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## **Experimental Evidence for Two Gaps in the High-Temperature La1***:***83Sr0***:***17CuO4 Superconductor**

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(Received 7 June 2006; published 2 February 2007)

The in-plane magnetic field penetration depth ( $\lambda_{ab}$ ) in single-crystal La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub> was investigated by muon-spin rotation ( $\mu$ SR). The temperature dependence of  $\lambda_{ab}^{-2}$  has an inflection point around 10– 15 K, suggesting the presence of two superconducting gaps: a large gap  $(\Delta_1^d)$  with *d*-wave and a small gap  $(\Delta_2^s)$  with *s*-wave symmetry. The zero-temperature values of the gaps at  $\mu_0H = 0.02$  T were found to be  $\Delta_1^d(0) = 8.2(1)$  meV and  $\Delta_2^s(0) = 1.57(8)$  meV.

DOI: [10.1103/PhysRevLett.98.057007](http://dx.doi.org/10.1103/PhysRevLett.98.057007) PACS numbers: 74.72.Dn, 74.25.Ha, 76.75.+i

It is mostly believed that the order parameter in cuprate high-temperature superconductors (HTS) has purely *d*-wave symmetry, as indicated by, e.g., tricrystal experiments [\[1\]](#page-3-0). There are, however, a wide variety of experimental data that support *s* or even more complicated types of symmetries  $(d + s, d + is, \text{ etc.})$  [\[2\]](#page-3-1). In order to solve this controversy, Müller suggested the presence of two superconducting condensates with different symmetries (*s*- and *d*-wave) in HTS [\[3](#page-3-2),[4](#page-3-3)]. This idea was generated partly because two gaps were observed in *n*-type  $SrTiO<sub>3</sub>$ [\[5\]](#page-3-4), the first oxide in which superconductivity was detected. In addition, it is known that the two-order parameter scenario leads to a substantial enhancement of the superconducting transition temperature in comparison to a single-band model  $[6,7]$  $[6,7]$  $[6,7]$ . The two-band model was successfully used to explain superconductivity in  $MgB_2$  [\[8\]](#page-3-7) and is considered also to be relevant to understand superconductivity in HTS [[7,](#page-3-6)[9\]](#page-3-8).

Important information on the symmetry of the order parameter can be obtained from magnetic field penetration depth  $(\lambda)$  measurements. In particular,  $\lambda(T)$ , which reflects the quasiparticle density of states available for thermal excitations, admits to probe the superconducting gap structure. Measurements of the field dependence of  $\lambda$  allow the study of the anisotropy of the superconducting energy gap [\[10\]](#page-3-9) and, in the case of two-gap superconductors, to obtain details on the relative contribution of each particular gap as a function of magnetic field [[11](#page-3-10)]. In this Letter we report a study of the in-plane magnetic penetration depth  $(\lambda_{ab})$  in slightly overdoped single-crystal La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub> by muon-spin-rotation  $(\mu SR)$ . At low magnetic fields  $(\mu_0 H \le 0.3 \text{ T}) \lambda_{ab}^{-2}(T)$  exhibits an inflection point at  $T \simeq$ 10–15 K. We interpret this feature as a consequence of the presence of two superconducting gaps. It is suggested that the large gap  $\left[\Delta_1^d(0) = 8.2(1) \text{ meV}\right]$  has *d*- and the small gap  $[\Delta_2^s(0) = 1.57(8) \text{ meV}]$  *s*-wave symmetry. With increasing magnetic field, the contribution of  $\Delta_2^s$  decreases substantially, in contrast to an almost constant contribution of  $\Delta_1^d$ . Both the temperature and the field dependences of

 $\lambda_{ab}^{-2}$  were found to be similar to what was observed in double-gap  $MgB_2$  [\[11](#page-3-10)[,12\]](#page-3-11).

The  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>$  single crystal was grown by the traveling solvent floating zone technique [\[13\]](#page-3-12). The transition temperature  $T_c$  and the width of the superconducting transition at  $\mu_0 H \approx 0$  T were found to be 36.2 and 1.5 K, respectively  $[14]$  $[14]$  $[14]$ . The  $\mu$ SR experiments were performed at the  $\pi$ M3 beam line at the Paul Scherrer Institute (Villigen, Switzerland). Typical counting statistics were  $\sim$ 16–18 million muon detections over three detectors. The sample was field cooled from above  $T_c$  to 1.6 K in a series of fields ranging from 20 mT to 0.64 T. The sample was aligned such that the  $c$  axis was parallel (within  $1^\circ$ , as measured by Laue x-ray diffraction) to the external magnetic field. In the transverse-field geometry, the local magnetic field distribution  $P(B)$  probed by  $\mu$ SR inside the superconducting sample in the mixed state is determined by the coherence length  $\xi$  and the penetration depth  $\lambda$ . In extreme type II superconductors ( $\lambda \gg \xi$ ) *P*(*B*) is almost independent of  $\xi$ , and the second moment of *P*(*B*) is proportional to  $1/\lambda^4$  $[15]$ .

Figure [1](#page-1-0) shows the magnetic field distributions  $P(B)$  for single-crystal  $\text{La}_{1.83}\text{Sr}_{0.17}\text{CuO}_4$  at  $T = 1.7$  K obtained by means of the maximum entropy Fourier transform technique. In order to extract the second moment of  $P(B)$  we used a similar procedure as described in Ref.  $[16]$ . All  $\mu$ SR time spectra were fitted by a three component expression:

$$
P(t) = \sum_{i=1}^{3} A_i \exp(-\sigma_i^2 t^2 / 2) \cos(\gamma_{\mu} B_i t + \phi).
$$
 (1)

Here  $A_i$ ,  $\sigma_i$ , and  $B_i$  are the asymmetry, the relaxation rate, and the mean field of the *i*th component, and  $\phi$  is the initial phase of the muon-spin ensemble. The first and the second moments of  $P(B)$  are [\[16\]](#page-3-15)

$$
\langle B \rangle = \sum_{i=1}^{3} \frac{A_i B_i}{A_1 + A_2 + A_3} \tag{2}
$$

and

<span id="page-1-0"></span>

FIG. 1 (color online). Local magnetic field distribution  $P(B)$  in the mixed state of single-crystal  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>$  (*T* = 1.7 K, field cooled) normalized to their maximum value at  $B = B_{\text{peak}}$ for 0.05 and 0.64 T. The inset shows theoretical  $P(B)$  distributions ( $\lambda = 220$  nm,  $\xi = 2$  nm, and  $\mu_0 H = 0.05$  T) for different values of the smearing parameter  $\sigma_B = 0, 0.3, 0.6,$  and 1.0 mT.

$$
\langle \Delta B^2 \rangle = \frac{\sigma^2}{\gamma_{\mu}^2} = \sum_{i=1}^3 \frac{A_i}{A_1 + A_2 + A_3} \{ (\sigma_i / \gamma_{\mu})^2 + [B_i - \langle B \rangle ]^2 \},\tag{3}
$$

where  $\gamma_{\mu} = 2\pi \times 135.5342 \text{ MHz/T}$  is the muon gyromagnetic ratio. The superconducting part of the square root of the second moment ( $\sigma_{\rm sc} \propto \lambda_{\rm ab}^{-2}$ ) was then obtained by subtracting the nuclear moment contribution  $(\sigma_{nm})$ measured at  $T > T_c$  according to  $\sigma_{\rm sc}^2 = \sigma^2 - \sigma_{\rm nm}^2$  [\[16\]](#page-3-15). To ensure that the increase of  $\sigma$  below  $T_c$  is attributed entirely to the vortex lattice, zero-field  $\mu$ SR experiments were performed. The experiments show no evidence for static magnetism in  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>$  down to 1.7 K.

In Fig. [2](#page-1-1) we plot the temperature dependences of  $\sigma_{\rm sc} \propto$  $\lambda_{ab}^{-2}$  for  $\mu_0 H = 0.02, 0.1$ , and 0.64 T (for clarity, data for 0.05 and 0.3 T are not shown). Most importantly, around 10–15 K an inflection point appears. It is well pronounced at  $\mu_0 H = 0.02$  T and almost absent at  $\mu_0 H = 0.64$  T. In Ref. [[17](#page-3-16)] it was pointed out that an inflection point in  $\lambda^{-2}(T)$  may appear in superconductors with two weakly coupled superconducting bands. Indeed, in  $MgB<sub>2</sub>$ , where the  $\sigma$ - and  $\pi$ -bands are almost decoupled, an upward curvature of  $\lambda^{-2}(T)$ , similar to the one observed for  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>$  at  $\mu_0H = 0.02$  T (Fig. [2\)](#page-1-1), was detected (see, e.g.,  $[12]$  $[12]$  $[12]$ ). Thus, in analogy to  $MgB<sub>2</sub>$ , we analyze our data by assuming that  $\sigma_{\rm sc}$  is a linear combination of two terms [[18](#page-3-17),[19](#page-3-18)]:

<span id="page-1-2"></span>
$$
\sigma_{sc}(T)/\sigma_{sc}(0) = \omega \delta \sigma[\Delta_1(0), T] + (1 - \omega) \delta \sigma[\Delta_2(0), T].
$$
\n(4)

Here  $\Delta_1(0)$  and  $\Delta_2(0)$  are the zero-temperature values of the large and the small gap, respectively, and  $\omega$  ( $0 \leq \omega \leq$ 

<span id="page-1-1"></span>

FIG. 2 (color online). Temperature dependence of  $\sigma_{sc} \propto \lambda_{ab}^{-2}$ of single-crystal La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub> measured at 0.02, 0.1, and 0.64 T (field cooled). Lines in the main figure and in the inset represent the fit with the two-gap model [Eq. ([4\)](#page-1-2)]. In the inset the contributions from the large *d*-wave gap and the small *s*-wave gap entering Eq. ([4\)](#page-1-2) are shown separately. See text for details.

1) is the weighting factor which measures their relative contributions to  $\lambda^{-2}$ . Note, that in contrast to MgB<sub>2</sub>, where both gaps are isotropic, in HTS at least one gap has *d*-wave symmetry [\[1\]](#page-3-0). Concerning the symmetry of the second gap, however, the situation is unclear. Based on the observation of a substantial *s*-wave contribution to the superconducting order parameter by Andreev reflection experiments [[2](#page-3-1)] and on the analysis of tunneling data [[3\]](#page-3-2), we assume that the second gap has isotropic *s*-wave symmetry. Thus, for the contribution to  $\sigma_{sc}$  arising from the *s*-wave gap, we used the standard relation [[19](#page-3-18)]

<span id="page-1-3"></span>
$$
\delta \sigma[T, \Delta^s(0)] = 1 + 2 \int_{\Delta^s(T)}^{\infty} \left(\frac{\partial f}{\partial E}\right) \frac{E}{\sqrt{E^2 - \Delta^s(T)^2}} dE. \quad (5)
$$

Here  $f = [1 + \exp(E/k_B T)]^{-1}$  is the Fermi function,  $k_B$  is the Boltzman constant, and  $\Delta^{s}(T) = \Delta^{s}(0)\tilde{\Delta}^{s}(T/T_c)$  represents the temperature dependence of the *s*-wave gap with the tabulated gap values  $\tilde{\Delta}^{s}(T/T_c)$  from [\[20\]](#page-3-19). For the *d*-wave gap contribution we take  $\Delta^d(T, \varphi) = \Delta^s(T) \times$  $cos(2\varphi)$  [[2\]](#page-3-1) and

<span id="page-1-4"></span>
$$
\delta \sigma[T, \Delta^d(0)] = 1 + 1/\pi \int_0^{2\pi} \int_{\Delta^d(T,\varphi)}^{\infty} \left(\frac{\partial f}{\partial E}\right)
$$

$$
\times \frac{E}{\sqrt{E^2 - \Delta^d(T,\varphi)^2}} dE d\varphi.
$$
(6)

In order to determine the symmetry of the two gaps, the field-cooled 0.05 T data were analyzed within " $d + s$ " and " $s + d$ " scenarios using Eq. [\(4](#page-1-2)). The analysis reveals for  $d + s$ :  $\Delta_1^d(0) = 9.0(2) \text{ meV}, \Delta_2^s(0) = 1.7(1) \text{ meV},$  $\omega = 0.69(3)$ , and for  $s + d$ :  $\Delta_1^s(0) = 6.2(2)$  meV,

$\mu_0H$ (T)	$T_c$ (K)	$\sigma_{sc}(0)$ ( $\mu s^{-1}$ )	$\omega$	$\Delta_1^d(0)$ (meV)	$\Delta_2^s(0)$ (meV)	$\frac{2\Delta_1^d(0)}{k_B T_c}$	$\frac{2\Delta_2^s(0)}{k_B T_c}$
0.02	36.3(1)	2.71(8)	0.68(3)	8.2(1)	1.57(8)	$5.24(7)^{a}$	$1.00(5)^{a}$
0.05	36.1(1)	2.20(7)	0.78(2)	8.2(1)	1.56(8)		
0.1	35.5(1)	2.07(7)	0.88(2)	8.0(1)	1.54(8)		
0.3	34.7(1)	1.82(6)	0.92(2)	7.8(1)	1.50(7)		
0.64	34.0(1)	1.71(5)	0.94(2)	7.7(1)	1.47(7)		

TABLE I. Summary of the two-gap analysis for single-crystal La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>. The meaning of the parameters is explained in the text.

a Common fit parameter for all fields.

 $\Delta_2^d(0) = 2.0(2) \text{ meV}, \quad \omega = 0.73(2).$  Comparison with  $\Delta(0) \approx 10$  meV obtained on a similar sample by tunneling experiments [\[21\]](#page-3-20), suggests that the large gap has *d*-wave symmetry. Another argument in favor of a ''large'' *d*-wave gap comes from the observation of a square vortex lattice in the same crystal as used in this work in fields higher than 0.4 T  $[14,22]$  $[14,22]$  $[14,22]$  $[14,22]$  $[14,22]$ , where, as shown below, the contribution from the large gap to  $\sigma_{sc}$  is dominant. A square vortex lattice is typical for *d*-wave superconductors [\[14\]](#page-3-13).

The solid lines in Fig. [2](#page-1-1) represent the global fit of Eq. [\(4\)](#page-1-2) to the data with contributions from the large and the small gaps described by Eqs.  $(5)$  $(5)$  and  $(6)$  $(6)$ , respectively. In the analysis all the  $\sigma_{\rm sc}(T)$  curves (0.02, 0.05, 0.1, 0.3, 0.64 T) were fitted simultaneously with  $\sigma_{\rm sc}(0)$ ,  $T_c$ , and  $\omega$  as individual parameters for each particular data set.  $\Delta_1^d(0)$  and  $\Delta_2^s(0)$  were assumed to scale linearly with  $T_c$  according to the relation  $2\Delta(0)/k_B T_c$  = const. The results are summa-rized in Table I and Fig. [3.](#page-2-0) It is seen in Fig.  $3(a)$  that the decrease of  $\sigma_{\rm sc}(0)$  is associated with an increase of the contribution of the large gap to  $\lambda^{-2}$ . Similar field dependences of  $\omega$  and  $\sigma_{sc}$  were observed in MgB<sub>2</sub> [[11](#page-3-10)[,23](#page-3-22)[,24\]](#page-3-23) and explained by the fact that superconductivity within the weaker  $\pi$ -band is suppressed at much lower fields than that within the stronger  $\sigma$ -band [\[24\]](#page-3-23). As shown in Fig. [3\(b\)](#page-2-1) this is also the case for  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>$ . Indeed, while the contribution from the large gap  $[\sigma_1(0) = \omega \sigma_{\rm sc}(0)]$ changes only slightly, the contribution from the small  $gap [\sigma_2(0) = (1 - \omega)\sigma_{sc}(0)]$  decreases by almost an order of magnitude in the field range  $0 < \mu_0 H \le 0.64$  T [Fig.  $3(b)$ ]. Thus, the temperature and the field dependences of  $\lambda_{ab}^{-2}$  in La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub> are similar to MgB<sub>2</sub>, and, consequently, demonstrate the existence of two gaps. This is the most obvious scenario, even though other gap dependences cannot be fully ruled out. Note that, the nuclear magnetic resonance [[25](#page-3-24)] and the inelastic neutron scattering data [[26](#page-3-25)] support this finding.

It is important to emphasize that the observation of an inflection point in  $\lambda^{-2}(T)$  is not restricted to MgB<sub>2</sub> and the particular sample used in this work. Indication of an inflection point in  $\lambda^{-2}(T)$  was also observed in hole-doped  $YBa_2Cu_3O_{7-\delta}$  [\[10,](#page-3-9)[27\]](#page-3-26),  $YBa_2Cu_4O_8$  [\[28\]](#page-3-27), and  $La<sub>1.85</sub>Sr<sub>0.15</sub>CuO<sub>4</sub>$  [\[29\]](#page-3-28), as well as in electron-doped  $Pr_{1.855}Ce_{0.145}CuO_{4y}$  [\[30\]](#page-3-29). In Ref. [\[27\]](#page-3-26) the increase of the second moment of  $P(B)$  observed in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$  at low temperatures was attributed to pinning effects. In order to investigate the role of pinning in our sample we compare the  $P(B)$  distributions for 0.05 and 0.64 T (Fig. [1\)](#page-1-0) with theoretical  $P(B)$  curves. A standard way to account for pinning is to convolute the theoretical  $P(B)$  for an ideal vortex lattice (black line in the inset of Fig. [1\)](#page-1-0) with a Gaussian distribution of fields [\[31\]](#page-3-30):

$$
P(B) = \frac{1}{\sqrt{2\pi}\sigma_B} \int \exp\left[-\frac{1}{2}\left(\frac{B-B'}{\sigma_B}\right)^2\right] P_{\rm id}(B')dB', \tag{7}
$$

where  $\sigma_B$  is the width of the Gaussian distribution and  $P_{\text{id}}(B)$  is the field distribution for an ideal vortex lattice [\[10\]](#page-3-9). For a stiff vortex lattice this convolution reflects how random disorder and distortions due to flux line pinning influence the ideal  $P_{\text{id}}(B)$  [[31](#page-3-30)]. The theoretical  $P(B)$  profiles for  $\sigma_B = 0, 0.3, 0.6,$  and 1.0 mT are shown in the inset of Fig. [1.](#page-1-0) The direct comparison of the  $P(B)$  data for  $\mu_0 H = 0.05$  T and 0.64 T with theoretical *P(B)* profiles clearly demonstrates that pinning is not the source of the increase of the second moment of  $P(B)$  at low temperatures. Indeed, pinning leads to an almost symmetric (around  $B_{\text{peak}}$ ) broadening of  $P(B)$  (see inset of Fig. [1\)](#page-1-0),

<span id="page-2-0"></span>

<span id="page-2-1"></span>FIG. 3 (color online). (a) Field dependences of  $\sigma_{\rm sc}(0)$  and  $\omega$ for single-crystal  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub> obtained from the fit of$ Eq. ([4\)](#page-1-2) to the data (see Table I). (b) Contribution from the large  $[\sigma_1(0)]$  and the small  $[\sigma_2(0)]$  superconducting gap to the total  $\sigma_{\rm sc}(0)$ .

while the experimental  $P(B)$  profiles very well coincide at low fields ( $\overline{B}$  <  $\overline{B}_{\text{peak}}$ ). Deviations only occur in the highfield tail of  $P(B)$  ( $B > B_{\text{peak}}$ ).

The obvious question which arises is where to locate the second superconducting gap in  $La_{2-x}Sr_xCuO_4$ . The phase diagram of cuprates is usually interpreted in terms of holes doped into the planar Cu $d_{x^2-y^2}$ -O $p_\alpha$  ( $\alpha = x, y$ ) antibonding band. In  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  it is assumed that one hole per Sr atom enters this band. However, recent *ab initio* calculations yielded additional features appearing on doping of  $La_{2-x}Sr_{x}CuO_{4}$  [\[32\]](#page-3-31). According to these calculations part of the holes occupy the Cu $d_{3z^2-r^2}$ -O $p_z$  orbitals. These re-sults are supported by neutron diffraction data [\[33\]](#page-3-32), showing that the doped holes appear in both the planar and the out-of-plane bands. In contrast to this finding, in angleresolved photoemission (ARPES) experiments on HTS only the planar band was observed, suggesting a quasitwo-dimensional electronic structure with negligible *intercell* coupling of  $CuO<sub>2</sub>$  layers (see, e.g.,  $[34]$ ). This is, however, inconsistent with in-plane and out-of-plane  $\lambda$ measurements [[35](#page-3-34)], optical conductivity [[36](#page-3-35)], and anisotropy parameter studies [\[37\]](#page-3-36). All these experiments demonstrate that with increasing doping cuprates become more and more three dimensional. Recently a 3D Fermi surface was observed in overdoped TlBa<sub>2</sub>CuO<sub>6+ $\delta$ </sub> [\[38\]](#page-3-37). Also, a careful analysis of ARPES data reveals that the finite dispersion of the energy bands along the *z* direction of the Brillouin zone  $(k_z$  dispersion) naturally induces an irreducible linewidth of the ARPES peaks which is unrelated to any scattering mechanism [[39](#page-3-38)] and implies that out-of-plane hybridized bands have to be incorporated.

In conclusion, we performed systematic  $\mu$ SR studies of the in-plane magnetic penetration depth  $\lambda_{ab}$  in singlecrystal  $La<sub>1.83</sub>Sr<sub>0.17</sub>CuO<sub>4</sub>$ . Both, the magnetic field and the temperature dependences of  $\lambda_{ab}^{-2}$  were found to be consistent with the presence of two gaps. The experimental data were analyzed by assuming that the large gap  $(\Delta_1^d)$  has *d*-wave and the small gap  $(\Delta_2^s)$  *s*-wave symmetry. Further  $\mu$ SR investigation of the penetration depth in  $La_{2-x}Sr_xCuO_4$  at various doping levels are in progress.

This work was partly performed at the Swiss Muon Source (S $\mu$ S), Paul Scherrer Institute (PSI, Switzerland). The authors are grateful to N. Momono, M. Oda, M. Ido, and J. Mesot for providing us the  $La<sub>183</sub>Sr<sub>017</sub>CuO<sub>4</sub>$  single crystal, J. Mesot for helpful discussions, and A. Amato, D. Herlach, and C.J. Juul for assistance during the  $\mu$ SR measurements. This work was supported by the Swiss National Science Foundation, in part by the NCCR program MaNEP, the EU Project CoMePhS, the SCOPES Grant No. IB7420-110784, and the K. Alex Müller Foundation.

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