Oxygen isotope effect on the superconductivity and stripe phase in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$

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The oxygen isotope effect on the superconductivity, stripe phase, and structure transition is systematically investigated in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with static stripe phase. Substitution of ^{16}O by ^{18}O leads to a decrease in superconducting transition temperature T_C while enhancing the temperature of the structural transition from low-temperature-orthorhombic phase to low-temperature-tetragonal phase. Compared to the Nd-free sample, a larger isotope effect on T_C is observed in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. These results indicate that the distortion of CuO_2 plane suppresses the superconductivity, giving a direct evidence of the competition of stripe phase and superconductivity because the distortion of CuO_2 plane enhances the stripe phase.

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High critical temperature (T_C) superconductor has been widely studied since the discovery of $La_{2-x}Ba_xCuO_4$ in 1986. However, the properties of these materials are hard to explain by BCS theory, which has been successful in explaining the properties of conventional superconductors. The mechanism of high-temperature superconductivity in cuprates is still an open question. Among numerous theoretical models, stripe phase has attracted considerable attention; according to this model, the spin and charge in high T_C superconductors distribute inhomogeneously and form "stripe."[1–3] It was experimentally observed by neutron scattering or other method in La_2CuO_4-based system[4–12] and $YBa_2Cu_3O_y$.[13–15] It generally appears that the fluctuating stripe promotes superconductivity, but static stripe may suppress superconductivity.[16] However, there is evidence that local magnetic order rather than charge-stripe order is responsible for the anomalous suppression of superconductivity.[17]

In La_2CuO_4 system, several structural phase transitions occur with doping of alkaline-earth and rare-earth metals.[17–22] With decreasing temperature, a transition from high-temperature-tetragonal phase to low-temperature-orthorhombic (LTO) phase, then to low-temperature-tetragonal (LTT) phase (or *Pccn* phase, depending on the hole concentration) was observed. The LTO and LTT phases involve distortion of CuO_2 planes due to the tilting of CuO_6 octahedras, producing stripe pinning potential. Here, we call the temperature T_{LT} at which the structure transition from LTO to LTT occurs. In $La_{2-x-y}Nd_ySr_xCuO_4$, the substitution of La by Nd enhances the pinning potential, which pins the stripe from fluctuating (Nd-free sample) to static.[7,8] When $x=1/8$, there is an anomalous suppression of superconductivity due to the stripe phase. In $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, neutron-diffraction experiment shows that the charge ordering and structural transition are essentially coincidental for $x=0.10$ and 0.12,[8,17] however, the spin ordering occurs significantly below the structural transition temperature T_{LT}.[17] Several groups have investigated the relationship among structural distortion, stripe phase, and superconductivity, using high pressure to control the structure transition and superconductivity.[22,23] It was found that the hydrostatic pressure lower than 5 GPa compresses the CuO_2 planes, which weakens the pinning potential, suppresses the LTT distortion, and enhances the superconductivity.

It is well known that the isotope effect study is very important in conventional superconductors, in which $\alpha_C = -d\ln T_C/d\ln M_O = 0.5$, and is the illation of BCS theory. Although many believe that antiferromagnetism is important for superconductivity, there has been renewed interest in the possible role of electron-lattice coupling.[24,25] Therefore, the research on isotope substitution is necessary to study the mechanism of high-T_C superconductivity. Several oxygen isotope substitution experiments have been done in $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Ba_xCuO_4$ with fluctuating stripe phase, and a large isotope exponent ($\alpha_C \sim 1$) on T_C was found near 1/8 doping.[26–29] To check a possible change in the isotope effect induced by Nd doping and investigate the relationship among structural distortion, stripe phase, and superconductivity, we systematically study the oxygen isotope effect in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with $x=0.10$, 0.125, 0.15, and 0.175. It is found that T_C is suppressed, while T_{LT} is enhanced with the substitution of ^{16}O by ^{18}O, indicating that the distortion of CuO_2 plane suppresses superconductivity. Because the charge ordering and structural transition are essentially coincidental for $x=0.10$ and 0.12, therefore, in this sense the results of isotope effect definitely provide an evidence for the competition between stripe phase and superconductivity. Compared to the Nd-free sample with fluctuating stripe phase, a larger isotope exponent α_C is observed in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with static stripe phase, suggesting a strong electron-lattice coupling in cuprates.

Polycrystalline samples $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ for $x=0.10$, 0.125, 0.15, and 0.175 were prepared by conventional solid-state reaction. All samples were characterized by x-ray diffraction and no observable impurity phase is found. One pellet for each sample with different x was cut into two pieces for oxygen isotope diffusion. The two pieces of each composition were put into an alumina boat, which were sealed in a quartz tube filled with oxygen pressure of 1 bar (one for $^{16}O_2$ and another for $^{18}O_2$) mounted in a furnace. The quartz tubes formed parts of two identical closed loops. They were first heated at 980 °C for 90 h, then slowly cooled to 500 °C, kept for 10 h, and finally cooled to room temperature with furnace. The obtained samples were reexamined by

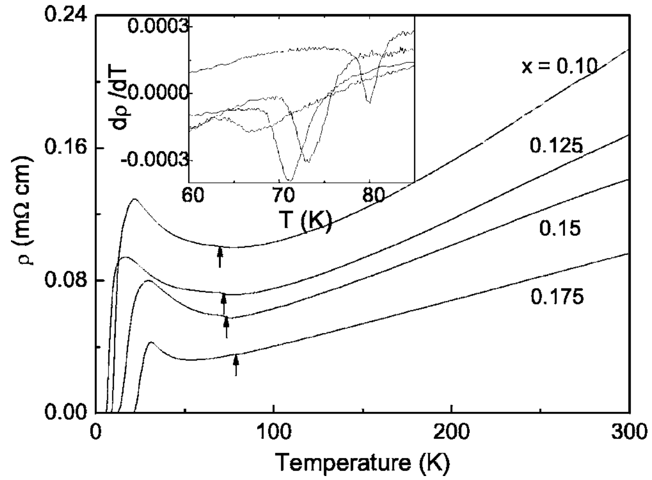

FIG. 1. Temperature dependence of resistivity for the samples $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with $x=0.10$, 0.125, 0.15, and 0.175 treated in ^{16}O. The arrows indicate the LTO-LTT transition temperature T_{LT}. Inset shows the derivative curves of resistivity near T_{LT}.

x-ray diffraction to confirm their single phase. The oxygen isotope enrichment is determined by the weight changes of both ^{16}O and ^{18}O samples. The ^{18}O samples have about $80\%(\pm5\%)$ ^{18}O and $20\%(\pm5\%)$ ^{16}O. To ensure the isotope exchange effect, back exchange of ^{18}O sample by ^{16}O was carried out in the same way and the weight change showed a complete back exchange. Resistance measurements were performed using the ac four-probe method with an ac resistance bridge system (Linear Research Inc., LR-700P). To reduce the experimental deviation, each couple of ^{16}O and ^{18}O samples is measured synchronously in a cooling process.

Temperature dependence of resistivity for the samples $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with $x=0.10$, 0.125, 0.15, and 0.175 treated in ^{16}O are shown in Fig. 1. T_C (defined as the midpoint of superconducting transition in resistivity) is 11, 7.9, 17.8, and 25 K for $x=0.10$, 0.125, 0.15, and 0.175, respectively, consistent with that reported in other literatures.[8,17,20,22] The suppression of T_C compared with $La_{2-x}Sr_xCuO_4$ (Ref. 32) is attributed to the static stripe phase induced by the substitution of Nd for La atoms.[7] The abnormal suppression of T_C near 1/8 doping can be clearly seen in our samples; that is, the T_C is least for the sample with $x=0.125$. A small resistivity jump appears at about 70 K, which is indicated by an arrow and regarded as the signal of structural transition from LTO to LTT.[17,19] To show this jump clearly, the temperature dependence of the derivative of resistivity is shown in the inset of Fig. 1. A dip can be seen clearly in the derivative curve. Here we define T_{LT} as the dip temperature: 66.7, 71, 73.1, and 80 K for $x=0.10$, 0.125, 0.15, and 0.175, respectively. T_{LT} increases with increasing Sr doping, consistent with that reported in Ref. 17.

Figure 2 shows the temperature dependence of resistivity near the superconducting transition for the sample with $x=0.125$. It should be pointed out that the superconducting transition is a little broad, which may be caused by the fluctuation of Sr, Nd, and/or O contents. T_C is 7.9 and 6.6 K for ^{16}O and ^{18}O samples, respectively. To ensure the change of T_C from isotope substitution, back exchange of ^{18}O sample

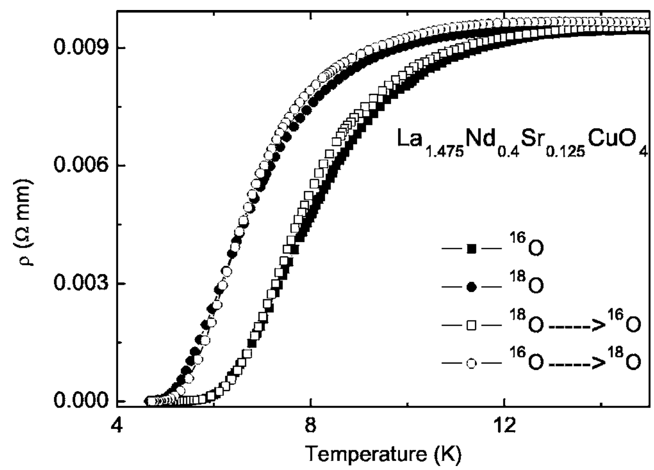

FIG. 2. Temperature dependence of resistivity near the superconducting transition for the ^{16}O and ^{18}O samples with $x=0.125$.

by ^{16}O was performed. Figure 3 shows the Raman spectra at room temperature for the sample with $x=0.125$. The apical O stretch mode is softened from $433(\pm1)$ to $413(\pm1)$ cm^{-1} by the substitution of ^{18}O for ^{16}O. This frequency shift of $4.6\%\pm0.3\%$ suggests about 79% ^{18}O substitution because Raman shift is in proportion to $1-\sqrt{16/M'}$.[30,31] The ^{18}O substitution estimated by Raman shift is consistent with the result obtained from weight change. For comparison, $\rho(T)$ of back-exchanged samples are also shown in Fig. 2. Two $^{16}O/^{18}O$ samples show the same T_C, which definitely indicates that the change of T_C arises from the oxygen isotope exchange. The isotope exponent on T_C in this sample, $\alpha_C=-d\ln T_C/d\ln M_O$, is 1.89, much larger than 0.5 deduced from BCS theory.

The phonon-mediated BCS theory shows that in the condition of weak electron-phonon coupling, the increase in the lattice mass enhances the effective mass of charge carriers $m^\star$, lowers the superconducting gap Δ, and finally suppresses

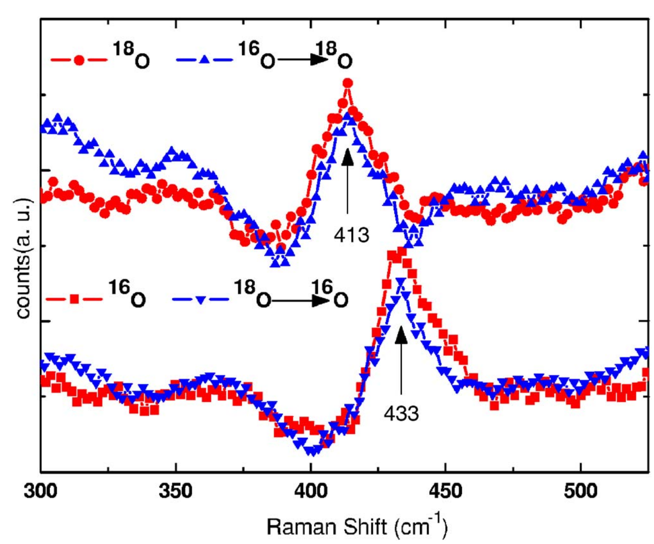

FIG. 3. (Color online) Raman spectra at room temperature for the ^{16}O and ^{18}O samples with $x=0.125$. A 514.5 nm Ar-laser line was used as the excitation line.

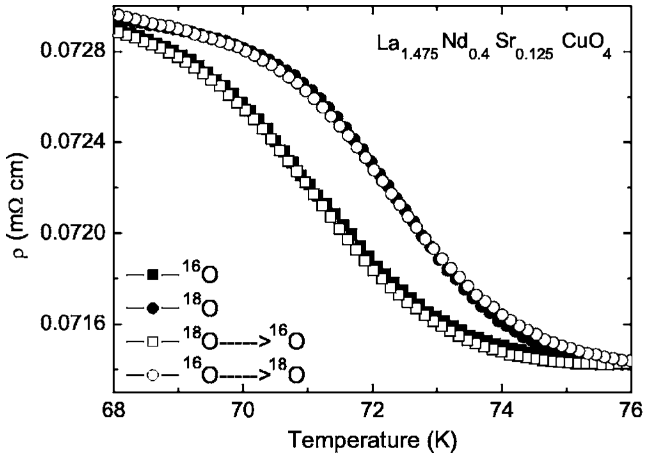

FIG. 4. Temperature dependence of resistivity near the structure transition for the ^{16}O by ^{18}O samples of x=0.125.

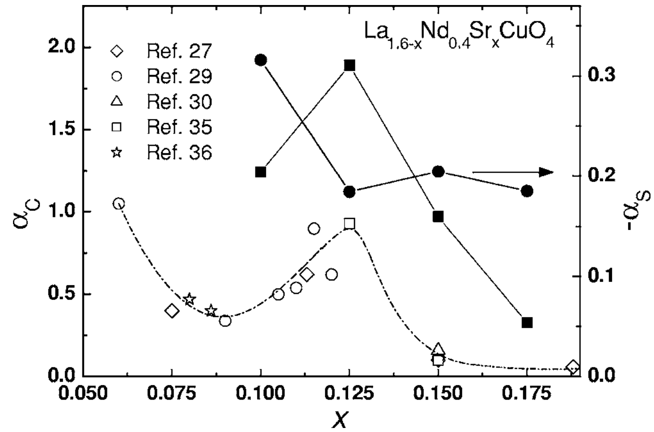

FIG. 5. α_C (solid squares) and α_S (solid circles) as a function of x for $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. For comparison, α_C from the previous works is also depicted for polycrystalline and single crystal $La_{2-x}Sr_xCuO_4$. The dashed line is a guide for the eyes.

T_C. This used to be successful in explaining the isotope effect in most of conventional superconductors, but failed in explaining the isotope effect in high T_C superconductors.[32] Particularly in $La_{2-x}Sr_xCuO_4$,[26–29] the isotope exponent around 1/8 doping is about 1. Zhao *et al.*[28,29] explained this with small polaron theory, wherein the effective mass of supercarriers depends strongly on the oxygen isotope mass in deeply underdoped regime, indicating strong electron-phonon coupling in it. The isotope exponent $\alpha_C \sim 1.89$ in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with x=0.125 is much larger than that (~1.0) in the Nd-free sample $La_{2-x}Sr_xCuO_4$. It indicates a stronger electron-lattice coupling in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. Nd doping induces a structural transition from LTO to LTT, which pins the stripe phase from fluctuating (Nd-free sample) to static[7,8] and enhances the distortion of CuO_2 plane.[20] It has been reported in manganites that lattice distortion tends to introduce stronger electron-phonon coupling.[33] Therefore, the electron coupled to the more distorted CuO_2 plane induced by Nd doping is responsible for the larger isotope exponent relative to the Nd-free sample.

Figure 4 shows the temperature dependence of resistivity near T_{LT} for the samples of x=0.125. T_{LT} is enhanced from 71 to 72.3 K with the substitution of ^{16}O by ^{18}O, the isotope exponent α_{LT} is about −0.19. As shown in Fig. 4, the resistivities are almost the same for the back-exchanged samples. It ensures that the change in T_{LT} arises from the isotope effect. The increase of T_{LT} indicates the enhancement of stabilization for LTT phase, suggesting the enhancement of distortion in CuO_2 plane by substitution of ^{16}O by ^{18}O. As shown in Figs. 2 and 4, the substitution of ^{16}O by ^{18}O leads to a decrease in T_C and an increase in T_{LT}. The oxygen isotope effect provides an evidence that the distortion of CuO_2 plane suppresses the superconductivity, consistent with the increase of T_C by lowering the impact of the disorder in Bi2212.[34] It has been reported that the charge ordering and structural transition are essentially coincidental for x=0.10 and 0.12.[8,17] Therefore, the increase of T_{LT} indicates the enhancement of charge stripe, suggesting the competition between the stripe phase and superconductivity.

For the samples $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with x=0.10, 0.15, and 0.175, the oxygen isotope exponents for superconducting transition α_C are 1.24, 0.98, and 0.33, while for structural transition, α_{LT} are −0.32, −0.20, and −0.17, respectively. All samples show that the substitution of ^{16}O by ^{18}O leads to a decrease in T_C and an increase in T_{LT}. Sr content dependence of oxygen isotope exponent for superconducting transition α_C and structural transition α_{LT} is shown in Fig. 5. The largest α_C is observed in the sample with x=0.125; such 1/8 anomaly for α_C has been reported in La_2CuO_4-based superconductors.[26–29] However, the α_{LT} decreases with increasing Sr content. For comparison, α_C reported in $La_{2-x}Sr_xCuO_4$ (Refs. 27, 29, 30, 35, and 36) is also shown in Fig. 5. It clearly shows a trend that α_C decreases with increasing Sr content except for an anomaly around x=0.125 in $La_{2-x}Sr_xCuO_4$. Our observation in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ shows a similar trend except that α_C for all Nd-doped samples is larger than that in $La_{2-x}Sr_xCuO_4$. It further indicates that the stronger distortion of CuO_2 plane induced by Nd doping leads to a stronger electron-lattice coupling. The sample with x=0.10 shows the largest α_{LT} value, which may be due to the approach to the phase boundary from LTO to LTT/*Pccn* for this Sr content.[17] The large isotope effect caused by the instability of the lattice around the phase boundary has also been found in cobalt.[37]

In the phase diagram of $La_{2-x-y}Nd_ySr_xCuO_4$ system, T_{LT} and the temperature of occurrence of stripe phase increase simultaneously while keeping Sr content constant and increasing Nd content.[17] Therefore, it can be believed that substitution of ^{16}O by ^{18}O leads to an increase of T_{LT}, and, consequently, to enhancement of the stripe phase. By keeping Nd content unchanged and increasing Sr doping level, the temperature where the stripe phase occurs shows a hump as a function of Sr doping level, while T_{LT} increases with increasing Sr doping level.[17] The suppression of the stripe phase for $x > 1/8$ is just caused by the deviation of hole concentration from 1/8. In our case, the oxygen isotope substitution does not change the hole concentration. Therefore, the increase of T_{LT} caused by the substitution of ^{16}O by ^{18}O for each composition indicates the enhancement of the stripe phase. These results show us a direct evidence of the competition between static stripe phase and superconductivity.

Recently, Reznik *et al.* found that a strong anomaly in Cu–O bond-stretching phonon in $La_{2-x}Sr_xCuO_4$ appears in superconducting doping level, while diappears in nonsuperconducting doping level. It suggests the importance of electron-phonon coupling to the mechanism of superconductivity.[38] The anomaly is strongest in the samples with static stripe phase: $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ and $La_{2-x}Ba_xCuO_4$. Our isotope substitution confirms the strong electron-phonon coupling in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ system and supports the importance of electron-phonon coupling to the mechanism of superconductivity for high-T_C superconductors.

In conclusion, oxygen isotope effect is systematically studied in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with static stripe phase. T_C is suppressed and T_{LT} is enhanced with the substitution of ^{16}O by ^{18}O. These results provide an evidence that the distortion of CuO_2 plane suppresses the superconductivity and that there exists a competition between static strip phase and superconductivity. α_C shows 1/8 anomaly, similar to that observed in Nd-free sample. A larger oxygen isotope effect on T_C is observed compared to the Nd-free samples. It indicates that a stronger distortion of CuO_2 plane leads to a stronger electron-phonon coupling. In addition, our results confirm the strong electron-phonon coupling in the $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. It is well known that the distortion of CuO_2 plane is a common feature shared by high-T_C cuprates. Therefore, electron-phonon coupling should play an important role in the mechanism of high-T_C superconductivity.

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