Oxygen-isotope effect on the superconducting gap in the cuprate superconductor \( Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta} \)

R. Khasanov,\(^1\) S. Strässle,\(^1\) K. Conder,\(^2\) E. Pomjakushina,\(^2,^3\) A. Bussmann-Holder,\(^4\) and H. Keller\(^1\)

\(^1\)Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland
\(^2\)Laboratory for Developments and Methods, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
\(^3\)Laboratory for Neutron Scattering, Paul Scherrer Institute & ETH Zürich, CH-5232 Villigen PSI, Switzerland
\(^4\)Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

The oxygen-isotope \( ^{16}\text{O}^{18}\text{O} \) effect (OIE) on the zero-temperature superconducting energy gap \( \Delta_0 \) was studied for a series of \( Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta} \) samples \((0.0 \leq x \leq 0.45)\). The OIE on \( \Delta_0 \) was found to scale with the one on the superconducting transition temperature. These experimental results are in quantitative agreement with predictions from a polaronic model for cuprate high-temperature superconductors and rule out approaches based on purely electronic mechanisms.

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Within BCS theory the isotope-effect exponents of the superconducting transition temperature \( T_c \) and of the zero-temperature superconducting energy gap \( \Delta_0 \) are constant and have the same values for both quantities. Strikingly different observations were reported for cuprate high-temperature superconductors (HTS’s), where the isotope effect on \( T_c \) is strongly doping dependent. The isotope-effect exponent is vanishingly small at optimum doping but increases nonlinearly when approaching the underdoped regime \( 0 < x < 0.45 \) where it even exceeds the BCS value of 0.5. Further additional distinctions arise in HTS’s caused by the fact that the superconducting order parameter is complex with a leading \( d \)-wave component in coexistence with a smaller \( s \)-wave one \( \mathbb{R} \mathbb{R} \mathbb{R} \mathbb{R} \mathbb{R} \) as suggested early on \( 10, 11 \). This intricacy does not allow to draw specific conclusions about any isotope effect on the zero-temperature superconducting gap \( \Delta_0 \), especially when considering that a \( d \)-wave gap could be a consequence of electronic effects. If high temperature superconductivity were caused by strong electronic correlations only, no isotope effect on the zero-temperature gap is expected. On the other hand, the coexisting \( s \)-wave gap could originate from electron-lattice interactions and carry an isotope effect. Thus, a methodic investigation of an isotope effect on the complex superconducting gap together with its relation to the one on \( T_c \) admits to conclude about the nature of the pairing mechanism.

In this paper we report the studies of the oxygen-isotope \( ^{16}\text{O}^{18}\text{O} \) effect (OIE) on the zero-temperature superconducting energy gap \( \Delta_0 \) in the cuprate superconductor \( Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta} \). A linear relation between \( \Delta_0 \) and \( T_c \) is found as predicted theoretically. The isotope effect on \( \Delta_0 \) scales linearly with the one on \( T_c \) and reverses sign around optimum doping, as anticipated from model calculations. Different doping levels of the isotope exchanged samples were ruled out by performing careful back exchange experiments.

Polycrystalline samples of \( Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta} \) \((x = 0.0, 0.2, 0.3, 0.45)\) were prepared by standard solid state reaction \( 12 \). In order to obtain fine grains needed for the determination of the magnetic field penetration depth \( \lambda \) by low field magnetization measurements, the ceramic samples were first grounded for approximately 20 minutes in air and then passed through 10 \( \mu \)m sieves. The oxygen-isotope exchange was performed by heating the samples in \( ^{18}\text{O}_2 \) gas. In order to ensure the same thermal history of the substituted \( ^{18}\text{O} \) and not substituted \( ^{16}\text{O} \) samples, both annealings \( (^{16}\text{O}_2 \) and \( ^{18}\text{O}_2 \) gas) were always made simultaneously. The \( ^{18}\text{O} \) content was determined from the weight change and found to correspond to a 90(2)% exchange. The samples containing Pr were \( c \)-axis oriented in a field of 9 T whereas samples with no Pr remained in non-oriented powder form.

AC and DC magnetization experiments were carried through in the temperature range 2–100 K by using Quantum Design Magnetoeters (MPMS and PPMS). The samples with no Pr were studied in DC experiments with a DC field amplitude of 0.5 mT. The oriented samples containing Pr were investigated with the AC field (field amplitude 0.3 mT and field frequency 333 Hz) applied parallel to the \( c \)-axis. The separation of the grains and the absence of weak links was tested by confirming the linear relation between the magnetization and the field at \( T = 10 \) K. The DC field variation ranged from 0.5 mT to 1.5 mT, and the AC fields from 0.1 mT to 1 mT with frequencies between 49 and 599 Hz. The magnetization data were corrected by subtracting the paramagnetic background.

The Meissner fractions \( f \) were obtained from the measured magnetization, sample masses, and their X-ray density inferring a spherical grain shape. In the inset of Fig. 1(a) \( f(T) \) is shown for the \( c \)-axis aligned \( ^{16}\text{O} \) and \( ^{18}\text{O} \) samples with \( x = 0.2 \). Since the Meissner fraction is substantially smaller than 1 it is ensured that the average size of the grains is comparable to \( \lambda \). This strong reduction in \( f \) is a consequence of the surface-field penetration in each individual grain. In addition, in the whole temperature range \( f \) is systematically larger in the \( ^{16}\text{O} \) samples than in the \( ^{18}\text{O} \) samples confirm-
penetration depth ($\lambda$) samples provide direct information on the penetration depth through the relation $\lambda_{ab} = 2\pi n_{ab} f(T)$ [18]. In order to account for the undetermined average grain size, the values of $\lambda_{ab}^2 (2\,K)$ for the $^{16}$O samples were normalized to those obtained from muon-spin rotation ($\mu$SR) experiments [19].

The experimental results for the in-plane magnetic field penetration depth $\lambda_{ab}$ for $^{16}$O/$^{18}$O substituted Y$_{1-x}$Pr$_{x}$Ba$_2$Cu$_3$O$_{y-\delta}$ with $x = 0.0, 0.2, 0.3, 0.45$ are shown in Fig. 1(a). In order to guarantee that for the $^{16}$O and $^{18}$O substituted samples the doping level remains the same, back-exchange experiments were performed for two representative compositions [see Fig. 1(b)]. It is important to note here, that these back-exchange experiments are absolutely essential since they guarantee that any compositional deviations or preparation errors are excluded. Only in this way real isotope effects can be observed in contrast to marginal ones caused by different doping levels [20].

The analysis of the data presented in Fig. 1(a) was made within the BCS scheme, extended to account for a $d$-wave gap and using tabulated values for the temperature dependence of the normalized gap (see Ref. 6). For $T > 10\,K$ all experimental data are well described by this approach with a nearly linear temperature dependence up to $T \approx 0.5T_c$. Below $10\,K$ the experimental data saturate in contrast to the calculated curves which continue to increase. This deviation between experiment and theory can be a consequence of impurity scattering [21], chemical and structural defects, or may be due to the above mentioned simplifying assumption that the gap is of $d$-wave symmetry only. From the data neither of these sources can be identified unambiguously. The values of $T_c$ and $\Delta_0$ obtained from the analysis of $\lambda_{ab}^2 (T)$ data are summarized in Table I. The relative isotope shifts of $T_c$ and $\Delta_0$ were determined from their relative percentage change with isotope substitution. The values of $\delta T_c/T_c$, and $\delta\Delta_0/\Delta_0 (\delta X/X = [^{18}X - ^{16}X] / ^{16}X, X = T_c$ or $\Delta_0$), corrected for the incomplete isotope exchange in the $^{18}$O samples, are summarized in Table I. The results for the OIE on $T_c$ are in accord with already published data [1, 2, 12, 13, 14, 15].

In Fig. 2 the values of the zero-temperature gaps $\Delta_0$ are shown as a function of their corresponding values of $T_c$. In order to demonstrate their consistency with earlier reported gap values, data for various HTS’s obtained by different methods [6, 7, 9] are included together with theoretical results discussed below [22, 24, 25]. It should be emphasized that the data presented in Fig. 2 follow the general trend, namely, $\Delta_0$ scales rather linearly with $T_c$. The linear relation between $\Delta_0$ and $T_c$ is absolutely nontrivial, since competing and coexisting energy scales dominate the phase diagram of HTS. Especially, the pseudogap which is in many experiments indistinguishable from the superconducting gap, has given rise to statements controversy to our findings, i.e., that $\Delta_0$ increases with decreasing $T_c$ (see e.g. Ref. 24 and references therein). The linear relation between $\Delta_0$ and $T_c$ was pre-
TABLE I: Values of $T_c$ and $\Delta_0$ for the $^{16}\text{O}/^{18}\text{O}$-substituted Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ samples investigated in this work. The relative isotope shifts of these quantities are displayed in the last two columns of the table.

| Sample                  | $T_c$  | $\Delta_0$ | $T_c$  | $\Delta_0$ | $\delta T_c/T_c$ | $\delta \Delta_0/\Delta_0$ |
|-------------------------|--------|------------|--------|------------|------------------|-----------------------------|
| Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ | 93.23(7) | 29.75(22) | 93.01(6) | 30.06(24) | -0.22(11) | 1.1(1.2) |
| Y$_{0.8}$Pr$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$ | 70.02(6) | 19.61(14) | 69.22(8) | 19.33(13) | -1.25(16) | -1.6(1.1) |
| Y$_{0.7}$Pr$_{0.3}$Ba$_2$Cu$_3$O$_{7-\delta}$ | 55.50(8) | 12.28(9) | 54.40(8) | 11.98(11) | -2.16(23) | -2.7(1.3) |
| Y$_{0.55}$Pr$_{0.45}$Ba$_2$Cu$_3$O$_{7-\delta}$ | 33.01(8) | 6.87(5) | 31.20(7) | 6.53(5) | -6.06(37) | -5.5(1.2) |
| Y$_{0.8}$Pr$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$ | 69.80(6) | 19.54(12) | 69.02(7) | 19.19(13) | -1.25(14) | -1.99(1.0) |
| Y$_{0.7}$Pr$_{0.3}$Ba$_2$Cu$_3$O$_{7-\delta}$ | 55.41(8) | 12.21(8) | 54.30(7) | 11.94(8) | -2.21(22) | -2.04(1.0) |

$^a$Results for back-exchanged $^{18}\text{O} \rightarrow ^{16}\text{O}$ samples

$^b$Results for back-exchanged $^{16}\text{O} \rightarrow ^{18}\text{O}$ samples

FIG. 2: (Color online) The zero-temperature superconducting gap $\Delta_0$ vs. the superconducting transition temperature $T_c$ for $^{16}\text{O}/^{18}\text{O}$-substituted Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ samples studied in the present work and for various hole-doped HTS’s studied by means of Andreev reflection (AR) and muon-spin rotation ($\mu$SR). The solid line corresponding to $2\Delta_0/k_BT_c = 5.34$ was predicted theoretically by using a two-component model with polaronic coupling. The dashed line corresponds to the BCS value $2\Delta_0/k_BT_c = 3.52$. The theoretical model considers two components where the doped holes lead to the formation of metallic regions in the otherwise insulating antiferromagnetic matrix (for the same analysis of the data sets of $^{16}\text{O}$ and $^{18}\text{O}$ samples. The OIE on $\Delta_0$ is compared to the one on $T_c$ in Fig. 3, together with theoretically derived results. Interestingly, the same linear relation between both is observed in consistency with a model, where polaronic renormalizations of the single particle energies were introduced. Of fundamental importance is the observation of a sign reversal of the isotope effect around optimum doping as predicted in Refs. 24, 25. This novel discovery provides substantial evidence that polaronic effects control the physics of HTS.

FIG. 3: (Color online) Comparison of the oxygen-isotope shift on the superconducting gap $\Delta_0$ with the one on the transition temperature $T_c$ for Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ ($x = 0.0, 0.2, 0.3, 0.45$). Circles refer to the present experiments, the solid line was obtained from model calculations as described in Refs. 23, 24, 25. The stars refer to the back-exchange data.
The locally strong electron-lattice interaction within the metallic regions causes an $s$–$d$ order parameter, in contrast to the embedding matrix with a $d$–$d$ order parameter. Interband interactions between both subsystems guarantee a single transition temperature together with coupled gaps which were calculated self-consistently. The important effect of the polaron formation is an exponential renormalization of the band width which carries an isotope effect, and an isotope independent level shift. Correspondingly, all hopping integrals are renormalized, but contribute in a very different way to the isotope effect [23]. By using different values for the polaronic coupling the average gap $\Delta_0 = \sqrt{\Delta_1^2 + \Delta_2^2}$ is attained as a function of the corresponding $T_c$ yielding $2\Delta_0/k_BT_c = 5.34$. The results are included in Fig. 2 as straight line. Also the $\mu$SR [4, 7] and Andreev reflection data [9] displayed in Fig. 3 refer to an average gap. The calculated oxygen-isotope shift of the average gap $\Delta_0$ vs. the one on $T_c$ is compared with the present data of $Y_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ in Fig. 3. A good agreement between both, experiment and theory is observed. Here it is worth mentioning that the isotope effect on the individual gaps, i.e., $\Delta_s$, $\Delta_d$, is of the same order of magnitude for both gaps, but always slightly enhanced for the $d$–$d$ wave gap as compared to the $s$–$s$ wave one [22]. It is important to emphasize, that conventional electron-phonon coupling does not lead to this doping dependent isotope effect but would always yield – within weak coupling – an isotope exponent of 0.5. Also, models based on a purely electronic approach cannot capture these effects.

In summary, from measurements of the in-plane magnetic field penetration depth the zero-temperature superconducting energy gaps $\Delta_0$ for $Y_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ ($0.0 \leq x \leq 0.45$) were determined. A nontrivial linear relation between $\Delta_0$ and $T_c$ was found as predicted theoretically. The OIE on the superconducting gap $\Delta_0$ scales linearly with the one on $T_c$ and reverses sign around optimum doping. This sign reversal is in agreement with predictions from model calculations and unexpected from other theories. Different doping levels of the isotope exchanged samples were ruled out by performing careful back-exchange experiments. The experimental results together with their theoretical analysis suggest that unconventional electron-lattice interactions play an important role in the physics of high-temperature superconductivity.

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