Direct observation of the oxygen isotope effect on the in-plane magnetic field penetration depth in optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$

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We report the first direct observation of the oxygen-isotope ($^{16}$O/$^{18}$O) effect on the in-plane penetration depth $\lambda_{ab}$ in a nearly optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$ film using the novel low-energy muon-spin rotation technique. Spin polarized low energy muons are implanted in the film at a known depth $z$ beneath the surface and precess in the local magnetic field $B(z)$. This feature allows us to measure directly the profile $B(z)$ of the magnetic field inside the superconducting film in the Meissner state and to make a model independent determination of $\lambda_{ab}$. A substantial isotope shift $\Delta \lambda_{ab}/\lambda_{ab} = 2.8(7)\%$ at 4 K is observed, implying that the in-plane effective supercarrier mass $m^*_{ab}$ is oxygen-isotope dependent with $\Delta m^*_{ab}/m^*_{ab} = 5.5(1.4)\%$.

PACS numbers: 76.75.+i, 74.72.Bk, 74.78.Bz, 82.20.Tr

One of the fundamental questions concerning the physics of cuprate high-temperature superconductors (HTSC) is whether the electron-phonon interaction plays an essential role in these systems or not. The high values of the superconducting transition temperature $T_c$ and the observation of only a tiny oxygen-isotope shift of $T_c$ in optimally doped HTSC (see, e.g. $^{[1]}$) were taken as important arguments to propose alternative pairing mechanisms of purely electronic origin. The conventional phonon-mediated theory of superconductivity is based on the Migdal adiabatic approximation in which the effective supercarrier mass $m^*$ is independent of the mass $M$ of the lattice atoms. However, if the interaction between the carriers and the lattice is strong enough, the Migdal adiabatic approximation breaks down and $m^*$ depends on $M$ (see, e.g. $^{[2]}$). The ideal experiment to explore a possible coupling of the supercarriers to the lattice is an isotope effect study of the magnetic field penetration depth. For HTSC, which are superconductors in the clean limit, the in-plane penetration depth $\lambda_{ab}$ obeys the relation:

$$1/\lambda_{ab}^2 \propto n_s/m^*_{ab},$$

(1)

where $n_s$ is the superconducting charge carrier density and $m^*_{ab}$ is the in-plane effective mass of the charge carriers. Note that there is no such simple relation for $T_c$. Since $n_s$ was found to be predominantly isotope independent $^{[3]}$, isotope effect experiments on the penetration depth turned out to be a unique tool to investigate unconventional lattice effects in HTSC. Previous oxygen-isotope effect studies of the penetration depth in YBa$_2$Cu$_3$O$_{7-\delta}$ $^{[3]}$, La$_{2-x}$Sr$_x$CuO$_4$ $^{[4]}$, Bi$_2$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10-\delta}$ $^{[7]}$ and Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ $^{[4]}$ showed indeed a pronounced oxygen isotope dependence of the supercarrier mass. In all these experiments, however, the penetration depth was determined indirectly from magnetization measurements $^{[3]}$, the Meissner fraction $^{[4]}$, magnetic torque $^{[4]}$, and muon-spin rotation ($\mu$SR) experiments $^{[8]}$. The expression “indirectly” means that all these techniques require a model to extract the absolute value of the penetration depth $\lambda$. In contrast, measurements of the exponential field decay ($B(z) = B_0 \exp(-z/\lambda)$) in the Meissner state allow to determine $\lambda$ in a direct way. Recently, such a direct measurement of the in-plane magnetic field penetration depth $\lambda_{ab}$ in a thin YBa$_2$Cu$_3$O$_{7-\delta}$ film was performed $^{[10]}$ at the Paul Scherrer Institute (PSI, Switzerland) by using the novel low-energy $\mu$SR (LE$\mu$SR) technique $^{[11]}$. In this experiment the muon implantation depth was controlled by the variation of the incoming muon energy and the magnitude of the field was monitored by the muon spin precession frequency, alike in standard $\mu$SR.

In this Letter, we report on the oxygen-isotope ($^{16}$O/$^{18}$O) effect (OIE) on the in-plane magnetic field penetration depth in a high-quality YBa$_2$Cu$_3$O$_{7-\delta}$ film near optimal doping measured directly by LE$\mu$SR. The experiments revealed that in the Meissner state the magnetic field $B(z)$ in the $^{16}$O substituted sample decreases stronger with distance $z$ than in the $^{18}$O substituted sample, clearly demonstrating that $^{16}\lambda_{ab} < ^{18}\lambda_{ab}$.

The samples used for the LE$\mu$SR experiments were high quality YBa$_2$Cu$_3$O$_{7-\delta}$ films with an area of 2x3 cm$^2$ and a thickness of 600 nm supplied by Theva $^{[12]}$. The films were grown by thermal coevaporation of the constituents onto a single-crystal barium titanate substrate. The oxygen-isotope exchange was performed by annealing the samples in $^{16}$O$_2$ gas. In order to ensure that the $^{16}$O and $^{18}$O substituted samples are subject of the same thermal history, the annealing (in $^{16}$O$_2$ and $^{18}$O$_2$) was performed simultaneously. The exchange process was
carried out at 600°C during 25 h, followed by slow cooling (20°C/h) in order to oxidize them completely. The fraction of the 18O content in the film was estimated to be 95%, corresponding to the 18O content in the initial gas used for the annealing process. The oxygen content (7-δ) and the quality of the films were estimated from 1mT field-cooled (FC) SQUID magnetization measurements. The observed $T_c$ onsets are 16$T_c = 89.3(1)$ K and 18$T_c = 89.1(1)$ K. The transition widths are ±0.9 K for both samples. A comparison of the $T_c$’s with the known experimental $T_c$ vs δ curves for 16O and 18O substituted YBa$_2$Cu$_3$O$_{7-δ}$[13] yields δ = 0.150(5) for both films. While δ indicates that the samples are slightly underdoped the narrow transition shows that the oxygen distribution over the films is quite homogeneous. The results of the OIE on $T_c$ are summarized in Table I.

| Method       | Sample   | 16$T_c$ (K) | 18$T_c$ (K) | $\Delta T_c/T_c$ (%) | $\Delta \lambda_{ab}/\lambda_{ab}$ (%) | $\Delta m_{ab}/m_{ab}$ (%) |
|--------------|----------|-------------|-------------|----------------------|----------------------------------------|---------------------------|
| LEµSR       | Thin film| 89.3(1)     | 89.1(1)     | -0.22(16)            | 2.8(7)                                 | 5.5(1.4)                  |
| Magnetization| Fine powder| 91.66(3)    | 91.42(3)    | -0.26(5)             | 3.0(1.1)                               | 6.0(2.2)                  |
|             |          | 91.71(3)°   | 91.45(1)°   | -0.28(5)             | 2.4(1.0)                               | 4.8(2.0)                  |

°results for the back-exchange (18O→16O) sample
°°results for the back-exchange (16O→18O) sample

The transverse-field LEµSR experiments were performed on the πE3 muon beam line at PSI. A weak external magnetic field $B_0 = 9.2$ mT was applied parallel to the sample surface after the sample was cooled in zero magnetic field from a temperature above $T_c$ to 4 K. Spin-polarized muons were implanted at a depth ranging from 20-150 nm beneath the surface of the film by varying the energy of the incident muons from 3 to 30 keV. For each implantation energy a time-differential µSR spectrum was measured. The muon implantation depth profile $n(z)$ for the given implantation energy was calculated using a Monte-Carlo code TRIM.SP[14]. The reliability of the calculated $n(z)$ was crosschecked by previous LEµSR experiments on thin metal layers[15].

The experimental data were analyzed in the following way. For each implantation energy the average value of the magnetic field $\bar{B}$ and correspondent average value of the stopping distance $\bar{z}$ were extracted. The value of $\bar{B}$ was taken from the fit of the time evolution of the muon-spin polarization spectrum by using the Gaussian relaxation function:

$$a(t) = a_0 \exp(-\sigma^2 t^2/2) \cos(\gamma \bar{B} t + \phi)$$
(2)

where $a_0$ is the initial asymmetry, $\sigma$ is the Gaussian relaxation rate, and $\gamma = 2\pi \cdot 135$MHz/T is the gyromagnetic ratio of the muon. Note that at our level of statistics ($\sim 5 \times 10^5$ events per spectrum) the fit of experimental data with Eq. 2 satisfies the $\chi^2$ criterium. The value of $\bar{z}$ was taken as the first moment of the emulated $n(z)$ distribution. Results of this analysis for the 16O and 18O substituted YBa$_2$Cu$_3$O$_{7-δ}$ films are shown in Fig. 1. The data points for the 18O film are systematically higher than those for the 16O film, showing that $16\lambda_{ab} < 18\lambda_{ab}$. The solid lines represent a fit to the $\bar{B}$ data by the function:

$$\bar{B}(z) = B_0 \frac{\cosh[(t - z)/\lambda_{ab}]}{\cosh(t/\lambda_{ab})}$$
(3)

This is the form of the classical exponential field decay in the Meissner state $B(z) = B_0 \exp(-z/\lambda_{ab})$, modified for a film with thickness $2t$ with flux penetrating from both sides. The value of $z$ was corrected by $z_0 = 8$ nm, corresponding to a "dead layer", which mainly arises from the surface roughness of the film[10]. Fits with Eq. 3 to the extracted $16\bar{B}(z)$ and $18\bar{B}(z)$ yield $16\lambda_{ab}(4K) = 151.85(75)$ nm and $18\lambda_{ab}(4K) = 155.82(68)$ nm. Taking into account a 18O content of 95%, the relative shift was found to be $\Delta \lambda_{ab}/\lambda_{ab} = (18\lambda_{ab} - 16\lambda_{ab})/16\lambda_{ab} = 2.8(7)\%$ at 4 K. This value is consistent with previous results for optimally doped YBa$_2$Cu$_3$O$_{7-δ}$[8] and Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10+δ}$[9] obtained from indirect magnetization measurements. Here we would like to mention two important points: (i) The good agreement between the experimental points and theoretical curves (Eq. 3) indicates that the films are homogenous over the whole thickness. (ii) $\lambda_{ab}$ obtained from LEµSR is model independent. It is extracted from the measured field profile in the Meissner state using only one fit parameter $\lambda_{ab}$.

We also performed additional OIE experiments on
\( \Delta \lambda_{ab} = 1.5185(75) \text{ nm} \)

\( \Delta \lambda_{ab} = 1.5582(68) \text{ nm} \)

\[ \Delta \lambda_{ab}/\lambda_{ab} = 2.8(7) \% \]

\( T = 4 \text{ K} \)

\( B(0) = 9.2 \text{ mT} \)

\( \lambda_{ab} \) based on measurements of the Meissner fraction in fine YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) powder. The powder samples were ground for about 60 min and then passed through a 10 \( \mu \text{m} \) sieve. The oxygen exchange procedure was analogous to the one described above for the thin film samples. The \( ^{18}\text{O} \) content in the sample, as determined from the change of the sample weight after the isotope exchange, was found to be 89(2)%. The value of the Meissner fraction \( f \) was calculated from 1 mT FC SQUID magnetization measurements. The absence of weak links between grains was confirmed by the linear magnetic field dependence of the FC magnetization measured at 5 K in 0.5 mT, 1 mT, and 1.5 mT. The temperature dependence of the magnetic field penetration depth \( \lambda_{\text{eff}} \) (powder average) was calculated from \( f \) under the assumption that the sample grains are spherical \(^{10}\). The in-plane penetration depth \( \lambda_{ab}(T) \) (Fig. 2) was determined from the measured \( \lambda_{\text{eff}}(T) \) using the relation \( \lambda_{\text{eff}} = 1.31 \lambda_{ab} \) which holds for highly anisotropic superconductors \((\lambda_{c}/\lambda_{ab} > 5)\) \(^{17}\). Taking into account a \( ^{18}\text{O} \) content of 89%, the relative shift at 4 K was found to be \( \Delta \lambda_{ab}/\lambda_{ab} = 3.0(1.1) \% \) in good agreement with the LE\&SR data (see Table I). In order to substantiate the intrinsic character of the observed OIE on \( \lambda_{ab} \), we performed a back exchange experiment. For this purpose the \( ^{16}\text{O} \) sample was annealed in \( ^{18}\text{O}_{2} \) gas \((^{16}\text{O} \rightarrow ^{18}\text{O})\) and the \( ^{18}\text{O} \) sample in \( ^{16}\text{O}_{2} \) gas \((^{18}\text{O} \rightarrow ^{16}\text{O})\). The annealing procedure was analogous to the one described above for the thin film samples. The results of the back exchange experiments are also shown in Fig. 2 (cross symbols). All the results of the OIE on \( T_c \) and \( \lambda_{ab} \) are summarized in Table II.

According to Eq. (1) the OIE on \( \lambda_{ab} \) is due to a shift in \( n_s \) and/or \( m^*_{ab} \):

\[ \frac{\Delta \lambda_{ab}}{\lambda_{ab}} = \frac{1}{2} \frac{\Delta m^*_{ab}}{m^*_{ab}} - \frac{1}{2} \frac{\Delta n_s}{n_s}. \]

Previous OIE studies in HTSC’s \(^3\) showed a pronounced OIE on \( m^*_{ab} \) with negligible OIE on \( n_s \). In the present work we provide further evidence of this scenario from measurements of the nuclear quadrupole resonance (NQR) frequency (\( \nu_Q \)) of the plane \((\nu_Q^{pl})\) and the chain \((\nu_Q^{ch})\) \(^{63}\text{Cu} \) in \(^{16}\text{O} \) and \(^{18}\text{O} \) substituted YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) powder samples. It is known \(^{18}\) that in HTSC \( \nu_Q^{pl} \) and \( \nu_Q^{ch} \) are very sensitive to changes in the density of mobile carriers \( n \) in the superconducting CuO\(_2\) planes \((n^{pl})\) and CuO chains \((n^{ch})\). Both \( \nu_Q^{pl} \) and \( \nu_Q^{ch} \) increase with doped hole concentration roughly 20 MHz per one doped hole per Cu atom \(^{20}\). The NQR measurements of \(^{63}\text{Cu} \) at 94 K show that within 5 kHz error bar limits the plane copper and the chain copper NQR frequencies are the same in \(^{16}\text{O} \) and \(^{18}\text{O} \) substituted YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) powder samples (see Table II). This implies that by the oxygen substitution \( n^{pl} \) and \( n^{ch} \) change for less than \( 3 \times 10^{-4} \) hole per Cu atom. In optimally doped YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) one has two plane and one chain Cu atom in the unit cell. From the NQR measurements we can thus conclude that the change of the hole number per unit cell is less than \( 3 \times 10^{-4} \% \). The NQR results are summarized in Table II. For YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) it was shown \(^{21}\) that \( n_s \approx n \) at \( T < 70 \text{ K} \). Based on this observation we thus can conclude that \( |1/2 \cdot \Delta n_s/n_s| < 0.05\% \) in Eq. (4). Consequently, the main contribution to the OIE on \( \lambda_{ab} \) has to come from the isotope dependence of the in-plane charge carrier mass \( m^*_{ab} \), so that \( \Delta \lambda_{ab}/\lambda_{ab} \approx (\Delta m^*_{ab}/m^*_{ab})/2. \) With \( \Delta \lambda_{ab}/\lambda_{ab} = 2.8(7) \% \) we obtain \( \Delta m^*_{ab}/m^*_{ab} = 5.5(1.4) \% \) at 4 K (see Table I). This result is remarkable in spite of the fact that the observed OIE on \( T_c \) in optimally doped YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) is rather small \((\Delta T_c/T_c = -0.26(5) \% \) at 4 K). Based on this observation we thus can conclude that \( |1/2 \cdot \Delta n_s/n_s| < 0.05\% \) in Eq. (4). Consequently, the main contribution to the OIE on \( \lambda_{ab} \) has to come from the isotope dependence of the in-plane charge carrier mass \( m^*_{ab} \), so that \( \Delta \lambda_{ab}/\lambda_{ab} \approx (\Delta m^*_{ab}/m^*_{ab})/2. \) With \( \Delta \lambda_{ab}/\lambda_{ab} = 2.8(7) \% \) we obtain \( \Delta m^*_{ab}/m^*_{ab} = 5.5(1.4) \% \) at 4 K (see Table I). This result is remarkable in spite of the fact that the observed OIE on \( T_c \) in optimally doped YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) is rather small \((\Delta T_c/T_c = -0.26(5) \% \) at 4 K).
TABLE II: Results of plane and chain $^{63}$Cu NQR in $^{16}$O and $^{18}$O substituted YBa$_2$Cu$_3$O$_{7-\delta}$ powder samples at 94 K (see text for an explanation).

|        | $^{16}$O   | $^{18}$O   |        |
|--------|------------|------------|--------|
| $\nu_Q$ Linewidth (MHz) | 31.580(5) | 31.580(5) | $\Delta \nu_Q/\nu_Q$ (%) |
| $\nu_Q$ Linewidth (KHz) | 336(10)  | 340(10)  | 0.00(2) |
| Chains | 22.050(5) | 22.050(5) | 161(5)  | 161(5)  | 0.00(3) |

In conclusion, we used LE$\mu$SR to measure directly the oxygen isotope ($^{16}$O/$^{18}$O) effect on the in-plane magnetic field penetration depth $\lambda_{ab}$ in optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$ films. The OIE on $\lambda_{ab}$ at 4 K was found to be $\Delta \lambda_{ab}/\lambda_{ab} = 2.8(7)\%$. The intrinsic character of the OIE on $\lambda_{ab}$ was confirmed by back exchange experiments on fine powders with low-field SQUID magnetization measurements. It is concluded that the OIE arises mainly from the isotope dependence of the in-plane charge carrier mass $m^*_a$ with $\Delta m^*_a/m^*_a = 5.5(1.4)\%$ at 4 K. This finding implies that even in optimally doped cuprate superconductors for which only a small isotope effect on $T_c$ is observed, the supercarriers are strongly coupled to the lattice.

This work was partly performed at the Swiss Muon Source (S$\mu$S) at the Paul Scherrer Institute (Villigen, Switzerland). The authors are grateful to A. Bussmann-Holder, K.A. Müller, T. Schneider, and Z.X. Shen for stimulating discussions. This work was supported by the Swiss National Science Foundation and by the NCCR program Materials with Novel Electronic Properties (MaNEP) sponsored by the Swiss National Science Foundation.

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