Universal Correlations between T_c and n_s/m^* (Carrier Density over Effective Mass) in High-T_c Cuprate Superconductors

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The muon-spin-relaxation rate σ has been measured in sixteen specimens of high-T_c cuprate superconductors (the 2:1:4, 1:2:3, 2:2:1:2, and 2:2:2:3 series). This has allowed us to study the magnetic field penetration depth λ and thus the superconducting carrier density n_s divided by the effective mass m^* ($\sigma \propto 1/\lambda^2 \propto n_s/m^*$). A universal linear relation between T_c and $\sigma(T \to 0) \propto n_s/m^*$ has been found with increasing carrier doping. In heavily doped samples, however, T_c shows saturation and suppression with increasing n_s/m^*. This saturation starts at different values of n_s/m^* for materials with different multiplicities of CuO planes.

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Following the discovery of superconductivity in $(La_{2-x}Ba_x)CuO_4$ (Ref. 1), several different series of cuprate systems have been found to show high-T_c superconductivity.[2-6] The transition temperature T_c, within a given series, for example, $(La_{2-x}Sr_x)CuO_4$, depends strongly on the substitution concentration x or the oxygen concentration[7] which governs the carrier (hole) density of the system. Each different series of cuprate has a different maximum T_c, e.g., $T_c \approx 38$ K for $(La_{1.85}Sr_{0.15})CuO_4$, $T_c \approx 90$ K for $YBa_2Cu_3O_7$, and $T_c \approx 125$ K for $Tl_2Ba_2Ca_2Cu_3O_{10}$.

We have performed muon-spin-relaxation (μSR) measurements[8] on sixteen different specimens of various high-T_c superconductors. Using μSR, one can determine the magnetic field penetration depth λ which is related to the superconducting carrier concentration n_s divided by the effective mass m^*. In this Letter, we report universal correlations between T_c and n_s/m^* based on our accumulated μSR results. For the initial increase of carrier doping, T_c increases linearly with increasing n_s/m^*; samples from different series fall on the same line. In heavily doped regions, where n_s/m^* becomes large, T_c deviates from this linear relationship, showing saturation and suppression with increasing n_s/m^*. This deviation from linearity occurs at different values of n_s/m^* for the various series of high-T_c superconductors.

Muon-spin-relaxation measurements have been applied extensively to high-T_c superconductors and related magnetic systems.[9-15] In order to measure the magnetic field penetration depth λ of type-II superconductors, an external magnetic field H_{ext} ($H_{c1} < H_{ext} < H_{c2}$) is applied perpendicular to the direction of initial muon spin polarization. The time histogram of muon-decay positrons, emitted from positive muons stopped in the specimen, exhibits a sinusoidal oscillation due to the muon spin precession around H_{ext} as

$$N(t) = N(0)\exp(-t/\tau_\mu)\{1 + AG(t)\cos(\omega t + \phi)\}, \qquad (1)$$

where $\tau_\mu = 2.2$ μsec is the muon lifetime and A is the initial asymmetry (typically $0.2 \le A \le 0.3$).

The relaxation function $G(t)$ represents the muon spin depolarization caused, in this case, by the distribution of local fields B in the vortex state of a type-II superconductor. The width $\Delta B \equiv \langle (B - \langle B \rangle)^2 \rangle^{1/2}$ of this distribution was originally studied by Pincus *et al.*[16] in conventional superconductors: ΔB is nearly independent of H_{ext} when the separation of adjacent vortices is smaller than λ (in high-T_c superconductors, this condition is satisfied for $H_{ext} \gtrsim 1$ kG); in this case $\Delta B \propto 1/\lambda^2$. To estimate ΔB, the μSR data are analyzed with the approximation $G(t) = \exp(-\frac{1}{2}\sigma^2 t^2)$. The relaxation rate σ can then be related to λ and n_s/m^* as

$$\sigma \propto \Delta B \propto 1/\lambda^2 \propto n_s/m^*. \qquad (2)$$

The μSR measurements were performed at TRIUMF using the M15 and M20 surface muon channels. Surface muons are stopped within 0.5 mm of material in condensed matter. Each specimen (typically a 2-cm-diam by 1 to 2-mm-thick disk) was mounted in a cryostat with its face perpendicular to the incident beam. The muons were injected with their spins perpendicular to the beam direction, and the transverse external field (typically 2.5 kG $\leq H_{ext} \leq$ 13 kG) was applied along the beam direction. Sample preparation details are described in Refs. 5, 17, 18, and elsewhere. Preliminary stages of the present study have been reported in Ref. 19 and at recent conferences.[9]

All measurements reported here were made on ceramic sintered pellet specimens with randomly oriented microcrystallites. The magnetic field penetration depth of CuO high-T_c superconductors is anisotropic, as demonstrated[9,17] in μSR studies of a c-axis aligned ceramic specimen of $YBa_2Cu_3O_7$ (Ref. 17 also reports the observation of an asymmetric line shape of the field distribution characteristic of an Abrikosov flux lattice). The results on nonaligned specimens therefore reflect an angular average over $\lambda_{\parallel}$ (for $\mathbf{H}_{ext} \parallel \mathbf{c}$ axis) and $\lambda_{\perp}$ (for $\mathbf{H}_{ext} \perp \mathbf{c}$ axis). However, the results from different specimens may be compared with the assumption that this angular averaging is essentially the same for different specimens. [For large anisotropy ($\lambda_{\parallel} \ll \lambda_{\perp}$), σ is sensitive only to $\lambda_{\parallel}$ (see Ref. 20).] To minimize systematic errors in the comparison, all the data reported here were taken using the same measuring configuration with specimens of approximately the same disk shape. In principle, the field broadening in μSR can be due to both (a) intrinsic effect of flux penetration and (b) spatial variation in the demagnetization field. In our measurements, we confirmed that the contribution of (b) is much smaller than that of (a), as described in Ref. 21. Therefore, Eq. (2) holds even when we take account of the effects of demagnetization and angular averaging.

The temperature dependence of the relaxation rate σ was measured in all the specimens; typical temperature dependences are shown in Fig. 1 for $(Tl_{0.5}Pb_{0.5})Sr_2CaCu_2O_7$ and $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$. The temperature dependence of σ and other details for $YBa_2Cu_3O_y$ and Bi(Pb)-Sr-Ca-Cu-O systems will be reported separately.[17] Here, we focus on the relation between $\sigma(T \to 0)$ and T_c. The low-temperature relaxation rate $\sigma(T \to 0)$, obtained by extrapolating the low-temperature values of $\sigma(T)$ to $T=0$, represents the ground-state value of n_s/m^*. By performing zero-field μSR measurements, we confirmed that there is no signature of magnetic order in most of the systems in the temperature range of our measurements $T \gtrsim 4$ K. In two specimens, $(La_{2-x}Sr_2)CuO_4$ with $x=0.08$ and 0.10, we have found a signature of static magnetic ordering for $T \lesssim 5$ K, the effect of which on $\sigma(T \to 0)$ has been corrected for (see Ref. 14 for a study of magnetic order at low temperatures). μSR provides a reliable method of determining T_c, as $\sigma(T)$ increases suddenly below T_c with decreasing temperature and because the μSR signal clearly reflects the superconducting volume fraction. Our results confirm that more than 90% of the volume of each specimen becomes superconducting with a sharp transition temperature T_c. In Fig. 2, the results thus obtained are shown with T_c on the vertical axis and $\sigma(T \to 0) \propto n_s/m^*$ on the horizontal axis.

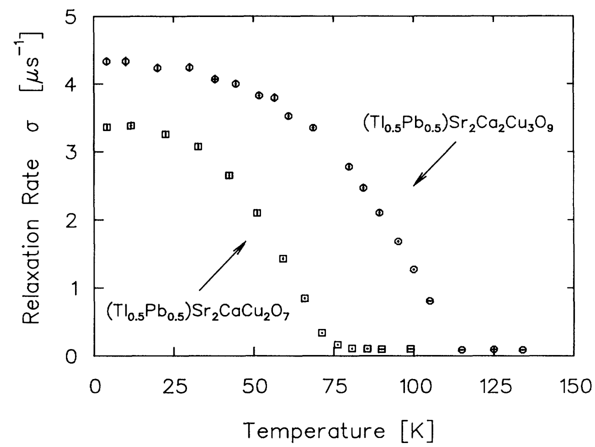

FIG. 1. The temperature dependence of the muon-spin-relaxation rate σ observed in $(Tl_{0.5}Pb_{0.5})Sr_2CaCu_2O_7$ (open squares) and in $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$ (open circles). The former material has the double CuO planes while the latter has the triple (see Ref. 5 for resistivity and crystal structures).

As shown in Fig. 2, a universal linear relation exists between T_c and $\sigma(T \to 0) \propto n_s/m^*$ which transcends differences of materials with single, double, and triple layers of CuO planes in the unit cell. Above certain values of n_s/m^*, deviations from linearity are seen with further doping and increasing $\sigma(T \to 0)$. This is most clearly shown in the case of the 2:1:4 compound, $(La_{2-x}Sr_x)CuO_4$. Up to $x \sim 0.10$, T_c increases with σ following the straight line. T_c then shows saturation and suppression with increasing x and increasing $\sigma \propto n_s/m^*$. The $x=0.1$ and 0.2 specimens have similar values of $T_c \sim 30$ K, while $\sigma(T \to 0)$ for the latter is about twice that for the former. These results demonstrate that n_s/m^* continues to increase with increasing x for the heavily doped region $x \gtrsim 0.15$ where T_c decreases with increasing x (Ref. 7). The suppression of T_c in this region is therefore *not* due to the disappearance of superconducting carriers by rather should be attributed to other causes. Around $x \sim 0.23$, we find that the superconducting volume fraction is sharply reduced with increasing doping x.

Similar relations between T_c and $\sigma(T \to 0) \propto n_s/m^*$ are also seen for the 1:2:3 compounds $YBa_2Cu_3O_y$ and $(Y_{0.7}Ca_{0.3})Ba_2Cu_3O_7$. Up to $y \sim 6.9$, both T_c and $\sigma(T \to 0)$ increase proportionally with increasing oxygen concentration y, at which point T_c starts to saturate. In the Ca-doped specimen, where additional carriers are provided by the substitution of Ca^{2+} for Y^{3+}, T_c de-

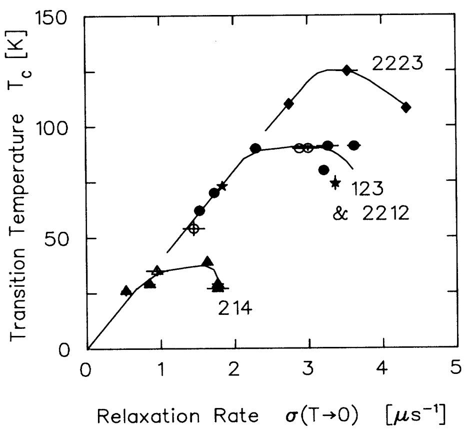

FIG. 2. The superconducting transition temperature T_c plotted vs low-temperature muon-spin-relaxation rate $\sigma(T\rightarrow 0)$ measured in sixteen different specimens of CuO high-T_c superconductors. The horizontal axis $\sigma(T\rightarrow 0)$ is proportional to $1/\lambda^2$ and consequently to n_s/m^*. The closed triangles represent points for the 2:1:4 system $La_{2-x}Sr_xCuO_4$, for $x=0.08$, 0.10, 0.15, 0.20, and 0.21 in the order of increasing σ. The first two points fall on the universal straight line. Closed circles denote the 1:2:3 systems $YBa_2Cu_3O_y$. In the order of increasing σ, points on the straight line are for $y=6.67$, 6.76, and 6.87. The two closed circle points at around $\sigma=3.1-3.5$ with $T_c=90$ K are obtained for two different 1:2:3 specimens with $y=7.0$. The closed circle at $\sigma=3.2$ and $T_c=80$ K represents $(Y_{0.7}Ca_{0.3})Ba_2Cu_3O_7$. The two stars at $T_c\sim 75$ K represent the 2:2:1:2 and similar systems, $Bi_2Sr_2CaCu_2O_8$ and $(Tl_{0.5}Pb_{0.5})Sr_2CaCu_2O_7$, in the order of increasing σ. The results for systems with triple CuO layers are shown by closed diamonds; $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_{10}$, $Tl_2Ba_2Ca_2Cu_3O_{10}$, and $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$, at $T_c=110-125$ K in the order of increasing σ. For the purpose of comparison, the four points reported in Ref. 19 are plotted by an open triangle ($La_{1.85}Sr_{0.15}CuO_4$) and open circles ($YBa_2Cu_3O_y$, with $y=6.66$, 6.95, and 7.0 in the order of increasing σ). Note that these four points are obtained with experimental conditions and specimens different from those in the present measurements. Error bars are within the size of each point unless specified. Solid lines are guides to the eye.

creases to 80 K, while the relaxation rate σ remains unchanged; i.e., the data point for the Ca-doped 1:2:3 compound in Fig. 2 stays apart from the initial straight line.

In the 2:2:1:2 and 2:2:2:3 compounds and systems with similar crystal structures, procedures for controlling the carrier concentration have not yet been established. To study the empirical trends, we have performed μSR measurements on two specimens of the double-layer systems $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2 compound) and $(Tl_{0.5}Pb_{0.5})Sr_2CaCu_2O_7$ (see Ref. 5) and on three specimens of the triple-layer systems $Tl_2Ba_2Ca_2Cu_3O_{10}$ (2:2:2:3 compound), $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_{10}$, and $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$. The results for these systems in Fig. 2 suggest that T_c also shows the saturation with increasing $\sigma(T\rightarrow 0)\propto n_s/m^*$ in the 2:2:1:2 and 2:2:2:3 systems. Thus, Fig. 2 clearly demonstrates that the saturation and suppression of T_c with increasing n_s/m^* occurs in all the different series of the CuO high-T_c superconductors.

Values of the carrier concentration in the normal state n_n can be derived from the Hall coefficient R_H, or in principle, directly from the chemical formulas, and n_n/m^* may be deduced from the plasma frequency. The Hall coefficient R_H of many high-T_c superconductors, however, shows substantial temperature dependence above T_c, making it difficult to estimate n_n. It is difficult to estimate the oxygen concentration accurately, especially for the three-layered 2:2:2:3 systems, making uncertain the values of the carrier concentration estimated from the stoichiometry. As seen in the 1:2:3 compounds, crystallographic ordering of the oxygen sites might also be a hidden variable which changes the effective carrier density. Evaluation of the plasma frequency from infrared absorption spectra is subject to uncertainty owing to the relatively continuous and small plasma edge.[22] (Trends seen in a plasma frequency measurement[22] are generally consistent with the present results.)

Compared to these other methods, μSR measurements have several advantages: (1) The concentration of superconducting carriers n_s can be directly studied. (2) μSR signals are volume proportional; the results are relatively insensitive to small impurity phases. (3) The extrapolated values of $\sigma(T\rightarrow 0)$ can be determined very accurately. Thus, the plot shown in Fig. 2 is a reliable way to study the relation between T_c and n_s/m^*. Both T_c and $\sigma(T\rightarrow 0)$ represent the experimentally measured quantities; the chemical composition, which is usually subject to a significant uncertainty, can be treated as an implicit variable.

For a typical value of $m^*=5m_e$ (m_e is the bare electron mass), for example, a relaxation rate $\sigma=1$ μsec^{-1} corresponds to a carrier density of $n_s=2\times 10^{21}$ cm^{-3} in isotropic type-II superconductors. Therefore, the results shown in Fig. 2 are consistent in order of magnitude with estimates of the carrier density based on the calculation of valency. We would like to note that n_s can be regarded either as the three-dimensional (3D) density per volume, or as the two-dimensional (2D) density on each CuO plane. The average interplane distance between adjacent CuO planes is about 6 ± 1 Å for each of the different series of high-T_c systems included in Fig. 2. The 2D and 3D densities are related with approximately the same conversion factor for different systems.

As we noted in Ref. 19, the linear relation between T_c and n_s/m^* cannot be expected in the weak-coupling limit of the BCS theory of superconductivity[23] where the Fermi energy ϵ_F, Debye frequency ω_D, and T_c are related as $T_c \ll \hbar\omega_D \ll \epsilon_F$ and $T_c \propto \hbar\omega_D$ (the Debye frequency represents the typical energy scale of the mediating boson). The Fermi energy of a noninteracting 2D electron gas is proportional to n_n/m^*. Therefore, one possible way to explain the observed linear relation is to view it as $T_c \propto \epsilon_F$, which is expected[24] when the energy scale of the mediating boson is comparable to or higher than ϵ_F. Of course, different types of theories may also explain the linear relation. In any event, the proportionality observed in Fig. 2 is universal to the single-layer 2:1:4, the double-layer 1:2:3 and 2:2:1:2, and the triple-layer 2:2:2:3 systems. It seems likely that this linear relation reflects intrinsic physical properties of the CuO planes. In contrast, the deviation from the linearity occurs at different values of $\sigma(T\rightarrow 0) \propto n_s/m^*$ for the various series of cuprates. The starting point for this deviation may be related to the different multiplicity of the CuO layers for different systems.

In conclusion, we have shown that universal correlations exist between T_c and n_s/m^* in all the planar cuprate high-T_c superconductors studied in this paper. These results provide a basic set of experimental data which must be explained by any successful theory of high-T_c superconductivity.

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[21]In type-II superconductors, the (fictitious) magnetic field H, magnetization M, demagnetization factor N $(0 \leq N \leq 1)$, and the averaged magnetic induction (i.e., observed internal field) $\langle B\rangle$ are related as $H = \langle B\rangle - 4\pi M = H_{ext} - 4\pi NM$. Since the μ^+ Knight shift in high-T_c systems is found to be negligibly small, the frequency shift of the μ^+ precession $F = \langle B\rangle - H_{ext} = 4\pi(1-N)M$ (in field units) directly reflects the demagnetization field. For the μ^+ in $(Y_{0.7}Ca_{0.3})Ba_2Cu_3O_7$, the observed shift was $F(T\rightarrow 0) = -6$ G, and the measured bulk magnetization was $4\pi M(T\rightarrow 0) = -50$ G. Therefore, we obtain an average value for N of 0.88 (in a reasonable agreement with the geometric value $N \sim 0.9$ calculated from the sample shape). The inhomogeneity ΔN of N is then estimated to be smaller than $1-0.88=0.12$; subsequent broadening $|4\pi\Delta NM|$ is therefore less than $|F| = 6$ G. This value is to be compared with the observed broadening $\Delta B = 40$ G. Since the intrinsic and demagnetization widths combine to give the total width as $(\Delta B)^2 = (\Delta B_{intrinsic})^2 + (\Delta B_{demag})^2$, the latter term can give, at most, only a 3% effect to the total width. We also obtained similar results of $|F/\Delta B|$ on the other specimens, thus confirming the predominant contribution of the intrinsic width due to the field penetration. In further support of this interpretation, we note that the absolute values of $\lambda_{\parallel}(T\rightarrow 0)$ in $YBa_2Cu_3O_7$ estimated by μSR using an unoriented sintered specimen (Ref. 17) (1400 ± 400 Å) and a **c**-axis-oriented sintered specimen (Ref. 9) (1500 Å) agree well with the results from a bulk magnetic measurement on a single-crystal specimen [1400 ± 500 Å; L. Krusin-Elbaum *et al.*, Phys. Rev. Lett. **62**, 217 (1989)].

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