PHYSICAL REVIEW B VOLUME 35, NUMBER 13 1 MAY 1987

Magnetic penetration depth and flux-pinning effects in high-T_c superconductor $La_{1.85}Sr_{0.15}CuO_4$

G. Aeppli and R. J. Cava
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

E. J. Ansaldo
University of Saskatchewan, Saskatoon, Canada S7N 0W0

J. H. Brewer, S. R. Kreitzman, G. M. Luke, and D. R. Noakes
The University of British Columbia, Vancouver, Canada V6T 2A3

R. F. Kiefl
TRIUMF, Vancouver, Canada V6T 2A3
and the University of British Columbia, Vancouver, Canada V6T 2A3
(Received 11 March 1987)

We have performed muon-spin-relaxation (μSR) measurements on $La_{1.85}Sr_{0.15}CuO_4$ above and below its superconducting transition temperature, $T_c = 37$ K. From transverse-field μSR at 6 K, the magnetic penetration depth is $\lambda \approx 2500$ Å, which, together with previous thermodynamic data and the London formula, implies a carrier density of 0.3×10^{22} cm^{-3}. The temperature dependence of λ in our sintered powder sample differs from that for an ordinary homogeneous superconductor. We also show that the longitudinal- and transverse-field μSR techniques are sensitive probes of H_{c1} and flux-pinning effects.

Compounds of the form $M_{2-x}Z_xCuO_4$ [M = La (Refs. 1-4) or Y (Ref. 5) and Z = Ba,[1,5] Sr,[2-4] or Ca (Ref. 2)] are superconductors with unprecedentedly high transition temperatures. For example, for M = La, Z = Sr, and $x = 0.15$, the bulk T_c is 37 K, and, correspondingly, the upper critical field H_{c2} exceeds 220 kG. In the present paper, we describe experiments in which positive muons are used as microscopic probes of the internal fields in $La_{1.85}Sr_{0.15}CuO_4$ below T_c. Muon spin relaxation (μSR) is especially suited to measuring quantities such as the magnetic penetration depth (λ) because it does not require special-purpose samples, such as thin films or spheres of controlled dimensions.[6]

We performed our μSR measurements at the $M15$ surface muon channel of TRIUMF. The sample, a sintered polycrystalline pellet, 1 cm in diameter and 2 mm thick, was prepared by means described elsewhere[3] and mounted perpendicular to the incident beam. In time-differential μSR experiments, muons are stopped one at a time in the sample, where they decay, emitting positrons preferentially along their final polarization.[7] Data are collected as a function of time t after arrival of the individual muons in the sample by counting the numbers $N_+(t)$ and $N_-(t)$ of positrons emitted in the directions parallel and antiparallel to the incident muon spin. The resulting ratio, $[N_+(t) - N_-(t)]/[N_+(t) + N_-(t)]$, is proportional to the time-dependent polarization of the muon ensemble.[8] A magnetic field can be applied either parallel (longitudinal) or perpendicular (transverse) to the initial muon polarization, yielding, respectively, a T_1-type relaxation function $G_{zz}(t)$ or a T_2-like relaxation envelope $G_{xx}(t)$ modulating a precessing muon decay asymmetry, just as for the free induction decay in NMR.[7]

Figure 1 shows the transverse-field (TF) μSR precession signals for a field of 80 G at room temperature and at 10 K after zero-field cooling (ZFC). The enhanced relaxation rate at 10 K is obvious. The solid lines are the results of fits made to a Gaussian relaxation function, $G_{xx}(t) = \exp[-(\Lambda t)^2]$, which corresponds to a Gaussian distribution of internal fields.[7] The low field in these spectra is convenient for illustrative purposes, but for measurements of λ, we used an external field $H_{\text{ext}} = 4$ kG, which is well above H_{c1} ($\cong 150$ G at 10 K, from other measurements[3,4] in addition to our data described below) and well below H_{c2} for $T < 35$ K.[4] Figure 2 displays the muon depolarization rate Λ as a function of increasing temperature after cooling in an external field of 4 kG. Above $T_c = 37$ K, the fitted Λ, which is due to static nuclear dipole relaxation, is small [$0.102(2) \times 10^6$ s^{-1}]; below T_c, Λ increases by an order of magnitude indicating that the internal field becomes more inhomogeneous as the superconducting state is entered. NMR (Ref. 9) and μSR (Ref. 10) experiments on other type-II superconductors have revealed the same effect, due to the vortex lattices formed when the external field penetrates the materials. The mean square inhomogeneity in the field sampled by the muons in μSR or nuclear spins in NMR is[9]

$$\langle |\Delta H|^2 \rangle = \frac{H_{\text{ext}}^2}{4\pi} \left(\frac{d}{\lambda} \right)^2 \left[1 + \left(\frac{2\pi\lambda}{d} \right)^2 \right]^{-1} , \quad (1)$$

where λ is the London penetration depth and d is the spac-

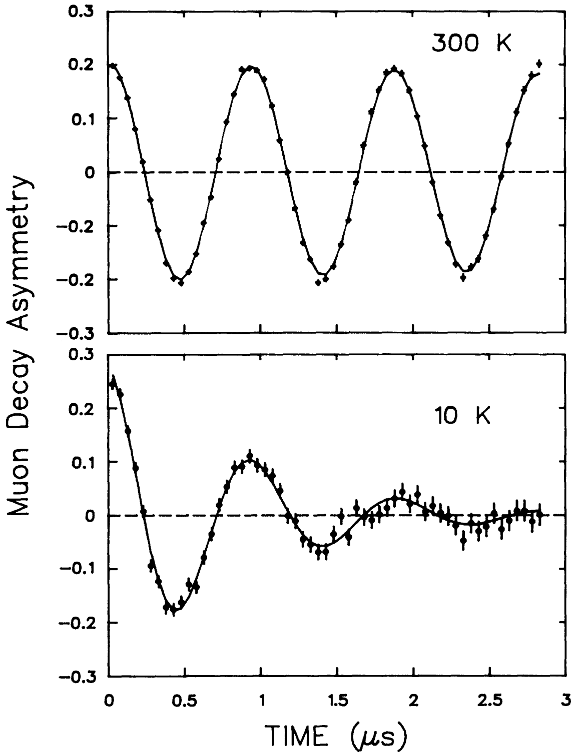

FIG. 1. Muon-spin-relaxation signal in a transverse field of 80 G for T above and below $T_c = 37$ K. The 10-K data were obtained after cooling in a 4-kG field.

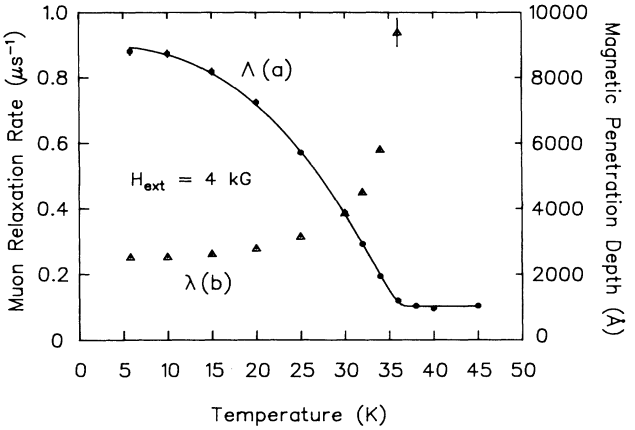

FIG. 2. Temperature dependence of (a) (circles) the Gaussian relaxation rate Λ obtained for a transverse field $H_{\text{ext}} = 4$ kG after cooling in zero applied field; and (b) (triangles) the magnetic penetration depth λ inferred from Λ using Eq. (2) after correcting for a fixed nuclear dipolar relation rate $\Lambda_0 = 0.102(2) \times 10^6\ \text{s}^{-1}$, assumed to add in quadrature with the depolarization rate due to the vortex lattice. The solid curve is a guide to the eye.

ing between vortices. For the square vortex array assumed in the derivation of Eq. (1), $d^2 = \phi/H_{\text{ext}}$, where ϕ is the magnetic flux quantum, 2.068×10^{-7} G cm^2, so that Eq. (1) can be rewritten as

$$\langle |\Delta H|^2 \rangle = \frac{H_{\text{ext}}\phi}{4\pi\lambda^2}\left[1 + \frac{4\pi^2\lambda^2 H_{\text{ext}}}{\phi}\right]^{-1} . \tag{2}$$

Because $\Lambda\sqrt{2} = \gamma_\mu \langle |\Delta H|^2 \rangle^{1/2}$ ($\gamma_\mu = 2\pi \times 13.55$ MHz/kG is the gyromagnetic ratio of the muon), Eq. (2) implies that λ can be extracted from μSR data. In particular, for high fields, $d \ll \lambda$ and Eq. (2) becomes $\Lambda(\mu\text{s}^{-1}) \approx [2364/\lambda(\text{Å})]^2$. For $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$, the corresponding value for λ at 6 K is 2500 Å, which is comparable to those of $A15$ compounds such as Nb_3Sn and V_3Si. It is also not far from what may be obtained using estimated values[11] of K ($= \lambda/\xi_0 \approx 100$) and ξ_0 ($\cong 30$ Å), where ξ_0 is the coherence length. In type-II superconductors, the London formula gives an approximate value for λ,

$$\lambda = \left[\frac{m^*}{4\pi n_s e^2 c^2}\right]^{1/2} = \left[\frac{m^*/m_e}{4\pi n_s r_e}\right]^{1/2} , \tag{3}$$

where $r_e = e^2/me^2 = 2.82 \times 10^{-5}$ Å is the classical radius of the electron, m^* is the effective carrier mass, and n_s is the superfluid density, which, for ordinary superconductors at $T = 0$, is identical to the carrier density (n) for $T > T_c$. The Sommerfeld constant can also be expressed in terms of m^* and n,

$$\gamma = k_B^2(\pi/3)^{2/3} m^* n^{1/3}/\hbar^2 . \tag{4}$$

Simultaneous solution of Eqs. (3) and (4), using the measured values of λ and γ (≈ 6 mJ/mole K^2 from Ref. 4) yields a small n, 0.3×10^{22} carriers/cm^3 ($\cong 0.3$ carriers/formula unit), and a sizable m^*/m_e of 7. For the band structure of Mattheiss,[12] n ($\cong 10^{22}$ carriers/cm^3) is larger and m^*/m_e (~ 3.5) somewhat smaller. Comparison of our result for m^*/m_e and that of Mattheiss indicates that the electron-phonon coupling strength $\lambda_{e\text{-ph}}$ in $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ is not unusually large; indeed, $\lambda_{e\text{-ph}}$ is of order unity, in agreement with a recent calculation.[13] We emphasize that our values for n and m^*/m_e are merely estimates based on Eqs. (3) and (4); it remains to be seen whether the uncorrected London formula is valid for samples such as ours.

Figure 2(b) shows the temperature dependence of λ computed from Λ and Eq. (2). Attempts to fit the present data to the form,

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

which generally describes the behavior of ordinary superconductors, failed. The failure can be due to inhomogeneities, which lead to a distribution of T_c's or to the pressed powder's granularity, which causes percolation, finite size, and weak link effects.[1,4] We note also that when the electronic mean free path l becomes comparable to ξ_0, which might well be the case for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$,[14] the standard expressions, Eqs. (3) and (5), for $\lambda(0)$ and $\lambda(T)$ become invalid.[6]

Figure 3 displays the field dependence of Λ for several temperatures and sample histories. Upon decreasing H_{ext}

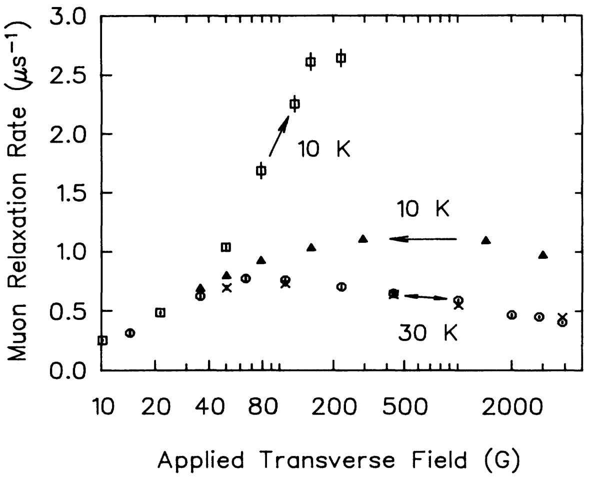

FIG. 3. Dependence of muon spin depolarization rate on transverse-field strength for various temperatures and sample histories. (Squares, ZFC at 10 K; triangles, FC at 10 K; circles, ZFC at 30 K; crosses, FC at 30 K.)

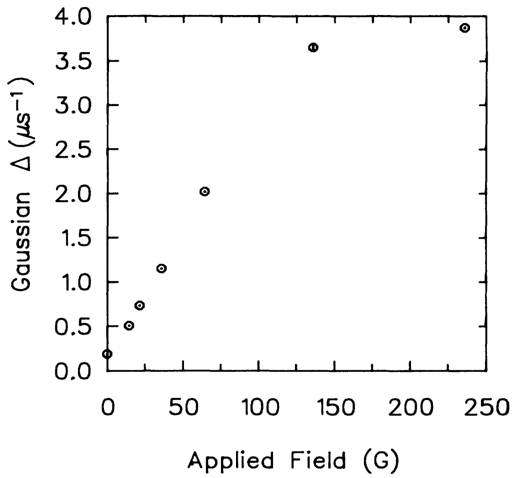

FIG. 4. Dependence of the fitted value of the Gaussian relaxation rate (a measure of the mean-squared internal field transverse to the applied field) found upon increasing the external field after zero-field cooling to $T=10$ K.

at $T=10$ K, Λ is essentially field independent until $H_{\mathrm{ext}} \approx H_{c1}=150$ G, as it should be according to Eq. (2). Subsequent reductions in H_{ext} lead to a monotonic decrease in Λ. Due to flux-pinning and finite-size effects, some flux still penetrates for $H_{\mathrm{ext}} < H_{c1}$. The hysteresis in Λ (see Fig. 3) is clear evidence for flux pinning at 10 K. The near reversibility of the 30-K results indicates that there is considerably less flux pinning at higher T.

The zero-field (ZF) and longitudinal-field (LF) μSR techniques measure $G_{zz}(t)$, the relaxation of the muon polarization component parallel to the initial muon-spin direction ($\hat{\mathbf{z}}$); muons are depolarized only by fields with components perpendicular to $\hat{\mathbf{z}}$. After cooling the sample to 10 K in zero field (ZFC), we observe a slowly decaying ZF relaxation function indistinguishable from that found at 60 K and well described by a Gaussian Kubo-Toyabe form.[15] Quite different behavior obtains after cooling the sample to 10 K (from 50 K) in a field of 4 kG applied along $\hat{\mathbf{z}}$ and subsequently removing the field (FC). Here, the zero-field relaxation is rapid and $G_{zz}(t)$ resembles a Lorentzian Kubo-Toyabe function.[15] We conclude that the ZF relaxation after ZFC is due to static nuclear dipole moments, while the ZF relaxation after FC is derived from trapped flux lines with components perpendicular to $\hat{\mathbf{z}}$. The LF measurements also demonstrate the existence of H_{ext}-induced internal fields transverse to $\hat{\mathbf{z}}$. To understand such results, it is important to recall that a vortex lattice in which the flux lines are parallel to H_{ext} cannot enhance the LF relaxation. Indeed, we expect that in the normal vortex core regions, H_{ext} will serve to quench any such relaxation. This familiar decoupling effect, where $\lim_{t\rightarrow\infty} G_{zz}(t)$ increases rapidly with H_{ext} because H_{ext} serves to maintain the initial muon polarization, was described by Kubo and Toyabe and has been observed in many LF μSR experiments on nonsuperconducting materials.[15-17] Comparison of the ZF and LF relaxation functions at 10 K shows that the expected decoupling also occurs for superconducting $\mathrm{La_{1.85}Sr_{0.15}CuO_4}$. However, as shown in Fig. 4 the initial Gaussian relaxation rate increases monotonically with H_{ext} until $H \approx H_{c1}=150$, beyond which it appears to be field independent. The latter result is anticipated for the vortex state, while the original increase is probably due to flux lines forced to bend around small superconducting grains, thus generating transverse local fields with amplitudes roughly proportional to H_{ext}.

In summary, we have measured the magnetic penetration depth in the high-T_c superconductor $\mathrm{La_{1.85}Sr_{0.15}CuO_4}$. The result at 6 K, $\lambda \approx 2500$ Å, taken together with earlier thermodynamic measurements,[4] indicates a low density of carriers in this oxide. The temperature dependence of λ is inconsistent with the standard form for ordinary superconductors, a result which is hardly surprising when the morphology and chemical nature of the sample are considered. Our LF μSR measurements demonstrate the existence of random internal fields, induced by $H_{\mathrm{ext}} < H_{c1}$, but with components perpendicular to H_{ext}. Thus, μSR is a useful probe not only of λ for $H \gg H_{c1}$, but also of internal field distributions for $H < H_{c1}$ in inhomogeneous samples such as our sintered powder specimen.

We thank B. Batlogg, D. Harshman, and W. Weber for helpful discussions. Research at TRIUMF is supported by the Natural Sciences and Engineering Research Council of Canada and, through TRIUMF, by the Canadian National Research Council. One of the authors (G.A.) is very grateful to TRIUMF for its kind hospitality when this experiment was performed.

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

[2] K. Kishio, K. Kitazawa, S. Kanbe, I. Yasuda, N. Sugii, H. Takagi, S. Ichida, K. Fueki, and S. Tanaka (unpublished).

[3] R. J. Cava, R. B. van Dover, B. Batlogg, and E. A. Rietman, Phys. Rev. Lett. **58**, 408 (1987).

[4] B. Batlogg, A. P. Ramirez, R. J. Cava, R. B. van Dover, and E. A. Rietman, Phys. Rev. B **35**, 5340 (1987).

[5] M. K. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. **58**, 908 (1987).

[6] For a review of early work on London penetration depths, see B. S. Chandresekhar, in *Superconductivity I,* edited R. Parks (Marcel Dekker, New York, 1969), p. 1; R. Meservey and B. B. Schwarz, *ibid.,* p. 117.

[7] A. Schenck, *Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics* (Hilger, Bristol, 1985).

[8] J. H. Brewer, S. R. Kreitzman, D. R. Noakes, E. J. Ansaldo, D. R. Harshman, and R. Keitel, Phys. Rev. B **33**, 7813 (1986).

[9] P. Pincus, A. C. Gossard, V. Jaccarino, and J. Wernick, Phys. Lett. **13**, 21 (1964).

[10] A. T. Fiory, D. E. Murnick, M. Leventhal, and W. J. Kossler, Phys. Rev. Lett. **33**, 969 (1974); Y. J. Uemura, W. J. Kossler, B. Hitti, J. R. Kempton, H. E. Schone, X. H. Yu, C. E. Stronach, W. F. Lanford, D. R. Noakes, R. Keitel, M. Senba, J. H. Brewer, E. J. Ansaldo, Y. Oonuki, T. Komatsubara, G. Aeppli, and E. Bucher, Hyperfine Interact. **31**, 413 (1986).

[11] B. Batlogg (private communication).

[12] L. F. Mattheiss, Phys. Rev. Lett. **58**, 1928 (1987).

[13] W. Weber, Phys. Rev. Lett. **58**, 1371 (1987).

[14] W. W. Kwok, G. W. Crabtree, D. G. Hinks, D. W. Capone, J. D. Jorgenson, and K. Zhang, Phys. Rev. B **35**, 5343 (1987).

[15] R. Kubo and T. Toyabe, in *Magnetic Resonance and Relaxation,* edited by R. Blinc (North-Holland, Amsterdam, 1967), p. 810.

[16] R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, T. Yamazaki, and R. Kubo, Phys. Rev. B **20**, 856 (1979).

[17] J. H. Brewer, S. R. Kreitzman, K. M. Crowe, C. W. Clawson, and C. Y. Huang, Phys. Lett. **120A**, 199 (1987).