Oxygen isotope effects on the superconducting transition and magnetic states within the phase Ddagram of Y1-xPrxBa2Cu3O7-delta

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Abstract: The various phases observed in all cuprate superconductors [superconducting (SC), spin-glass (SG), and antiferromagnetic (AFM)] were investigated with respect to oxygen-isotope (16O/18O) effects, using here as a prototype system of cuprates Y1-xPrxBa2Cu3O7-delta. All phases exhibit an isotope effect which is strongest where the respective phase terminates. In addition, the isotope effects on the magnetic phases (SG and AFM) are sign reversed as compared to the one on the superconducting phase. In the coexistence regime of the SG and SC phase a two-component behavior is observed where the isotope induced decrease of the superfluid density leads to a corresponding enhancement in the SG related density.

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Oxygen isotope effect on superconductivity and magnetism in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$.

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Oxygen isotope ($^{16}O/^{18}O$) effects (OIE's) on the superconducting transition (T_c), the spin-glass ordering (T_g), and the antiferromagnetic ordering (T_N) temperatures were studied for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ as a function of Pr content ($0.0 \leq x \leq 1.0$). The OIE on T_c increases with increasing x up to $x \approx 0.55$, where superconductivity disappears. For decreasing x the OIE's on T_N and T_g increase down to $x \approx 0.7$ where antiferromagnetic order and down to $x \approx 0.3$ where spin-glass behavior vanish, respectively. The OIE's on T_g and T_N are found to have *opposite signs* as compared to the OIE on T_c. All OIE's are suggested to arise from the isotope dependent mobility (kinetic energy) of the charge carriers.

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High-temperature cuprate superconductors (HTS's) exhibit a rich phase diagram as a function of doping (see *e.g.* Fig. 2). The undoped parent compounds are characterized by a long range 3D antiferromagnetic (AFM) order which is rapidly destroyed when holes are doped into the CuO_2 planes. The short-range AFM correlations survive, however, well in the superconducting (SC) region of the phase diagram by forming a spin-glass (SG) state. The issues of the interplay of magnetism and superconductivity in HTS's and the nature of doping-induced charge carriers within the antiferromagnetic CuO_2 planes are still controversial. Understanding these fundamental questions can help to clarify the pairing mechanism of high-temperature superconductivity.

In conventional superconductors, key experimental evidence for a phonon mediated pairing mechanism was provided by measurements of the isotope effect on the transition temperature T_c. In contrast, in HTS's a number of unconventional oxygen-isotope ($^{16}O/^{18}O$) effects (OIE's) on various physical quantities, including among others the transition temperature T_c, the in-plane magnetic penetration depth $\lambda_{ab}(0)$, the pseudogap temperature T^*, and the spin-glass temperature T_g, were observed which cannot be explained by standard BCS theory [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. For instance, it was found that the OIE's on T_c and $\lambda_{ab}(0)$ are strongly doping dependent [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. In particular, close to optimal doping the OIE on T_c is almost zero [1, 2, 3], while the OIE on $\lambda_{ab}(0)$ is still substantial [7, 8, 9, 10]. With decreasing doping both OIE's on T_c and $\lambda_{ab}(0)$ increase and, for highly underdoped materials, even exceed the value of the BCS isotope exponent $\alpha_{T_c}^{\rm BCS} = 0.5$ [2, 4, 5, 7, 9, 10]. In order to obtain a more global view of cuprate superconductors a detailed study of the isotope dependence of magnetic quantities is needed. So far, to our knowledge only little work has been reported on this subject. This includes experimental studies of the OIE on the antiferromagnetic ordering temperature (T_N) [11] and the OIE on the spin-glass ordering temeprature (T_g) [12], as well as a theoretical investigation of the OIE on T_N [13]. Here we report a systematic study of the OIE on T_c, T_g, and T_N in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ as a function of Pr content x ($0.0 \leq x \leq 1.0$) by means of magnetization and muon-spin rotation (μSR) experiments. The OIE's on T_c and T_N were found to be vanishingly small at $x = 0.0$ and $x = 1.0$, respectively, and increase for the intermediate x. In the range $0.3 < x < 0.6$ where superconductivity and spin-glass magnetism coexist both T_c and T_g exhibit a large OIE. The OIE's on T_N and T_g are *sign reversed* compared to the OIE on T_c.

Polycrystalline samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($0 \leq x \leq 1.0$) were prepared by standard solid state reaction [14]. Oxygen isotope exchange was performed during heating the samples in $^{18}O_2$ gas. To ensure the same thermal history of the substituted (^{18}O) and not substituted (^{16}O) samples, both annealings (in $^{16}O_2$ and $^{18}O_2$ gas) were always performed simultaneously. The ^{18}O content in the samples, as determined from a change of the sample weight after the isotope exchange, was found to be 80(5)% for all ^{18}O substituted samples.

The OIE on T_c was obtained by field-cooled magnetization (M_{FC}) experiments performed with a SQUID magnetometer in a field of 1.0 mT and at temperatures between 1.75 K and 100 K. The values of T_c were defined as the temperatures where the linearly extrapolated $M_{FC}(T)$'s intersect the zero line [see Fig. 1 (a)].

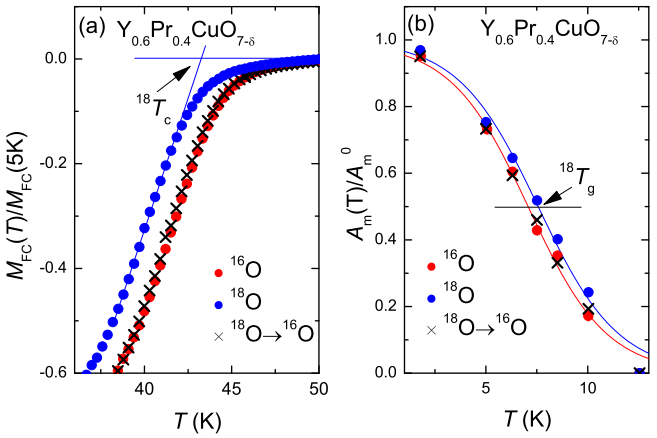

FIG. 1: (a) Temperature dependences of the field–cooled magnetization M_{FC} normalized to its value at $T = 5$ K, and (b) A_m/A_m^0 for ^{16}O, ^{18}O, and backexchanged ^{18}O→^{16}O samples of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_{7-\delta}$. The solid lines in (b) represent the fits of $A_m(T)$ for the ^{16}O/^{18}O substituted samples by means of Eq. (2). The arrows indicate T_c and T_g for the ^{18}O substituted sample.

The OIE's on the magnetic ordering temperatures (T_g and T_N) were extracted from the zero-field μSR data. No magnetism down to $T \simeq 1.7$ K was detected for the ^{16}O/^{18}O substituted samples with $x = 0.0$ and $x = 0.2$. For $x = 0.3$ and 0.4 magnetism was identified as a fast decrease of the asymmetry at $T < 10$ K and $T < 5$ K, respectively. For $x \geq 0.45$ damped oscillations due to muon-spin precession in local magnetic fields were observed. The μSR asymmetry spectra for $x = 0.8$ and 1.0, *i.e.* in the deep antiferromagnetic phase, were analyzed by using the following expression:

$$A(t) = A_n \exp(-\sigma^2 t^2/2) + A_m[\omega \exp(-\lambda_1 t)\cos(\gamma_\mu B_\mu t) + (1-\omega)\exp(-\lambda_2 t) J_0(\gamma_\mu B_\mu t)]. \quad (1)$$

Here A_m and A_n represent the oscillating (magnetic) and nonoscillating amplitudes, respectively, ω is a weighting factor, B_μ is the mean internal magnetic field at the muon site, $\gamma_\mu = 2\pi \times 135.5342$ MHz/T is the muon gyromagnetic ratio, and J_0 is the zeroth-order Bessel function. We used the damped Bessel function J_0 together with the cosine oscillating term in order to account for the unphysically large values of the initial phase $\phi \simeq 20^o - 45^o$ which has to be introduced close to T_N in order to fit the data by using the cosine term only [$\cos(\gamma_\mu B_\mu t + \phi)$]. For $0.45 \leq x \leq 0.7$ and for $x = 0.4$ and 0.3 the fit was simplified by taking from the second part of Eq. (1) only the damped Bessel term and the exponential damping term with $B_\mu = 0$, respectively. The magnetic ordering temperatures (T_g and T_N) were then determined by fitting the temperature dependence of A_m by means of the phenomenological function:

$$A_m(T)/A_m^0 = (1 + \exp[(T - T_m)/\Delta T_m])^{-1}. \quad (2)$$

Here A_m^0 is the maximum value of the asymmetry, T_m is the magnetic ordering temperature ($m = g, N$) and ΔT_m is the width of the magnetic transition [see Fig. 1 (b)].

In order to confirm the intrinsic origin of the OIE's on T_c and T_m, back-exchange OIE experiments were carried out for the samples with $x = 0.0$ and $x = 0.4$. As shown in Fig. 1, the ^{16}O oxygen back exchange of the ^{18}O sample of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_{7-\delta}$ results within error in almost the same $M_{FC}(T)$ [panel (a)] and $A_m(T)$ [panel (b)] as for the ^{16}O sample. The results of the OIE's on T_c, T_g and T_N are summarized in Table I and Fig. 2. The doping dependences of T_c, T_g, and T_N for the ^{16}O substituted samples are in agreement with the results of Cooke *et al.* [15]. The second magnetic transition at $T_{N_2} \simeq 17$ K (observed for $x = 0.7 \div 1.0$) which is associated with the ordering of the Pr sublattice is not considered here.

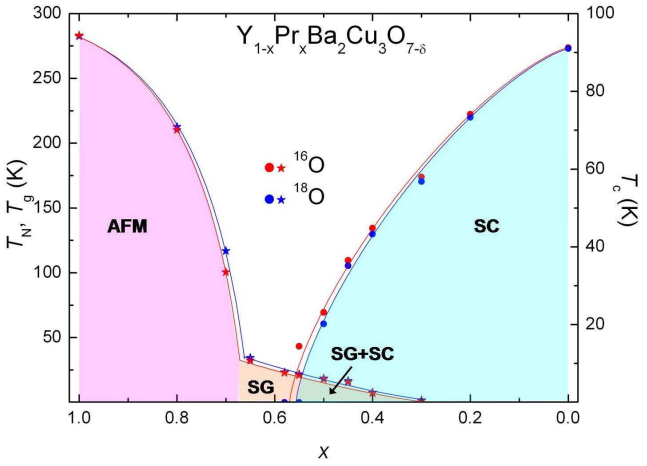

FIG. 2: Dependence of the superconducting transition (T_c), the spin-glass ordering (T_g), and the antiferromegnetic ordering (T_N) temperatures for ^{16}O/^{18}O substituted $Y_xPr_{1-x}Ba_2Cu_3O_{7-\delta}$ on the Pr content x. The solid lines for $^{16}T_g(x)$ [$^{18}T_g(x)$] and $^{16}T_N(x)$ [$^{18}T_g(x)$] are guides to the eye and those for $^{16}T_c(x)$ [$^{18}T_c(x)$] result from the power law fit (see text for details). The areas denoted by "AFM", "SG", and "SC" represent the antiferromagnetic, the spin-glass and the superconducting regions, respectively. "SG+SC" corresponds to the region where spin-glass magnetism coexist with superconductivity.

It is convenient to quantify the OIE's on the transition temperatures T_y (y denotes c, g, or N) in terms of the isotope effect exponents defined by:

$$\alpha_{T_y} = -\frac{d\ln T_y}{d\ln M_O} = -\frac{\Delta T_y/T_y}{\Delta M_O/M_O} = -\frac{(^{18}T_y - {}^{16}T_y)/{}^{16}T_y}{(^{18}M_O - {}^{16}M_O)/{}^{16}M_O}, \quad (3)$$

where M_O is the mass of the oxygen isotope (^{16}O/^{18}O). The values of α_{T_c} and α_{T_m} ($m = g$, N) are listed in Table I and shown in Fig. 3 as a function of Pr content x. For $0.0 \leq x \leq 0.5$ the values of α_{T_c} are in agreement with previous results [2]. In order to estimate α_{T_c} for $x = 0.55$, we assume for $^{18}T_c$ the conservative value $^{18}T_c = 1.7(1.7)$ K, yielding $\alpha_{T_c} = 7(1)$. In the

TABLE I: Summary of the OIE studies on T_c and T_m ($m = g$, N) for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. The meaning of the parameters is – $^{16}T_c/^{18}T_c$ and $^{16}T_m/^{18}T_m$: the superconducting transition temperature and the magnetic ordering temperature for the $^{16}O/^{18}O$ substituted samples, respectively, $\alpha_{T_c} = -\mathrm{d}\ln T_c/\mathrm{d}\ln M_O$: the OIE exponent of T_c, $\alpha_{T_m} = -\mathrm{d}\ln T_m/\mathrm{d}\ln M_O$: the OIE exponent of T_m.

x	$^{16}T_c$ (K)	$^{18}T_c$ (K)	α_{T_c}	$^{16}T_m$ (K)	$^{18}T_m$ (K)	α_{T_m}
0.00	91.19(5)	90.99(4)	0.018(5)	–	–	–
0.20	74.07(2)	73.27(2)	0.086(3)	–	–	–
0.30	57.97(8)	56.79(7)	0.163(15)	1.13(11)	1.66(12)	-3.8(1.2)
0.40	44.80(2)	43.25(3)	0.277(7)	7.07(9)	7.56(9)	-0.55(31)
0.45	36.50(6)	35.12(6)	0.302(19)	15.54(13)	16.58(11)	-0.54(9)
0.50	23.12(4)	20.16(4)	1.024(21)	17.82(11)	18.46(12)	-0.29(7)
0.55	14.4(2)	< 1.7	7(1)[a]	21.05(18)	21.65(21)	-0.23(9)
0.58	–	–	–	22.8(2)	23.2(2)	-0.13(9)
0.65	–	–	–	32.3(4)	34.4(4)	-0.50(14)
0.70	–	–	–	100.5(1.4)	116.8(1.3)	-1.30(14)
0.80	–	–	–	210.2(4)	212.6(4)	-0.09(2)
1.00	–	–	–	283.2(7)	282.5(7)	0.02(3)
0.00	91.35(4)[b]	91.16(4)[c]	0.017(5)	–	–	–
0.40	44.63(3)[b]	–	0.247(8)	7.10(9)	–	-0.52(30)

[a]Estimated value (see text)
[b]Back exchanged $^{18}O \rightarrow ^{16}O$ sample
[c]Back exchanged $^{16}O \rightarrow ^{18}O$ sample

SG phase a high value of $\alpha_{T_m} = -3.8(1.2)$ for $x = 0.3$ was found, in accordance with a previous study of the SG behavior in Mn doped $La_{2-x}Sr_xCuO_4$ [12]. Both α_{T_c} and α_{T_m} exhibit unusual features, *i.e.*: (i) α_{T_c} and $\alpha_{T_m}(m = g, N)$ depend strongly on x, being small in amplitude at "extreme" Pr content ($x = 0.0$ for T_c and $x = 1.0$ for T_N) and strongly increase upon approaching $x = 0.55$ and $x = 0.3$, respectively (see Fig. 3). For $0.5 \leq x \leq 0.55$, α_{T_c} exceeds considerably the BCS isotope-exponent $\alpha_{T_c}^{\mathrm{BCS}} = 0.5$. (ii) α_{T_c} increases monotonically with increasing x, while α_{T_m} has a pronounced maximum around $x \simeq 0.7$. This maximum is a consequence of the fact that T_N depends much stronger on x than T_g (see Fig. 2). (iii) α_{T_c} and α_{T_m} have *opposite* sign, *i.e.*, T_c decreases with increasing oxygen-isotope mass ($^{16}T_c > ^{18}T_c$), whereas T_g and T_N increases ($^{16}T_g < ^{18}T_g$, $^{16}T_N < ^{18}T_N$, except for $x = 1.0$). This is particulary interesting in the region of the phase diagram where superconductivity (SC) and the spin-glass (SG) magnetism coexist (see Figs. 2 and 3). (iv) The strong increase of both α_{T_c} and α_{T_m} by approaching $x = 0.55$ and $x = 0.3$, respectively (see Fig. 3) suggests that the critical concentrations where superconductivity ($x_{T_c}^{crit}$) and magnetism ($x_{T_m}^{crit}$) disappear are different for ^{16}O and ^{18}O substituted samples. An analysis of the $^{16}O/^{18}O$ data for $T_c(x)$ presented in Fig. 2 by means of the power law $T_c(x) = T_c(x = 0)[1 - (x/x_{T_c}^{crit})^{\delta}]^{\beta}$ yields: $^{16}x_{T_c}^{crit} = 0.570(1)$, $^{16}\delta = 1.33(4)$, $^{16}\beta = 0.72(3)$ and $^{18}x_{T_c}^{crit} = 0.556(2)$, $^{18}\delta = 1.33(2)$, $^{18}\beta = 0.73(3)$. A linear extrapolation of $^{16}T_g(x)$ and $^{18}T_g(x)$ in the region $x \simeq 0.3 \div 0.5$ to $T_g = 0$ yields: $^{16}x_{T_m}^{crit} \simeq 0.287(2)$ and $^{18}x_{T_m}^{crit} \simeq 0.278(2)$. It is interesting to note that the relative oxygen-isotope shifts of the critical concentrations $x_{T_c}^{crit}$ and $x_{T_m}^{crit}$, defined by $\Delta x/x = (^{18}x - ^{16}x)/^{16}x$, are the same within experimental error: $\Delta x_{T_c}^{crit}/x_{T_c}^{crit} = -2.5(4)\%$ and $\Delta x_{T_m}^{crit}/x_{T_m}^{crit} = -3.1(7)\%$.

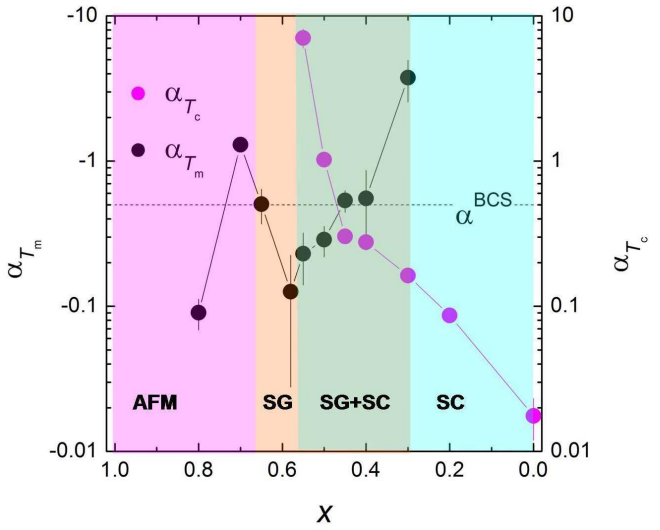

FIG. 3: OIE exponents α_{T_c} and α_{T_m} ($m = g$, N) for $^{16}O/^{18}O$ substituted $Y_xPr_{1-x}Ba_2Cu_3O_{7-\delta}$ as a function of the Pr content x. The dashed line corresponds to $\alpha_{T_c}^{\mathrm{BCS}} = 0.5$. The meaning of the areas denoted by "AFM", "SG", "SG+SC", and "SC" are the same as in Fig. 2.

Recently, Ofer *et al.* [16] studied a series of $Ca_xLa_{1-x}Ba_{1.75-x}La_{0.25+x}Cu_3O_y$ HTS's with various doping levels – from undoped to highly overdoped. They concluded that all characteristic temperatures T_N, T_g,

and T_c are controlled by the in-plane magnetic exchange energy J of the undoped (parent) compound, thus inferring that the isotope effects on T_N, T_g, and T_c are driven by the isotope dependence of J. In order to get a rough estimate of the OIE on J we employed the method proposed by Zhao *et al.* [5, 11]. For the undoped antiferromagnetic parent compounds the following relation holds: $\Delta T_N/T_N \simeq \Delta J/J \cdot B/(B+1)$, where $B = 2J/T_N \simeq 10$. For $x = 1.0$ with $^{16}T_N = 283.2(7)$ and $^{18}T_N = 282.5(7)$ (see Table I) one obtains $\Delta J/J \simeq 0.3(4)\%$, *i.e.*, the OIE on J is zero within experimental uncertainty. Therefore, J is very unlikely the source of the observed OIE's on T_N, T_g, and T_c.

Opposite to the above interpretation, we argue that the observed OIE's on T_g and T_N are directly related to the different charge carrier mobilities (renormalized kinetic energies [17]) in the ^{16}O/^{18}O substituted samples caused by the isotope dependence of the charge carrier mass m^*. As shown in Refs. [4, 5, 6, 7, 8] the charge carriers in the ^{18}O substituted samples are heavier and, consequently, less mobile than in ^{16}O substituted samples. The recent studies of Hücker *et al.* [18] clearly demonstrate that increasing the hole mobility in $La_{2-x}Sr_xCuO_4$ rapidly suppresses the antiferromagnetic ordering temperature T_N. A similar conclusion was also reached by Shengelaya *et al.* [12] based on the observation of a giant OIE on the spin-glass ordering temperature T_g in Mn doped $La_{2-x}Sr_xCuO_4$.

As for OIE on T_c, the observation of the onset of superconductivity at different $x_{T_c}^{crit}$ (see Fig. 2) implies that the number of carriers condensed into Cooper pairs are larger for ^{16}O substituted samples then for ^{18}O ones. Consequently, we propose that the different hole mobilities in ^{16}O/^{18}O substituted samples are also responsible for the oxygen isotope shift of the critical concentration $x_{T_c}^{crit}$ and then necessarily also for the OIE on T_c. This statement is further supported by the following facts: (i) The relative isotope shifts of $x_{T_c}^{crit}$ and $x_{T_m}^{crit}$ are roughly the same $\Delta x_{T_c}^{crit}/x_{T_c}^{crit} \simeq \Delta x_{T_m}^{crit}/x_{T_m}^{crit}$ (see above), indicating that the OIE's on T_c, T_g, and T_N are of similar origin. (ii) In the region where superconductivity and spin-glass magnetism coexist (see Fig. 2 and Table I) the increase of T_g in the ^{18}O substituted sample (decrease of the hole mobility) is associated with a corresponding decrease of T_c.

In conclusion, oxygen isotope (^{16}O/^{18}O) effects on the superconducting transition (T_c), the spin-glass ordering (T_g), and the antiferromagnetic ordering (T_N) temperatures were studied for a series of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples as a function of Pr content ($0.0 \leq x \leq 1.0$). The OIE exponent α_{T_c} increases with increasing x (decreasing doping) reaching a maximum at $x_{T_c}^{crit} \approx 0.55$, where superconductivity disappears. At $x = 1.0$ (undoped case) $\alpha_{T_N} \simeq 0$ within experimental uncertainty. For decreasing x (increasing doping) α_{T_m} increases, reaching a maximum at $x_{T_m}^{crit} \approx 0.3$ where spin-glass behavior vanishes. In the range of $0.3 \leq x < 1.0$ the OIE's on T_g and T_N are *sign reversed* as compared to the one on T_c. The relative isotope shift of the critical Pr concentration where superconductivity ($x_{T_c}^{crit}$) and magnetism ($x_{T_m}^{crit}$) disappear are the same $\Delta x_{T_c}^{crit}/x_{T_c}^{crit} \simeq \Delta x_{T_m}^{crit}/x_{T_m}^{crit}$, indicating that the OIE's on T_c, T_g, and T_N are interrelated. These OIE's are suggested to arise from the isotope dependent mobility of the charge carriers as proposed in a model where polaronic renormalization of the single particle energies are introduced [17]. The formation of polaronic charge carriers may be caused by a strong Jahn-Teller effect [19], in close analogy to doped perovskite manganites [20]. The unconventional isotope effects presented here clearly demonstrate that lattice effects play a significant role in the physics of cuprates in both the magnetic and the superconducting state.

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