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Muon-spin-rotation measurements of the penetration depth in the $YBa_2Cu_4O_8$ family of superconductors

A. Shengelaya, C. M. Aegerter, S. Romer, and H. Keller
Physik-Institut der Universität Zürich, CH-8057 Zürich, Switzerland

P. W. Klamut,* R. Dybzinski, and B. Dabrowski
Physics Department, Northern Illinois University, DeKalb, Illinois 60115

I. M. Savić
Faculty of Physics, University of Belgrade, 11001 Belgrade, Yugoslavia

J. Klamut
Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 937, 50-950 Wroclaw, Poland

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We report muon-spin-rotation (μSR) measurements of the in-plane magnetic penetration depth (λ_{ab}) in polycrystalline Ca- and La-doped $YBa_2Cu_4O_8$ superconductors with T_c ranging from 72 to 88 K. In all samples the temperature dependence of the μSR depolarization rate $\sigma(T)\propto 1/\lambda_{ab}^2(T)$ is linear below ~25 K. This is similar to what was observed in recent μSR and surface impedance experiments on $YBa_2Cu_3O_{7-\delta}$ single crystals. The low-temperature behavior of $\sigma(T)/\sigma(0)=\lambda_{ab}^2(0)/\lambda_{ab}^2(T)$ shows practically no dependence on doping when plotted versus $t=T/T_c$. This observation puts important constraints on models proposed to explain the linear temperature dependence of λ_{ab} in cuprate superconductors at low temperatures.
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I. INTRODUCTION

The magnetic field penetration depth λ is one of the fundamental lengths of a superconductor that in the clean limit is given by the London formula

$$1/\lambda^2=\frac{\mu_0 e^2 n_s}{m^*}, \tag{1}$$

where m^* is the effective mass of the superconducting carriers and n_s is the superconducting carrier density. The temperature dependence $\lambda(T)$ reflects the quasiparticle density of states available for thermal excitations and therefore probes the superconducting gap structure. In addition, the zero-temperature value $\lambda(0)$ sets a scale for the screening of an external magnetic field. Therefore, a systematic and comprehensive study of the magnetic penetration depth and its temperature dependence in various families of cuprates is of considerable importance for understanding the occurrence of superconductivity in these materials.

The muon-spin-rotation (μSR) technique is a unique tool to probe the microscopic magnetic flux distribution in the bulk of a type-II superconductor. Detailed μSR investigations of polycrystalline high-T_c superconductors (HTSC) have demonstrated that λ can be obtained from the muon spin depolarization rate $\sigma(T)\sim 1/\lambda^2(T)$, which probes the magnetic field distribution in the mixed state.[1] One of the most interesting results of μSR investigations in HTSC is a remarkable empirical relation between T_c and the zero-temperature depolarization rate $\sigma(0)$ that seems to be universal for many underdoped HTSC.[2,3] This experimental finding was taken as evidence for a Bose-Einstein condensation of preformed real-space pairs.[2] It is important to note that the value of $\sigma(0)$ is reliable and can be determined to absolute values especially well by μSR. However, the temperature dependence of σ and hence of the penetration depth as determined by μSR has proved to be controversial. In s-wave BCS theory the temperature dependence of depolarization rate at low temperatures ($T\ll T_c$) is expected to be

$$\Delta\sigma(T)/\sigma(0)=-2\Delta\lambda(T)/\lambda(0)$$
$$\sim(\Delta_0/kT)^{1/2}\exp(-\Delta_0/kT), \tag{2}$$

where Δ_0 is the isotropic energy gap. Early μSR studies on polycrystalline HTSC have concluded that σ has a weak temperature dependence for $T\ll T_c$, suggesting there is an energy gap in the spectrum of excitations,[4,5] as expected for conventional s-wave pairing. On the other hand, microwave surface impedance measurements on high-quality single crystals of $YBa_2Cu_3O_{7-\delta}$ (Y123),[6] $Bi_2Sr_2CaCu_2O_8$,[7,8] and $Tl_2Ba_2CuO_{6+\delta}$ (Ref. 9) have found a linear temperature dependence in λ at low temperatures, suggesting a pairing state with nodes in the gap. More recent μSR measurements on high-quality Y123 single crystals[10] also revealed a linear temperature dependence of λ up to $0.4T_c$, in accord with microwave measurements. It is not clear why μSR measurements in polycrystalline and single-crystal samples give different $\lambda(T)$ dependencies at low temperatures. It was suggested that small amounts of impurities or other crystalline imperfections can change the low-temperature behavior $\lambda(T)$ from a linear to a quadratic dependence,[10] that is difficult to distinguish from the BCS behavior without precise low-temperature data.

To shine more light on this problem, we decided to perform μSR measurements of the penetration depth in $YBa_2Cu_4O_8$ (Y124) with a special attention to the low-temperature behavior. The structure of Y124 is distinguished by its high thermal stability and fixed oxygen stoichiometry with well-ordered double CuO chains. Early μSR measurements of $\lambda(T)$ in Y124 gave controversial results. Zimmermann *et al.*[11] found that $\lambda(T)$ can be described by the weak-coupling BCS model, whereas Ansaldo *et al.*[12] observed a significant deviation from BCS behavior. In both cases the statements about the pairing mechanism were made on the basis of the overall behavior of $\lambda(T)$ rather than from the low-temperature region.

II. EXPERIMENTAL DETAILS

Polycrystalline samples were synthesized from stoichiometric mixtures of Y_2O_3, $BaCO_3$, and CuO. Powder samples were fired in air at 820–900 °C with frequent intermediate grinding and pelletizing. The 124 phase was obtained after final high oxygen pressure synthesis for 10 h in 20% of oxygen in argon at a total pressure of 3 kbar (600 bars O_2 pressure) at 1050 °C followed by slow cooling to room temperature. The crystal structure was examined by x-ray powder diffraction. All samples were proven to be single phased with the *Ammm* symmetry. The samples used in this study were sintered polycrystalline disks of $YBa_2Cu_4O_8$ (Y124, $T_c=81$ K), $Y_{0.94}Ca_{0.06}Ba_2Cu_4O_8$ (Y124-Ca0.06, $T_c=88$ K), $YBa_{1.925}La_{0.075}Cu_4O_8$ (Y124-La0.075, $T_c=74$ K), and $YBa_{1.9}La_{0.1}Cu_4O_8$ (Y124-La0.10, $T_c=72$ K). The transition temperature (onset of superconductivity as measured by dc magnetization) is changed with La and Ca doping due to hole filling or hole doping, respectively.

The transverse-field μSR (TF-μSR) measurements were performed on beam-line $\pi M3$ at the Paul Scherrer Institute (PSI, Switzerland) using low-momentum muons (29 MeV/c) in the temperature range 2–300 K. A detailed discussion of the TF-μSR technique is given in Ref. 5 where details of the application of the technique to determine λ can be found.

The quantity measured in TF-μSR experiment is the time evolution of spin polarization $P(t)$ of implanted (originally 100% polarized) muons. The muon-spin-polarization signal in an applied field consists of a precession function modulated by the spin depolarization function $G(t)$, whose Fourier transform gives the field probability distribution $p(B)$ in the bulk of the sample. In the mixed state of a type-II superconductor the probability distribution $p(B)$ is given by the spatially varying fields from the vortex lattice and therefore depends on the magnetic penetration depth. For instance, for an isotropic extreme type-II superconductor with a perfect triangular vortex lattice, the second moment $\langle \Delta B^2 \rangle$ of the field distribution is directly related to the penetration depth via

$$\langle \Delta B^2 \rangle = 0.00371 \Phi_0^2 \lambda^{-4}, \qquad (3)$$

where Φ_0 is the magnetic flux quantum.[13] However, in practice, conventional Fourier transform methods exhibit several problems. Significant noise in the time signal at long times, due to finite count rates, is distributed over the whole of frequency space. Furthermore, only a finite time window is available to the experiment, leading to further aberrations in the frequency spectra.

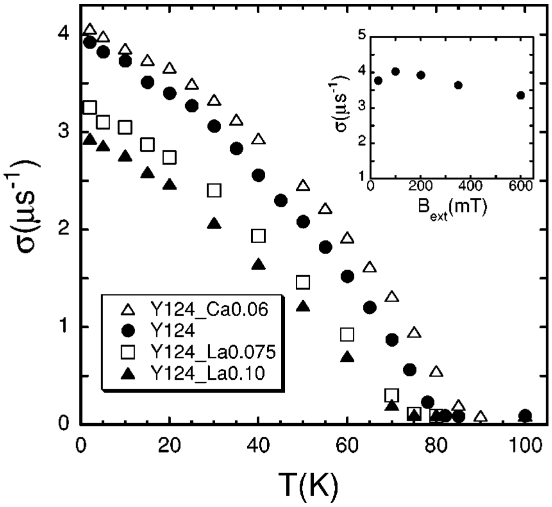

FIG. 1. Temperature dependence of the μSR depolarization rate σ for pure, Ca- and La-doped Y124 superconductors. Inset: depolarization rate as a function of the external field B_{ext} for a Y124 sample at 2 K.

Detailed μSR experiments in polycrystalline cuprate superconductors have shown that the internal field distribution in the mixed state can be well approximated by a Gaussian distribution.[5] In this case, the time dependence of the muon spin polarization $P(t) \propto \exp(-\sigma^2 t^2/2)$ and the effective penetration depth λ_{eff} (powder average) can be extracted from the μSR depolarization rate $\sigma \sim \lambda_{eff}^{-2}$. It was shown[14,15] that in polycrystalline samples of highly anisotropic systems such as the HTSC ($\lambda_c/\lambda_{ab} > 5$), λ_{eff} is dominated by the shorter penetration depth λ_{ab} due to the supercurrents flowing in the CuO_2 planes: $\lambda_{eff} = 1.31 \lambda_{ab}$.[16] The constant of proportionality between σ and λ_{ab}^{-2} depends on the symmetry and regularity of the flux lattice distribution of the demagnetization factors due to varying grain sizes and grain shapes in polycrystalline samples of highly anisotropic type-II superconductors. In the following analysis, we will be interested in the temperature dependence of σ (and hence λ) and that the exact value of the constant of proportionality is not crucial.

In the present study we analyzed μSR spectra using both Gaussian approximation and direct Fourier transform based on a maximum entropy algorithm[17] with no prior assumptions on the form of the distribution. It was found that a Gaussian distribution of local fields gives a reasonable estimate of $\langle \Delta B^2 \rangle$ in agreement with previous studies.[5] Therefore, in this paper we use the Gaussian approximation of $p(B)$ that has the advantage that no uncontrolled systematic errors are introduced in the data evaluation procedure, so that the results from different samples and workers can easily be compared.

Equation (3) is only valid for high magnetic fields ($B_{ext} > 2\mu_0 H_{c1}$), where the second moment of the field distribution $\langle \Delta B^2 \rangle$ is field independent.[13] To check for this, we measured $\sigma \propto \langle \Delta B^2 \rangle^{1/2}$ as a function of the applied field at $T=2$ K. As can be seen from the inset of Fig. 1, σ was practically field independent above $B_{ext} \simeq 30$ mT. Based on these measurements, we studied the temperature dependence of σ in a

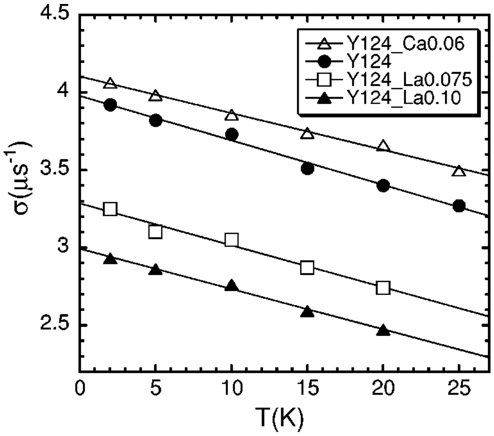

FIG. 2. The low-temperature part of the depolarization rate $\sigma(T)$ depicted in Fig. 1, showing a linear decrease of σ with temperature. The solid lines are fits to the linear relation $\sigma(T) = \sigma(0)(1-\alpha T)$.

magnetic field of 200 mT. The samples were always cooled to low temperatures in a field, to induce a homogeneous vortex lattice.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the transverse-field muon spin depolarization rate σ for the different samples. It can be seen that all samples exhibit a linear term below ~25 K, in contrast to a flat behavior expected from an isotropic BCS-like gap. This is more clearly shown in Fig. 2 where the low-temperature behavior of σ is presented. The solid lines are fits to $\sigma(T) = \sigma(0)(1-\alpha T)$. The value of α is slightly increasing with decreasing T_c from 5.8×10^{-3} K^{-1} to 8.7×10^{-3} K^{-1} (see Table I) and is close to the value of 7.5×10^{-3} K^{-1} obtained from μSR measurements on single-crystal $YBa_2Cu_3O_{6.95}$.[10] To check whether the observed linear low-temperature behavior of σ is intrinsic or is due to changes in the behavior of the flux lattice, we repeated the measurements after slow and fast cooling, under warming and cooling, and also in different magnetic fields. These experiments show no change in the low-temperature behavior of σ, confirming the intrinsic character of a linear temperature dependence of σ.

In Fig. 3(a) we plot the normalized $\sigma(T)/\sigma(0)$ versus the reduced temperature $t = T/T_c$ so that the temperature dependencies for samples with different doping levels can be compared directly. It is seen from Fig. 3(a) that all samples exhibit quite a similar temperature dependence. Particularly, at low temperatures below $\sim 0.35T_c$ all data points collapse onto a single line [see Fig. 3(b)] that can be well fitted by $\sigma(T)/\sigma(0) = 1 - at$ with the same value $a = 0.60(5)$ for all samples. A similar scaling behavior has recently been found by Hardy *et al.*[18] on single-crystal $YBa_2Cu_3O_{7-\delta}$ with $\delta = 0.01$, 0.05, and 0.40 from surface impedance measurements. Their data at low temperatures are well fitted by $\lambda^2(0)/\lambda^2(T) = 1 - 0.50(5)t$. The quantity $\lambda^2(0)/\lambda^2(T)$ represents the normalized superfluid density n_s/n_0. Thus our results on Y124 and those of Hardy *et al.* on Y123 indicate that in HTSC n_s is decreasing linearly with temperature with $dn_s/dt = -(0.5-0.6)$, independent of doping. Usually, the linear variation of $\lambda^2(0)/\lambda^2(T)$ at low temperatures is interpreted as a consequence of quasiparticle excitations near the nodes of a superconducting gap, suggesting an unconventional order parameter in the HTSC.[19] It was shown,[20] however, that phase fluctuations of the superconducting order parameter, as proposed by Emery and Kivelson,[21] can also lead to such a T dependence without invoking an unconventional order parameter. Any successful theory of HTSC should be able to explain not only the linear T dependence, but also the observed independence of dn_s/dt on doping.

TABLE I. T_c and $\sigma(0)$of the samples listed. The $\sigma(0)$ values were derived by fitting the low-temperature dependence $\sigma(T) = \sigma(0)(1-\alpha T)$. Also listed is the parameter α, which characterizes the slope of $\sigma(T)$ dependence at low temperatures.

Sample	T_c (K)	$\sigma(0)$ (μs^{-1})	α (K^{-1})
$Y_{0.94}Ca_{0.06}Ba_2Cu_4O_8$	88	4.10	0.0058
$YBa_2Cu_4O_8$	81	3.97	0.0072
$YBa_{1.925}La_{0.075}Cu_4O_8$	74	3.28	0.0082
$YBa_{1.9}La_{0.1}Cu_4O_8$	72	2.99	0.0087

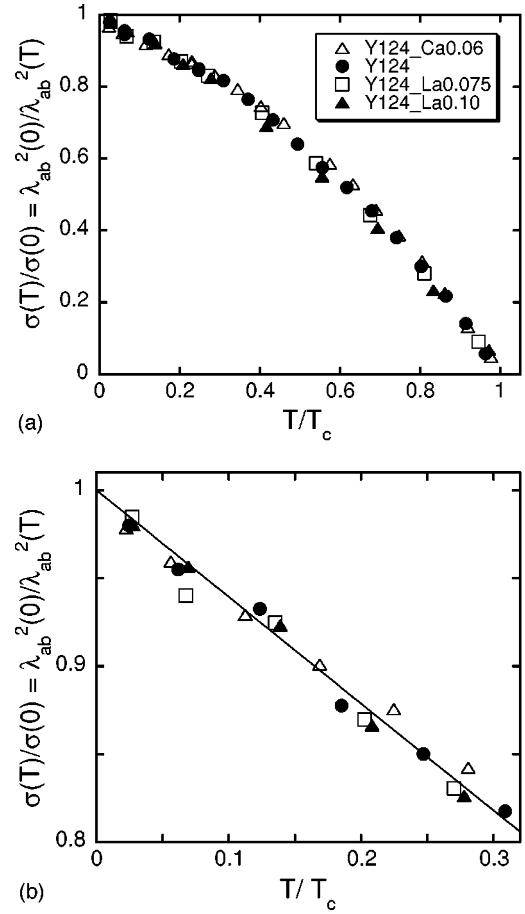

FIG. 3. (a) Normalized depolarization rate $\sigma(T)/\sigma(0)$ as a function of reduced temperature T/T_c. (b) A closeup of the low-temperature region of $\sigma(T)/\sigma(0)$ vs T/T_c depicted in (a). The solid line is a fit to the linear relation $\sigma(T)/\sigma(0) = 1 - 0.6T/T_c$.

Now let us discuss the zero-temperature values of the depolarization rate $\sigma(0)$ obtained for our samples. Figure 4 shows T_c plotted versus $\sigma(0)$ (Uemura plot[2,3]) for our Y124 samples together with previous results on Y124 (Ref. 22)

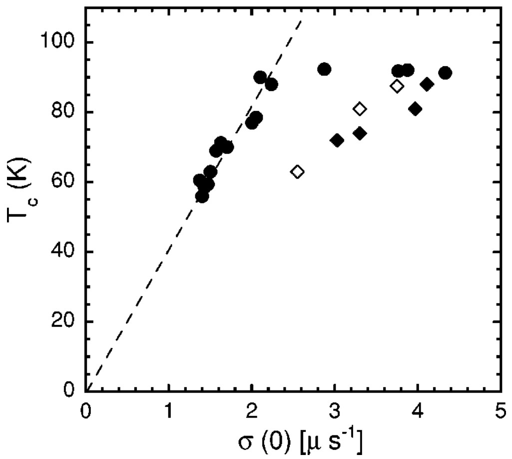

FIG. 4. T_c versus the zero-temperature depolarization rate $\sigma(0)$ for various Y123 and Y124 samples. The dashed line is the universal Uemura line. Solid circles: Y123 data from Zimmermann *et al.* (Ref. 11). Open diamonds: 124 data for $YBa_{1.85}La_{0.15}Cu_4O_8$ (T_c=63 K), $YBa_2Cu_4O_8$ (T_c=81 K) and $Y_{0.9}Ca_{0.1}Ba_2Cu_4O_8$ (T_c=87.5 K) from Bernhard *et al.* (Ref. 22). Solid diamonds: 124 data obtained in the present study for $YBa_{1.9}La_{0.1}Cu_4O_8$ (T_c=72 K), $YBa_{1.925}La_{0.075}Cu_4O_8$ (T_c=74 K), $YBa_2Cu_4O_8$ (T_c=81 K), and $Y_{0.94}Ca_{0.06}Ba_2Cu_4O_8$ (T_c=88 K).

and Y123 (Ref. 11) compounds. In the underdoped regime T_c scales linearly with $\sigma(0)$ on a single universal line for most HTSC families as shown by the dashed line in Fig. 4. This is a generic behavior expected for HTSC with CuO_2 planes *only*. It was found that several HTSC systems containing CuO chains exhibit enhanced values of $\sigma(0)$ compared with the ''Uemura line'' for ''plane-only'' systems.[22,23] This deviation from Uemura scaling was explained by an additional contribution to the superfluid density from disorder-free CuO chains.[22,23] Indeed, several experiments from various techniques such as NMR,[24] infrared,[25] and thermal conductivity[26] indicate that well-ordered CuO chains, although not intrinsically superconducting, become superconducting due to coupling with the CuO_2 planes. While T_c is practically unaffected, the additional condensate density from the superconducting chains enhances $\sigma(0)$, giving rise to the deviation from Uemura scaling.

In Fig. 4 it is seen that our $\sigma(0)$ values are significantly larger than those reported previously for Y124.[22] Since even sintered powder samples differ in grain size, density, and other qualities, the $\sigma(0)$ values obtained by different groups may differ slightly unless the samples came from the same source and were prepared the same way. However, in our opinion, these extrinsic effects are not strong enough to explain our larger $\sigma(0)$ values compared to the results of other groups, especially for the pure Y124 sample. Therefore, below we consider the possibility that large $\sigma(0)$ in our samples are due to the enhanced contribution of chain carrier superconducting condensate. Following Bernhard *et al.*,[22] we assume that the condensate in the chains contributes only to the shielding currents along the direction of the chains. Then the measured depolarization rate $\sigma(0)$ is the geometric mean of the components parallel and perpendicular to the CuO chains: $\sigma(0)=\sqrt{\sigma_{pl}(\sigma_{pl}+\sigma_{ch})}$, where σ_{pl} and σ_{ch} are the plane and chain depolarization rates, respectively. Assuming that σ_{pl} follows the Uemura scaling, we obtain $\sigma_{pl}\approx 2.15$ μs^{-1} for pure Y124 sample. With the measured value of the depolarization rate of $\sigma(0)=3.97$ μs^{-1} we thus obtain $\sigma_{ch}=5.18$ μs^{-1}. This is about two times larger than $\sigma_{ch}=2.44$ μs^{-1} derived by Bernhard *et al.*[22] for Y124. They found that the slightest amounts of disorder in the CuO chains led to a dramatic suppression of the chain condensate in Y123.[22,23] In contrast to Y123, Y124 has well-ordered CuO chains and the crystal structure tolerates only minor changes of the oxygen content between -0.14 and 0.1 per unit formula.[27] We suggest that although the oxygen nonstoichiometry effects are less dramatic in Y124 they can still lead to a significant reduction of chain condensate in this compound. Further experiments are required to clarify this problem.

Based on their small value of $\sigma_{ch}=2.44$ μs^{-1} for Y124 compared to $\sigma_{ch}=5.3$ μs^{-1} for Y123, Bernhard *et al.*[22] concluded that only part of the carriers in the double CuO chains of Y124 contribute to the superconducting condensate in contrast to the fully oxygenated chains in Y123. Our value of $\sigma_{ch}=5.18$ μs^{-1} for Y124, which is comparable to that observed in Y123, suggests that in disorder-free CuO chains of Y124 also practically *all* carriers condense to Cooper pairs.

To summarize, we have studied the in-plane penetration depth in pure as well as Ca- and La-doped polycrystalline Y124 samples by TF-μSR. The measurements on all samples reveal a clear linear temperature dependence of $\sigma(T)$ $\sim\lambda_{ab}^{-2}(T)$ at low temperatures, similar to that observed in recent μSR and surface impedance studies on Y123 single crystals. The low-temperature variation of $\sigma(T)/\sigma(0)$ $=\lambda_{ab}^2(0)/\lambda_{ab}^2(T)$ shows practically no dependence on doping when plotted versus $t=T/T_c$. This observation puts an important constraint on models proposed to explain the linear low-temperature dependence of λ_{ab} in cuprates.

The measured zero-temperature depolarization rates $\sigma(0)\sim n_s/m^*$ are larger than those previously reported for Y124 samples. This may be attributed to an enhanced contribution from the chain-carrier superconducting condensate and indicates a lower degree of disorder of the CuO chains in the samples investigated in this work.

To the best of our knowledge, the presented results are the first to show a clear linear $\sigma(T)$ dependence at low temperatures in unoriented polycrystalline HTSC samples. Previously such a behavior was observed only in high-quality single crystals, which restricted work in this field to the few materials from which such samples can be obtained in sufficient size to permit μSR measurements. The possibility to reveal the characteristic linear $\sigma(T)$ dependence in polycrystalline samples motivates us to perform more comparative studies on different high-temperature superconducting compounds in the future.

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[*] Also at Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P.O. Box 937, 50-950 Wroclaw, Poland.

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