Closing of the pseudogap in $\text{La}_{2-z-x}\text{Nd}_z\text{Sr}_x\text{CuO}_4$ ($z = 0, 0.4$)

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(Dated: October 10, 2021)

Abstract

Doping of $\text{La}_2\text{CuO}_4$ and $\text{La}_{1.6}\text{Nd}_{0.4}\text{CuO}_4$ with Sr gives rise to holes that locate pairwise at lattice-site $O$ atoms. Such $O$ atoms reside as lattice defects only in the $\text{CuO}_2$ planes if the doping level $x$ is below a watershed value, $x < \hat{x}$, but also in each bracketing $\text{LaO}$ layer if $x > \hat{x}$. The $O$ atoms form a 2D charge order of incommensurability $q_{\text{CO}}^{\text{CuO}_2}(x)$ and $q_{\text{CO}}^{\text{LaO}}(x)$. At the doping $x^*$ (quantum critical point) that causes the closing of the pseudogap at $T = 0$, $q_{\text{CO}}^{\text{CuO}_2}(x^*) = 2q_{\text{CO}}^{\text{LaO}}(x^*)$ holds.
I. CHARGE ORDER IN La$_{2-x}$Sr$_x$CuO$_4$

When $La_2CuO_4$ is doped with strontium, substitution of $La \rightarrow La^{3+} + 3e^-$ by $Sr \rightarrow Sr^{2+} + 2e^-$ causes a lack of electrons, or equivalently, a creation of holes. Coulomb repulsion spreads the holes, which then attach pairwise to oxygen, $O^{2-} + 2e^+ \rightarrow O$. The $O$ atoms form a charge order (CO) by a 2D superlattice in each $CuO_2$ plane of $La_{2-x}Sr_xCuO_4$. The reciprocal of the superlattice spacing, $A_0(x) = B_0(x)$, is called the incommensurability\[1\]
\[
\frac{1}{A_0(x)} \equiv q_{CO}^{CuO_2}(x) = \frac{\Omega^\pm}{2} \sqrt{x - \tilde{p}} , \quad x < \hat{x} .
\] (1)

In the medium doping range, $0.10 \lesssim x \lesssim 0.15$, the charge order manifests as unidirectional stripes, at higher doping as nematicity. The formula, in reciprocal lattice units (r.l.u.), is valid for doping up to a “watershed” concentration $\hat{x}$, which depends on the species of doping. The stripe-orientation factor is $\Omega^+ = \sqrt{2}$ for $x > x_6 = 2/\sqrt{3} \approx 0.356$ when stripes are parallel to the $a$ or $b$ axis, but $\Omega^- = 1$ for $x < x_6$ when stripes are diagonal. The offset value $\tilde{p}$ under the radical is the hole concentration necessary to keep three-dimensional antiferromagnetism (3D-AFM) suppressed. Their skirmisher task keeps those “suppressor holes” from participating in charge order.

Also because of Coulomb repulsion, the doped-hole concentration saturates in the $CuO_2$ planes at $\hat{x}$, causing the square-root curve from Eq.(1) to level off, with increased doping, to a constant plateau (depending on dopant and co-dopant species),\[1\]
\[
q_{CO}^{CuO_2}(x) = \frac{\sqrt{2}}{2} \sqrt{\hat{x} - \tilde{p}} , \quad x \geq \hat{x} .
\] (2)

(see Fig. 1). Additional doped holes then overflow to the $LaO$ layers that sandwich the $CuO_2$ planes, where they also reside pairwise in (apical) $O$ atoms. Again, Coulomb repulsion spreads the double holes to a planar superlattice of lattice-defect $O$ atoms in each $LaO$ layer, with attending charge order of incommensurability\[1\]
\[
q_{CO}^{LaO}(x) = \frac{\sqrt{2}}{2} \sqrt{\frac{x - \hat{x}}{2}} , \quad x \geq \hat{x} .
\] (3)

The denominator 2 under the radical is due to the two $LaO$ layers per unit cell.

The $Sr$ doping that causes the closing of the pseudogap at $T = 0$ is denoted as $x^*$. It is widely regarded a quantum critical point. Inserting the observed data of $\hat{x}$, $\tilde{p}$, and $x^*$ of $La_{2-x}Sr_xCuO_4$ from Table I into Eqs. (2, 3) gives incommensurabilities
\[
2q_{CO}^{LaO}(x^*) = q_{CO}^{CuO_2}(\hat{x}) = q_{CO}^{CuO_2}(x^*) .
\] (4)
For the sake of graphic visualization we combine the defect-charge order of the upper LaO layer (LaO) and lower LaO layer (LaO), staggered by half a superlattice spacing, $\frac{1}{2}A^{LaO}_0 = \frac{1}{2}/q^{CO}_LaO$ (see Fig. 2), such that together they form a charge order of incommensurability

$$q^{LaO+LaO}_CO(x) = 2q^{LaO}_CO(x) = \sqrt{x - \tilde{x}}.$$  (5)

FIG. 1. Incommensurability of O superlattice charge order, $q = q_c$, and O superlattice magnetization, $q = 2q_m$, in the CuO$_2$ planes (solid curves and horizontal lines) and in the LaO layers (hatched curves) of La$_{2-z-x}$Ln$_z$AexCuO$_4$ (Ln = Nd, Eu; z = 0, 0.4, 0.2) due to doping with Ae = Sr or Ba. Commensurate doping concentrations are denoted by $x_n \equiv 2/n^2$. Circles show data from X-ray diffraction and scattering or neutron scattering. For La$_{1.6}$Nd$_{0.4}$Sr$_x$CuO$_4$ the $2q_m$ data are experimental, the horizontal $q_c$ line is inferred (see text). The broken black curve is a graph of Eq. (1), calculated with a constant offset value, $\tilde{p} = x_{10} = 0.02$. Its discontinuity at $x_6 \simeq 0.056$ is caused by a change of stripe orientation, relative to the planar crystal axes, from diagonal for $x < x_6$ to parallel for $x > x_6$. The solid (colored) horizontal lines are graphs of Eq. (2) and the hatched colored curves are graphs of $2q^{LaO}_c(x)$, Eqs. (3, 5). The condition for closing of the pseudogap, Eq. (4), is illustrated by the dashed down arrows.
TABLE I: Watershed concentration \( \hat{x} \) of doped Sr where the square-root curve of charge-order incommensurability in the \( CuO_2 \) plane, \( q_{CO}^{CuO_2}(x) \), levels off; concentration of 3D-AFM suppressor holes \( \hat{p} \); and Sr doping \( x^* \) at the closing of the pseudogap at \( T = 0 \). The charge-order incommensurability in each LaO layer is denoted as \( q_{LaO}^{LaO} \).

| Compound                  | \( \hat{x} \) | \( \hat{p} \) | \( x^* \) | \( q_{CO}^{CuO_2}(\hat{x}) \) | \( q_{CO}^{CuO_2}(x^*) \) | \( 2q_{CO}^{LaO}(x^*) \) |
|---------------------------|---------------|--------------|-----------|-------------------------------|---------------------------|-----------------------------|
| \( La_{2-x}Sr_xCuO_4 \)  | 0.125         | 0.015        | 0.18      | 0.235                         | 0.235                     | 0.235                       |
| \( La_{1.6-x}Nd_{0.4}Sr_xCuO_4 \) | 0.16 | 0.019 | 0.23 | 0.266 | 0.266 | 0.266 |

FIG. 2. Cartoon illustrating charge-order stripes in the \( CuO_2 \) plane (red), upper LaO layer (LaO, green), and lower LaO layer (LaO, blue) in side view (top part) and down view (center and bottom part). At doping \( x^* \), shown here, each LaO layer has an incommensurability \( q_{LaO}^{LaO}(x^*) = \frac{1}{2} q_{CO}^{CuO_2}(x^*) \). When their stripes are staggered and combined, both bracketing LaO layers together have the same incommensurability as the bracketed \( CuO_2 \) plane, \( \frac{q_{CO}^{LaO}+q_{LaO}^{LaO}}{2}(x^*) = q_{CO}^{CuO_2}(x^*) \). The stripes of the combined LaO layers are staggered against those of the \( CuO_2 \) plane such that they interlace.

Solving Eq. (4) for \( x^* \) relates the Sr-doping that causes closing of the pseudogap (at \( T = 0 \)) with the watershed doping \( \hat{x} \) and the offset concentration \( \hat{p} \),

\[
x^* = \frac{3\hat{x} - \hat{p}}{2}.
\]
II. SELECTION OF DATA

The condition of Eq. (6) for the three parameters $\hat{x}$, $\tilde{p}$ and $x^*$ is satisfied, within error bars, for the data of $La_{2-x}Sr_xCuO_4$, listed in Table I. The offset value $\tilde{p} = 0.015 \pm 0.002$ is confirmed, through Eq. (1), by many measurements of charge-order and magnetization stripes in $La_{2-x}Sr_xCuO_4$ near $x = 0.125 = 1/8$, cited in Ref. 1. Visually, it can be inferred from the accumulation of data points slightly above the square-root curve in Fig. 1 (drawn with constant $\tilde{p} = x_{10} = 0.02$) for $x > 0.09$ but coincident with a slightly upshifted curve in that range (not shown) when $\tilde{p} = 0.015$ is used. The value of the watershed doping in Table I, $\hat{x} = 0.125$, is obtained by Eq. (2) from a constant charge-order incommensurability, $q_{CO}^{CuO_2} = 0.235, x \geq \hat{x}$, being within error bars of $q_{CO}^{CuO_2} = 0.235 \pm 0.005$ r.l.u., observed with non-resonant hard X-rays, and $q_{CO}^{CuO_2} = 0.231 \pm 0.005$ r.l.u., observed with resonant inelastic soft X-ray spectroscopy. The doping level where the pseudogap closes (at $T = 0$), $x^* = 0.18 \pm 0.01$, has been determined with resistivity and Nernst-effect measurements.

Turning to the data for $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, an offset value $\tilde{p} = 0.019$ is obtained with Eq. (1) from $q_{CO}^{CuO_2} = 0.23 \pm 0.005$ r.l.u. of $La_{1.475}Nd_{0.4}Sr_{0.125}CuO_4$. Measurements of resistivity, Hall effect, Nernst effect, and thermoelectric power give $x^* = 0.23 \pm 0.01$. If the relationship between charge order and magnetic order, $q_{CO}^{CuO_2}(x) = 2q_{M}^{CuO_2}(x)$, holds, as for $La_{2-x}Ae_xCuO_4$ ($Ae = Sr, Ba$), then $q_{CO}^{CuO_2} = 0.278$, together with Eq. (2), would give a value of $\hat{x} = 0.17 \pm 0.005$. Although close, the values of $\tilde{p}$, $x^*$ and $\hat{x}$ thusly obtained do not satisfy the condition of Eq. (6) within error bars. What could be the reason?

It is known that the magnetic moment of the co-dopant, $m(Nd^{3+}) \neq 0$ — as opposed to $m(La^{3+}) = 0$ — couples with the magnetic moment of the host $Cu^{2+}$ ions, $m(Cu^{2+}) \neq 0$, at temperatures $T < 5 K$. This is the reason why magnetization stripes are observable in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ at such high doping, in contrast to $La_{2-x}Ae_xCuO_4$. The $m(Nd^{3+})$ moments may also couple with the magnetic moments of the lattice-defect $O$ atoms that give rise to the magnetization stripes, $m(O) \neq 0$, such that the relation of Eq. (7) is slightly violated in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. If we instead assume that Eq. (6) is valid, we...
obtain \( \hat{x} = (2x^* + \tilde{p})/3 = 0.16 \), listed in Table I, and a charge-order incommensurability \( q_{CO}^{CuO_2}(\hat{x}) = 0.266 \) instead of \( 2q_{M}^{CuO_2} = 0.278 \) (see Fig. 1), in slight violation of Eq. (7).

Besides \( La_{2-x}Sr_xCuO_4 \) and \( La_{1.6-x}Nd_{0.4}Sr_xCuO_4 \), treated here, there remain two more heterovalent-metal doped cuprates with high-\( T_c \) superconductivity: \( La_{2-x}Ba_xCuO_4 \) and \( La_{1.8-x}Eu_{0.2}Sr_xCuO_4 \). Because of difficulties in crystal growth, few data beyond \( x = 0.125 \) have been observed for \( La_{2-x}Ba_xCuO_4 \). With \( \hat{x} = 0.14 \) and \( \tilde{p} = 0.015 \) determined by Eqs. (2, 1), a prediction of \( x^* = 0.20 \) can be made with Eq. (6), listed in Table II. Likewise, with \( \tilde{p} = 0.015 \) and \( x^* = 0.23 \) of \( La_{1.8-x}Eu_{0.2}Sr_xCuO_4 \), the watershed doping for that compound is predicted as \( \hat{x} = 0.16 \) and the constant incommensurability of charge order, \( q_{CO}^{CuO_2} = 0.266 \) — both in agreement with \( La_{1.6-x}Nd_{0.4}Sr_xCuO_4 \).

| Compound                  | \( \hat{x} \) | \( q_{CO}^{CuO_2}(\hat{x}) \) | \( \tilde{p} \) | \( x^* \) |
|---------------------------|---------------|-------------------------------|---------------|--------|
| \( La_{2-x}Ba_xCuO_4 \)  | 0.14          | 0.25                          | 0.015         | 0.20   |
| \( La_{1.8-x}Eu_{0.2}Sr_xCuO_4 \) | 0.16         | 0.266                         | 0.015         | 0.23   |

TABLE II: Same notations as in Table I. Predictions with Eqs. (6, 2) are marked bold.

### III. SHIFT OF \( x^* \) BY Nd CO-DOPING

A question often asked is: Why is the high-doping end of the pseudogap phase different for \( La_{2-x}Sr_xCuO_4 \) and \( La_{1.6-x}Nd_{0.4}Sr_xCuO_4 \), with \( x^* = 0.18 \) for the former but \( x^* = 0.23 \) for the latter? If \( x^* \) is affected by the watershed doping \( \hat{x} \), as can be inferred from Eq. (4), then the question is better rephrased as: Why is \( \hat{x} = 0.125 \) in \( La_{2-x}Sr_xCuO_4 \) but \( \hat{x} = 0.16 \) in \( La_{1.6-x}Nd_{0.4}Sr_xCuO_4 \)?

It is likely that the difference is due both to different magnetic moments of co-dopant and host ions, \( m(Nd^{3+}) \neq m(La^{3+}) = 0 \), and to different ions sizes, \( r(Nd^{3+}) = 1.26 \) Å < \( r(La^{3+}) = 1.30 \) Å. The former gives rise to repulsive magnetic interaction between \( m(Nd^{3+}) \) and \( m(O) \) moments in the \( (La/Nd)O \) layers. The latter causes a slightly smaller lattice constant \( c_0 \) of the \( Nd \)-codoped compound, and consequently a slightly smaller unit-cell volume \( v_0 \) (see Table III). Conceptually, this makes it harder for doped holes to enter the \( (La/Nd)O \) layers and form lattice-defect atoms, \( O^2^- + 2e^+ \rightarrow O \). The necessary higher internal pressure is achieved by Coulomb repulsion of doped holes in the \( CuO_2 \) planes at a.
higher density, to wit by $\hat{x} = 0.125 \rightarrow 0.16$.

| Doping $\rightarrow$ | $x = 0$ | $x = 0$ | $x = 0$ | $x = 0$ | $x = 0.12$ | $x = 0.12$ | $x = 0.12$ | $x = 0.12$ |
|---------------------|--------|--------|--------|--------|--------|--------|--------|--------|
| ↓ Compound          | $a_0$ (Å) | $b_0$ (Å) | $c_0$ (Å) | $v_0$ (Å³) | $a_0$ (Å) | $b_0$ (Å) | $c_0$ (Å) | $v_0$ (Å³) |
| $La_{2-x}Sr_xCuO_4$ | 5.34   | 5.43   | 13.12  | 380    | 5.33   | 5.36   | 13.18  | 377    |
| $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ | 5.34 | 5.40 | 13.03  | 376    | 5.32   | 5.32   | 13.13  | 372    |

TABLE III: Lattice constants $a_0$, $b_0$, $c_0$ and unit-cell volume, $v_0 = a_0b_0c_0$, of $La_{2-x}Sr_xCuO_4$ and $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ without Sr doping, $x = 0$, and at $x = 0.12$ (Refs. 15, 16).

IV. Sr-DOPING BEYOND $x^*$

What happens with further Sr-doping of the lanthanum/neodymium cuprates, $x > x^*$? The constancy of $q_{CO}^{CuO_2}(x)$ for $x > x^*$, shown in Fig. 1, implies that none of the additional doped holes generate more defect $O$ atoms in the $CuO_2$ plane. Neither do they generate more defect $O$ atoms in the bracketing $LaO$ layers — otherwise the condition of Eq. (4) would fail and the pseudogap would open again. This leaves as the most probable scenario that the additional doped holes will be delocalized. The assessment agrees with the notion of a Fermi-liquid phase beyond the quantum critical point, $x^*$, where the half-occupied $Cu d_{x^2-y^2}$ orbitals—localized in the Mott insulator ($x < 0.02$) and in the pseudogap phase ($0.02 < x < x^*$), except when part of Fermi arcs—are now delocalized.

A transition of hole density (per Cu atom) from

$$n^+ \simeq x \quad \rightarrow \quad n^+ \simeq 1 + x$$

(8)

has been shown by measurements of electrical resistivity $\rho$, Hall coefficient $R_H$, and Seebeck coefficient $S$ (thermopower) in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$. It can be interpreted as massive charge delocalization. From transport measurements in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ under pressure, a doping condition for a Lifshitz transition, $x^* \leq x^\dagger$, was obtained (A Lifshitz transition is a change of the Fermi surface between hole-like and electron-like, centered in cuprates at the $M = (\frac{1}{2}, \frac{1}{2}, 0)$ and $\Gamma = 0$ point of the Brillouin zone, respectively.) At a Lifshitz
transition the transport charges can be expected to change from holes to electrons \cite{transition} with density

$$n^+ \simeq x \quad \rightarrow \quad n^- \simeq 1 - x$$

at \( x < x^0 < x \).

It has been pointed out that the charge-density relation (8), if obtained from Hall coefficient \( R_H \) and magnetoresistivity \( \rho_{xx} \), holds only in the high-magnetic-field limit.\cite{variable} This may explain possible discrepancies between relations (8) and (9). Another issue, not completely resolved, is the Seebeck coefficient, \( S > 0 \), being at odds with the electron-like Fermi surface in the highly overdoped range.\cite{variable2}

In conclusion, no change of charge order is observed beyond closing of the pseudogap. Instead, a significant change of the band structure can be expected, along with a massive reconstruction of the Fermi surface—details of which await clarification—and entrance to the Fermi-liquid phase.

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