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Fermi Surface reconstruction and anomalous low temperature resistivity in electron-doped $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$

Tarapada Sarkar^{1,2}, P. R. Mandal^{1,2}, J. S. Higgins^{1,2}, Yi Zhao^{1,2}, Heshan Yu³, Kui Jin³, Richard L. Greene^{1,2}

¹*Center for Nanophysics & Advanced Materials, University of Maryland, College Park, Maryland 20742, USA.*

²*Department of Physics, University of Maryland, College Park, Maryland 20742, USA.*

³*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

We report ab-plane Hall Effect and magnetoresistivity measurements on $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ thin films as a function of doping for magnetic fields up to 14T and temperatures down to 1.8K. A dramatic change in the low temperature (1.8 K) normal state Hall coefficient is found near a doping $\text{Ce}=0.14$. This, along with a nonlinear Hall resistance as a function of magnetic field, suggests that the Fermi surface reconstructs at a critical doping of $\text{Ce}= 0.14$. A competing antiferromagnetic phase is the likely cause of this FSR. Low temperature linear-in-T resistivity is found at $\text{Ce}=0.14$, but anomalously, also at higher doping. We compare our data with similar behavior found in hole-doped cuprates at a doping where the pseudogap ends.

Correspondence: rickg@umd.edu

The mechanism responsible for the high-temperature superconductivity in the cuprates, and the nature of the normal state from which it evolves, is a major unsolved problem in condensed matter physics. Most of the research on cuprates has focused on hole-doped materials, which are more numerous. However, the few examples of electron-doped cuprates offer many advantages for a possible solution to the high- T_c superconductivity problem. The doping phase diagram is much simpler for n-type cuprates. The superconductivity evolves from an antiferromagnetic (AFM) state without the mysterious “pseudogap” state found in the hole-doped cuprates [1,2]. Moreover, the critical magnetic field needed to suppress the superconductivity is much lower for electron-doped cuprates so that the fundamentally important non-superconducting ground state can be probed by experiment. In recent work [3] on $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ (LCCO) a surprising linear in temperature normal state resistivity was discovered at low temperature (30mK to 10K) over a range of Ce doping. The strength of the T-linear resistivity was proportional to the superconducting transition temperature (T_c), which suggested that antiferromagnetic spin fluctuations were responsible for both. Theory suggests that AFM should end at a quantum critical point (QCP) and only at the QCP might a T-linear resistivity be found [4-6]. It is also thought that quantum fluctuations associated with a QCP can lead to superconductivity. In LCCO long range AFM ends at a doping near $\text{Ce}=0.09$ [7] where no T-linear resistivity is found. Short range magnetism persists to higher doping, but where it ends is unknown [8]. Superconductivity exists over the approximate doping range 0.08 to 0.17, with conventional metallic (Fermi liquid) behavior at higher doping. These prior results raise several important questions of relevance to the origin of high-temperature superconductivity (HTSC) in the cuprates: 1) can short range magnetic order produce QCP-like behavior, and 2) can short range order cause a FSR. In this paper we present new low temperature transport measurements on electron-doped LCCO that show the answer to these questions is yes. These surprising experimental conclusions will need new theoretical ideas to reconcile them with the extended range of T-linear resistivity found previously [3].

Our new results on LCCO are also of significance in comparison with recent studies of hole-doped cuprates at very high magnetic fields. In particular, normal state Hall effect measurements done at fields up to 90T on hole-doped YBCO [9] and LSCO [10] have received much attention because they suggested that a Fermi surface reconstruction (FSR) occurs at a

critical doping (p^*) under the superconductivity (SC) dome. This critical doping is also where the mysterious pseudogap (PG) ends. It was found that a large Fermi surface (FS) at $p > p^*$ transitions to a small Fermi surface at $p < p^*$ corresponding to a Hall number n_H which goes from $1+p$ to p . This recent transport work agrees with prior SI-STM [12] and other experiments [13] which suggested a FSR at a doping near 0.19 in hole-doped cuprates. Although the exact cause of the pseudogap is unknown the p^* end point has recently been suggested to be related to the end of spiral AFM [14] or a novel “topological” phase transition [15]. As shown in Fig. 3 (b) of this work, we find a very similar change in Hall number at our suggested FSR in LCCO at a critical doping (p^*) of $Ce=0.14$. But, in our case it is almost certainly short range magnetism that ends at p^* . Since the physics that drives the FSR and the SC is likely to be the same on both sides of the cuprate phase diagram, our results appear to be of considerable significance for a deeper understanding of the HTSC in the cuprates.

LCCO is unique among n-doped cuprates because it can be prepared in thin film form over a wider range of doping, in particular beyond the superconducting dome. However, some prior work on other n-type cuprates has suggested that an AFM QCP exists in n-doped cuprates. For example, in $(Nd,Ce)_2CuO_4$ (NCCO), ARPES [16,17] and Shubnikov quantum oscillation (QO) experiments [18,19] suggest a FSR at $Ce= 0.17$. In contrast, a normal state Hall Effect critical doping is reported to be near optimal doping ($Ce= 0.145$) [20], very close to where the long range order AFM ends, but rather different from where quantum oscillation (QO) experiments suggest that the FS reconstructs (i.e., $Ce =0.17$). In $(Pr,Ce)_2CuO_4$ (PCCO) the Hall Effect shows a critical doping at $Ce= 0.17$ [21] but no QO or ARPES studies have been done on PCCO over an extended doping range. Also, T-linear resistivity is only found at one doping in NCCO and PCCO. This behavior of NCCO and PCCO is not fully understood and is another significant motivation for our present transport study of LCCO.

Figure 1(a) displays the ab -plane resistivity (ρ_{xx}) versus temperature T for six LCCO c -axis oriented films at $H=0$. The resistive superconducting transition T_c has the similar trend as reported earlier [3]. Figure 1(b) illustrates the temperature dependent resistivity to show the normal state behavior of $x = 0.13$ and 0.14 compositions at an applied magnetic field of $H > H_{c2}$. The 13% doped sample shows an upturn at low temperatures starting from 17 K and tends to saturation at low temperatures as observed for other dopings ($x=0.11, 0.10$). The sample 0.08 has an upturn at low temperatures, however it does not saturate at low temperatures unlike the

samples $0.10 \geq x \geq 0.13$ (see ref. 36 (Fig.S1)). The minima of the normal state resistivity at low temperatures are defined as $T_{\rho_{\min}}$ (T at ρ minima) shown in figure 1(b). The 14% doped sample does not show any upturn down to 400 mK. As found previously [3], a low temperature T-linear resistivity is found for Ce doping above 0.14 for doping within the superconducting dome. Our similar data for Ce=0.15 and 0.16 is shown in Fig.1(c)

In Fig. 2 we show the normal state Hall coefficient of LCCO films as a function of temperature (measured from 100 K to 1.8 K) for different Ce doping. The absolute value of the Hall coefficient measured at 14 T jumps dramatically between 13 % and 14% doping. The Hall coefficient of the films with doping $x \geq 0.14$ shows a positive value which is constant below 10 K, and there is a sign change at 1.8 K between doping 0.13 and 0.14. The Hall coefficient for samples $0.10 < x < 0.13$ as a function of temperature shows a peak ($T_{RH_{\max}}$) and starts to fall at a temperature which depends on the doping. The dotted black lines are an extrapolation to $T=0$ under an assumption of no FSR and that all the samples have behavior similar to the overdoped samples ($x \geq 0.14$.)

Figure 3(a) displays the temperature vs doping (Ce) phase diagram of $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$. The hatched regime is the AFM measured by in-plane angular magnetoresistance ending at $x=0.14$ [8]. The yellow regime is the superconducting dome. The normal state in-plane resistivity minima, $T_{\rho_{\min}}$ is determined from the derivative ($d\rho/dT$). The normal state in-plane Hall resistivity maxima, $T_{RH_{\max}}$, ends at $x=0.14$. The estimated FSR line T_{FSR} (solid blue line) separates the large Fermi surface region from the reconstructed FS as a function of doping. The dotted blue line is the extrapolation of T_{FSR} assuming that $T_{\rho_{\min}}$ is due to only to the FSR.

Figure 4 displays the in-plane electrical resistivity ρ of two LCCO samples as a function of temperature, with doping x as indicated. The red curve is data taken in zero magnetic field ($H = 0$). The black curve is the fitted data of the red curve using $\rho(T) = \rho_0 + AT^n$ (ρ_0 is the residual resistivity (45 $\mu\Omega\text{-cm}$ for 0.11, 23 $\mu\Omega\text{-cm}$ for 0.13), $n=2$) above T_c and has been extrapolated to $T \rightarrow 0$ to get ρ_0 assuming there is no upturn. The green line is the normal state resistivity measured at 10 T with $\rho(0)$ (73 $\mu\Omega\text{-cm}$ for 0.11, 27 $\mu\Omega\text{-cm}$ for 0.13) its extrapolation to $T=0$.

In electron doped cuprates commensurate (π, π) spin density wave (SDW) order has been detected by muon spin rotation and neutron diffraction [1]. This SDW order (long range or short range) exists over a wide range of doping starting at the undoped AFM state and vanishing at a critical doping x_c , where the resistivity minima [21,22] and in-plane angular magnetoresistance also vanish [23]. Theory [24,6] suggests that there should be a quantum critical point separating the overdoped paramagnetic state, with a large Fermi surface, from the SDW state with a reconstructed Fermi surface of small electron and hole pockets. This is experimentally suggested in electron doped NCCO and PCCO near optimal doping by low temperature QO [19,20] and ARPES measurements [17,18]. A FSR was also suggested by earlier normal state Hall measurement on PCCO, where an abrupt drop of the Hall coefficient and sign change was found at 300 mK as one approached optimal electron doping from the overdoped side [21].

As shown in figure 2 the normal state Hall coefficient at 1.8 K for LCCO as a function of doping suddenly drops and changes sign between 0.13 and 0.14, which in analogy with PCCO, strongly suggests a Fermi surface reconstruction at $x=0.14$. The 2D Fermi surface of most cuprates is well established from ARPES and QO experiments. For n-type at higher doping, the FS is a large hole-like cylinder and for underdoped the FS has electron pockets. From theory [26] the Hall number ($n_H=V/eR_H$) in the electron doped cuprates should follow $n_H=1-x$ at doping above SDW reconstruction and $n_H=-x$ for the under doped regime well below the FSR. Our data for LCCO, shown in Fig. 3 (b), is in good qualitative agreement with this, however QO and ARPES experiment have not yet been done on LCCO. This is the same behavior as found recently in hole-doped cuprates at very high magnetic fields, where the Hall coefficient goes from $1+p$ in the overdoped region to p in the lower doped region [9,11]. This suggested a low temperature ($T=0K$) FSR at a critical doping of p^* , the doping where the pseudogap state ends. Since the FSR in the n-type cuprates is caused by the onset of short range AFM (when coming from the overdoped side), it may well be that a related short range order can reconstruct the FS in hole-doped cuprates.

As also shown in Fig 3(b) the Hall number deviates from the $1-x$ line for the higher-doped samples. The carrier density has been calculated assuming one band transport, which is supported by a linear in field Hall resistivity for over doped and heavily underdoped samples (see Ref 35 and 36 (Fig. SI)). But, we can fit the data with $n_H=1-bx$, where b is a correction parameter of 1.74. We have defined the b as a correction factor in the doping concentration. In

electron-doped cuprates the doping dependence depends on the Ce content and the oxygen content. To achieve the optimal properties the n-type cuprates are annealed in vacuum, which can create oxygen vacancies, so that $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ should really be written as $\text{La}_{2-x}\text{Ce}_x\text{CuO}_{4-\square}$. Hence, we are changing two parameters to get the optimal superconductivity. Any change in \square will affect the true carrier concentration. The oxygen vacancy effectively adds electrons to the system, i.e, x become bx. So the actual doping in the system could be higher than that of the Ce content (x). We take b as a correction factor to the carrier density due to any contribution from oxygen vacancies.

The difference between $n_H (=1-x)$ and $n'_H (=1-bx)$ is the change in the Hall number due to oxygen vacancies. Now if we take the 15 % sample to calculate the difference in Hall Number we find $\Delta n_H = (n_H - n'_H) = 0.11$. If we convert this number to a change in Hall coefficient we find $\Delta R_H = (R'_H - R_H) = (V/en'_H - V/en_H) = 1.0 \times 10^{-10} \Omega\text{m/T}$. Is this reasonable? Higgins et al. [25] reported that changes in the oxygen content in over doped Ce=0.17 PCCO can change the value of R_H from 5.5×10^{-10} to 7.5×10^{-10} ($\Omega\text{m/T}$), which is about 2 times higher than what we estimate for LCCO. Thus it is quite reasonable that our change in carrier number 1-x to 1-bx could be caused by oxygen vacancies.

Another possible origin of the deviation from 1-x carrier number is the shape of the Fermi surface for doping above the FSR. In the theory of Lin and Millis [24] for the Hall effect of n-type cuprates they found the Fermi surface shape could affect the value of the Hall number, but not the slope b. Our data suggests that the oxygen deficiency is the more likely explanation for the deviation in R_H at higher doping. The Hall number for under doped samples $0.08 \leq x \leq 0.11$ follows $n_H = -x$. The deviation of the measured Hall coefficient from the $n_H = -x$ line is negligibly small, i.e, no oxygen vacancy correction needed. The reason for this is not clear, but it could be that below the FSR the oxygen vacancy formation energy is higher when electron carriers are dominant. The doping near the FSR (Ce=0.13) gives a very high negative value of n_H . But at this doping LCCO has two types of carriers. So we do not expect a simple one carrier R_H for this doping to fit on either line in main text Fig. 3(b).

The Hall coefficient of 0.13 samples goes through a maximum at 17.5 K (where the short range AFM regime starts for this doping) and starts to drop from positive to negative. The behavior of the Hall coefficient strongly suggests that if there was no Fermi surface

reconstruction the Hall coefficient would roughly follow the black dotted line shown in Fig. 2(b). The difference between the black dotted line and the measured solid line is caused by loss of carriers below the FSR. So one can surmise that the Fermi surface reconstruction starts at temperature 17.5 K for $x=0.13$. We can use the Hall coefficient maxima as the temperature where the FSR starts for each doping (0.11 at 27.5 K and 0.10 at 35 K) as temperature decreases. This low temperature drop of Hall coefficient, seen in samples with $x < 0.14$ can be attributed to the FSR due to SDW (AFM) order below T_{FSR} in the hatched regime shown in Fig. 3(a). All over doped samples ($x \geq 0.14$) should have a large hole like-FS at low temperatures. This needs to be confirmed by ARPES and/or QO experiments in the future. One could argue that AFM fluctuations modify the current direction from the Fermi velocity direction, which could result in a deviation of the Hall coefficient [26]. So the actual carrier number could be different than the Hall measured carrier number. However at low temperatures, Hall coefficient is independent of temperatures as shown in figure 2 and shows linear behavior as a function of magnetic field (except $x=0.13$), which invalidates any significant role of AFM fluctuation in the Hall coefficient. The dramatic doping dependent change that we observe in the low temperature Hall number is more likely due to a Fermi surface reconstruction than the AFM fluctuation mechanism. The AFM fluctuations are there at all dopings, as shown by E. M. Motoyama et al. in NCCO[2], so one would not expect any dramatic change in Hall number at $x=0.14$.

We now discuss some features of the normal state resistivity. As shown in Fig. 1(c), we find a normal state low temperature linear in T resistivity for a range of Ce doping at, and above, the FSR. Our data here is in accord with resistivity measured previously to even lower temperatures [3]. This is a very anomalous and unexplained resistivity behavior. A T -linear resistivity at the FSR doping can be understood as scattering associated with the fluctuations at temperatures above a QCP, but similar very low temperature behavior at higher doping can not be explained by the usual quantum critical theory [5,6]. Our results suggest that the FSR and the T -linear resistivity are closely connected, but the exact relation is a mystery. A doping range of T -linear resistivity has also been observed in hole-doped cuprates [27,28] at, and above, the pseudogap end point. However in contrast to n-type LCCO, it has not been possible to apply large enough magnetic fields to probe the normal state at very low temperatures, i.e, to access the ground state. Nevertheless, the very similar behavior in electron and hole-doped cuprates,

suggests that the close connection between a FSR and T linear resistivity are crucial to understanding the HTSC. The temperature dependent *ab*-plane resistivity exhibits a resistivity minimum at low temperatures for samples $x \leq 0.13$ and no minimum for higher doping. This is a well-known feature of all cuprate superconductors. In very under doped ($x=0.05$ and 0.10) PCCO the low-temperature resistivity upturn was attributed to 2D weak localization [29]. However, the resistivity tends to saturate as the temperature approaches zero for samples near the FSR. This low temperature saturation cannot be explained by 2D weak localization where the resistivity should obey $\rho \propto \log T$. Later, the upturn observed in PCCO and NCCO was attributed to a Kondo effect due to scattering of conduction electrons by unpaired Cu^+ spins [30]. But, Dagan et al. [31] found that for PCCO all doping below the FSR show an anisotropic magnetoresistance. Since this rules out Kondo scattering Dagan et al. suggested another form of spin scattering, with the spin linked to the AFM, as the cause of the upturn. This explanation has received support in a theoretical proposal by Chen et al. [32].

Here, we suggest an alternative explanation for the doping close to the FSR. We note that the $T_{RH_{\max}}$ of the Hall coefficient as a function temperature and $T_{p\min}$ of the resistivity of LCCO are at the same temperature for the samples $0.010 \leq x \leq 0.13$ as shown in Fig. 3(a). This correlation strongly suggests that the low temperature resistivity upturn is due to carrier and mobility changes below the Fermi surface reconstruction. For the doping near the FSR we try an analysis similar to that done recently in hole-doped cuprates [10]. We take $1/\rho = ne\mu$ for one carrier transport and we assume that the mobility does not change due to the FSR. As $T \rightarrow 0$ $n_p(\text{with FSR})/n(\text{without FSR}) = \rho_0 / \rho(0)$ where ρ_0 is the residual resistance assuming no FSR at $T \rightarrow 0$ and $\rho(0)$ is the resistivity due to loss of carriers associated with the FSR (see Fig. 4). So $n_p = n(\rho_0 / \rho(0))$. For the large Fermi surface $n=1-x$, thus $n_p = (1-x)(\rho_0 / \rho(0))$. This n_p should be the Hall number below the FSR. The experimental value of $\rho_0/\rho(0)$ is 0.62 and 0.85 for $x=0.11$ and $x=0.13$ samples respectively. Calculating n_p using the above expression gives 0.55 for $x=0.11$ and 0.74 for $x=0.13$. The measured values of n_H are 0.13 for $x=0.11$ and 6.2 for $x=0.13$, where $n_H = V/eR_H$ (V is volume per copper, e charge of the carrier and R_H the measured Hall coefficient). If the size of the upturn only depended on the loss of carriers then the values of n_H and n_p should be the same. Here we show an alternate calculation to correlate the change of the resistivity with the drop of Hall coefficient (difference between the dotted black line at $T \rightarrow 0$ ($R_{H(0)}$) assuming no FSR and measured solid line $T \rightarrow 0$ ($R_H(0)$) as shown in Fig. 2). If we consider the change in the

resistivity is only due to a loss of carriers then, $1/\rho = ne\mu = V/\mu R_H$. So $R_{H(0)}/R_H(0) = \rho_0/\rho(0)$. The value of $R_{H(0)}/R_H(0)$ (0.067 for $x=0.11$) is one order smaller than the $\rho_0/\rho(0)$ (0.616). So the resistivity upturn at low temperatures cannot be explained only by loss of carriers. There must also be a mobility change. This experimental result is supported by a recent theory paper from Sachdev's group [33]. This is not at all surprising for the $x=0.13$ doping since this doping clearly has two types of carriers and cannot be explained by a one band model (see SI). Thus the size of the upturn in the normal state resistivity in electron-doped cuprates is more complex than its counterpart hole-doped materials whose resistivity upturn has been explained only by a drop of carrier density [10].

The low temperature upturn seen in heavily under doped n-type samples cannot be explained by the FSR alone. The heavily under doped samples, unlike optimal and slightly under doped samples, do not show a low temperature resistivity saturation as temperatures approaches to zero (see ref 34 and 36(Fig. S2)). The resistivity of these samples is two orders of magnitude higher than optimal or slightly higher doped samples at low temperatures. For these samples the upturn in normal state resistivity is probably a combination of the FSR and disorder localization which gives logarithmic increase of resistivity as temperatures tends to zero.

In conclusion, we have performed low temperature, normal state ($H > H_{c2}$), ab-plane resistivity and Hall effect measurements on electron-doped $La_{2-x}Ce_xCuO_4$ as a function of doping. Our results give very strong evidence for a Fermi surface reconstruction (FSR) at $x=0.14$. The low temperature resistivity shows an upturn below $x=0.14$ and the Hall number as a function of doping drops at 0.14 from $1-x$ to $-x$. The Hall resistivity at $0.18 \geq x \geq 0.14$ and $0.11 \geq x \geq 0.08$ is linear with magnetic field and at $x=0.13$ becomes nonlinear, more evidence for a change in the FS and the existence of two types of carriers at this doping. We find a low temperature linear-in-T resistivity for an extended range of doping beyond the FSR doping. This anomalous behavior is unexplained, but it appears to impact the high $-T_c$ superconductivity found in zero magnetic field. The low temperature resistivity upturn found for doping below 0.14 can be explained by a change in carrier number and mobility below the FSR. Our work shows that there are the striking experimental similarities between the transport properties of electron and hole-doped copper oxides and provides evidence that the normal state near the FSR doping

is similar in all the cuprates. The cause of the FSR is a commensurate spin density wave in the n-doped cuprates but is yet to be determined in the hole-doped cuprates.

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Figure Captions

Figure 1. (a) (color online). *ab*-plane resistivity versus temperature for $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ films with various Ce doping. (b) The normal state *ab*-plane resistivity versus temperature in a magnetic field of $H > H_{c2}$ applied parallel to the *c* axis for $x=0.13$ (8 T) and $x=0.14$ (6 T), inset shows linear fit for $x=0.14$ (5 K to 400mK). (c) Normal state resistivity below 20 K for $x=0.15$ and 0.16 with linear fit.

Figure 2. (a), (b). The Hall coefficient versus temperature for $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$ films with various Ce doping (x) measured at a magnetic field of 14 T (solid lines). The dotted black lines are an extrapolation assuming no Fermi surface reconstruction (FSR).

Figure 3. (a) Temperature vs doping (Ce) phase diagram of $\text{La}_{2-x}\text{Ce}_x\text{CuO}_4$. The hatched regime is the AFM region measured by in-plane magnetoresistance ending at $x=0.14$ (ref-[8]). Yellow regime is the superconducting dome. $T_{\rho_{\min}}$ (Black filled circle) is the normal state in-plane resistivity minima ending at $x=0.14$. $T_{R_{H\max}}$ (Hollow red circle) is the normal state in-plane Hall resistivity maxima ending at $x=0.14$. T_{FSR} is the FSR line (solid blue line) which separate the large Fermi surface from the reconstructed Fermi surface. Dotted blue line is the extrapolation of T_{FSR} . (b) Hall number $n_H = V/eR_H$ at 1.8 K as a function of Ce doping with single carrier fitting $n_H = 1-x$ and $n_H = -x$. Red solid line is the $n_H = 1-bx$ fitting where b is a fitting parameter. The gray data points of 0.08 and 0.09 Ce doping are taken from (ref [35]). Error bars are coming from the error in the film thickness measurement

Figure 4. In-plane electrical resistivity (ρ) of two LCCO samples as a function of temperature, with doping x as indicated. The red curve is data taken in zero magnetic field ($H = 0$). The black curve is the fitted data of The red curve above T_c and is extrapolated to $T \rightarrow 0$ to get ρ_0 . The green line is the normal state resistivity measured at 10 T with $\rho(0)$ the normal state resistivity at $T \rightarrow 0$.

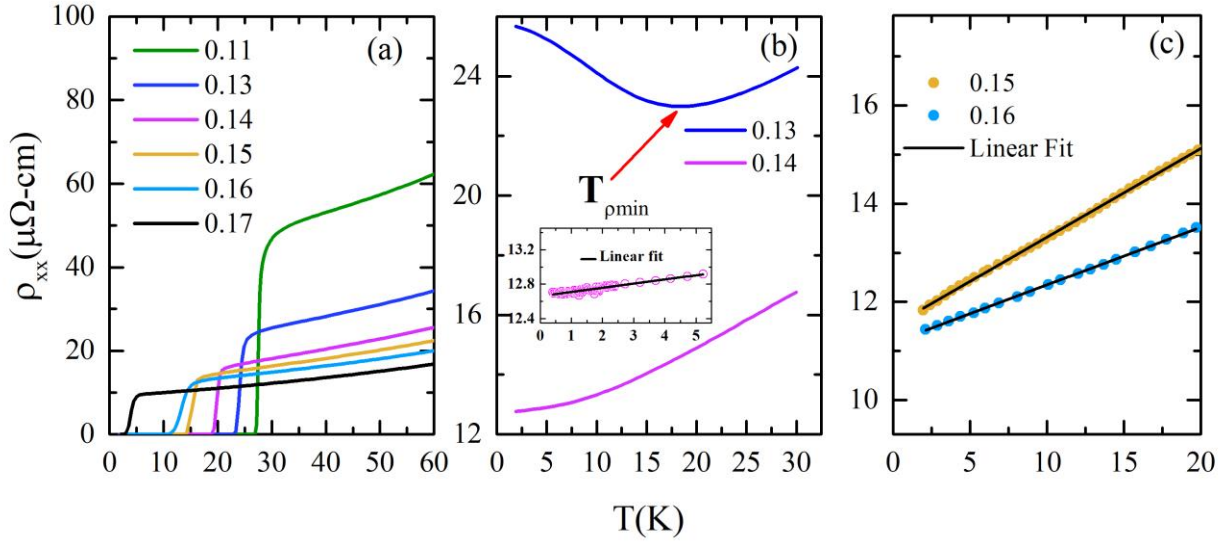


Figure 1. Tarapada Sarkar *et al.*

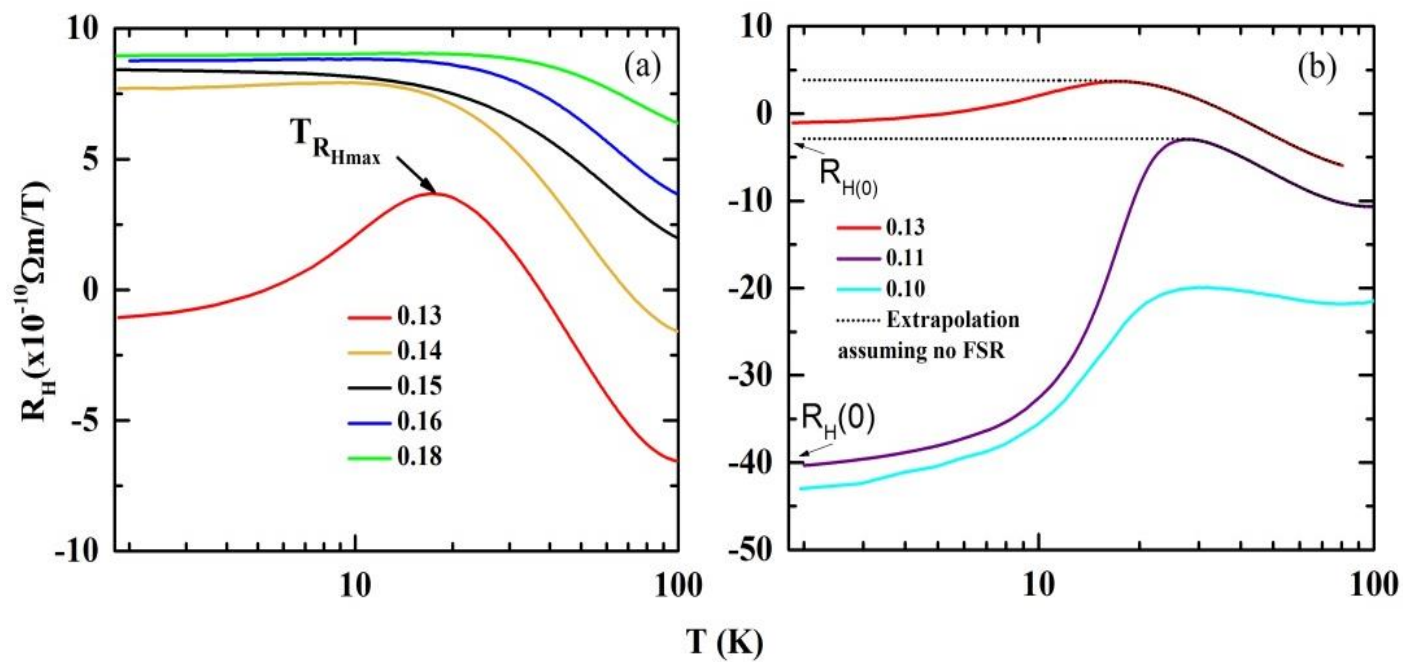


Figure 2. Tarapada Sarkar *et al.*

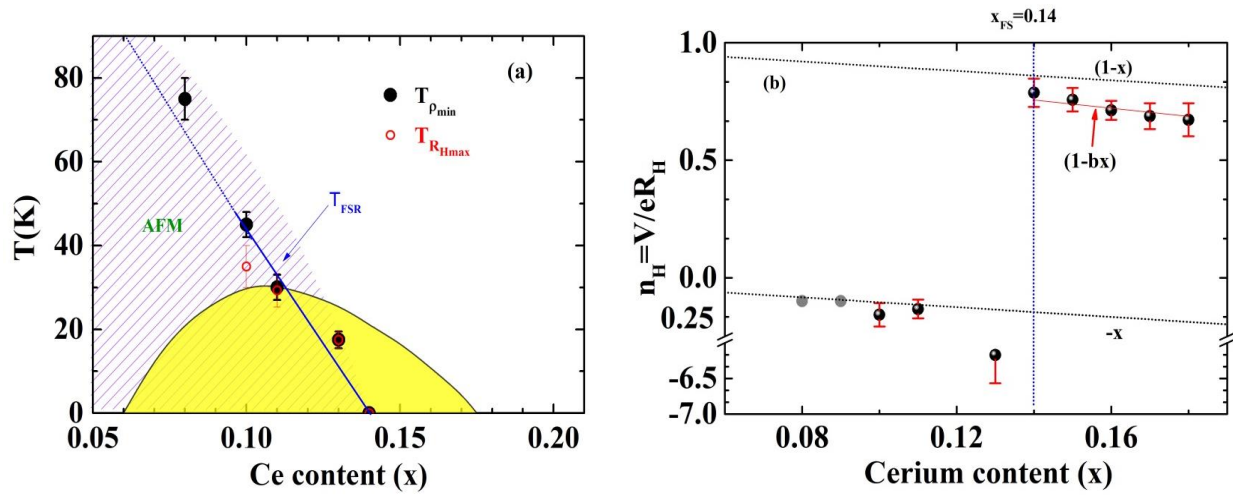


Figure 3. Tarapada Sarkar *et al.*

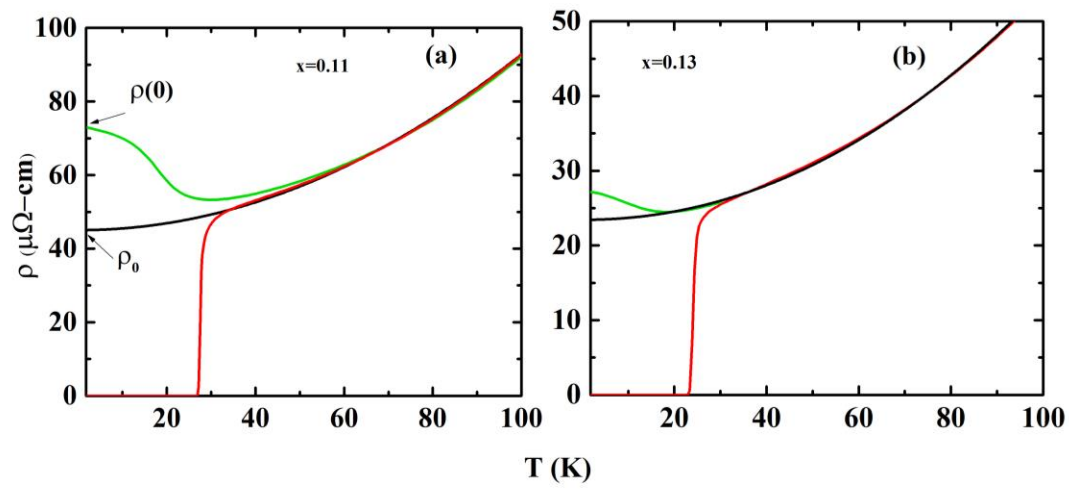


Figure 4. Tarapada Sarkar *et al.*