Oxygen-isotope effect on the in-plane penetration depth in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ as revealed by muon-spin rotation

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The oxygen-isotope ($^{16}\text{O}/^{18}\text{O}$) effect (OIE) on the in-plane penetration depth $\lambda_{ab}(0)$ in underdoped $Y_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ was studied by muon-spin rotation. A pronounced OIE on $\lambda_{ab}^{-2}(0)$ was observed with a relative isotope shift of $\Delta\lambda_{ab}^{-2}/\lambda_{ab}^{-2}=-5(2)\%$ for x=0.3 and -9(2)% for x=0.4. It arises mainly from the oxygen-mass dependence of the in-plane effective mass m_{ab}^* . The OIE exponents of T_c and of $\lambda_{ab}^{-2}(0)$ exhibit a relation that appears to be generic for cuprate superconductors.

PACS numbers: 76.75.+i, 74.72.-h, 82.20.Tr, 71.38

The pairing mechanism responsible temperature superconductivity remains elusive in spite of the fact that many models have been proposed since its discovery. A fundamental question is whether lattice effects are relevant for the occurrence of high-temperature superconductivity. In order to clarify this point a large number of isotope-effect studies were performed since 1987 [1]. The first oxygen-isotope effect (OIE) studies on the transition temperature T_c were performed on optimally doped samples, showing no significant isotope shift [2]. However, later experiments revealed a small but finite dependence of T_c on the oxygen-isotope mass $M_{\rm O}$ [3, 4, 5, 6], as well as on the copper-isotope mass $M_{\rm Cu}$ [7, 8]. Moreover, a general trend in the dependence of the OIE exponent $\alpha_{\rm O} = -d \ln T_{\rm c}/d \ln M_{\rm O}$ on the doping level was found which appears to be generic for all cuprate superconductors [1, 5, 8, 9, 10]: In the underdoped region $\alpha_{\rm O}$ is large, even exceeding the conventional BCS-value $\alpha = 0.5$ and becomes small in the optimally doped and overdoped regime.

There is increasing evidence that a strong electron-phonon coupling is present in cuprate superconductors, which may lead to the formation of polarons (bare charge carriers accompanied by local lattice distortions) [11, 12]. One way to test this hypothesis is to demonstrate that the effective mass of the supercarriers m^* depends on the mass M of the lattice atoms. This is in contrast to conventional BCS superconductors, where only the 'bare' charge carriers condense into Cooper pairs, and m^* is essentially independent of M. For cuprate superconductors (clean limit) the in-plane penetration depth λ_{ab} is simply given by $\lambda_{ab}^{-2}(0) \propto n_s/m_{ab}^*$, where n_s is the superconducting charge carrier density, and m_{ab}^* is the in-plane effective mass of the superconducting charge carriers. This implies that the OIE on λ_{ab} is due to a shift in n_s and/or

 m_{ab}^* :

$$\Delta \lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = \Delta n_s/n_s - \Delta m_{ab}^*/m_{ab}^*. \tag{1}$$

Therefore a possible mass dependence of m_{ab}^* can be tested by investigating the isotope effect on λ_{ab} , provided that the contribution of n_s to the total isotope shift is known.

Previous OIE studies of the penetration depth in $YBa_2Cu_3O_{7-\delta}$ [13], $La_{2-x}Sr_xCuO_4$ [10, 14, 15], and $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10+\delta}$ [16] indeed showed a pronounced oxygen-mass dependence on the supercarrier mass. However, in all these experiments the penetration depth was determined indirectly from the onset of magnetization [13, 16], from the Meissner fraction [10, 14], and from magnetic torque measurements [15]. The muonspin rotation (μSR) technique is a direct and accurate method to determine the penetration depth λ in type II superconductors. In this Letter, we report μSR measurements of in-plane penetration depth λ_{ab} in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (x = 0.3 and 0.4) with two different oxygen isotopes (16 O and 18 O). A large OIE on λ_{ab} was observed which mainly arises from the oxygen-mass dependence of m_{ab}^* .

Polycrystalline samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (x=0.3 and x=0.4) were prepared by standard solid state reaction [17]. Oxygen isotope exchange was performed during heating the samples in $^{18}O_2$ gas. In order to ensure the same thermal history of the substituted (^{18}O) and not substituted (^{16}O) sample, two experiments (in $^{16}O_2$ and $^{18}O_2$) were always performed simultaneously. The exchange and back exchange processes were carried out at $600^{\circ}C$ during 25 h, and then the samples were slowly cooled ($20^{\circ}C/h$) in order to oxidize them completely. The ^{18}O content in the samples, as determined from a change of the sample weight after the isotope exchange, was found to be 78(2)% for both samples. The total oxygen content of the samples was deter-

mined using high-accuracy volumetric analysis [17]. To examine the quality of the samples low-field (1mT, field-cooled) SQUID magnetization measurements were performed (see Fig. 1). For both concentrations the T_c onset for the ¹⁶O samples was higher than for ¹⁸O with nearly the same transition width. An oxygen back exchange of the ¹⁸O sample (x = 0.4) resulted within error in almost the same magnetization curve as for the ¹⁶O sample, confirming that the back exchange is almost complete. The results of the OIE on T_c are summarized in Table I. Taking into account an isotope exchange of 78%, we found $\alpha_{\rm O} = 0.22(4)$ for x = 0.3 and $\alpha_{\rm O} = 0.37(5)$ for x = 0.4, in agreement with previous results [9, 18].

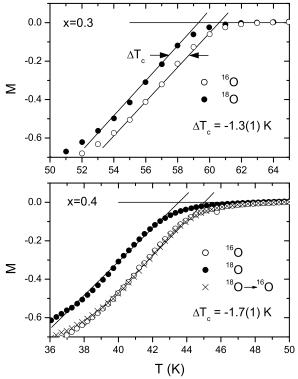


FIG. 1: Section near T_c of the low-field (1mT, field-cooled) magnetization curves (normalized to the value at 10K) for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (x = 0.3 and 0.4).

The μ SR experiments were performed at the Paul Scherrer Institute (PSI), Switzerland, using the π M3 μ SR facility. The samples consisted of sintered pellets (12 mm in diameter, 3 mm thick) which were mounted on a Fe₂O₃ sample holder in order to reduce the background from muons not stopping in the sample. The polycrystalline Y_{1-x}Pr_xBa₂Cu₃O_{7- δ} samples were cooled from far above T_c in a magnetic field of 200 mT perpendicular to the sample disk. Time-differential μ SR spectroscopy was employed, from which one can deduce the probability distribution of the local magnetic field p(B) of the vortex state by measuring the time evolution of the muon-spin polarization [19]. In a powder sample the magnetic penetration depth λ can be extracted from the

muon-spin depolarization rate $\sigma(T) \propto 1/\lambda^2(T)$, which probes the second moment $\langle \Delta B^2 \rangle^{1/2}$ of p(B) in the mixed state [19, 20]. For highly anisotropic layered superconductors (like the cuprate superconductors) λ is mainly determinated by the in-plane penetration depth λ_{ab} [20]: $\sigma(T) \propto 1/\lambda_{ab}^2(T) \propto n_s/m_{ab}^*$.

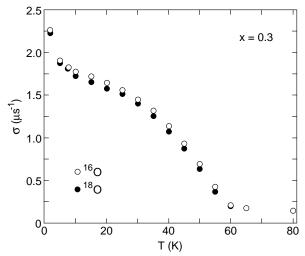


FIG. 2: Temperature dependence of the μ SR depolarization rate σ of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ for x=0.3, measured in a field 200 mT (field-cooled).

The depolarization rate σ was extracted from the μSR time spectra using a Gaussian relaxation function $R(t) = \exp[-\sigma^2 t^2/2]$. Figure 2 shows the temperature dependence of the measured σ for the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with x=0.3. Similar results were obtained for the samples with x = 0.4. It is evident that the values of σ for ¹⁸O are systematically lower than those for ¹⁶O. As expected for a type II superconductor in the mixed state, σ continuously increases below T_c with decreasing temperature [20]. The sharp increase of σ below $\simeq 10$ K is due to antiferromagnetic ordering of the Cu(2) moments [21]. Above T_c a small temperature independent depolarization rate $\sigma_{nm} \simeq 0.15 \ \mu \text{s}^{-1}$ is seen, arising from the nuclear magnetic moments of Cu and Pr. Therefore, the total σ is determined by three contributions: a superconducting (σ_{sc}) , an antiferromagnetic (σ_{afm}) , and a small nuclear magnetic dipole (σ_{nm}) contribution. Because σ_{afm} is only present at low temperatures, data points below 10 K were not considered in the analysis. The superconducting contribution σ_{sc} was then determined by subtracting σ_{nm} measured above T_c from σ . In Fig. 3 we show the temperature dependence of σ_{sc} for the $\mathrm{Y}_{1-x}\mathrm{Pr}_{x}\mathrm{Ba}_{2}\mathrm{Cu}_{3}\mathrm{O}_{7-\delta}$ samples with x=0.3and 0.4. It is evident that for both concentrations a remarkable oxygen isotope shift on T_c as well as on σ_{sc} is present.

The data in Fig. 3 were fitted to the power law $\sigma_{sc}(T)/\sigma_{sc}(0) = 1 - (T/T_c)^n$ [20] with $\sigma_{sc}(0)$ and n as free parameters, and T_c fixed. The values of T_c were taken from the magnetization measurements (see Table I). The

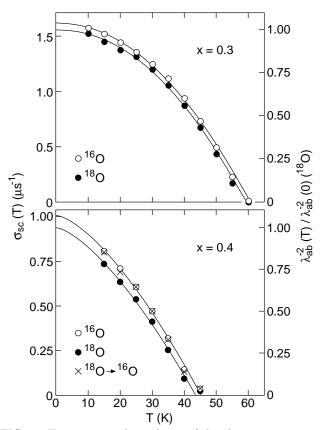


FIG. 3: Temperature dependence of depolarization rate σ_{sc} in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ for x=0.3 and 0.4 (200 mT, field-cooled). On the right axis the normalized in-plane penetration depth $\lambda_{ab}^{-2}(T)/\lambda_{ab}^{-2}(0)(^{18}O)$ is plotted for comparison with Ref. [15]. The solid lines correspond to fits to the power law $\sigma_{sc}(T)/\sigma_{sc}(0) = 1 - (T/T_c)^n$.

values of $\sigma_{sc}(0)$ obtained from the fits are listed in Table I and are in agreement with previous results [21]. The exponent n was found to be n = 2.0(1) for x = 0.3 and n = 1.5(1) for x = 0.4, which is typical for underdoped YBCO [20]. Moreover, n is within error the same for ¹⁶O and ¹⁸O. This implies that σ_{sc} has nearly the same temperature dependence for the two isotopes (see Fig. 3). In order to proof that the observed OIE on $\lambda_{ab}(0)$ are intrinsic, the ¹⁸O sample with x = 0.4 was back exchanged ($^{18}O \rightarrow ^{16}O$). As seen in Fig. 3, the data points of this sample (cross symbols) indeed coincide with those of the ¹⁶O sample. From the values of $\sigma_{sc}(0)$ listed in Table I the relative isotope shift of the in-plane penetration depth $\Delta \lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = [\sigma_{sc}^{18O}(0) - \sigma_{sc}^{16O}(0)]/\sigma_{sc}^{16O}(0)$ was determined. Taking into account an isotope exchange of 78%, one finds $\Delta \lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = -5(2)\%$ and -9(2)% for x = 0.3 and 0.4, respectively (Table I). For the OIE exponent $\beta_{\rm O} = -d \ln \lambda_{ab}^{-2}(0)/d \ln M_{\rm O}$, one readily obtains $\beta_{\rm O} = 0.38(12)$ for x = 0.3 and $\beta_{\rm O} =$ 0.71(14) for x=0.4 (Table I). This means that in underdoped $\mathbf{Y}_{1-x}\mathbf{Pr}_{x}\mathbf{Ba}_{2}\mathbf{Cu}_{3}\mathbf{O}_{7-\delta}$ the OIE on λ_{ab}^{-2} as well as on T_c increase with increasing Pr doping x (decreas-

TABLE I: Summary of the OIE results for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ extracted from the experimental data (see text for an explanation).

	¹⁶ O		¹⁸ O				
\overline{x}	T_c	$\sigma_{sc}(0)$	T_c	$\sigma_{sc}(0)$	α_{O}	$\frac{\Delta \lambda_{ab}^{-2}(0)}{\lambda_{ab}^{-2}(0)}$	β_{O}
	[K]	$[\mu \mathrm{s}^{-1}]$	[K]	$[\mu \mathrm{s}^{-1}]$		[%]	
	60.6(1)						0.38(12)
0.4	45.3(1)	1.01(2)	43.6(1)	0.94(2)	0.37(5)	-9(2)	0.71(14)
0.4	$45.1(1)^a$	$1.01(4)^a$					

^aresults for the back-exchange ($^{18}O \rightarrow ^{16}O$) sample

ing T_c). This finding is in excellent agreement with the recent magnetic torque measurements on underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ [15].

According to Eq. (1) the observed $\Delta \lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0)$ is due to a shift of n_s and/or m_{ab}^* . For $La_{2-x}Sr_xCuO_4$ several independent experiments [10, 14, 15] have shown that the change of n_s during the exchange procedure is negligibly small. In the present work we provide further evidence: (i) The fully oxygenated $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples $(\delta \simeq 0)$ were all prepared under identical conditions, either in a ¹⁶O₂ or ¹⁸O₂ atmosphere [17], and the Pr content x did not change. It is very unlikely that n_s changes significantly upon ¹⁸O substitution, and after the back-exchange ($^{18}O\rightarrow$ ¹⁶O) the same results are obtained (see Figs. 1, 3 and Table I). (ii) According to a model [22] that describes the suppression of T_c in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$, the number of supercarriers decreases linearly with increasing x in the range of 0.05 < x < 0.5, and consequently $\Delta n_s/n_s = -\Delta x/x$. Moreover, for 0.1 < x < 0.5 the transition temperature T_c decreases linearly with x, with $\Delta T_c/\Delta x \simeq -150$ K/Pr atom [9]. Combining this two relations one obtains: $\Delta T_c \simeq -150 \cdot x \cdot \Delta n_s/n_s$. Assuming that the observed OIE on λ_{ab}^{-2} is only due to a change of n_s ($\Delta m_{ab}^*/m_{ab}^* \simeq 0$), one can estimate the corresponding shift of T_c . For x = 0.3 and x = 0.4 one finds $\Delta T_c \simeq -1.8(4)$ K and -4.2(6) K, respectively. The experimental values, however, are much lower (see Fig. 1): $\Delta T_c = -1.3(1) \text{ K} (x = 0.3) \text{ and } \Delta T_c = -1.7(1) \text{ K}$ (x = 0.4). We thus conclude that any change in n_s during the exchange procedure must be small, and that the change of λ_{ab} is mainly due to the OIE on the in-plane effective mass m_{ab}^* with $\Delta m_{ab}^*/m_{ab}^* \simeq 5(2)$ % and 9(2) % for x = 0.3 and x = 0.4, respectively. This implies that the effective supercarrier mass m_{ab}^* in this cuprate superconductor depends on the oxygen mass of the lattice atoms, which is not expected for a conventional phononmediated BSC superconductor.

In Fig. 4 the exponent $\beta_{\rm O}$ versus the exponent $\alpha_{\rm O}$ for $Y_{1-x} Pr_x Ba_2 Cu_3 O_{7-\delta}$ is plotted. For comparison we also included the recent magnetic torque results of underdoped $La_{2-x} Sr_x Cu O_4$ [15]. It is evident that these

exponents are linearly correlated: $\beta_{\rm O} = A \cdot \alpha_{\rm O} + B$. A best fit yields A = 1.8(4) and B = -0.01(12), so that $\beta_{\rm O} \simeq A \cdot \alpha_{\rm O}$. This empirical relation appears to be generic for cuprate superconductors. Quantitatively one can understand this behavior in terms of an empirical relation between T_c and the $\mu{\rm SR}$ depolarization rate $\sigma_{sc}(0)$ [23, 24]. It was shown [24] that for most families of cuprate superconductors the simple parabolic relation $\overline{T}_c = 2\overline{\sigma}(1-\overline{\sigma}/2)$ describes the experimental data rather well (here $\overline{T}_c = T_c/T_c^m$, $\overline{\sigma} = \sigma_{sc}(0)/\sigma_{sc}^m(0)$, and T_c^m and $\sigma_{sc}^m(0)$ are the transition temperature and depolarization rate of the optimally doped system). Using this parabolic Ansatz, one readily obtains the linear relation between $\beta_{\rm O}$ and $\alpha_{\rm O}$: $\beta_{\rm O}/\alpha_{\rm O} = 1 + 1/2$ [$(1 - (1 - \overline{T}_c)^{1/2})/(1 - \overline{T}_c)^{1/2}$]. In the heavily underdoped regime

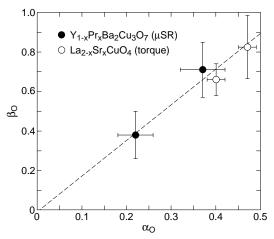


FIG. 4: Plot of the OIE exponents $\beta_{\rm O}$ versus $\alpha_{\rm O}$ for ${\rm Y}_{1-x}{\rm Pr}_x{\rm Ba}_2{\rm Cu}_3{\rm O}_{7-\delta}$ (x=0.3 and 0.4) and ${\rm La}_{2-x}{\rm Sr}_x{\rm Cu}{\rm O}_4$ (x=0.080 and 0.086) from [15]. The dashed line represents a best fit to the empirical relation $\beta_{\rm O}=A\cdot\alpha_{\rm O}+B$.

 $(\overline{T}_c \to 0) \ \beta_{\rm O}/\alpha_{\rm O} \to 1$. For the underdoped samples shown in Fig. 4 the reduced critical temperature \overline{T}_c is in the range 0.5 to 0.7, yielding $\beta_{\rm O}/\alpha_{\rm O}=1.2-1.4$, in agreement with A=1.8(4) obtained from the experimental data. Very recently, it was pointed out [25] that the unusual doping dependence of the OIE on T_c and on $\lambda_{ab}^{-2}(0)$ naturally follows from the doping driven 3D-2D crossover and the 2D quantum superconductor to insulator transition in the underdoped limit. It is predicted that in the underdoped regime $\beta_{\rm O}/\alpha_{\rm O} \to 1$, which is consistent with the parabolic Ansatz.

In summary, we performed μ SR measurements of the in-plane penetration depth λ_{ab} in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (x=0.3,0.4) for samples with two different oxygen isotopes (^{16}O and ^{18}O). A pronounced OIE on both the transition temperature T_c and $\lambda_{ab}^{-2}(0)$ was observed, which increases with decreasing T_c . The isotope shift on $\lambda_{ab}^{-2}(0)$ is attributed to a shift in the inplane effective mass m_{ab}^* . For x=0.3 and 0.4 we find $\Delta m_{ab}^*/m_{ab}^*=-5(2)\%$ and -9(2)%, respectively. Furthermore, an empirical relation between the OIE exponents β_O and α_O was found that appears to be generic for various classes of cuprate superconductors. The OIE on m_{ab}^* implies that the superconducting carriers have polaronic character, and that lattice effects play an essential role in the occurrence of high-temperature superconductivity.

We are grateful to G.M. Zhao, T. Schneider, and K.A. Müller for many fruitful discussions and to A. Amato, U. Zimmermann, and D. Herlach from PSI for technical support during the μ SR experiments. This work was partly supported by the Swiss National Science Foundation.

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