Negative Oxygen Isotope Effect on the Static Spin Stripe Order in Superconducting La$_{2-x}$Ba$_x$CuO$_4$($x = 1/8$) Observed by Muon-Spin Rotation

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Large negative oxygen-isotope ($^{16}$O and $^{18}$O) effects (OIEs) on the static spin-stripe-ordering temperature $T_{so}$ and the magnetic volume fraction $V_m$ were observed in La$_{2-x}$Ba$_x$CuO$_4$($x = 1/8$) by means of muon-spin-rotation experiments. The corresponding OIE exponents were found to be $\alpha_{T_m} = -0.57(6)$ and $\alpha_{V_m} = -0.71(9)$, which are sign reversed to $\alpha_c = 0.46(6)$ measured for the superconducting transition temperature $T_c$. This indicates that the electron-lattice interaction is involved in the stripe formation and plays an important role in the competition between bulk superconductivity and static stripe order in the cuprates.

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La$_{2-x}$Ba$_x$CuO$_4$ (LBCO) was the first cuprate system where high-$T_c$ superconductivity was discovered [1]. This compound holds a unique position in the field since the bulk superconducting (SC) transition temperature $T_c$ exhibits a deep minimum at $x = 1/8$ [2], which is known as the 1/8 anomaly [3,4]. At this doping level neutron and X-ray diffraction experiments revealed two-dimensional static charge and spin (stripe) order [5–8]. A central issue in cuprates is the microscopic origin of stripe formation and its relation to superconductivity. Given the fact that the amplitudes of the spin and charge orders as well as the ordering temperatures have maximum values at $x = 1/8$ [8], where $T_c$ is strongly suppressed, one might conclude that stripes and bulk (three-dimensional) superconductivity are incompatible types of order. This conclusion is also supported by high-pressure muon-spin-rotation experiments ($\mu$SR) in La$_{1.575}$Ba$_{0.125}$CuO$_4$ (LBCO-1/8) [9], demonstrating that static stripe order and bulk superconductivity occur in mutually exclusive spatial regions. On the other hand, recent investigations of the relation between superconductivity and stripe order show that the situation is more complex, indicating quasi-two-dimensional superconductivity in LBCO-1/8, coexisting with static stripe order, but with frustrated phase order between the layers [10–14]. The frustrated Josephson coupling was explained in terms of sinusoidally modulated [pair-density-wave (PDW)] SC order as proposed in Ref. [15]. However, at present it is unclear to what extent PDW order is a common feature of cuprate systems where stripe order occurs. While the relevance of stripe correlations for high-temperature superconductivity remains a subject of controversy, the collected experimental data indicate that the tendency toward uni-directional striplike ordering is common to cuprates [3,4,16]. Exploring the mechanism of stripe formation will help to clarify its role for the occurrence of high-temperature superconductivity in the cuprates. The stripe phase may be caused by a purely electronic and/or electron-lattice interaction. There is increasing experimental evidence for a strong electron-lattice interaction to be essential in the cuprates (see, e.g.,[17–20]). However, it is not clear whether this interaction is involved in the formation of the stripe phase.

Isotope effect experiments played a crucial role for understanding superconductivity, since for conventional superconductors they clearly demonstrated that the electron-phonon interaction is responsible for the electron pairing [21,22]. In the cuprate high-temperature superconductors (HTSs) unconventional oxygen isotope ($^{16}$O and $^{18}$O) effects (OIEs) on various quantities were observed, such as the superconducting transition temperature $T_c$, the SC energy gap $\Delta(0)$, the magnetic penetration depth $\lambda(0)$, the Néel temperature $T_N$, the spin glass transition temperature $T_g$, and the pseudogap onset temperature $T^*$ [17,18,23–26]. So far, a large OIE on $T_c$ was observed in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ [27] and La$_{1.8-x}$Eu$_{0.2}$Sr$_2$CuO$_4$ [28] showing stripe order at $x = 1/8$ [29,30]. However, no OIE investigation on the charge and spin order in the stripe phase of cuprates has been reported.

In this Letter we present OIE investigations of the static spin-stripe order in LBCO-1/8 by means of $\mu$SR experiments. The main reason to use LBCO-1/8 here, instead of La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ [27] or La$_{1.8-x}$Eu$_{0.2}$Sr$_2$CuO$_4$ [28], is to avoid a strong magnetic response of the Eu and Nd 4f moments in the $\mu$SR signal, which does not allow a reliable OIE study of the spin-stripe phase in these systems. In this work substantial OIEs were found on magnetic quantities characterizing the static spin-stripe phase in LBCO-1/8.
demonstrating that the electron-lattice interaction is essential in the stripe formation mechanism of cuprates. In addition, we also studied the OIE on $T_c$ in LBCO-1/8 by magnetization measurements. Remarkably, it was found that the OIEs have opposite signs for the magnetic and superconducting states in the stripe phase of LBCO-1/8. These findings reveal that lattice vibrations play an important role in the competition between superconductivity and static spin-stripe order in LBCO-1/8.

A polycrystalline sample of La$_{2-x}$Ba$_x$CuO$_4$ with $x = 1/8$ was prepared by the conventional solid-state reaction method using La$_2$O$_3$, BaCO$_3$, and CuO. The single-phase character of the sample was checked by powder x-ray diffraction. All the measurements were performed on samples from the same batch. For the oxygen isotope exchange the sample was divided into two parts. To ensure that the substituted (18)O and not substituted (16)O samples were subject of the same thermal history, both parts were annealed simultaneously in separate chambers (in 16O$_2$ and 18O$_2$ gas, respectively) under exactly the same conditions. The oxygen isotope enrichment of the samples was determined in situ using mass spectrometry. The 18O enriched samples contain $\approx$82% 18O and $\approx$18% 16O. In a first step the OIE on the superconducting transition temperature $T_c$ was determined by magnetization experiments performed with a SQUID magnetometer (Quantum Design MPMS-XL) in a field of 0.5 mT. The temperature dependence of the zero-field-cooled (ZFC) diamagnetic moment $m_{ZFC}$ for the 16O, 18O, and back-exchanged (18O $\rightarrow$ 16O) samples of LBCO-1/8 is shown in Fig. 1. The diamagnetic moment exhibits a two-step SC transition in all samples, similar to our previous work [9]. The first transition appears at $T_{c1} \approx 30$ K and the second transition at $T_{c2} \approx 5$ K with a larger diamagnetic response. Detailed investigations performed on single crystalline samples of LBCO-1/8 provided an explanation for this two-step SC transition [11]. The authors interpreted the transition at $T_{c1}$ as due to the development of 2D superconductivity in the CuO$_2$ planes, while the interlayer Josephson coupling is frustrated by static stripes. A transition to a 3D SC phase takes place at a much lower temperature $T_{c2} \ll T_{c1}$. The values of $T_{c1}$ and $T_{c2}$ were defined as the temperatures where the linearly extrapolated magnetic moments intersect the zero line (see Fig. 1). Both $T_{c1}$ and $T_{c2}$ decrease by $\approx$1.4 and $\approx$1.2 K, respectively, upon replacing 16O with 18O. To ensure that the observed changes of $T_{c1}$ and $T_{c2}$ are indeed due to isotope substitution, magnetization measurements were also carried out on a back-exchanged (18O $\rightarrow$ 16O) sample (see Fig. 1). Note that the OIE on $T_{c1}$ is very well reproducible (inset of Fig. 1). However, at low temperatures $m_{ZFC}(T)$ for the back-exchanged sample does not follow the one for the 16O sample. This is due to the fact that the SC transition at $T_{c2}$ is extremely sensitive to the thermal history (oxygen annealing time) of the samples, which is about a factor of 2 longer for the back-exchanged sample. Therefore, we only discuss the OIE on $T_{c1}$ further.

The following values for the OIE on $T_{c1}$ were found: $T_{c1} = 29.7(1)$ K, $T_{c1} = 28.3(1)$ K, $\Delta T_{c1} = 18^\circ T_{c1} - 16^\circ T_{c1} = -1.4(2)$ K, and for the OIE exponent $\alpha_{T_{c1}} = -d\ln T_{c1}/d\ln M_0 = 0.46(6)$ ($M_0$ is the oxygen isotope mass). Note that this value is comparable to that found for La$_{2-x}$Ba$_x$CuO$_4$ ($\alpha = 0.10 - 0.15$) [31], but is much smaller than $\alpha_{T_{c1}} = 1.89$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ ($x = 1/8$) [27] and $\alpha_{T_{c1}} = 1.09$ for La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_4$ ($x = 0.16$) [28].

Finally, the OIE on the static spin-stripe order in LBCO-1/8 was studied by means of zero-field (ZF) and transverse-field (TF) $\mu$SR experiments. In a $\mu$SR experiment positive muons implanted into a sample serve as an extremely sensitive local probe to detect small internal magnetic fields and ordered magnetic volume fractions in the bulk of magnetic materials. Note that the appearance of static magnetic order below $\approx 30$ K in LBCO-1/8 was originally observed by $\mu$SR [32]. The $\mu$SR experiments were carried out at the πM3 beam line at the Paul Scherrer Institute (Switzerland) using the general purpose instrument (GPS) with a standard veto setup providing a low-background $\mu$SR signal. The $\mu$SR time spectra were analyzed using the free software package MUSRFIT [33].

Figure 2 shows the TF$\mu$SR asymmetry $A$ (normalized to its maximum value $A_0$), extracted from the $\mu$SR spectra following the procedure given in Ref. [34], as a function of temperature for the 16O, 18O, and back-exchanged (18O $\rightarrow$ 16O) samples of LBCO-1/8 in an applied field of $\mu_0H = 3$ mT. Above 40 K, $A$ saturates at a maximum value for both 16O and 18O, indicating that the whole sample is in the paramagnetic state, and all the muon spins precess in the applied magnetic field. Below 40 K, $A$ decreases with decreasing temperature and reaches an almost constant value at low temperatures. The reduction of $A$ signals the appearance of magnetic order in the spin-stripe phase,
This demonstrates that the observed negative OIE on phenomenological function \(I\), is intrinsic. Note that in order to exclude any doping differences in the oxygen-towards higher temperatures as compared to one for the data by means of Eq. (1).

where the muon spins experience a local magnetic field larger than the applied magnetic field. As a result, the fraction of muons in the paramagnetic state decreases. Note that \(A(T)\) for the \(^{18}\)O sample is systematically shifted towards higher temperatures as compared to one for the \(^{16}\)O sample, indicating that the static spin-stripe-ordering temperature \(\Delta T_{SO}\) for \(^{18}\)O is higher than \(\Delta T_{SO}\) for \(^{16}\)O. The values of \(\Delta T_{SO}\) and \(\Delta T_{SO}\) were determined by using the phenomenological function [24],

\[
A(T)/A_0 = a \left[ 1 - \frac{1}{\exp[(T - T_{SO})/\Delta T_{SO}] + 1} \right] + b, \quad (1)
\]

where \(\Delta T_{SO}\) is the width of the transition, and \(a\) and \(b\) are empirical parameters. Analyzing the data in Fig. 2 with Eq. (1) yields \(\Delta T_{SO} = 32.9(3)\) K and \(\Delta T_{SO} = 34.8(2)\) K with a large negative OIE exponent \(\alpha_{t_{SO}} = -0.56(9)\). A back exchange experiment \((^{18}\mathrm{O} \rightarrow ^{16}\mathrm{O})\) was carried out in order to exclude any doping differences in the oxygen-isotope exchanged samples. As shown in Fig. 2 the oxygen back-exchanged sample of LBCO-1/8 exhibits within experimental error almost the same \(A(T)\) as the \(^{18}\)O sample. This demonstrates that the observed negative OIE on \(T_{SO}\) is intrinsic. Note that that \(\alpha_{t_{SO}} = -0.56(6)\) and \(\alpha_{t_{SO}} = 0.46(6)\) have almost the same magnitude, but sign reversed.

In order to explore the OIE on the magnetic volume fraction \(V_m\) as well as on \(T_{SO}\), ZF \(\mu\)SR experiments (no external magnetic field applied) were carried out. Figure 3 shows representative ZF \(\mu\)SR time spectra for the \(^{16}\)O and \(^{18}\)O samples of LBCO-1/8. Below \(T \approx 30\) K damped oscillations due to the presence of a local magnetic field at the muon site are observed, indicating long range static spin-stripe order [32,35]. The \(\mu\)SR signals in the whole temperature range were analyzed by decomposing the signal into a magnetic and a nonmagnetic contribution [35]:

\[
P(t) = V_m \left[ \frac{2}{3} e^{-\lambda_I t} J_0(\gamma_p B_p t) + \frac{1}{3} e^{-\lambda_L t} \right] + (1 - V_m) e^{-\lambda_{nm} t}.
\]

Here, \(P(t)\) is the muon spin polarization function, \(V_m\) denotes the relative magnetic volume fraction, and \(\gamma_p/(2\pi) = 135.5\) MHz/T is the muon gyromagnetic ratio. \(B_p\) is the average internal magnetic field at the muon site. \(\lambda_I\) and \(\lambda_L\) are the depolarization rates representing the transversal and the longitudinal relaxing components related to the spin-stripe-ordered regions of the sample, respectively. \(J_0\) is the zeroth-order Bessel function of the first kind. This is characteristic for an incommensurate spin-density wave and has been observed in cuprates with static spin-stripe order [35]. \(\lambda_{nm}\) is the relaxation rate related to the nonmagnetic part of the sample, where spin-stripe order is absent.

The temperature dependence of the average internal magnetic field \(B_{\mu}\) for the \(^{16}\)O, \(^{18}\)O, and back-exchanged samples of LBCO-1/8 is shown in Fig. 4(a). It is evident that in the \(^{18}\)O sample \(B_{\mu}\) appears at a higher temperature than in the \(^{16}\)O sample, showing that \(\Delta T_{SO}\) is higher than \(\Delta T_{SO}\). The solid curves in Fig. 4(a) are fits of the data to the power law \(B_{\mu}(T) = B_{\mu}(0)(1 - (T/T_{SO}))^\delta\), where \(B_{\mu}(0)\) is the zero-temperature value of \(B_{\mu}\). \(\gamma\) and \(\delta\) are phenomenological exponents. The analysis yields \(\Delta T_{SO} = 30.1(3)\) K, \(18\Delta T_{SO} = 31.8(3)\) K, and the OIE exponent of \(T_{SO}\) obtained from \(B_{\mu}(T) = \alpha T_{SO} = -0.55(11)\).

\(\mu\)SR also allows us to determine the magnetic volume fraction \(V_m\) in magnetically ordered materials. Figure 4(b) shows the temperature dependence of \(V_m\) for the \(^{16}\)O and \(^{18}\)O samples. The solid lines in Fig. 4(b) are fits of the data to the same empirical power law as used for \(B_{\mu}(T)\) discussed above. The OIE exponent of \(T_{SO}\) obtained from
$V_m(T)$ is $\alpha_{T_m} = -0.61(7)$, in excellent agreement with $\alpha_{T_{SO}} = -0.55(11)$ and $\alpha_{T_{SO}} = -0.56(9)$ obtained from the temperature dependence of the $\mu$SR parameters $B_\mu$ and $A$, respectively. This demonstrates that the two independent $\mu$SR experiments, TF and ZF $\mu$SR, give consistent results for $\alpha_{T_{SO}}$, although the values of $T_{SO}$ are systematically different (see Table I) [36]. For further discussions we use the average value $\langle \alpha_{T_{SO}} \rangle = \alpha_{T_{SO}} = -0.57(6)$ determined from the three measured values. It is also clear from Fig. 4(b) that $V_m$ in the $^{18}$O sample is significantly larger than in the $^{16}$O sample in the whole temperature range, indicating a higher volume fraction of the static stripe order phase in the $^{18}$O sample. The zero-temperature values of the magnetic volume fraction were found to be $^{16}V_m(0) = 0.82(1)$ and $^{18}V_m(0) = 0.88(1)$, yielding an OIE exponent of $\alpha_{V_m} = -d\ln V_m/d\ln M_0 = -0.71(9)$. As shown in Fig. 4(b) the intrinsic OIE on $V_m(0)$ was confirmed by back-exchange ($^{18}$O $\rightarrow$ $^{16}$O) experiments. The obtained results show that the quantities $T_{SO}$ and $V_m(0)$ characterizing the static stripe state exhibit a large and negative OIE. To our knowledge this is the first study reporting a substantial OIE on the static stripe order state in a LBCO-1/8-doped cuprate.

The values of $T_{SO}$ and $V_m(0)$ related to the static stripe phase of $^{16}$O and $^{18}$O exchanged LBCO-1/8 obtained in this work as well as the corresponding OIE exponents are summarized in Table I. The average value of the static stripe order temperature $T_{SO} \approx 33$ K is in agreement with the previous values $T_{SO} \approx 30$–$34$ K obtained from $\mu$SR [32,35] and comparable to the value of the superconducting transition temperature $T_c \approx 30$ K. However, the value of $T_{SO}$ determined by $\mu$SR is smaller than $T_{SO} \approx 40$ K determined by neutron scattering [11] due to the different time window of the two techniques. One should point out that the values of $T_{SO}$ and $V_m(0)$ increase with increasing oxygen-isotope mass (Figs. 2 and 4), whereas $T_c$ decreases (Fig. 1). This demonstrates a competition between bulk superconductivity and static stripe order in LBCO-1/8, and that the electron-lattice coupling is involved in this competition.

In conclusion, oxygen isotope effects on magnetic and superconducting quantities related to the static stripe phase of LBCO-1/8 were investigated by means of $\mu$SR and magnetization experiments. The static stripe order temperature $T_{SO}$ and the magnetic volume fraction $V_m(0)$ exhibit a large negative OIE which is novel and unexpected. Furthermore, the observed oxygen-isotope shifts of the superconducting transition temperature $T_c$ and the spin-ordering temperature $T_{SO}$ have almost the same magnitude, but opposite signs. This provides clear evidence that bulk superconductivity and static spin-order are competitive phenomena in the stripe phase of LBCO-1/8, and that the electron-lattice interaction is a crucial factor controlling this competition. At present the role of the electron-lattice coupling for stripe formation is not known. Further experiments are needed to clarify this point. Our results may contribute to a better understanding of the complex microscopic mechanism of stripe formation and of high-temperature superconductivity in the cuprates in general.

**TABLE I.** The values of $T_{SO}$, $\Delta T_{SO} = 18T_{SO} - 16T_{SO}$, $V_m(0)$, and $\Delta V_m(0) = 18V_m(0) - 16V_m(0)$ of the $^{16}$O and $^{18}$O samples of LBCO-1/8 determined from various measured $\mu$SR parameters. The OIE exponents $\alpha_{T_{SO}}$ and $\alpha_{V_m}$ are corrected for the incomplete $^{18}$O exchange of $82(5)$%.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$^{16}T_{SO}$</th>
<th>$^{18}T_{SO}$</th>
<th>$\Delta T_{SO}$</th>
<th>$\alpha_{T_{SO}}$</th>
<th>$^{16}V_m(0)$</th>
<th>$^{18}V_m(0)$</th>
<th>$\Delta V_m(0)$</th>
<th>$\alpha_{V_m}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A(T)$</td>
<td>32.9(3)</td>
<td>34.8(2)</td>
<td>1.9(4)</td>
<td>$-0.56(9)$</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>$B_\mu(T)$</td>
<td>30.1(3)</td>
<td>31.8(3)</td>
<td>1.7(5)</td>
<td>$-0.55(11)$</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>$V_m(T)$</td>
<td>35.2(2)</td>
<td>37.4(2)</td>
<td>2.2(3)</td>
<td>$-0.61(7)$</td>
<td>0.82(1)</td>
<td>0.88(1)</td>
<td>0.06(1)</td>
<td>$-0.71(9)$</td>
</tr>
</tbody>
</table>

As shown in Fig. 4(b) the intrinsic OIE on $V_m(0)$ was confirmed by back-exchange ($^{18}$O $\rightarrow$ $^{16}$O) experiments. The obtained results show that the quantities $T_{SO}$ and $V_m(0)$ characterizing the static stripe state exhibit a large and negative OIE. To our knowledge this is the first study reporting a substantial OIE on the static stripe order state in a LBCO-1/8-doped cuprate.
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[36] The reason for this might be that close to the static stripe phase-transition magnetic fluctuations give rise to a depolarized μSR signal with a nonzero value for the magnetic fraction. In the static spin-stripe-ordered phase a well-defined field is sensed by the muon spin.