Electrostatic Modulation of the Superfluid Density in an Ultrathin $La_{2-x}Sr_xCuO_4$ Film

A. Rüfenacht,[1] J.-P. Locquet,[2] J. Fompeyrine,[2] D. Caimi,[2] and P. Martinoli[1]
[1]*Institut de Physique, Université de Neuchâtel, CH-2000 Neuchâtel, Switzerland*
[2]*IBM Research Division, Zurich Research Laboratory, CH-8803 Rüschlikon, Switzerland*
(Received 6 March 2006; published 9 June 2006)

By capacitively charging an underdoped ultrathin $La_{2-x}Sr_xCuO_4$ film with an electric field applied across a gate insulator with a high dielectric constant, relative changes of the areal superfluid density $n_{s\square}$ of unprecedented strength were observed in measurements of the film kinetic inductance. Although $n_{s\square}$ appears to be substantially reduced by disorder, the data provide, for the first time on the same sample, direct compelling evidence for the Uemura relation $T_c \propto n_{s\square}(T=0)$ in the underdoped regime of copper-oxide superconductors.

DOI: 10.1103/PhysRevLett.96.227002 PACS numbers: 74.62.Yb, 74.25.Nf, 74.78.Bz, 74.90.+n

It is widely accepted that copper-oxide superconductors in the underdoped regime are doped charge transfer insulators whose properties strongly depend on the concentration of the ''free'' electrical carriers. If the doping level is measured by a parameter x expressing the number of free carriers per Cu site in the CuO_2 planes, in the hole-doped materials superconductivity typically sets in at $x \approx 0.05$, reaches its maximum strength at $x \approx 0.15$, and disappears above $x \approx 0.30$. Usually, x is changed by nonisovalent chemical substitution of the antiferromagnetic insulating parent compound ($x = 0$): for instance, by substituting a Sr^{+2} ion for one of the La^{+3} ions in the $La_{2-x}Sr_xCuO_4$ (LSCO) compound studied in this work. However, changing x by chemical substitution implies that the microstructure unavoidably varies from sample to sample. Because of the d-wave symmetry of the order parameter in copper-oxide superconductors, disorder effects may then become relevant and, in thin films, even very pronounced, thereby making the interpretation of experiments probing the doping dependence of various quantities a difficult task. An elegant way to overcome this difficulty relies on the electric-field effect [1], which allows to modulate the free-carrier concentration in the superconducting electrode of a capacitorlike heterostructure by varying the electrostatic field applied across a gate insulator. So far, tunable electric-field-effect devices incorporating a copper-oxide superconductor were almost exclusively studied with conventional transport measurements probing the resistive region above the critical temperature T_c [2–5].

A powerful method to investigate the superconducting state of thin films from the fluctuation-dominated region near T_c down to $T \ll T_c$ is to measure their sheet kinetic inductance L_k [6]. In a two-dimensional (2D) superconductor $L_k^{-1}(T)$ is proportional to the temperature-dependent areal superfluid density $n_{s\square}(T)$: $L_k^{-1}(T) = e^2 n_{s\square}(T)/m^*$, where m^* is the carrier effective mass. Although not all the free carriers may participate to the superconducting condensate, one can nevertheless expect that varying their areal concentration $n_\square$ will somehow affect $n_{s\square}$ and, consequently, L_k^{-1}. Changes of L_k^{-1} reflecting changes of $n_{s\square}$ were indeed observed by Fiory *et al.* [7] in the first electric-field-effect experiment performed on a copper-oxide film. However, since the film thickness was much larger than the Thomas-Fermi charge screening length, the relative changes $\Delta L_k^{-1}/L_k^{-1}$ turned out to be extremely small, typically of the order of $\sim 10^{-5}$. Field-induced relative changes of L_k^{-1} in the range 10^{-4}–10^{-3} were also observed in surface impedance measurements at microwave frequencies [8], whose interpretation, however, was less transparent because of the nonlinear response of the $SrTiO_3$ gate insulator to the applied electric field.

In this Letter we describe an experiment in which the temperature and electric-field dependences of L_k^{-1} were investigated by capacitively charging an epitaxially grown ultrathin (two-unit-cell-thick) LSCO film in the underdoped regime ($x = 0.1$) with an electrostatic field E applied across a gate insulator with a high dielectric constant. We find that the field-induced change $\Delta L_k^{-1}(T, E)$ is a nonmonotonic function of T which, for the highest fields accessible to the experiment ($E \approx 2 \times 10^8$ V/m), reaches a maximum corresponding to $\sim 20\%$ of $L_k^{-1}(0, 0)$ at $T/T_c \approx 0.7$ and saturates to $\sim 10\%$ of $L_k^{-1}(0, 0)$ at very low temperatures. These large modulations offer new interesting opportunities to explore the intriguing superconducting behavior of the copper oxides. As an illustration, the field-induced relative changes $\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0)$ measured at very low temperature were correlated, for the first time on the same sample, with the corresponding relative variations $\Delta T_c(E)/T_c(0)$. Both quantities were found to vary linearly with E and, quite remarkably, $\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0) = \Delta T_c(E)/T_c(0)$, a result providing direct compelling evidence for the validity of Uemura's relation [9] $T_c \propto n_{s\square}(0)/m^*$ for underdoped copper-oxide superconductors. We consider this observation as the central result emerging from this work.

To achieve a substantial electrostatic modulation of $n_{s\square}$ in a thin film, it is essential that its thickness d is of the order of the Thomas-Fermi charge screening length λ_{TF}, the field-induced charges being confined in a layer of thickness $\sim\lambda_{TF}$ near the superconductor-insulator interface. If one relies on a 2D free-electron gas to estimate λ_{TF} for metallic LSCO in the quasi-2D underdoped regime, one finds $\lambda_{TF} = \phi_0(\epsilon_s\epsilon_0 d_s/\pi m_e)^{1/2}$, where d_s is the CuO_2-interlayer distance and ϵ_s the dielectric constant of LSCO (ϕ_0 is the superconducting magnetic flux quantum). Using $d_s = 0.66$ nm and $\epsilon_s \approx 29$ [10] for LSCO, one obtains $\lambda_{TF} \approx 0.5$ nm. This means preparing epitaxial films only a few unit cells thick in the c-axis direction, one unit-cell (UC) corresponding to a thickness of 1.33 nm. Even when grown on an epitaxially optimized substrate like $SrLaAlO_4$ (SLAO) [11], such ultrathin LSCO films have a strongly reduced T_c, superconductivity being completely suppressed in 1-UC-thick films. We have recently demonstrated [12], however, that it is possible to grow with block-by-block molecular beam epitaxy [13] (1–2)-UC-thick LSCO films having T_c in the range (10–20) K by inserting a *normal* (i.e., nonsuperconducting) metallic LSCO buffer layer between the superconducting film and the SLAO substrate, a procedure which apparently helps minimizing the degradation of the film structure at the interface. Being confined to a region of thickness ξ_c (the c-axis coherence length) near the superconductor-normal metal contact, proximity effects weakening superconductivity in the LSCO film are expected to be irrelevant for the 2-UC-thick ($d = 2.66$ nm) LSCO film studied in this work, since $\xi_c \ll d$ (typically, $\xi_c \approx 0.1$–0.2 nm for LSCO in the quasi-2D underdoped regime). Relying on this almost homoepitaxial method, in a first step a trilayer heterostructure consisting of a 12-UC-thick normal LSCO buffer layer ($x = 0.4$), a 2-UC-thick superconducting LSCO film in the underdoped regime ($x = 0.1$), and an amorphous HfO_2 film of thickness $D \approx 15$ nm [14] was grown *in situ* on a monocrystalline SLAO substrate. Hafnium oxide was chosen as gate insulator on account of its high static dielectric constant ϵ, for which values up to $\epsilon \approx 25$ are reported [15], and its large breakdown fields, typically $(6–8) \times 10^8$ V/m [16]. In five subsequent photolithographic steps the trilayer was then patterned in the capacitorlike "mesa structure" shown in Fig. 1. To avoid short circuits, before depositing the top platinum (Pt) gate electrode, a SiO_2 layer was deposited around the mesa after the Pt metallization of the bottom electrodes. In the gate-voltage range $|V_G| \leq 3$ V covered by the experiments, corresponding to electrostatic fields $|E| = (|V_G|/D) \leq (2 \times 10^8)$ V/m, the leakage current was found to be independent of polarity, the maximum leakage current density being $\sim 10^{-4}$ A/cm^2 at $V_G = \pm 3$ V. The inverse sheet kinetic inductance L_k^{-1} of the superconducting LSCO film was extracted from measurements (down to $T \approx 0.5$ K) of the mutual-inductance change of a drive-receive two-coil system caused by the screening supercurrents flowing in the film in response to a small ac excitation at a frequency of 33 kHz [6].

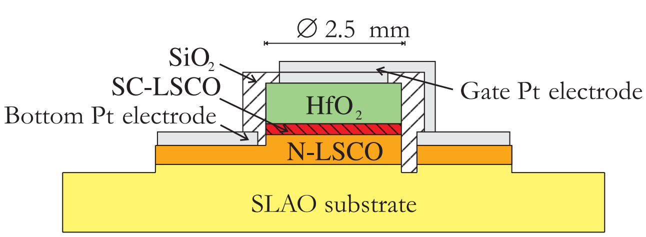

FIG. 1 (color online). Cross section of the multilayer electric-field-effect device. N: normal; SC: superconducting.

In Fig. 2(a) the temperature dependence of L_k^{-1} is shown for $V_G = 0, \pm 3$ V. The data reveal a marked electrostatic modulation of $n_{s\square}(T)$ over the whole temperature range, $L_k^{-1}(T)$ exhibiting a substantial enhancement for hole injection (corresponding to $V_G < 0$) and an almost identical reduction for hole depletion (corresponding to $V_G > 0$). This is qualitatively consistent with the fact that the LSCO film is in the underdoped regime, where $n_{s\square}(T)$ is expected to rise with increasing hole concentration. When plotted against the reduced temperature $t \equiv T/T_c(E)$, where $T_c(E)$

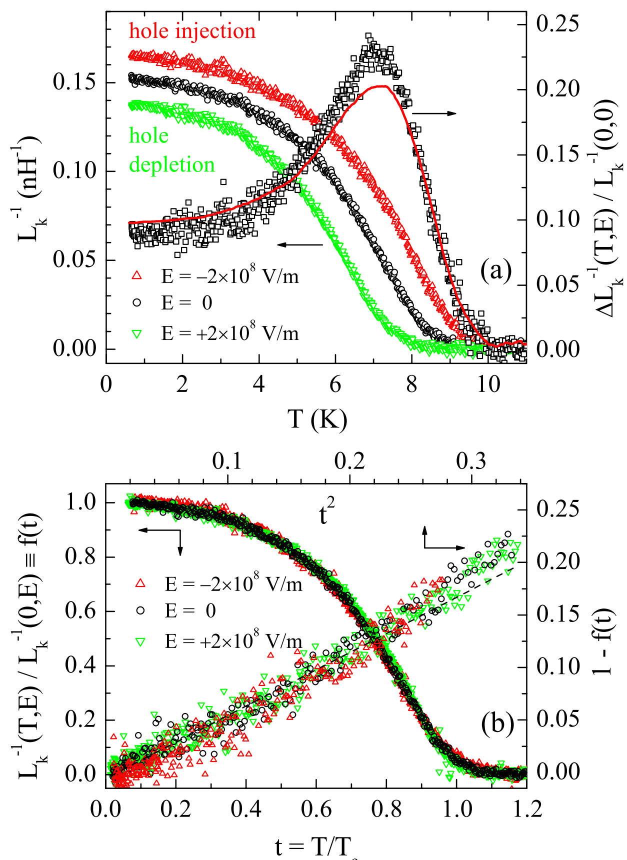

FIG. 2 (color online). (a) Temperature dependence of the inverse kinetic inductance at three values of the gate voltage ($V_G = 0, \pm 3$ V) and of its normalized deviation for $|V_G| = 3$ V from the $V_G = 0$ data of an underdoped ($x = 0.1$) 2-UC-thick LSCO film. The full curve is the prediction of Eq. (3); (b) universal temperature dependence $f(t)$ of the normalized L_k^{-1} vs T curves of (a). Notice the t^2 dependence of $[1 - f(t)]$ at low temperatures ($t < 0.45$) highlighted by the dashed straight line.

was defined *ad hoc* by extrapolating to zero the linear high-temperature portion of the L_k^{-1} versus T curves, the normalized data $L_k^{-1}(T, E)/L_k^{-1}(0, E)$ all collapse on the universal curve $f(t)$ shown in Fig. 2(b). This demonstrates that, at least in the limited doping range explored in this work, $f(t)$ is independent of the carrier concentration, a conclusion unaffected by the choice of the procedure adopted to define $T_c(E)$. In other words, within the framework of a two-fluid model of superconductivity, this means that the ratio $n_{n\square}(T)/n_{s\square}(T)$ between the areal normal-fluid density $n_{n\square}(T)$ of the thermally excited quasiparticles and $n_{s\square}(T)$ is independent of doping. In Fig. 2(b) we also show that, at low temperatures, $[1 - f(t)]$, which is proportional to the change of $n_{n\square}(T)$ with temperature, follows the t^2 dependence predicted by the theory of disordered d-wave superconductors [17].

Deeper insight into the nature of the superconducting state of the copper oxides in the underdoped regime is provided by studying the relationship between the field-induced relative shift $\Delta T_c(E)/T_c(0)$ of the transition temperature and the relative change $\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0)$ of the inverse kinetic inductance at very low temperature, in the limit $T \rightarrow 0$. As shown in Fig. 3, both quantities vary linearly with E in the field-range $E = \pm 2 \times 10^8$ V/m. Most remarkable, however, is the observation that, within experimental accuracy, $\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0) = \Delta T_c(E)/T_c(0)$, a result pointing unambiguously, at least in the limited carrier concentration range covered by our experiment, to the relation $T_c \propto n_{s\square}(0)/m^*$ proposed by Uemura *et al.* [9], but in striking contrast with recent L_k measurements performed on underdoped yttrium-barium-copper-oxide films, where T_c was found roughly proportional to $[n_{s\square}(0)/m^*]^{1/2}$ [18]. We think that this discrepancy might result from sample-to-sample varying disorder in the experiments of Ref. [18], although other explanations can be envisaged. Agreement of our result with a recently proposed alternative scaling relation [19] would imply that the normal-state conductivity at T_c is independent of $n_\square$, which is definitely not the case [20]. We emphasize that the message conveyed by our observation is clearly distinct from that emerging from the field-effect experiments of Ref. [21], in which the relation $T_c \propto n_\square$ was inferred from transport measurements. As a matter of fact, $n_{s\square}(0)$ and $n_\square$ are likely to be related, but not necessarily identical, quantities.

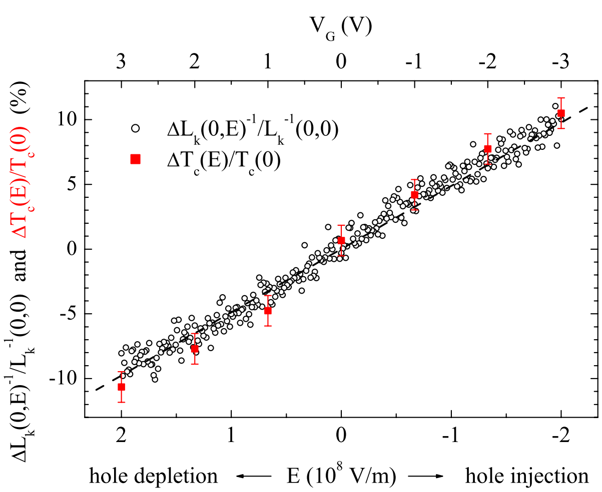

FIG. 3 (color online). Comparison of the field-induced relative changes of the critical temperature and of the inverse kinetic inductance at very low temperature ($T = 0.6$ K, $T/T_c \approx 0.07$) for an underdoped ($x = 0.1$) 2-UC-thick LSCO film as a function of the applied electrostatic field. The dashed straight line is a fit according to Eq. (2).

At this point, it is instructive to compare our results with existing theoretical expressions relating $L_k^{-1}(0)$ to T_c. Assuming $n_{s\square}(0) = n_\square$, the quasiparticle approach [22] predicts $L_k^{-1}(0)/T_c = (k_B e^2/m^* a^2)(3J/t_h)\Delta_0^{-1}$. Typical values for cuprates are $J \approx 130$ meV for the exchange energy and $J/t_h \approx 0.3$ for the ratio between J and the tight-binding hopping integral t_h [22]. Then, since the maximum value Δ_0 of the d-wave energy gap is roughly $J/3 \approx 45$ meV [22], $L_k^{-1}(0)/T_c \approx 0.2$ nH^{-1}/K, using $m^* \approx 2m_e$ [22] and $a = 0.38$ nm for the cell side of the square CuO_2 lattice. An alternative prediction, based on a phase-fluctuation mechanism [23,24], leads, in two dimensions, to $L_k^{-1}(0)/T_c = Ank_B(2\pi/\phi_0)^2$, where A is a constant of order unity and n, the number of CuO_2 planes in the LSCO film, reflects the fact that the measured L_k is the parallel connection of the kinetic inductances of the individual CuO_2 planes. For our 2-UC-thick LSCO film ($n = 4$) this gives $L_k^{-1}(0)/T_c \approx 0.5$ nH^{-1}/K. Both theoretical scenarios lead to values which are at least 1 order of magnitude larger than that actually observed in our experiment [$L_k^{-1}(0)/T_c \approx 0.02$ nH^{-1}/K]. This strongly suggests that disorder not only considerably reduces $n_{s\square}(0)$, but also tends to suppress $n_{s\square}(0)$ more efficiently than T_c, however without altering the proportionality between $L_k^{-1}(0)$ and T_c. In this connection, we notice that the different sensitivity of $L_k^{-1}(0)$ and T_c to disorder is consistent with previous experimental observations and can be explained by a mean-field theory of d-wave superconductors in which the order-parameter spatial variations caused by disorder in short-coherence-length materials are adequately taken into account [25]. For the very same reason (i.e., T_c more robust against disorder than $n_{s\square}$), we argue that the universal "superfluid jump" at T_c predicted by the Berezinskii-Kosterlitz-Thouless theory tends to be suppressed by disorder, a conjecture consistent with the absence of any rapid superfluid drop near T_c in the L_k^{-1} versus T curves of Fig. 2.

In order to explain the $\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0)$ data of Fig. 3 in more quantitative terms, we make the reasonable assumption that the fraction $n_{s\square}(0)/n_\square$ of holes participating to the superconducting condensate at $T = 0$ is independent of their concentration and, consequently, of E. Then, $\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0) = \Delta n_\square(E)/n_\square$. To determine the field-induced change $\Delta n_{\square}(E)$ of the areal hole density $n_{\square}$, we ignore the discrete layered structure of LSCO and assume that it behaves like a 2D metallic continuum, whose charge screening properties are described by the 2D expression of λ_{TF} given above. Relying on this simple model, elementary electrostatics leads to:

$$\Delta n_{\square}(E) = (CD/e)[1 - \exp(-d/\lambda_{\mathrm{TF}})]E, \qquad (1)$$

where $C^{-1} = (\epsilon\epsilon_0/D)^{-1} + (\epsilon_0/\lambda_{\mathrm{TF}})^{-1}$, an expression showing that the capacitance C (per unit surface) of the field-effect structure is the series connection of the ''geometrical'' capacitance $\epsilon\epsilon_0/D$ and the ''interface'' capacitance $\epsilon_0/\lambda_{\mathrm{TF}}$. It should be noticed, however, that other contributions to the interface capacitance [see, for instance, Ref. [26]] are not included in this treatment. In our experiment, $d/\lambda_{\mathrm{TF}} \approx 5$ and it turns out, *a posteriori*, that $\lambda_{\mathrm{TF}} \lesssim D/\epsilon$, so that:

$$\Delta L_k^{-1}(0, E)/L_k^{-1}(0, 0) = \epsilon\epsilon_0 E/en_{\square}. \qquad (2)$$

Expressing $n_{\square}$ in terms of the Sr-concentration $[n_{\square} = (x/a^2 d_s)d = 2.78 \times 10^{18}\ \mathrm{m}^{-2}]$, Eq. (2) can be fitted to the data of Fig. 3 taking $\epsilon \approx 24$, a value in good agreement with that reported for HfO_2 in Ref. [15]. Notice, incidentally, that in deriving Eq. (1) the dielectric constant of HfO_2 was assumed to be independent of E, which is consistent with the linear field dependence of the data of Fig. 3.

Finally, Uemura's relation $L_k^{-1}(0, E) \propto T_c(E)$ and the universality of $f(t)$, the two basic results of this work, can be combined to describe the temperature dependence of the field-induced change $\Delta L_k^{-1}(T, E)$. Expanding $L_k^{-1}(T, E) = L_k^{-1}(0, E) f[T/T(E)]$ about $E = 0$, one obtains:

$$\frac{\Delta L_k^{-1}(T, E)}{L_k^{-1}(0, 0)} = \left[f(t) - t\frac{df}{dt} \right] \frac{\Delta T_c(E)}{T_c(0)}, \qquad (3)$$

where $t = T/T_c(0)$. In Fig. 2(a) the temperature dependence of $\Delta L_k^{-1}(T, E)/L_k^{-1}(0, 0)$, as deduced by averaging the deviations of $L_k^{-1}(T, E)$ [corresponding to $E = \pm(2 \times 10^8)$ V/m] with respect to $L_k^{-1}(T, 0)$, is compared to Eq. (3), where the expression in the brackets was calculated from the universal function $f(t)$ shown in Fig. 2(b). The agreement is very good, thereby demonstrating the overall consistency of our observations.

In conclusion, large electrostatic modulations of the areal superfluid density $n_{s\square}$ of an ultrathin LSCO film in the underdoped regime were observed by charging the film in an accurately devised capacitor structure. The central result emerging from our investigations is that the proportionality between T_c and $n_{s\square}(T = 0)$, empirically proposed by Uemura, was verified, for the first time, on the same sample and is thus unaffected by the uncertainties resulting from sample-to-sample varying disorder of other experimental approaches. When adequately normalized, the $n_{s\square}(T, E)$ data exhibit a universal temperature dependence, which, at low temperatures, follows the theoretical prediction for disordered d-wave superconductors.

We thank H. Beck, A. M. Goldman, A. F. Hebard, S. E. Korshunov, A. E. Koshelev, J. Mannhart, and J.-M. Triscone for clarifying discussions and H. Siegwart for technical assistance. This work was supported by the Swiss National Science Foundation through the National Center of Competence in Research ''Materials with Novel Electronic Properties''.

[1] For a review, see C. H. Ahn *et al.*, Nature (London) **424**, 1015 (2003), and references quoted therein.
[2] J. Mannhart *et al.*, Z. Phys. B **83**, 307 (1991).
[3] X. X. Xi *et al.*, Appl. Phys. Lett. **59**, 3470 (1991).
[4] V. C. Matijasevic *et al.*, Physica (Amsterdam) **235–240C**, 2097 (1994).
[5] D. Matthey *et al.*, Appl. Phys. Lett. **83**, 3758 (2003).
[6] B. Jeanneret *et al.*, Appl. Phys. Lett. **55**, 2336 (1989).
[7] A. T. Fiory *et al.*, Phys. Rev. Lett. **65**, 3441 (1990).
[8] A. T. Findikoglu *et al.*, Appl. Phys. Lett. **63**, 3215 (1993); J. Appl. Phys. **76**, 2937 (1994).
[9] Y. J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).
[10] C. Y. Chen *et al.*, Phys. Rev. B **43**, 392 (1991).
[11] J.-P. Locquet *et al.*, Nature (London) **394**, 453 (1998).
[12] A. Rüfenacht *et al.*, Solid-State Electron. **47**, 2167 (2003).
[13] J.-L. Locquet *et al.*, Appl. Phys. Lett. **64**, 372 (1994).
[14] Z. M. Rittersma *et al.*, J. Appl. Phys. **99**, 024508 (2006).
[15] J. Roberston, Eur. Phys. J. Appl. Phys. **28**, 265 (2004).
[16] M. J. Biercuk *et al.*, Appl. Phys. Lett. **83**, 2405 (2003).
[17] P. J. Hirschfeld and N. D. Goldenfeld, Phys. Rev. B **48**, 4219 (1993).
[18] Y. Zuev *et al.*, Phys. Rev. Lett. **95**, 137002 (2005).
[19] C. C. Homes *et al.*, Nature (London) **430**, 539 (2004).
[20] H. Sato *et al.*, Phys. Rev. B **61**, 12 447 (2000).
[21] X. X. Xi *et al.*, Phys. Rev. Lett. **68**, 1240 (1992).
[22] P. A. Lee and X.-G. Wen, Phys. Rev. Lett. **78**, 4111 (1997); P. A. Lee, Physica (Amsterdam) **317–318C**, 194 (1999).
[23] V. J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995).
[24] T. Schneider, in *The Physics of Superconductors*, edited by K. H. Benneman and J. B. Ketterson (Springer-Verlag, Berlin, 2004), Vol. II, p. 111.
[25] M. Franz *et al.*, Phys. Rev. B **56**, 7882 (1997), and references quoted therein.
[26] G. Singh-Bhalla *et al.*, Appl. Phys. Lett. **83**, 2417 (2003).