

Systematic variation of magnetic-field penetration depth in high- T_c superconductors studied by muon-spin relaxation

Y. J. Uemura, V. J. Emery, A. R. Moodenbaugh, and M. Suenaga
Brookhaven National Laboratory, Upton, New York 11973

D. C. Johnston,* A. J. Jacobson, and J. T. Lewandowski
Corporate Research Laboratories, Exxon Research and Engineering Company, Annandale, New Jersey 08801

J. H. Brewer, R. F. Kiefl, S. R. Kreitzman, G. M. Luke, and T. Riseman
TRIUMF and Department of Physics, University of British Columbia, Vancouver,
British Columbia, Canada V6T2A3

C. E. Stronach
Virginia State University, Petersburg, Virginia 23803

W. J. Kossler, J. R. Kempton,† X. H. Yu, D. Opie, and H. E. Schone
College of William and Mary, Williamsburg, Virginia 23185

(Received 10 February 1988; revised manuscript received 18 April 1988)

The muon-spin relaxation rate σ has been measured in the high- T_c superconductors $\text{YBa}_2\text{Cu}_3\text{O}_x$ for $x=6.66, 6.95, 7.0$, and $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ in transverse external magnetic fields $\sim 1-4$ kG. We find a simple relation which connects the transition temperature T_c , the magnetic-field penetration depth λ_L , the carrier concentration n_s , and the effective mass m^* as $T_c \propto \sigma \propto 1/\lambda_L \propto n_s/m^*$. The linear dependence $T_c \propto n_s/m^*$ suggests a high-energy scale for the coupling between superconducting carriers.

The discovery^{1,2} of the layered oxide high- T_c superconductor systems $\text{La}_{2-y}\text{Sr}_y\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$ has triggered extensive experimental activity³ and renewed theoretical interest^{4,5} in the search for a novel mechanism for superconductivity. Muon-spin relaxation (μSR) is a direct method⁶ for measuring magnetic-field penetration depths in superconductors.⁷ μSR has been applied to the high- T_c oxide superconductors⁸⁻¹¹ and related antiferromagnets.^{12,13} Because of recent technological developments in sample preparation, it has now become possible¹⁴ to study single-phase specimens with the oxygen concentration controlled to within ± 0.02 /formula unit (f.u.). In this paper, we present μSR measurements on $\text{YBa}_2\text{Cu}_3\text{O}_x$ superconductors with averaged oxygen concentrations $x=7.0, 6.95$, and 6.66 f.u. We combine these results with the earlier work⁹ on $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$, and focus on the systematic dependence of the observed muon relaxation rate σ and the derived penetration depth λ_L . The results indicate that the superconducting transition temperature T_c is approximately proportional to the superconducting carrier concentration n_s , divided by the effective mass m^* . We discuss the implication of this relation on energy scales of the coupling between the carriers.

The sintered-pellet specimen of $\text{YBa}_2\text{Cu}_3\text{O}_7$ was prepared using a method described in Ref. 15. The powder specimens of $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6.66}$ were prepared by using another method described in Ref. 14 which reports Meissner effect and susceptibility measurements on a series of $\text{YBa}_2\text{Cu}_3\text{O}_x$ specimens ranging from $x=6.0$ to 7.0 . The specimens with $x \geq 6.5$ show superconductivity. The μSR experiments on $\text{YBa}_2\text{Cu}_3\text{O}_7$

were performed at the alternating-gradient-synchrotron muon channel of Brookhaven National Laboratory with a transverse external magnetic field H_{ext} of 1 kG applied perpendicular to the initial direction of muon-spin polarization. The measurements on $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6.66}$ were carried out at the M15 muon channel of TRIUMF (Vancouver) with $H_{\text{ext}}=4$ kG. In both cases, the data were taken by cooling the specimen in external field from $T \geq T_c$ to lower temperatures.

In the transverse-field $\mu^+\text{SR}$ experiments, one observes the decay-time histogram of positive muons stopped in the specimen

$$N(t) \propto \exp(-t/\tau_\mu)[1 + AG_x(t)\cos(\omega_\mu t)], \quad (1)$$

where τ_μ is the muon lifetime $2.2 \mu\text{sec}$, A is the initial precession asymmetry, ω_μ is the muon precession frequency, and the relaxation function $G_x(t)$ represents the time evolution of the muon-spin polarization. At all measuring temperatures ($5.0 \leq T \leq 300$ K), the observed precession amplitude A indicates that, within experimental error, all the muons stopped in the specimen contribute to the precession signal. The frequency ω_μ was approximately equal to $\gamma_\mu H_{\text{ext}}$ ($\gamma_\mu = 2\pi \times 1.355 \times 10^4 \text{ Oe}^{-1}$ is the gyromagnetic ratio of μ^+) above T_c . ω_μ decreased slightly with decreasing temperature below T_c , due to the partial exclusion of the external field H_{ext} in the type-II superconductors at $H_{\text{ext}} \geq H_{c1}$. For simplicity, here we assume a Gaussian shape for $G_x(t)$:

$$G_x(t) = \exp\left[-\frac{\sigma^2 t^2}{2}\right], \quad (2)$$

where σ is the muon-spin relaxation rate.

Figure 1 shows the temperature dependence of σ obtained for the present $\text{YBa}_2\text{Cu}_3\text{O}_x$ compounds together with the earlier results⁹ on $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. The very small values of σ observed in all the specimens above T_c can be accounted for by nuclear dipolar broadening. Combining this feature with the full amplitude for A and the reduction of ω_μ below T_c , one can conclude that there is no static magnetic ordering in these superconducting specimens either above or below T_c . This aspect was confirmed in the zero-field μSR measurements on $\text{YBa}_2\text{Cu}_3\text{O}_7$. Below T_c , the value of σ increases rapidly with decreasing temperature. This is due to the inhomogeneity of the static local field at the muon site in the type-II superconducting state where H_{ext} penetrates as a lattice of flux vortices. We notice here that the four different specimens in Fig. 1 have reasonably similar shapes for the curvature of the temperature dependences $\sigma(T)$. This implies that T_c is approximately proportional to $\sigma(T \rightarrow 0)$, as demonstrated in Fig. 2 for the four different specimens. [In details, T_c gets somewhat lower than the linear relation for large $\sigma(T \rightarrow 0)$, which may hint a possible saturation behavior.]

Pincus *et al.*¹⁶ used the London equation to calculate the distribution of magnetic fields in the vortex state, and obtained the second moment

$$\sqrt{\langle \Delta H^2 \rangle} \cong \frac{\phi_0}{\lambda_L^2 \sqrt{16\pi^3}}, \quad (3)$$

with the flux quanta ϕ_0 , for the square lattice of the vortex when the second moment becomes independent of the external field H_{ext} , i.e., when λ_L is comparable to or greater than the distance between adjacent vortices. The present condition, with $H_{\text{ext}} \sim 1\text{--}4$ kG, satisfies this criterion. For a triangular lattice, one needs to multiply¹⁰

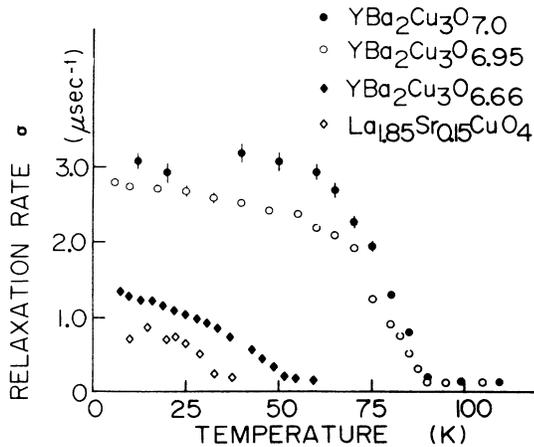


FIG. 1. Temperature dependence of the relaxation rate σ of the muon spin polarization, as defined in Eq. (2), observed in four different specimens of high- T_c superconductors. Data on $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$ were obtained in a transverse external magnetic field of 1 kG, while the measurements on the other two specimens were performed with a field of 4 kG.

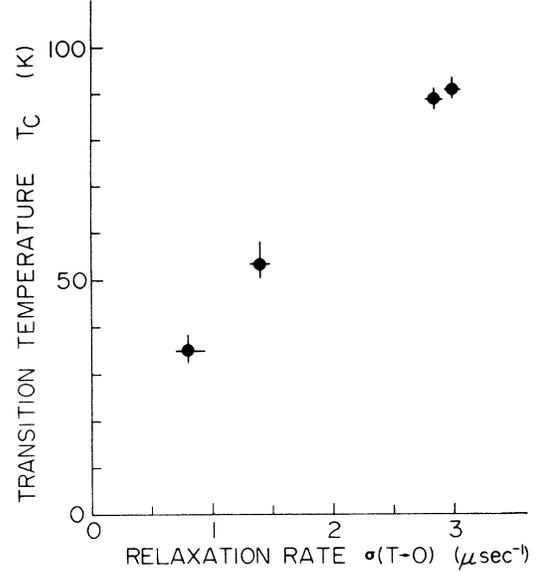


FIG. 2. Superconducting transition temperature T_c , as determined by the μSR measurement, plotted vs the values of the muon relaxation rate σ at $T \rightarrow 0$ for the four different specimens of high- T_c superconductors.

the right-hand side of Eq. (3) by 0.93. Then, one can deduce the value of the penetration depth λ_L directly from the observed relaxation rate σ , which corresponds to $\gamma_\mu \sqrt{\langle \Delta H^2 \rangle}$. Figure 3 shows the temperature dependence of λ_L thus obtained for the triangular vortex lattice. In the field-cooled measurements, the density of the magnetic flux is kept almost constant above and below T_c . If one changes the external field in the superconducting state, in contrast, the flux vortices have to move within the sample to change the spatial flux density, and, thus, the experimental results become sensitive to the flux pinning.¹⁷ Therefore, it is important to measure the penetration depth in the field-cooled condition, as in the present experiment.

In actual systems, we noticed that the functional form of $G_x(t)$ is somewhat in between Gaussian and exponential. This is due to the complicated distribution of magnetic fields for the vortex lattice as well as to the effect of anisotropy on the penetration depth λ_L . Correcting for the former effect would reduce the resulting values of λ_L by about 30%. For the case of maximum anisotropy where the penetration depth λ in the soft direction (H_{ext} applied parallel to the CuO plane) is infinite, Celio *et al.*¹⁸ find that the value of λ for $H_{\text{ext}} \perp (\text{CuO plane})$ to be about half the powder-average value. These corrections make it difficult to deduce the absolute values of λ_L accurately. However, we stress here that the relation $\sigma \propto 1/\lambda_L^2$ holds for any of the above calculations. Therefore, the systematic and temperature variations of λ_L can be discussed based on Figs. 1–3.

The London penetration depth λ_L is given as a function of effective mass m^* and the carrier density n_s as

$$\lambda_L = \sqrt{m^* c^2 / 4\pi n_s e^2}. \quad (4)$$

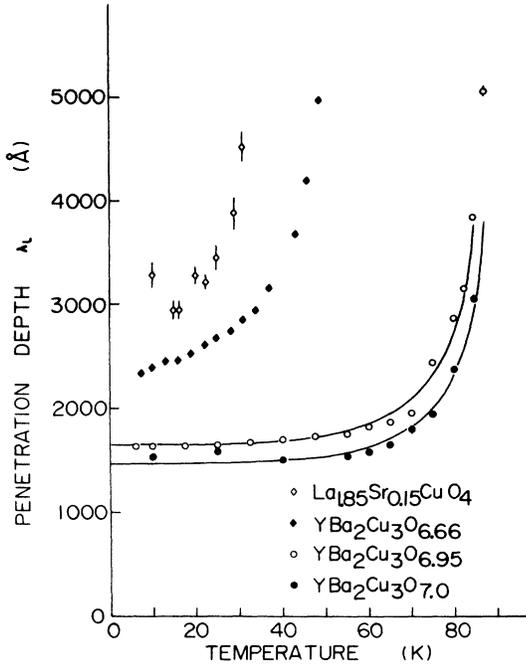


FIG. 3. Temperature dependence of the magnetic field penetration depth λ_L derived from the muon spin-relaxation rate shown in Fig. 1. λ_L was calculated using a simple approximation for the triangular vortex lattice as described in the text. The solid lines represent fits of the data to Eq. (7), with $\lambda_L(T=0) = 1656$ Å and $T_c = 89.9$ K for $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$, and $\lambda_L(T=0) = 1472$ Å and $T_c = 91.1$ K for $\text{YBa}_2\text{Cu}_3\text{O}_{7.0}$.

Combining this with the relation $\sigma = \gamma_\mu \sqrt{\Delta H^2}$, Eq. (3), and the approximate experimental result $\sigma(T \rightarrow 0) \propto T_c$, we obtain a simple relation

$$\sigma \propto \frac{1}{\lambda_L^2} \propto \frac{n_s}{m^*} \propto T_c. \quad (5)$$

Thus, the transition temperatures T_c of the four different samples are simply proportional to the carrier concentration n_s , divided by the effective mass m^* regardless of the crystallographic differences of the samples.

The relation $T_c \propto n$ (n : normal-state carrier density) has been found by Hall constant and related measurements for $\text{La}_{2-y}\text{Sr}_y\text{CuO}_4$ between $y = 0-0.15$ (Ref. 19) and $\text{YBa}_2\text{Cu}_3\text{O}_x$ between $x = 6.5-7.0$ (Ref. 20). This linear relation can also be obtained in a calculation of the number of holes for a formula unit, assuming charge neutrality for La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_{6.4}$ and adding one hole for the substitution of Sr to La and two holes for additional oxygen per formula unit. These results suggest that the carrier concentration n_s , rather than the effective mass m^* , plays the major role in changing T_c in Eq. (5). Indeed, the Sommerfeld constant γ of the low-temperature specific heat, which is proportional to m^* for two-dimensional systems, does not depend much on the differences in material,²¹ supporting the above viewpoint. The present work has shown that the linear relation holds for the superconducting carrier concentration n_s .

We now discuss the implications of the relation

$T_c \propto n_s/m^*$. In the Bardeen-Cooper-Schrieffer (BCS) theory²² with the phonon-mediated coupling of electrons, T_c is given in the so-called weak coupling limit as

$$k_B T_c \sim 2 \hbar \omega_D \exp \left[- \frac{2}{VD(\epsilon_f)} \right], \quad (6)$$

where k_B is the Boltzmann constant, ω_D is the Debye frequency, V represents the effective attractive interaction, and $D(\epsilon_f)$ is the density of states at the Fermi energy ϵ_f . To obtain this equation, one assumes $\epsilon_f \gg \hbar \omega_D$ and solves the gap equation by integrating the energy range of the coupling interaction $0 \rightarrow \hbar \omega_D$, which results in the preexponential factor $\hbar \omega_D$. It is difficult to reconcile the relation $T_c \propto n_s/m^*$ with Eq. (6), because $D(\epsilon_f)$ does not depend on n in the two-dimensional noninteracting electron gas. Recent single-crystal measurements²³ on Hc_2 and on the transport properties suggest a highly two-dimensional character for the electron system. Moreover, the magnitude of the electron-phonon interaction V , inferred from the temperature dependence of linear resistivity in the normal state,²⁴ is too small to explain the high transition temperature in the standard phonon-mediated mechanism.

In contrast, when the energy scale of the attractive interaction which couples the carriers is larger than that of ϵ_f , the energy integration in the gap equation runs over the range 0 to ϵ_f . This would put, roughly speaking, ϵ_f in the preexponential factor of Eq. (6) instead of $\hbar \omega_D$. In a two-dimensional noninteracting electron gas, the Fermi energy ϵ_f is proportional to the quantity n/m^* . Then one could expect the simple relation $T_c \propto n_s/m^*$. This argument also works without essential change for three-dimensional systems where $\epsilon_f \propto n^{2/3}/m^*$. Thus, the relation $T_c \propto n_s/m^*$ suggests a high-energy scale of the interaction which mediates the coupling between superconducting carriers in high- T_c superconductors. Such a high-energy scale may be found in models based on the large transfer integral of a carrier between the oxygen and neighboring copper atoms.⁵

We study the temperature dependence of λ_L with examples of $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7.0}$. The sharp changes of $\sigma(T)$ near T_c observed for these samples suggest a good homogeneity in the oxygen concentration.²⁵ As shown by the solid lines in Fig. 3, the experimental data agree well with the empirical formula²²

$$\lambda_L(T) = \frac{\lambda_L(T=0)}{\sqrt{1-(T/T_c)^4}}. \quad (7)$$

This result is consistent with earlier μSR works,^{8,10} but disagrees with a recent bulk measurement.²⁶ Equation (7) is calculated for λ_L much smaller than the coherence length ξ by assuming an isotropic energy gap at the Fermi surface. For $\lambda_L \gg \xi$, the BCS theory²² predicts that $\lambda_L(T)$ increases more rapidly than Eq. (7) with increasing temperature at $T \leq 0.7T_c$. The anomalous zeros of the energy gap at some point or line of the Fermi surface would change the theoretical curves for $\lambda_L(T)$ to increase faster with increasing temperature in the low-temperature region. Therefore, the present results suggest that the energy gap is predominantly finite.

In summary, based on the muon-spin relaxation experiments, we have shown that the approximate proportionality $T_c \propto n_s/m^*$ holds universally for different high- T_c oxide superconductor systems. This feature suggests the high-energy scale of the coupling between superconducting carriers. Further μ SR measurements on single-crystal specimens will be very helpful in eliminating uncertainties due to anisotropy of λ_L .

Note added. After this paper was submitted, additional measurements on a few more specimens of $\text{YBa}_2\text{Cu}_3\text{O}_{6.6-7.0}$ were performed at TRIUMF. The resulting values of $\sigma(T \rightarrow 0)$ and T_c smoothly interpolate the present points in Fig. 2. This confirms the smooth and monotonic variation of T_c as a function of n_s/m^* . Re-

cently, Uchida *et al.*²⁷ measured plasma edge frequencies ω_p of the infrared absorption (cf. $\omega_p \propto 1/\lambda_L$) in $(\text{LaSr})\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_{6.5-7.0}$, as well as in the newly found Bi-Sr-Ca-Cu-O. They found an approximate linear relation between T_c and ω_p^2 for various specimens, which is consistent with the present results.

This work is supported by the Division of Materials Sciences, U.S. Department of Energy under Contract No. 76-AC02-CH00016, the U.S. National Science Foundation under Grant No. DMR 8503223, NASA under Grant No. NAG-1-416, and by the Natural Sciences and Engineering Research Council of Canada.

*Present address: Department of Physics and Ames Laboratory, (U.S. Department of Energy), Iowa State University, Ames, IA 50011.

†Present address: TRIUMF, University of British Columbia, Vancouver, British Columbia, Canada V6T 2A3.

¹G. J. Bednorz and K. A. Müller, *Z. Phys. B* **64**, 189 (1986).

²S. Uchida *et al.*, *Jpn. J. Appl. Phys. Lett.* **26**, L1 (1987); C. W. Chu *et al.*, *Phys. Rev. Lett.* **58**, 405 (1987); R. J. Cava *et al.*, *ibid.* **58**, 408 (1987); M. K. Wu *et al.*, *ibid.* **58**, 908 (1987).

³See, for example, papers published in *Phys. Rev. Lett.* **58**, **59** (1987), *Phys. Rev. B* **35**, **36** (1987), and *Jpn. J. Appl. Phys. Lett.* **26** (1987).

⁴P. W. Anderson, *Science* **235**, 1196 (1987).

⁵V. J. Emery, *Phys. Rev. Lett.* **58**, 2794 (1987).

⁶For general aspects of muon spin rotation, see proceedings of the four previous international conferences, *Hyperfine Interact.* **6** (1979), **8** (1981), **17-19** (1984), **31** (1986).

⁷A. T. Fiory *et al.*, *Phys. Rev. Lett.* **33**, 969 (1974); F. N. Gygax *et al.*, *Hyperfine Interact.* **8**, 623 (1981).

⁸G. Aeppli *et al.*, *Phys. Rev. B* **35**, 7129 (1987).

⁹W. J. Kossler *et al.*, *Phys. Rev. B* **35**, 7133 (1987).

¹⁰F. N. Gygax *et al.*, *Europhys. Lett.* **4**, 473 (1987).

¹¹D. R. Harshman *et al.*, *Phys. Rev. B* **36**, 2386 (1987).

¹²Y. J. Uemura *et al.*, *Phys. Rev. Lett.* **59**, 1045 (1987).

¹³N. Nishida *et al.*, *Jpn. J. Appl. Phys.* **26**, L1856 (1987).

¹⁴D. C. Johnston *et al.*, in *Chemistry of High- T_c Superconductors*, ACS Symposium Series No. 351, edited by D. L. Nelson, M. S. Whittingham, and T. F. George (American Chem-

ical Society, Washington, DC, 1987), Chap. 14, p. 136; A. J. Jacobson *et al.* (unpublished).

¹⁵R. J. Cava *et al.*, *Phys. Rev. Lett.* **58**, 1676 (1987); D. E. Cox *et al.*, *J. Phys. Chem. Solids* **49**, 47 (1988).

¹⁶P. Pincus *et al.*, *Phys. Lett.* **13**, 21 (1964).

¹⁷Some examples of the flux pinning effect can be seen in Refs. 8 and 10. According to a private communication with A. Schenck, μ SR data taken in the zero-field cooling procedure are significantly different from specimen to specimen, even for the same nominal chemical formula.

¹⁸M. Celio, T. M. Riseman, J. H. Brewer, R. F. Kiefl, and W. J. Kossler (unpublished).

¹⁹M. W. Shafer *et al.*, *Phys. Rev. B* **36**, 4047 (1987).

²⁰H. Takagi *et al.* (unpublished).

²¹K. Kitazawa *et al.*, *Jpn. J. Appl. Phys. Lett.* **26**, L748 (1987); **26**, L751 (1987).

²²J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Phys. Rev.* **108**, 1175 (1957).

²³Y. Hidaka *et al.*, *Jpn. J. Appl. Phys. Lett.* **26**, L377 (1987); **26**, L726 (1987); T. R. Dinger *et al.*, *Phys. Rev. Lett.* **58**, 2687 (1987); T. K. Worthington *et al.*, *ibid.* **59**, 1160 (1987).

²⁴M. Gurevitch and A. T. Fiory, *Phys. Rev. Lett.* **59**, 1337 (1987).

²⁵Gradual changes of $\sigma(T)$ and $\lambda_L(T)$ as $T \rightarrow 0$ observed on $\text{YBa}_2\text{Cu}_3\text{O}_{6.66}$ may be due to a finite spread of oxygen concentration within the specimen.

²⁶J. R. Cooper *et al.*, *Phys. Rev. B* **37**, 638 (1988).

²⁷S. Uchida, *Bull. Am. Phys. Soc.* **33**, 507 (1988).