Negative Oxygen Isotope Effect on the Static Spin Stripe Order in Superconducting $La_{2-x}Ba_xCuO_4(x = 1/8)$ 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 $La_{2-x}Ba_xCuO_4(x = 1/8)$ by means of muon-spin-rotation experiments. The corresponding OIE exponents were found to be $\alpha_{T_{so}} = -0.57(6)$ and $\alpha_{V_m} = -0.71(9)$, which are sign reversed to $\alpha_{T_c} = 0.46(6)$ 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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$La_{2-x}Ba_xCuO_4$ (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 (μSR) in $La_{1.875}Ba_{0.125}CuO_4$ (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 stripelike 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 $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ [27] and $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ [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 μSR experiments. The main reason to use LBCO-1/8 here, instead of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ [27] or $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ [28], is to avoid a strong magnetic response of the Eu and Nd $4f$ moments in the μSR 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 $La_{2-x}Ba_xCuO_4$ with $x = 1/8$ was prepared by the conventional solid-state reaction method using La_2O_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 $^{16}O_2$ and $^{18}O_2$ gas, respectively) under exactly the same conditions. The oxygen isotope enrichment of the samples was determined in situ using mass spectrometry. The ^{18}O enriched samples contain $\simeq 82\%$ ^{18}O and $\simeq 18\%$ ^{16}O.

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 ^{16}O, ^{18}O, and back-exchanged ($^{18}O \rightarrow {}^{16}O$) 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 $T_{c1} \simeq 30$ K and the second transition at $T_{c2} \simeq 5$ 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 $\simeq 1.4$ and $\simeq 1.2$ K, respectively, upon replacing ^{16}O with ^{18}O. 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 ($^{18}O \rightarrow {}^{16}O$) 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 ^{16}O 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

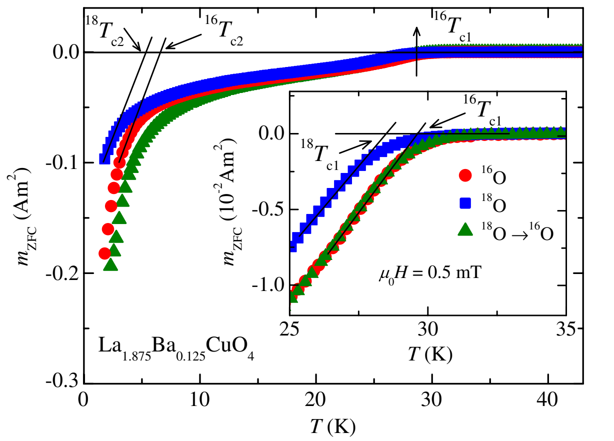

FIG. 1 (color online). Temperature dependence of the diamagnetic moment m_{ZFC} for the ^{16}O, ^{18}O, and back-exchanged ($^{18}O \rightarrow {}^{16}O$) samples of LBCO-1/8. The arrows denote the superconducting transition temperatures T_{c1} and T_{c2} (see text for an explanation). The inset shows the SC transition near T_{c1}.

sample. Therefore, we only discuss the OIE on T_{c1} further. The following values for the OIE on T_{c1} were found: $^{16}T_{c1} = 29.7(1)$ K, $^{18}T_{c1} = 28.3(1)$ K, $\Delta T_{c1} = {}^{18}T_{c1} - {}^{16}T_{c1} = -1.4(2)$ 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 $La_{2-x}Ba_xCuO_4 (x = 0.10 - 0.15)$ [31], but is much smaller than $\alpha_{T_c} \simeq 1.89$ for $La_{1.6-x}Nd_{0.4}Sr_xCuO_4 (x = 1/8)$ [27] and $\alpha_{T_c} \simeq 1.09$ for $La_{1.8-x}Eu_{0.2}Sr_xCuO_4 (x = 0.16)$ [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) μSR experiments. In a μSR 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 $\simeq 30$ K in LBCO-1/8 was originally observed by μSR [32]. The μSR 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 μSR signal. The μSR time spectra were analyzed using the free software package MUSRFIT [33].

Figure 2 shows the TF μSR asymmetry A (normalized to its maximum value A_0), extracted from the μSR spectra following the procedure given in Ref. [34], as a function of temperature for the ^{16}O, ^{18}O, and back-exchanged ($^{18}O \rightarrow {}^{16}O$) samples of LBCO-1/8 in an applied field of $\mu_0 H = 3$ mT. Above 40 K, A saturates at a maximum value for both ^{16}O and ^{18}O, 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,

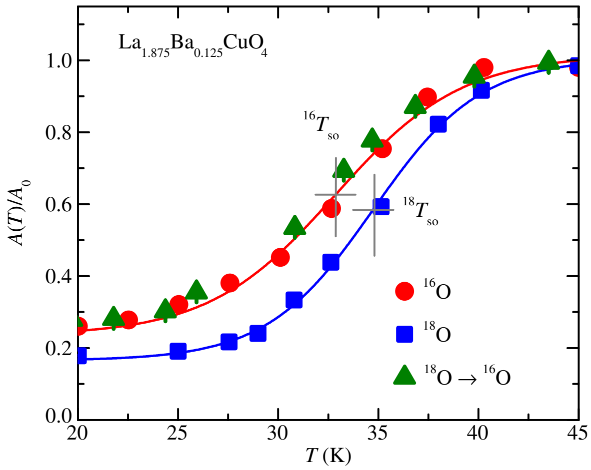

FIG. 2 (color online). The normalized TF asymmetry A/A_0 plotted as a function of temperature for the ^{16}O, ^{18}O, and back-exchanged (^{18}O → ^{16}O) samples of LBCO-1/8. The crosses mark the spin-stripe order temperatures $^{16}T_{SO}$ and $^{18}T_{SO}$ for the ^{16}O and ^{18}O sample, respectively. The solid lines represent fits to 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 $^{18}T_{SO}$ for ^{18}O is higher than $^{16}T_{SO}$ for ^{16}O. The values of $^{16}T_{SO}$ and $^{18}T_{SO}$ 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 ΔT_{SO} is the width of the transition, and a and b are empirical parameters. Analyzing the data in Fig. 2 with Eq. (1) yields $^{16}T_{SO} = 32.9(3)$ K and $^{18}T_{SO} = 34.8(2)$ K with a large negative OIE exponent $\alpha_{T_{so}} = -0.56(9)$. A back exchange experiment (^{18}O → ^{16}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 ^{16}O sample. This demonstrates that the observed negative OIE on T_{so} is intrinsic. Note that $\alpha_{T_{SO}} = -0.56(6)$ and $\alpha_{T_{c1}} = 0.46(6)$ 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 μSR experiments (no external magnetic field applied) were carried out. Figure 3 shows representative ZF μSR 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 μSR signals in the whole

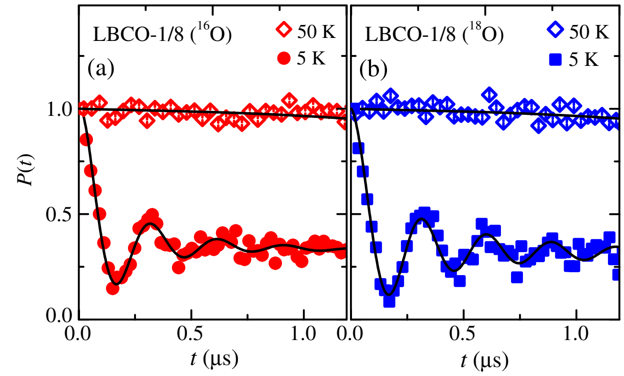

FIG. 3 (color online). ZF μSR signal $P(t)$ for the ^{16}O (a) and ^{18}O (b) samples of LBCO-1/8 recorded at $T = 5$ K and $T = 50$ K. The solid lines represent fits to the data by means of Eq. (2).

temperature range were analyzed by decomposing the signal into a magnetic and a nonmagnetic contribution [35]:

$$P(t) = V_m\left[\frac{2}{3}e^{-\lambda_T t}J_0(\gamma_\mu B_\mu t) + \frac{1}{3}e^{-\lambda_L t}\right] + (1 - V_m)e^{-\lambda_{nm} t}. \quad (2)$$

Here, $P(t)$ is the muon spin polarization function. V_m denotes the relative magnetic volume fraction, and $\gamma_\mu/(2\pi) \simeq 135.5$ MHz/T is the muon gyromagnetic ratio. B_μ is the average internal magnetic field at the muon site. λ_T and λ_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]. λ_{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_μ 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_μ appears at a higher temperature than in the ^{16}O sample, showing that $^{18}T_{SO}$ is higher than $^{16}T_{SO}$. The solid curves in Fig. 4(a) are fits of the data to the power law $B_\mu(T) = B_\mu(0)[1 - (T/T_{so})^\gamma]^\delta$, where $B_\mu(0)$ is the zero-temperature value of B_μ. γ and δ are phenomenological exponents. The analysis yields $^{18}T_{SO} = 30.1(3)$ K, $^{18}T_{SO} = 31.8(3)$ K, and the OIE exponent of T_{SO} obtained from $B_\mu(T)$ is $\alpha_{T_{SO}} = -0.55(11)$.

μSR 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

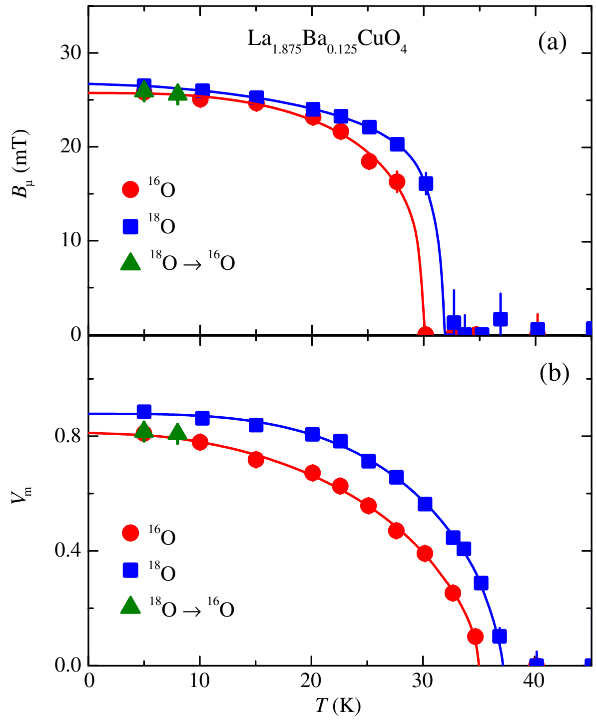

FIG. 4 (color online). (a) Temperature dependence of the average internal magnetic field B_μ at the muon site for ^{16}O, ^{18}O, and back-exchanged ($^{18}O \rightarrow {}^{16}O$) samples of LBCO-1/8. The solid lines represent fits of the data to the power law described in the text. (b) The temperature dependence of the magnetic volume fraction V_m for ^{16}O, ^{18}O, and back-exchanged ($^{18}O \rightarrow {}^{16}O$) samples of LBCO-1/8. The solid lines are fits of the data to the same empirical power law as used for $B_\mu(T)$ in (a).

$V_m(T)$ is $\alpha_{T_{so}} = -0.61(7)$, in excellent agreement with $\alpha_{T_{SO}} = -0.55(11)$ and $\alpha_{T_{SO}} = -0.56(9)$ obtained from the temperature dependence of the μSR parameters B_μ and A, respectively. This demonstrates that the two independent μSR experiments, TF and ZF μSR, 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 spin-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 spin-stripe state exhibit a large and negative OIE. To our knowledge this is the first study reporting a substantial OIE on the static spin-stripe order state in a LBCO-1/8-doped cuprate.

The values of T_{so} and $V_m(0)$ related to the static spin-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 spin-stripe order temperature $T_{SO} \simeq 33$ K is in agreement with the previous values $T_{SO} \simeq 30$–34 K obtained from μSR [32,35] and comparable to the value of the superconducting transition temperature $T_{c1} \simeq 30$ K. However, the value of T_{SO} determined by μSR is smaller than $T_{SO} \simeq 40$ 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_{c1} decreases (Fig. 1). This demonstrates a competition between bulk superconductivity and static spin-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 μSR and magnetization experiments. The static spin-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_{c1} 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}, $\Delta T_{SO} = {}^{18}T_{SO} - {}^{16}T_{SO}$, $V_m(0)$, and $\Delta V_m(0) = {}^{18}V_m(0) - {}^{16}V_m(0)$ of the ^{16}O and ^{18}O samples of LBCO-1/8 determined from various measured μSR parameters. The OIE exponents $\alpha_{T_{SO}}$ and α_{V_m} are corrected for the incomplete ^{18}O exchange of 82(5)%.

Parameter	$^{16}T_{SO}$	$^{18}T_{SO}$	ΔT_{SO}	$\alpha_{T_{SO}}$	$^{16}V_m(0)$	$^{18}V_m(0)$	$\Delta V_m(0)$	α_{V_m}
$A(T)$	32.9(3)	34.8(2)	1.9(4)	−0.56(9)	…	…	…	…
$B_\mu(T)$	30.1(3)	31.8(3)	1.7(5)	−0.55(11)	…	…	…	…
$V_m(T)$	35.2(2)	37.4(2)	2.2(3)	−0.61(7)	0.82(1)	0.88(1)	0.06(1)	-0.71(9)

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