PHYSICAL REVIEW B VOLUME 35, NUMBER 13 1 MAY 1987

Magnetic field penetration depth of $La_{1.85}Sr_{0.15}CuO_4$ measured by muon spin relaxation

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(Received 23 March 1987)

Muon-spin-relaxation measurements have been performed on a high-T_c superconductor $La_{1.85}Sr_{0.15}CuO_4$. In an external transverse magnetic field of 500 G, a magnetic field penetration depth $\lambda = 2000$ Å at $T = 10$ K has been determined from the muon-spin-relaxation rate which increased with decreasing temperature below T_c. From this λ and the Pauli susceptibility we estimate the superconducting carrier density to be $n = 3\times10^{21}/\mathrm{cm}^3$. The zero-field relaxation rates above and below T_c were equal, which suggests that the superconducting state in this sample is not associated with detectable static magnetic ordering.

The oxide superconductors La (or Y or Lu)–Ba (or Sr or Ca)–Cu–O have remarkable superconducting transition temperatures T_c,[1-3] as high as 90 K. Measurements on the basic transport and magnetic properties of these systems have just been started.[4,5] The mechanism for the high T_c as well as the detailed characteristics of these superconductors are yet to be understood. In this paper we report muon-spin-relaxation (μSR) measurements on $La_{1.85}Sr_{0.15}CuO_4$ ($T_c = 37$ K) carried out at the alternating-gradient synchrotron muon channel of Brookhaven National Laboratory. In zero-field μSR (ZF-μSR) measurements, we confirmed that the superconducting state is not associated with any detectable static magnetic ordering. The magnetic field penetration depth λ is determined from the muon-spin-relaxation rate measured in the transverse external field of 500 Oe. From our observed result for λ and the value for the normal-state susceptibility $\chi(T = T_c)$, we obtain an estimate for the superconducting carrier density n. The present experiment is a new example of μSR applied to type-II superconductors.[6]

The sample was prepared from dried La_2O_3, fully oxidized CuO, and high-purity $BaCO_3$. The powders were thoroughly ground together in an agate mortar. The mixture was fired overnight in air at 1000 °C. This intermediate product was promptly ground and pelletized, then fired at 1100 °C for 20 h in oxygen. The sintered pellet of 2.5 cm diameter and 0.5 cm thickness was mounted in a ^{4}He cryostat with its face perpendicular to the direction of the muon beam. Positive muons, with the spin polarized along the beam direction, were stopped in the sample, and the time histograms $F(t)$ and $B(t)$ of decay positrons were measured by counters placed forward and backward to the sample.

After normalization for the counting efficiency of the two counters, $F(t)$ and $B(t)$ in zero magnetic field are given simply as

$$F(t) = N\exp(-t/\tau_\mu)[1 + AG_z(t)] \ ,$$
$$B(t) = N\exp(-t/\tau_\mu)[1 - AG_z(t)] \ , \quad (1)$$

where N is a normalization constant; τ_μ is the muon's lifetime, 2.2 μsec; A is the decay asymmetry of the μ^+ ranging in practice between 0.1–0.3; and the relaxation function $G_z(t)$ represents the depolarization of muon spins within the specimen. Figure 1(a) shows $G_z(t)$ observed in zero field at $T = 50$ K and at $T = 13$ K. Both results are identical within the experimental accuracy. The observed small relaxation rate is about that expected from the Cu nuclear moments. When polycrystalline magnetic material undergoes magnetic ordering, it is known that the muon relaxation function $G_z(t)$ changes and the relaxation rate increases in the ordered state except for the unlikely case where the local field cancels from crystal symmetry. Indeed, $G_z(t)$ is quite sensitive to small static random local fields as shown for example for the case of spin glasses.[7] The present results on $La_{1.85}Sr_{0.15}CuO_4$, with very slow depolarization, indicate that the upper limit of the ordered magnetic moment existing in the sample is $\lesssim 0.01\mu_B$/unit cell. This implies that there is no detectable magnetic order in this system, but does not preclude the existence of rapidly fluctuating moments. Zero-field μSR is particularly suitable for identifying the small magnetic ordering in the superconducting polycrystal samples, because it has much better sensitivity than neutron scattering, and because NMR or ESR measurements in superconductors are complicated by the penetration of external and rf fields.

When an external magnetic field is applied perpendicular to the beam direction, the muon spin precesses around the external field [transverse-field μSR (TF-μSR)]. The time histogram in this case is obtained by substituting $AG_x(t)\cos(\omega t)$ for $AG_z(t)$ of Eq. (1). $G_x(t)$ is the transverse relaxation function, and ω is the muon precession frequency, which is approximately equal to the product of the external magnetic field H_{ext} and the gyromagnetic factor of the muon: $\gamma_\mu = 2\pi$ 13.55 MHz/kOe. Figures 1(b) and 1(c) show the oscillation pattern $G_x(t)\cos(\omega t)$ observed at $T = 38$ K and $T = 15$ K with the external field $H_{\mathrm{ext}} = 500$ Oe. Above T_c the relaxation rate is independent of temperature. When cooled in field below T_c, the relaxation rate increases with decreasing temperature. By assuming an approximate form $G_x(t)=\exp(-\Lambda^2t^2)$, we obtained the relaxation rate Λ as plotted in Fig. 2(a). It is demonstrated that Λ increases upon the onset of superconductivity.

In general, there are three possible factors which contribute to the muon spin relaxation in transverse field below T_c: (1) inhomogeneity of the external field due to the vortex structure and imperfect field penetration in the type-II superconductor; (2) static random local fields if the superconductivity is associated with magnetic ordering; (3) dynamic $(1/T_1)$ processes due to fluctuating local fields. ZF-μSR is sensitive only to (2) and (3). Since we saw no change of relaxation rate between $T=50$ K and $T=10$ K in ZF-μSR, possibilities (2) and (3) can be eliminated in the present case. Thus, we attribute the observed temperature dependence of the transverse relaxation function $G_x(t)$ to the inhomogeneous penetration of the external field.

To obtain the penetration depth λ from $G_x(t)$ we have used the high Ginzburg-Landau kappa approximation which allows one to use the London equations for the fields and currents.[8] The magnetic field distribution is then $h(\mathbf{r})=B\sum_q e^{i\mathbf{q}\cdot\mathbf{r}}/(1+\lambda^2q^2)$, where the $\mathbf{q}$ are reciprocal lattice vectors. If λ were much greater than the vortex lattice spacing a, we could replace the sum for Δh^2 with an integral which would lead to

$$\Delta H^2=B^2\frac{1}{2\pi\sqrt{3}}\left\{\left[\frac{\lambda}{\sqrt{3}/2a}\right]^2\left[1+4\pi^2\left[\frac{\lambda}{\sqrt{3}/2a}\right]^2\right]\right\}^{-1}$$

for a triangular vortex lattice.[9] However, $\lambda\approx a$ and the

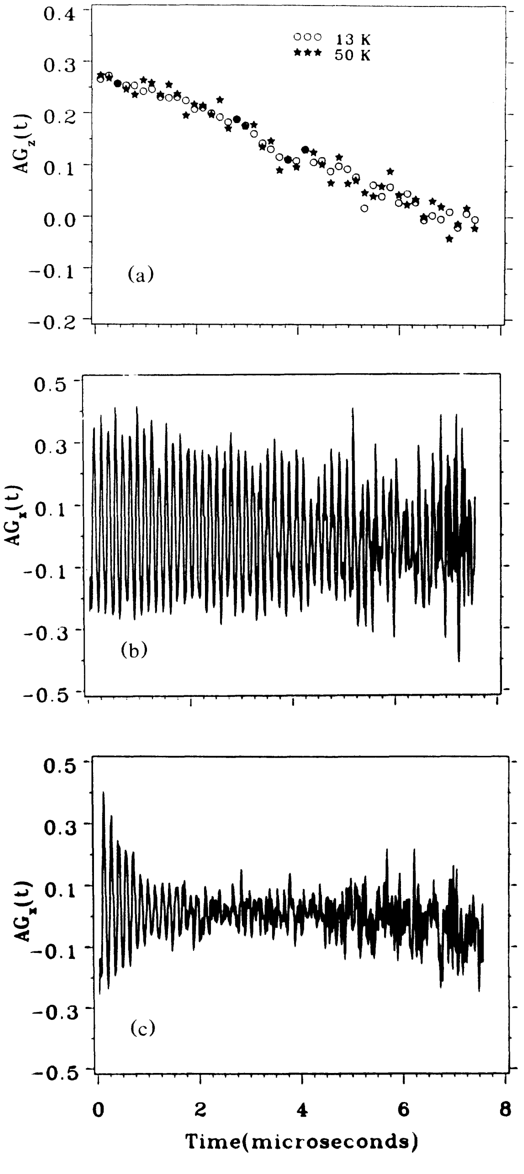

FIG. 1. The muon decay asymmetries as a function of time. (a) Zero applied field; stars are for $T=50$ K, open circles for $T=13$ K. (b) 50-mT transverse field asymmetry, $T=38$ K. (c) 50-mT transverse field asymmetry, $T=15$ K.

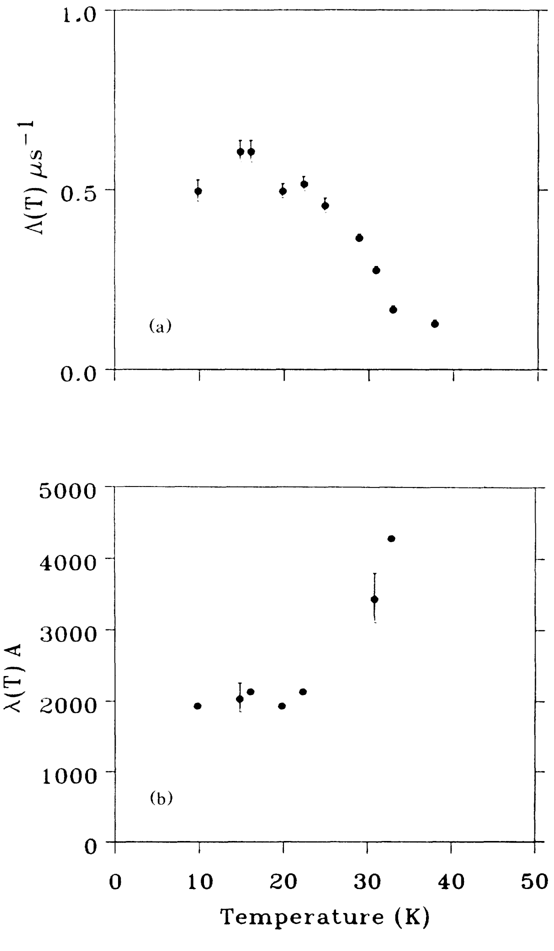

FIG. 2. (a) The depolarization rate Λ as a function of temperature. (b) The magnetic field penetration depth $\lambda(T)$ as obtained by comparison between experimental and model asymmetry functions.

field distribution is not at all Gaussian and possesses wings which are missed in fitting. Thus a more direct approach was used. We obtained $h_z(\mathbf{r})$ for a series of values for λ/a. From these we derived magnetic field distributions $P(B)(\lambda/a)$ and in turn the model functions $G_x^m(t)(\lambda/a)$. Then we matched our experimental $G_x(t)$ to these model functions. The $\lambda(T)$ so obtained are shown in Fig. 2(b). $\lambda \approx 2000$ Å at 10 K. Had we simply used ΔH above we would have obtained $\lambda \approx 3000$ Å.

Since these oxide superconductors are known to be not quite homogeneous, it is useful to have some bulk magnetic or transport properties measured on the same specimen. In Fig. 3 we present the temperature dependence of the electric resistivity together with the diamagnetic susceptibility measured via the low-frequency mutual inductance. These results show the onset of superconductivity at $T_c = 37$ K for the present specimen. From the ac diamagnetic susceptibility, we confirmed that more than $\frac{3}{4}$ of the sample becomes superconducting below $T = 10$ K.[10] μSR is a microscopic probe, and the signal represents the simple average over the different regions of the specimen. The effect of inhomogeneity is then less serious in comparison to transport measurements like resistivity. In view of this, the observed value of λ should represent the superconducting property of the bulk sample to first approximation, and corrections due to the inhomogeneity should be less than about a factor of 1.5.

The London penetration depth of superconductors is given by

$$\lambda_L = (m^* c^2 / 4\pi n e^2)^{1/2} ,$$

where m^* is the effective electron mass and n is the superconducting electron carrier density. Since the reported values for the mean free path l and the coherence length ξ are not far apart, we assume here that the correction factor $(\xi/l)^{1/2}$ for the dirty limit of type-II superconductors is about unity. Thus, the observed value of λ directly represents the London penetration depth λ_L. When we assume the effective mass m^* to be the bare electron mass m, we obtain $n = 7 \times 10^{20}$ as the superconducting carrier density of the present system. In order to separate the effects from m^* and n, one needs the value for another quantity related to the density of states at the Fermi level. Either the normal-state Pauli paramagnetic susceptibility χ or the linear term γ of the electronic specific heat would serve this purpose. Both χ and γ are proportional to

$$D(E_f) = (V/\pi^2)(m^*/\hbar^2)(3\pi^2 n)^{1/3} \sim m^* n^{1/3} .$$

Here we have used the value χ measured for a similar $La_{1.85}Sr_{0.15}CuO_4$ specimen,[4] which corresponds to $\gamma \approx 6.0$ mJ/K^{2}mole formula-unit. This value is roughly consistent with other reported values of γ and from it we obtain $n = 3 \times 10^{21}$ and $m^*/m = 4.7$.

Thus the large penetration depth of the present system is due mainly to the small carrier concentration n. This is in contrast to the case of the heavy-fermion superconductors where the large penetration depth is due mainly to the enhanced effective mass m^*. It is interesting to compare the present result on λ with the estimate from some other methods. Perhaps the most direct information to be compared is the plasma frequency ω_p which is related to λ_L by $\omega_p \lambda_L = c$. Tajima, Kitizawa, and Tanaka[11] measured the plasma edge for infrared reflection from $La_{1.82}Sr_{0.18}CuO_4$ to be at the wavelength 1.5 μm, which implies $\lambda_L = 2400$ Å. The optical measurements reflect the normal-state carrier density, whereas the present μSR experiment measures the superconducting carrier density. The good agreement between the optical and μSR results suggests that most of the carriers contribute to the superconductivity below T_c. λ has also been derived from some other bulk magnetic measurements[4,5] and the results 2000–3000 Å agree well with the present μSR study.

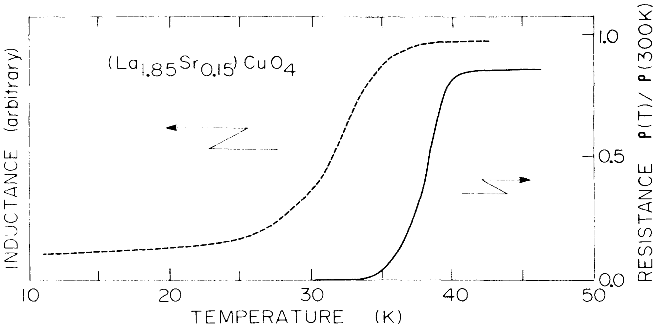

FIG. 3. Electric resistivity $\rho(T)/\rho(T = 300$ K) (solid line) and the change in inductive voltage (broken line) measured on the present specimen of $La_{1.85}Sr_{0.15}CuO_4$.

In conclusion, we have measured the magnetic field penetration depth $\lambda = 2000$ Å of $La_{1.85}Sr_{0.15}CuO_4$ by the transverse-field μSR technique, and derived the carrier density $n = 3 \times 10^{21}/\text{cm}^3$ using the values of λ and the normal-state susceptibility. We have also demonstrated by zero-field μSR that there is no detectable static magnetic order in the superconducting state. Muon spin relaxation is very suitable for the study of these oxide superconductors because one does not require single-crystal samples. One can use a pellet specimen as is because μSR does not use an rf resonance field. We are planning to extend the present study to similar oxide superconductors with higher transition temperatures.

We would like to thank Professor S. Tanaka for sending us the results of his optical measurements prior to publication. Discussions with Dr. V. J. Emery are gratefully acknowledged. We would also like to thank A. Goland, K. Lynn, and C. Pearson of BNL for aiding this experiment. During the course of this experiment we were informed by G. Aeppli that a similar work was being carried out at TRIUMF. This work is supported by the Division of Materials Science of the U.S. Department of Energy under Contract No. 76-AC02-CH00016, the National Science Foundation under Contract No. NSF-DMR 85-03223, and NASA under Grant No. NAG-1-416.

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[8] See, for instance, M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 152.

[9] This result for the triangular lattice is given in A. Schenck, *Muon Spin Rotation Spectroscopy* (Adam Hilger, Bristol, England, 1985), p. 209.

[10] One can visualize a $La_{1.85}Sr_{0.15}CuO_4$ superconductor as consisting of granular superconducting particles, i.e., islands (approximately several μm in diameter) of high-T_c material separated by nonsuperconducting layers (see Ref. 2). Therefore the change in the inductive voltage below T_c can be used to estimate the volume of superconducting material in the specimen by comparing with that for a known superconductor such as Nb.

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