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Superfluid density in overdoped cuprates: Thin films versus bulk samples

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Recent study of overdoped $La_{2-x}Sr_xCuO_4$ cuprate superconductor thin films by Božović *et al.* has revealed several unexpected findings, most notably the violation of the BCS description which was believed to adequately describe overdoped cuprates. In particular, it was found that the superfluid density in $La_{2-x}Sr_xCuO_4$ films decreases on the overdoped side as a linear function of critical temperature T_c, which was taken as evidence for the violation of Homes' law. We show explicitly that the law is indeed violated, and as the main reason for violation we find that the superfluid density in Božović's films is suppressed more strongly than in bulk samples. Based on the existing literature data, we show that the superfluid density in bulk cuprate samples does not decrease with doping, but instead tends to saturate on the overdoped side. The result is also supported by our recent measurement of a heavily overdoped bulk $La_{2-x}Sr_xCuO_4$ sample. Moreover, this saturation of superfluid density might not be limited to cuprates, as we find evidence for similar behavior in two pnictide superconductor families. We argue that quantum phase fluctuations play an important role in suppressing the superfluid density in thin films.

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I. INTRODUCTION

A recent finding by Božović *et al.* [1] that behavior of cuprate superconductors on the overdoped side of their phase diagram deviates strongly from the expected BCS behavior has generated a lot of attention. Božović *et al.* analyzed thousands of $La_{2-x}Sr_xCuO_4$ (LSCO) thin films, with thicknesses ranging from 0.66 nm to over 100 nm, and found that on the overdoped side the superfluid density decreases as a linear function of superconducting critical temperature T_c, and eventually goes to zero at the quantum critical point. This is in stark contrast with Homes' law [2]:

$$\rho_s \propto T_c\,\sigma_{\rm dc}, \tag{1}$$

where ρ_s is the superfluid density and $\sigma_{\rm dc}$ is dc conductivity just above T_c. Equation (1) predicts that when dc conductivity increases as a result of doping, the superfluid density should increase as well, assuming that the same percentage of charge carriers condenses. This is in sharp contrast with the results of Božović *et al.*

In Fig. 1 we display the updated Homes plot [2], which includes all the previous data [3], along with Božović's new data on LSCO films shown with open magenta circles. The plot clearly shows that Božović's data does indeed violate Homes' law. The optimally doped film as well as the films close to that doping level are on the scaling line. However, as doping increases on the overdoped side, the points move off the scaling line and progress *perpendicular* to the line. At the highest doping levels, the points take an additional downturn.

Kogan recently offered an explanation of Homes' scaling [4], as well as the deviations from it. He argued that Homes' law is a direct consequence of BCS theory and applies not only to dirty, but also to moderately clean superconductors. Moreover, Kogan showed that for clean superconductors, for which the ratio of superconducting coherence length ξ_0 and the mean free path l is on the order $\xi_0/l \sim 1$ or smaller, deviations from the scaling are expected. He predicted that clean superconductors should move *below* the scaling line. That behavior had indeed been observed previously [3] in Sr_2RuO_4 (three different samples are shown with orange flakes in Fig. 1), which has been known to be a clean-limit superconductor ($\xi_0/l \ll 1$). Similarly, it was shown that elemental niobium in the clean limit (when recrystallized in ultrahigh vacuum) also moves below the scaling line [5]. Božović's LSCO films have been known to be ultraclean, with a mean free path $l \gtrsim 4\ \mu$m (Ref. [1]), exceeding their in-plane coherence length (on the order of 20–30 Å, Ref. [6]) by at least two orders of magnitude. Therefore, it is not surprising that Božović's films violate Homes' law and move *below* the scaling line. However, as we show below, there could be other reasons for the violation of Homes' law.

II. SUPERFLUID DENSITY

In order to explore the violation of Homes' law systematically we have conducted an extensive literature search for the relevant experimental parameters from Eq. (1) (ρ_s, $\sigma_{\rm dc}$, and T_c). Even though studies of the overdoped side are

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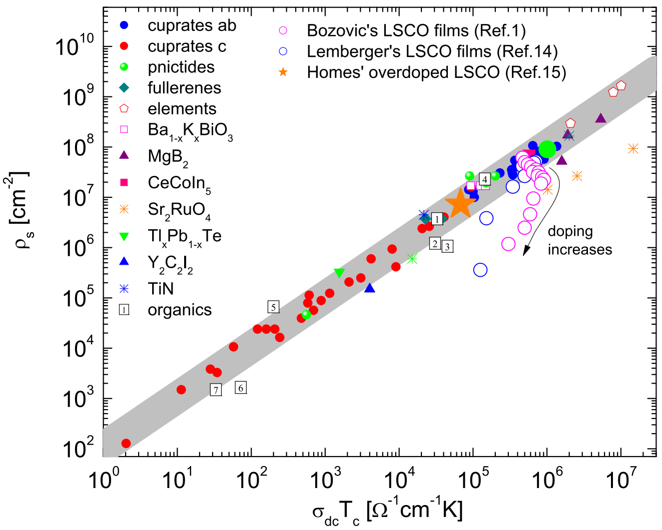

FIG. 1. Updated Homes' plot [2,3], which includes the data on Božović's LSCO films from Ref. [1]. We notice that Božović's data violate the scaling [Eq. (1)]: The points progress perpendicular to the scaling line as doping increases on the overdoped side of the phase diagram. The plot also includes several overdoped LSCO films from Lemberger's group [14], which also violate the scaling. A big orange star represents our recent measurement on a heavily overdoped LSCO single crystal [15]. Chemical formulas for all organic superconductors can be found in Ref. [3].

scarce, we have collected enough data to unveil the trends. Early reports of measurements performed on ceramic, sintered, polycrystalline, or powder samples were not considered here [7]. In Fig. 2(a) we plot the superfluid density ρ_s for two families of cuprate superconductors: LSCO [1,14–17] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) [18]. The superfluid density is shown as a function of reduced critical temperature $T_c/T_{c,\max}$, where $T_{c,\max}$ is the maximal critical temperature (i.e., optimal doping) for a given family [19]. Only the overdoped side of the phase diagram is shown, with $T_c/T_{c,\max} = 1$ being the optimal doping and T_c decreases as doping increases. We point out that Fig. 2(a) includes the data for both thin films (shown with open circles) and bulk single crystals (shown with full circles). The plot also includes our recent infrared (IR) measurement [15] on a heavily overdoped bulk single crystal LSCO with $T_c = 15$ K, combined with a previous IR measurement on optimally doped LSCO [16]. We note that this heavily overdoped sample does not violate the Homes scaling (see Fig. 1).

As shown previously [1] the superfluid density of Božović's films [1] decreases as a linear function of T_c, except at the highest dopings where the dependence becomes parabolic. For comparison, Fig. 2(a) also includes the results on LSCO films from Lemberger's group [14]. Even though only several overdoped films were measured [14], they show very similar absolute values and doping dependence as Božović's films, and they also violate Homes' scaling (see Fig. 1). However, the most striking finding revealed by Fig. 2(a) is that the superfluid density in bulk samples (full symbols) does not seem to decrease with doping. In bulk single crystal LSCO samples the superfluid density tends to saturate [15,16] or increase on the overdoped side [17].

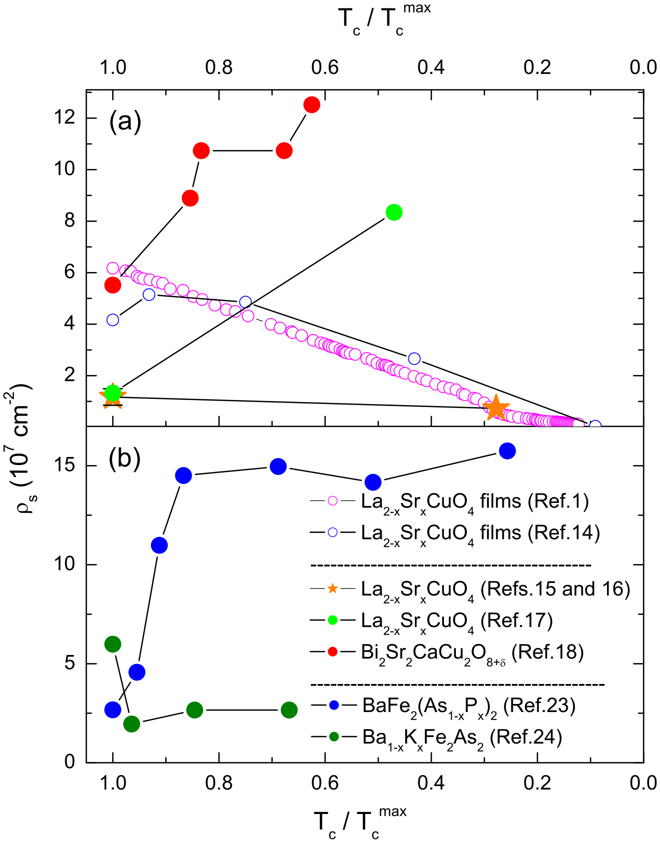

FIG. 2. (a) Superfluid density ρ_s of LSCO and Bi2212 from several different measurements as a function of critical temperature, normalized to the maximum value for a given family $T_c/T_c^{\max}$ (Ref. [19]). Only the overdoped side of the phase diagram is shown and $T_c/T_c^{\max} = 1$ corresponds to optimal doping. We notice that in films (Božović [1] and Lemberger [14] data displayed with open circles) the superfluid density decreases with doping and eventually goes to zero. On the other hand, in bulk samples (full circles) the superfluid density tends to saturate [15,16] or even increase [17]. In Bi2212 the superfluid density also increases with doping [18]. (b) Superfluid density ρ_s of two pnictide families of superconductors [23,24]. They are all bulk samples and they all show saturation of superfluid density.

The superfluid density in Bi2212 also increases with doping [18]. It is clear from Fig. 2(a) that the trend in the doping dependence is different for bulk samples and thin films: the superfluid density in bulk samples, contrary to thin films, does not decrease on the overdoped side.

III. PNICTIDES

The saturation of superfluid density observed in bulk samples [Fig. 2(a)] might not be limited to cuprates [20]. There is evidence that a similar effect is also present in at least two pnictide families. In Fig. 2(b) we show recent data on bulk single crystal pnictides $BaFe_2(As_{1-x}P_x)_2$ [23] and $Ba_{1-x}K_xFe_2As_2$ [24]. Both families reveal saturation of superfluid density on the overdoped side [25]. It can be seen that in $BaFe_2(As_{1-x}P_x)_2$ the saturation persists up to very high doping levels ($T_c/T_c^{\max} \approx 0.25$). We also point out that pnictides are multiband systems with dramatically different

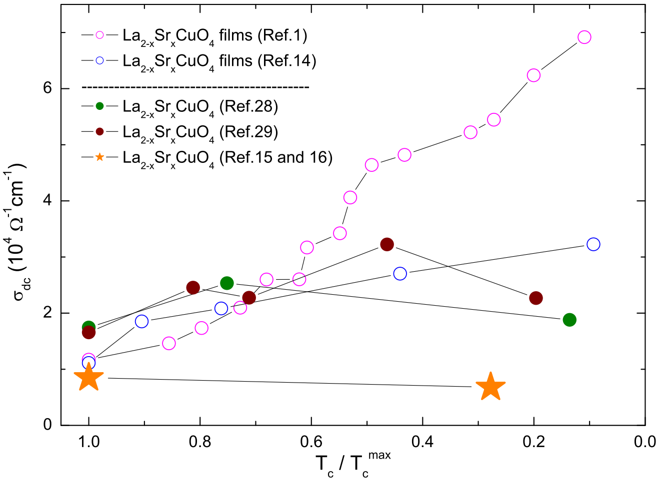

FIG. 3. Normal state conductivity at T_c, $\sigma_{\rm dc}$, for several LSCO bulk samples and films. $\sigma_{\rm dc}$ is shown as a function of critical temperature, normalized to the maximum value for a given family $T_c/T_c^{\rm max}$ (Ref. [19]). Only the overdoped side of the phase diagram is shown and $T_c/T_c^{\rm max} = 1$ corresponds to optimal doping. We notice that Božović films [1] have the strongest doping dependence, and at the highest doping levels their values are several times greater compared to other films or bulk samples. Lemberger films [14] have significantly lower conductivity, even though they have comparable superfluid density [Fig. 2(a)]. On the other hand, bulk samples (full circles) display little doping dependence of their $\sigma_{\rm dc}$. The values of $\sigma_{\rm dc}$ obtained from IR spectroscopy [15,16] (orange stars) are slightly lower.

scattering rates associated with transport in different bands, so they can effectively be superconductors that are in both the clean and dirty limit at the same time [27]. Measurements of pnictide thin films are currently not available, but we hypothesize that on the overdoped side of the phase diagram, just like in the cuprates, their superfluid density might also be reduced compared to bulk samples.

IV. NORMAL STATE CONDUCTIVITY

In this section we analyze and compare the values of normal state conductivity for different LSCO samples. In Fig. 3 we display the values of dc conductivity just above T_c, $\sigma_{\rm dc}$, from several different measurements on LSCO. The plot includes both thin films (open circles) from Božović [1] and Lemberger [14] groups, as well as two sets of measurements on bulk single crystals [28,29] (full circles). We also plot the results of IR measurements on an optimally and a heavily overdoped LSCO single crystals [15,16]. Similar to the superfluid density (Fig. 2) the conductivity is shown as a function of reduced critical temperature $T_c/T_{c,\rm max}$ and only on the overdoped side of the phase diagram. It is immediately clear that Božović's films are superior in terms of their conductivity. They show the strongest doping dependence and at the highest doping levels ($T_c/T_c^{\rm max} \approx 0.1$) their conductivity is approximately three times higher compared to all other samples (bulk or film). We also note that overdoped bulk samples might have issues with inhomogeneities [15], which can result in their conductivity being lower compared to films.

In spite of this finding, we argue that this high normal state conductivity of Božović's films is not the main reason for the violation of Homes' law. Namely, Fig. 1 also includes the data for Lemberger films (open blue circles) which also violate the scaling, i.e., as doping increases they move below the scaling line. Even though their conductivity is several times smaller compared with Božović's films, their superfluid density is comparable and it follows similar doping dependence [Fig. 2(a)]. Therefore, we can conclude that the main reason for Homes' law violation is the suppression of superfluid density in thin films compared with bulk crystals.

V. DATA VARIABILITY

We have previously argued [3] that for accurate scaling the data for Eq. (1) (ρ_s, T_c, and $\sigma_{\rm dc}$) should be taken on the same sample, using the same experimental technique, such as infrared or microwave spectroscopy. Using the data from different sources can lead to conflicting results [3,16,30]. The experimental values for optimally doped LSCO that we have compiled from the literature fully support this argument. To illustrate the point, in Table I we list the values of superfluid density ρ_s for optimally doped LSCO from several different sources. It can be seen that Božović's film has the highest superfluid density ($\rho_{s,\rm max}$), and that other values can be smaller by as much as 80%. We also note that the value of ρ_s obtained in Ref. [31] on Božović's film, but using a different experimental technique, is about 30% smaller. Similarly, Tajima's IR and muon spectroscopy measurements have resulted in superfluid densities that differ by almost a factor of three [16,32]. Therefore, one must not compare the absolute values of superfluid density obtained using different experimental techniques, even when taken on the same sample or film. However, we point out that the measurements in Ref. [1] were performed on thousands of samples, grown and measured using the same procedure, which assures that their relative values (i.e., their *doping* dependence) are reliably extracted and are an intrinsic property of these LSCO films.

VI. DISCUSSION

In this section we discuss possible scenarios that might be able to account for the effects observed above. It has been known for a long time that in thin films of conventional superconductors (such as Sn [33]) the so-called microscopically granular superconductivity might arise. These systems were modeled as Josephson junction arrays and it was shown that quantum fluctuations play an important role in suppressing superconductivity in them [34,35]. Similar ideas were also discussed in relation to superconductivity in the cuprates [36] and they might also apply to overdoped LSCO films. Further support comes from a recent experimental study of heavily overdoped LSCO [37].

More recently, quantum phase fluctuations have been argued to explain strong suppression of superfluid density in Božović's overdoped LSCO films. Schneider employed finite size scaling analysis [38] which uncovered that suppression is consistent with a finite length limited 3D-XY transition

TABLE I. Superfluid density ρ_s of optimally doped LSCO from six different sources. The first three are from thin films, whereas the last three are from bulk single crystals. $\rho_{s,\max}$ is the value from Ref. [1]. Note significant differences in superfluid density for the same film or bulk sample, extracted using two different experimental techniques.

Sample type	Experimental technique	ρ_s ($\times\ 10^7$ cm^{-2})	$\rho_s/\rho_{s,\max}$	Reference
Božović's film	Mutual inductance	6.17	100%	Ref. [1]
Božović's film	THz spectroscopy	4.53	73%	Ref. [31]
Lemberger's film	Mutual inductance	5.97	97%	Ref. [14]
Tajima's single crystal	IR spectroscopy	1.17	19%	Ref. [16]
Tajima's single crystal	Muon spectroscopy	3.05	49%	Ref. [16]
van der Marel's single crystal	IR spectroscopy	1.32	21%	Ref. [17]

[39]; in some films this limiting length is set by the film thickness and in others by inhomogeneities. Moreover, the analysis reveals a crossover from thermal to quantum critical regime as $T_c \to 0$, and Schneider argues that in Božović's overdoped LSCO films the suppression of superfluid density is driven by quantum phase fluctuations.

Additional experimental support for quantum phase fluctuations' driven suppression of superfluid density comes from the recent terahertz spectroscopy measurements on Božović's LSCO films by Mahmood *et al.* [41]. They discovered that below T_c a significant fraction of charge carriers remains uncondensed in a wide Drude-like peak [41] and argued that quantum phase fluctuations play an important role in suppressing the superfluid density.

Based on all these findings we suggest that in thin films quantum confinement (i.e., reduced dimensionality) enhances quantum phase fluctuations, making them more efficient in preventing pair formation and reducing the superfluid density. This might result in suppression of superfluid density in overdoped thin films, compared with bulk samples. If this suggestion is correct, one might expect that similar behavior could also be observed in pnictide thin films.

The final issues that we discuss here are what happens as the quantum critical point ($T_c \to 0$) is approached. In thin films the superfluid density is continuously suppressed and goes to zero as $T_c \to 0$. Based on Fig. 2 we hypothesize that in bulk samples the superfluid density will not be continuously suppressed to zero, but instead will experience an abrupt drop once the critical doping is reached. Measurements on bulk samples with such high doping levels are currently not available.

VII. SUMMARY

In summary, we have explicitly shown that Božović's overdoped LSCO films do indeed violate Homes' law. Analyzing the existing literature data, we have unraveled that the main reason for this violation is the stronger suppression of superfluid density in thin films, compared to bulk single crystals. We hypothesize that in thin films superfluid density is suppressed by quantum phase fluctuations. Our results have uncovered a fundamental difference between the superfluid density in bulk samples and thin films. These findings call for measurements of superfluid density in bulk cuprate samples close to the quantum critical point, i.e., in the region where $T_c \to 0$, as well as on overdoped pnictide films.

Note added in proof: We only recently became aware of two studies Refs. [42,43] reporting superfluid density in bulk single crystals of overedoped Tl-2201. The results of these papers are consistent with our main findings.

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