a variety of phases in the ground state, which depends on the carrier doping levels, and on the degree of disorder. In order to sort through the complications of the HTSC, it is necessary to identify some of universal characteristics among these superconductors in the experiments.

In all of these superconductors such as $YBa_2Cu_3O_{7-\delta}$ (YBCO), $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) and $TlBa_2Ca_2Cu_3O_{9\pm\delta}$ (TBCCO), copper oxide planes form a common structural element, which is thought to dominate the superconducting properties. Depending on the choice of stoichiometry, the crystallographic unit cell contains varying numbers of CuO_2 planes. In addition, YBCO compounds contain CuO "chains" (Fig. 1.1(b)) [20, 21], which are thought to serve largely as reservoirs to control the carrier density in the planes. The exact T_c depends on particular details in each system but, roughly speaking, the highest T_c achieved in the YBCO, BSCCO, and TBCCO systems are 93, 110, and 130 K [22, 23], respectively. From the practical application viewpoint, as well known, YBCO superconducting thin films possess not only high T_c value but also high critical current density (Jc $>10^6$ A/cm^2 at 77K) [24, 25]. The YBCO superconducting thin film is therefore considered to be one of the most viable candidates for device applications in the newly discovered high-Tc superconductors [26-29]. Recently, Gou et al. have developed an efficient method to fabricate high-quality YBCO superconducting thin films [30-34]. Such method provides the opportunity to accurately probe the physical properties in these materials. In order to control carrier dopings in the YBCO compound, on the one hand, holes can be doped by varying the oxygen content δ. On the other hand, calcium, a divalent alkaline-earth ion, substituting preferentially for trivalent Yttrium in the YBCO compound can also result in excessive carriers. Since the ionic radius of the Ca^{2+} ion (~1.12Å) is approximately equal to that of the Y^{3+} ion (~1.019Å) one expects that the carrier or hole concentration could be varied in a controlling manner without introducing any significant distortion in the YBCO lattice [35, 36]. Also, given that the Y-site is located midway between the superconducting CuO_2 planes, Ca substitution should be an effective means of increasing the carrier concentration on the CuO_2 planes. As a result, for oxygenated YBCO, it is established that Ca doping leads to an increase in the hole concentration only in the CuO_2 planes (Fig. 1.2) [37], in contrast to the global change of the hole concentration in all the CuO_2 plane, CuO chain, and apical oxygen sites induced by varying the oxygen content. Nevertheless, substituting Ca^{2+} for Y^{3+} in the YBCO cuprate has allowed us to access the overdoped regime of the YBCO cuprate, and it can help us to perform a complete knowledge of doping mechanism and the effect of doped carriers on T_c.

With the concern of the full knowledge of the fundamental nature of the quasiparticles in the HTSC, one must delineate its energy gap [38-40] and thus its density of states in the pairing states even in the existence of disorder. In fact, it was already confirmed by tricrystal phase-sensitive experiment [41, 42], angle-resolved photoemission spectroscopy (ARPES) [43-47] that all the HTSC's are having the d-wave order parameters. In addition, the measurements of temperature dependence of the penetration depth λ(T) also indirectly yield a consistent picture of the d-wave pairing state of the high-T_c superconducting materials. Among the many methods, the measurements of the ac susceptibility [48, 49], muon spin relaxation (μSR) [50-52], and microwave techniques are commonly employed to extract the information of the penetration depth complementary [53-73]. The μSR is unique in determining the absolute value of λ(0), but is cumbersome in extracting λ(T). Although microwave technique, on the other hand, is unable to yielding absolute value of λ(0), it is one of the most sensitive methods in measuring the change of the penetration depth, Δλ, since a small shift of resonance frequency, Δf(T), due to the variation in λ(T), could be measured with high resolution.

Whatsoever high-precision microwave measurement of the temperature dependence of the surface impedance $Z_s(T) = R_s(T) + jX_s(T)$ of HTSC is so far the most powerful technique to shed light on the basic nature of superconductivity under the influences of external electromagnetic fields. Its real part gives the surface loss and the imaginary part reflects the surface reactance. Then, the real-part and the imaginary-part of the microwave complex conductivity can be extracted from the microwave measurements of the surface impedance $Z_s(T)$. The real-part conductivity $\sigma_1(T)$ contains the important information about the characteristic behaviors of the low-energy excitations (quasiparticles) [74, 75]. As to the imaginary part conductivity $\sigma_2(T)$, the physical properties of superfluid density in the superconductors can be revealed. In other words, the microwave conductivity measurement not only exactly reveals rich information for understanding the physical properties of the quasiparticles and the superconducting carriers of the HTSC, but also provides a quantitative test on different proposed underlying microscopic theories related to the basic nature of the quasiparticles in the superconducting state.

In the microwave experiment, up to the present, the real-part conductivity $\sigma_1(T)$ of detwinned, high-purity, slightly overdoped $YBa_2Cu_3O_{6.993}$ single crystal was measured by Hosseini et al. [76] with five frequencies between 1 and 75 GHz for temperatures from 1 to 95 K. High-purity of single crystal sample, by its own meanings, is in the clean limit. Below 20 K, they found that the conductivity has an approximately linear temperature dependence and a near-Drude frequency dependence. Fitting the data to the Drude form, they extracted a spectral weight that is linear in temperature (in agreement with the measured sperfluid density) and an effective scattering rate, $1/\tau$, that is approximately constant over the low-temperature regime (1 to 20 K). However, some microwave experiments in single crystals or thin films did not show the linear temperature dependence in σ_1 at $T<T_c$ ($2GHz<f<35GHz$) [74, 77-79], presenting contradictions to high-purity single crystal data. To our knowledge, all of these results are clearly evident that the underlying physical natures of quasiparticles, in light of σ_1, are so complicated due to the sensitively characteristic features of the sample qualities. In fact, the microwave conductivities σ_1 of $YBa_2Cu_3O_{7-\delta}$ (YBCO) and $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ (Ca-YBCO) thin films have been measured by us, and some samples do show the linear temperature dependence in σ_1 at $T<T_c$ but others do not. In order to solve this puzzle, some theoretical models will be introduced in the following. These models may help us to construct a clearer physical picture of the nature of quasiparticles and

the condensation mechanism of Cooper pairs in the HTSC.

As to the theoretical aspect related to the nature of quasiparticles in the disordered samples, Lee [80] in 1993 predicted that even the impurity concentration is small, the wave functions of quasiparticles in the normal state are essentially extended and the quasiparticles in the superconducting state would then become strongly localized because of a short coherence length *d*-wave superconductor. These low energy quasiparticles are produced by impurity scatters in the unitary limit in a two-dimensional d-wave superconductor with an effective mobility gap. And thus it leads to thermally activated behavior for the microwave conductivity and possibly for the London penetration depth. He also predicted that these localized quasiparticles lead the conductivity σ_1(T) to approach 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. He strongly suggested that samples with more disorder are more promising candidates to test the effects of quasiparticle localization in the microwave conductivity $\sigma_1(\omega,T)$, which would be much proper to study in the samples of thin films. Subsequently, Durst and Lee [81] further proposed a theoretical model based on the Lee's model that the universal value $e^2/2\pi\hbar \times \xi_0/a$ should be further 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. In fact, α is a Fermi liquid parameter inherited from the quasiparticle-quasiparticle interactions. Based on Bardeen-Cooper-Schrieffer (BCS) Fermi liquid theory describing the nature of quasiparticles, Wen and Lee [82, 83] further developed a model which argued that the linear temperature dependence of superfluid density can be well described by the thermally excited quasiparticles near the d-wave nodes. And the linear slope of the superfluid density with respect to temperature should be corrected with the factor α^2. They predicted that α is small or equal to 1 and must be independent of doping. These parameters, α and β, will be tested by the transport properties in the microwave measurement in the dissertation.

Moreover, Vishveshwara and Fisher [84] theoretically explored the quasiparticle transport properties which are greatly influenced by the disordered state in the samples. They found that the low-energy quasiparticles can either be delocalized and free to move through the sample as an extended state, or can be localized by the disorder. These two possibilities correspond to two distinct superconducting phases –the thermal metal with delocalized quasiparticle excitations and the thermal insulator with localized quasiparticles- in the nature of quasiparticle transport in the disordered samples. Perhaps to understand the nature of quasiparticles related to the existence and properties of these multiple phases in the superconducting states in the context of dirty *d*-wave superconductors should be of great interest [85-87], which may give an important role of inhomogeneous phases in the play of superconducting mechanism indirectly.

Because of their high transition temperatures, small coherence lengths, and low superfluid densities, the cuprate superconductors are easily influenced by thermal fluctuations of the superconducting order parameter, as predicted by Emery and Kivelson [88]. On the basis of classical phase fluctuation model, Emery and Kivelson have sketched a phase diagram, from which two energy scales, $T_\theta^{\max}$ and T_{MF}, were revealed, where $T_\theta^{\max}$ is the upper bound on the phase ordering temperature and T_{MF} is the mean field transition temperature. They suggested that $T_\theta^{\max}$ has played a more prominent role in determining the critical temperature in the underdoped regime due to the effects of classical phase fluctuations. Furthermore, such fluctuations are largely responsible for the effects of flux lattice melting [89] and vortex glass [90] transitions in the presence of a finite magnetic field as observed in numerous experiments currently. In addition, they must strongly affect the zero-field transition in such materials. Phase fluctuations due to thermal effects thus influence the transport properties of the high-T_c materials as well. For example, the finite-frequency conductivity shows a fluctuation-induced peak near T_c for YBCO single crystal [91].

Recent measurements in the most high-T_c cuprates, such as $Bi_2Sr_2CaCu_2O_{8+\delta}$, (BSCCO) have found that the real part of the complex conductivity, $\sigma_1(\omega)$, not only has a peak near T_c but also remains quite large even far below T_c at frequencies in the 30-200 GHz range [92, 93]. In some cases, $\sigma_1(\omega,T)$ at such low temperatures exceeds the peak values observed near T_c [93]. These large values occur over a broad concentration range varying from overdoped to optimally doped to underdoped. In fact, Barabash et al. [94] have interpreted the large low-temperature values of $\sigma_1(\omega,T)$ with the in-plane superfluid density $n_s(0)$ at T = 0 in terms of the model of classical phase fluctuations in the d-wave superconductors.

During the past years, a consensus has begun to emerge that the phenomenon of high temperature superconductivity in cuprates is associated with inhomogeneity [95, 96]. Perhaps the best way to understand the origin of the superconductivity in high temperature superconductors is to understand how it is related with the basic features of inhomogeneity, which are intrinsic or extrinsic. The inhomogeneity may point to be a microscopic phase separation, i.e., superconducting grains, embedded in a nonsuperconducting matrix [97]. Such inhomogeneity has been observed, and there is neutron spectroscopic evidence for nanoscale cluster formation and percolative superconductivity in various cuprates [98]. The issue of inhomogeneity (either in the static or dynamical sense) is very important in view of recent experimental and theoretical developments that find phase separation to be an integral part of high-Tc scenario.

In experiments, firstly, the weak but continuous decrease of the normal state conductivity seen in LaSrCuO at low temperatures under the application of high magnetic fields [99] has been observed, and it immediately raises the following question: how the transition to a superconducting state in an insulating sample is. Evidently, this could not be explained in the framework of BCS-like theories, which assume a metallic normal state to start with. In fact, this question is not new. Before the high-T_c cuprates were discovered, it was shown that granular superconducting materials can be insulating in their normal state, and still undergo a transition to a coherent superconducting state via the quantum tunneling effect [100]. This behavior has been confirmed by Gerber et al. [101]. In that case, the transition to the superconducting state by the condensation of Cooper pairs in the small metallic grains is somewhat independent of the inter-grain coupling, and the transition to a coherent superconducting state can be understood as resulting from the Josephson coupling between them. This result, obtained on a "conventional" superconductor, suggests that the superconducting transition seen in "insulating" cuprates is not be due to an exotic mechanism, but rather to some degree of granularity, related to sample inhomogeneity (intrinsic (phase separation), or extrinsic).

Secondly, we have known that the superconducting CuO_2 planes of cuprates are intrinsically granular in the nm scale [102]. A striking evidence was recently provided by high-resolution scanning tunneling spectroscopy on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) single crystals [95] and confirmed on thin films [103]. The emerging picture is that hole-rich and hole-poor domains, with size of the order of the in-plane superconducting coherence length, coexist in the superconducting planes. The former domains are expected to be metallic and superconducting, while the latter ones may be nonsuperconducting and antiferromagnetic or with strong residual antiferromagnetic fluctuations. Such electronic inhomogeneities are not to be confused with chemical inhomogeneities.

There are microstrip ring and line superconducting resonators made by $YBa_2Cu_3O_{7-\delta}$ (YBCO) and $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ (Ca-YBCO) thin films deposited on both sides of $LaAlO_3$ 10 x 10 x 0.5 mm^3 (100) oriented substrates in this work. Using these samples with various oxygen contents as easily changing the hole concentration, a number of intrinsic and/or extrinsic properties of the high-T_c superconducting cuprates in all doping levels for both YBCO and Ca-YBCO thin films have been explored in detail. Especially, the temperature dependence of the superconducting carrier density, the penetration depth and the complex conductivity can be estimated from the measurement of the surface impedance with the proper geometrical factor, respectively. All these findings not only allow us to test the current models related to the nature of the superconducting mechanism possibly, but also provide a number of viable data to support the model construction of the superconductivity mechanism in an exact manner. As regards, the theoretical models mentioned previously were used as a complementary tool to help us delineate the basic nature of quasiparticles and condensation mechanism of Cooper pairs in the high-T_c cuprates.

As revealed, we have found that the basic nature of the quasiparticles in the superconducting state can be described well in terms of Fermi-liquid by means of Lee and Wen's model. In analyzing the microwave complex conductivity of the superconducting YBCO and Ca-YBCO thin films, the thermal fluctuations play an important role on it, as predicted by Lee's model, Wen and Lee's model and Emery and Kivelson's model. By plotting the phase diagram of some characteristic energy scales extracted from the complex conductivity, we have found that the phase diagram was very similar to that predicted by Emery and Kivelson's model. And this may give evident that classical phase fluctuations would play an important role on physical properties of high-Tc superconductivity. Furthermore, by systematically analyzing the real part conductivity σ_1 of various sample qualities, we found that the quasiparticles are thoroughly localized due to disorder in some samples. But the other samples have both the localized quasiparticles and delocalized quasiparticles as these sample qualities (or degrees of disorder states) in the clean limit. According to the Vishveshwara and Fisher's model, the localized quasiparticles are in the thermal insulator phase. And the delocalized quasiparticles are in the thermal metal phase. So, as the sample qualities (or degrees of disorder states) are in the clean limit, there are two phases coexistence. On the other hand, the superconducting properties, such as superfluid density and T_c, are independent of the degrees of disorder states. At last, we have found that the real part conductivity σ_1 and imaginary part conductivity σ_2 are due to the macroscopic quantum tunneling of quasiparticles and superfluid density via thermal activation, respectively. So, the high-Tc superconductivity seen by us is due to the quantum tunneling effect of these granular materials, although the Cooper pairs have preformed at $T>T_c$.

This dissertation is organized as follows. The introduction is given in chapter 1. We herein will focus on the YBCO and Ca-YBCO thin film measurements only. In chapter 2, we give a description for the growth of the high-quality (001)-oriented YBCO and Ca-YBCO thin films by pulsed laser deposition. All films were examined by X-ray diffraction for crystal structures and orientations. The d.c. electric transport properties were measured with standard four-probe techniques, and the surface morphology was investigated by atomic force microscope (AFM). The a.c.

electromagnetic properties were measured with the microstrip ring and line resonators made of double-sided YBCO and Ca-YBCO thin films. For each sample, the hole concentration was varied by controlling the oxygen content of it. In chapter 3, the theoretical models on measuring the microwave properties in superconductors will be given. Then, in chapter 4 the direct experimental results, such as resistivity, resonance frequency and the indirect results, such as penetration depth, quality factor, surface impedance and complex conductivity, which are derived from the above theoretical considerations, are discussed in detail. Finally, in chapter 5, some remark and conclusion related to the basic nature of the high-T_c superconductivity in the superconducting state in the electromagnetic field will be proposed.

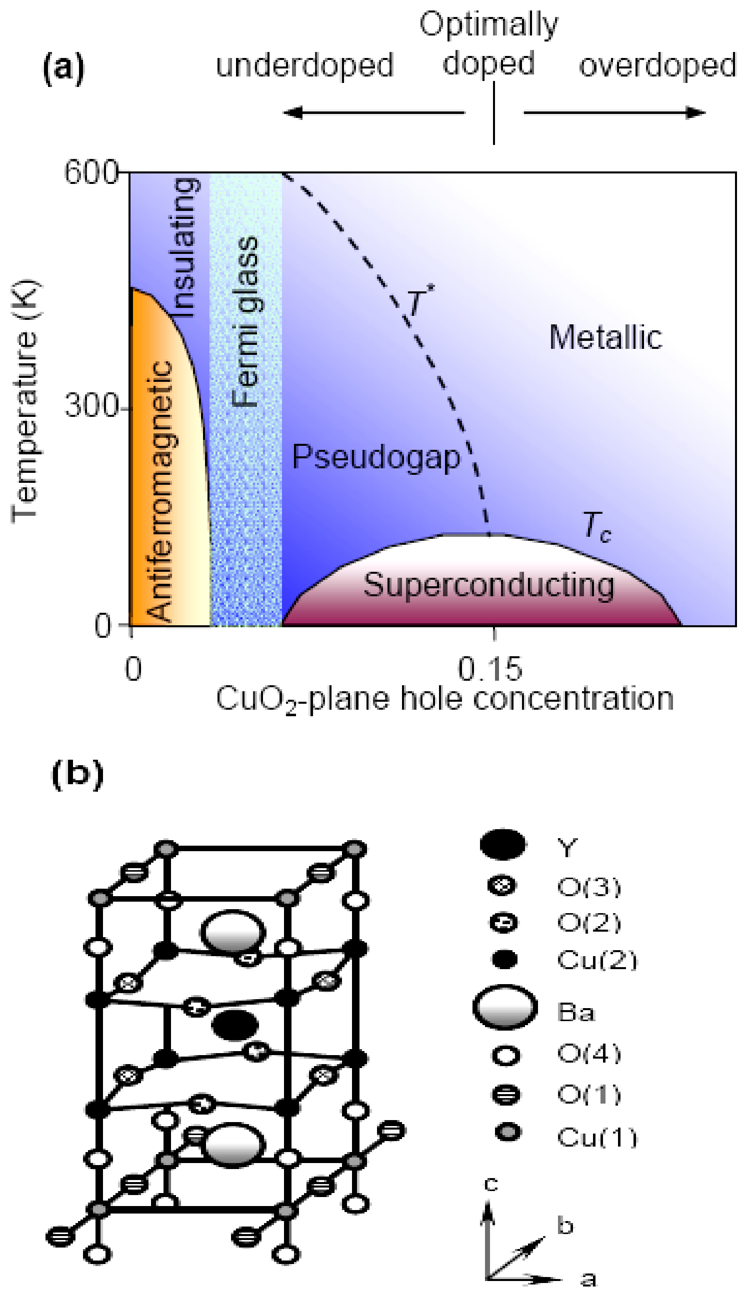

Fig. 1.1. (a) Schematic phase diagram of HTSC which shows the evolution of various phases as a function of the in-plane hole concentration. (b) Schematic atomic structure of $YBa_2Cu_3O_{7-\delta}$. The dimensions of the unit cell are 3.82 Å (a-xais) × 3.89 Å (b-axis) × 11.68 Å(c-axis) [12].

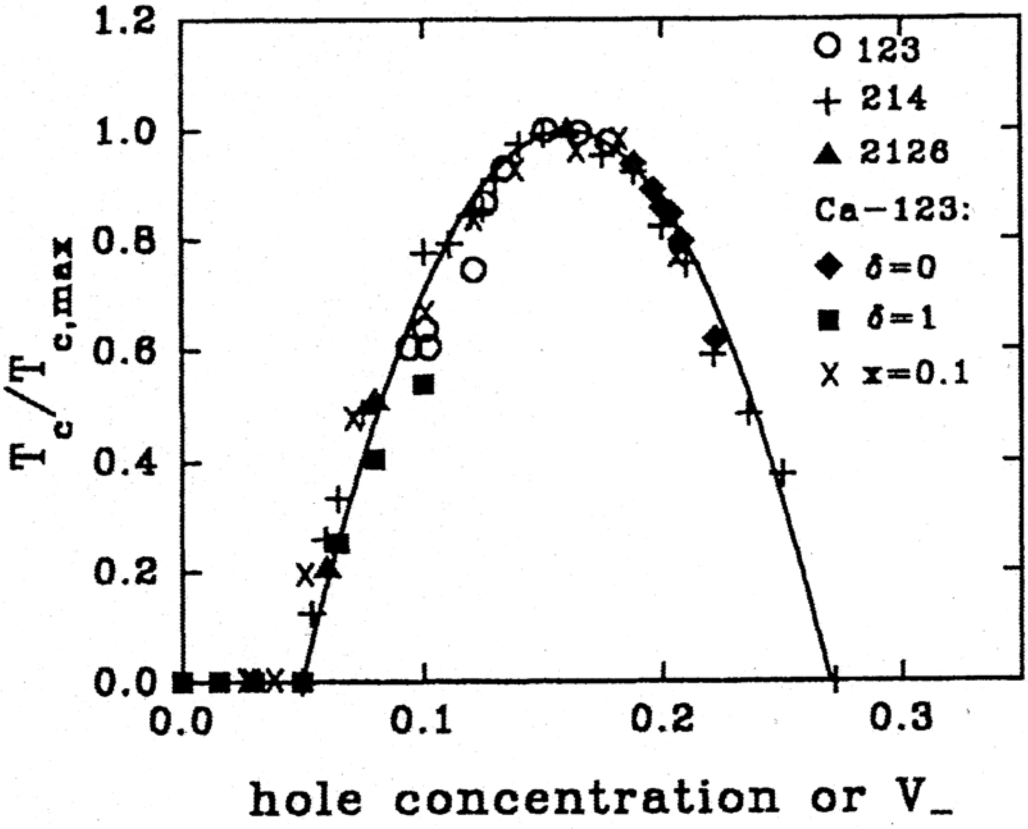

Fig. 1.2. T_c, normalized to $T_{c,max}$, plotted as a function of hole concentration, p determined (i) from $p=x/2$ for $Y_{1-x}Ca_xBa_2Cu_3O_6$ (solid squares), (ii) from p=V_ for $YBa_2Cu_3O_{7-\delta}$ with different δ (open circles), (iii) from p=V_ for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ with $\delta \approx 0.04$ and different x (solid diamonds), and (iv) from p=V_ for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ with x=0.1 and different δ (crosses, ×). The solid curve is determined by $T_c/T_{c,max}=1-82.6(p-0.16)^2$, the "plus" symbols (+) are T_c vs x data for $La_{2-x}Sr_xCuO_4$ and solid triangles for $La_{2-x}Sr_xCaCu_2O_6$.