Anomalous Fermi liquid behavior of overdoped high- T_c superconductors

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According to a generic temperature versus carrier-doping (T-p) phase diagram of high-temperature superconductors (HTSC), it has been proposed that as doping increases to the overdoped region, they approach gradually a conventional (canonical) Fermi liquid. However, Hall effect measurements in several systems reported by different authors show a still strong *T* dependence in overdoped samples. We report here electrical transport measurements of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films presenting a temperature dependence of the Hall constant, R_H , which does not present a gradual transition towards the *T*-independent behavior of a canonical Fermi liquid. Instead, the *T* dependence passes by a minimum near optimal doping and then increases again in the overdoped region. We discuss the theoretical predictions from two representative Fermi liquid models and show that they cannot give a satisfactory explanation to our data. We conclude that this region of the phase diagram in YBCO, as in most HTSC, is not a canonical Fermi liquid, therefore we call it an anomalous Fermi liquid.

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I. INTRODUCTION

A. Phase diagram

High-temperature superconductors (HTSC) are known to undergo fundamental changes in some of their properties as the carrier density (doping, p) is changed. At low doping we have an insulating antiferromagnetic phase (AFM), possibly a Mott-Hubbard insulator, which gradually disappears upon increase of doping, allowing for an anomalous metallic phase presenting superconductivity above a certain doping level. Figure 1 presents such a generic phase diagram of HTSC for hole doping.^{1,2} Apart from the intensive research on the superconductive (SC) region initiated after the discovery of HTSC in 1986, the surrounding normal phases began to receive special attention during the past decade. The existence of a pseudogap (PG) below the temperature $T^*(p)$ is today a well established fact in the underdoped regime $p < p_{op}$ (p_{op} ≈ 0.15), confirmed by different experimental techniques, such as nuclear magnetic resonance (NMR), angle-resolved photoemission spectroscopy (ARPES), specific-heat measurements, tunneling, etc., as reviewed in Refs. 3 and 4. The "strange metal" or marginal Fermi liquid (MFL) phase has been also well characterized experimentally, and given a detailed theoretical explanation.⁵ However, the canonical Fermi liquid (FL) phase, as defined in Ref. 6, which is supposed to exist below a temperature $T_f(p)$ (Refs. 1, 2, and 7–9) in the overdoped region $(p > p_{op})$, still rests on speculative grounds with contradictory experimental data and no clear theoretical understanding. We want to provide additional experimental data and discussion in order to shed more light on the nature of this region. Our main conclusion is that this phase is not a canonical FL.

B. Resistivity

The temperature T^* can be determined from resistivity versus temperature measurements as the point where the resistivity, ρ , departs (decreases) from a linear behavior in un-

derdoped samples.¹⁰ The drop of ρ is understood to be a consequence of the opening of a PG in the interaction at the origin of carrier scattering, for instance spin fluctuation.^{11,12} In a similar way, T_f can be obtained from resistivity measurements as the departure from linearity (increase) in overdoped samples.⁸ The existence of a certain anomaly along the line T_f has been theoretically suggested by Friedel and Kohmoto,⁹ and a new phase below the same line was demonstrated by ARPES measurements.⁸ The upturn in ρ versus T has been fitted to $\rho = a + bT^2$,¹³ which would imply the emergence of FL behavior assuming a dominant fermionfermion scattering.⁶ However, this conclusion is not quite clear since the upturn occurs at temperatures much higher than in usual metals. Furthermore, deviations from the above quadratic law have been observed, which contradict the FL assumption. For instance, Proust et al.14 have reported a behavior $\rho = \rho_o + \alpha T + \beta T^2$ for Tl-2201 with a substantial linear term ($\alpha T > \beta T^2$). Naqib *et al.*¹⁵ also reported a dependence



FIG. 1. General T-p phase diagram for hole doping in HTSC. Phases: Antiferromagnetic (AFM), pseudogap (PG), marginal Fermi liquid (MFL), superconductor (SC), and Fermi liquid (FL). The question mark in the FL phase indicates it is not confirmed.

 $\rho = \rho_o + aT^m$ for their measured samples $Y_{1-x}Ca_xBa_2(Cu_{1-y}Zn_y)_3O_{7-\delta}$, as well as in samples measured (LSCO),18 other authors: $La_{2-x}Sr_{x}CuO_{4}$ by $Bi_2Sr_{1.6}La_{0.4}CuO_y$,¹⁹ YBa₂Cu₃O_{7- δ} (YBCO-123),¹⁶ and $Tl_2Ba_2CuO_6$ (TBCO-221).²⁰ In all cases, *m* ranges from 1.1 at $p \sim 0.2$ to 1.3 for $p \sim 0.25$. A fractional power law would be a clear indication of non-FL behavior and has been associated with the hypothesis of a quantum critical point (QCP),¹⁵ in analogy with the situation in heavy fermions.²¹ We argue in Sec. V that the increase of ρ over linearity in overdoped samples is not necessarily related to a phase transition or a crossover, but instead may be a consequence of the physical requirement of non-negative resistivity at low temperatures.

C. Hall constant

In contrast to the T-independent canonical FL behavior, measurements of R_H in different HTSC materials present a strong T dependence. R_H rises as temperature goes down below room temperature, presenting a peak near T_c , before the normal-superconductor transition. The temperature dependence, excluding the peak itself, can be fitted to the equation $R_H(T) = R_H^{\infty} + \beta/T$,^{22,23} where the fit parameters depend on doping. FL theories of the normal state of the cuprates assume that the T dependence of R_H decreases continuously with doping in all HTSC.^{6,13} However, an objective observation of published data shows, on the one hand, that evolution of $R_H(T)$ with doping presents important differences between different HTSC systems (see discussion in Sec. V). On the other hand, none of them fits into the canonical FL picture. The case of YBCO-123,²⁵ systematically presented in this report in Sec. III B, shows a peculiar nonmonotonic behavior.

D. Hall angle

The Hall cotangent $\cot(\theta_H) = \rho_{xx}/\rho_{xy}$ presents experimentally a robust *T* dependence, almost independent of doping, which can be fitted by $\cot(\theta_H) = A + BT^2$.^{17,25} Some deviations from the quadratic *T* dependence have been observed, especially for doping different from optimal. Wuyts *et al.*¹⁶ present results for YBCO-123 films showing that *B* increases with doping above the optimal level. This result is contrary to what would be expected if the system were to approach the canonical FL. Konstantinovic *et al.*¹⁹ reported measurements of $\cot(\theta_H)$ in BSCCO-2212 and Bi₂Sr_{1.6}La_{0.4}Cu₁O, where they find the dependence $\cot(\theta_H) = A + BT^{\gamma}$, with γ varying with doping from ~2 for underdoped samples, going down continuously to ~1.7 in the overdoped region.

This broad spectrum of results suggests that HTSC may not approach continuously a canonical FL as doping increases and that evolution with doping varies from system to system. To our knowledge, no conclusive experimental evidence exists of a canonical FL behavior in overdoped samples, except perhaps in TlBaCuO, where $R_H(T)$ presents a weak T dependence and additionally the Wiedeman-Franz law has been verified.¹⁴ In order to explore further the anomalous overdoped state, we have measured resistivity and Hall effect in $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ samples with different Ca and O content (doping) at temperatures above T_c . We compare our results to theoretical predictions from two representative FL models and show that they cannot give a satisfactory explanation to our data.

II. FERMI LIQUID MODELS

A FL phenomenological model based on the assumption of an anisotropic scattering rate along the FS was proposed by Carrington et al.²⁶ A further development led to the nearly antiferromagnetic Fermi liquid (NAFL) of Stojkovic and Pines¹³ (hereafter referred to as SP-NAFL model). This model is supported by ARPES measurements²⁷ and bandstructure calculations,²⁸ which show that the FS of holedoped HTSC looks like a square with rounded corners centered at the Γ point (π, π) . Two regions with different scattering rates are assumed:²⁶ hot regions corresponding to the large flat surfaces where magnetic interactions [spin fluctuation scattering (SFS)] are stronger, with $(\omega_h \tau_h)^{-1} \propto T$, and cold regions near the corners of the FS, with $(\omega_c \tau_c)^{-1} \propto T^2$; with $\tau_c > \tau_h$. For overdoped samples it is assumed that the FS grows with doping and that the scattering anisotropy reduces, therefore weakening the $R_H(T)$ dependence. But there are no specific quantitative predictions, in this as in most models, for overdoped samples.

A second FL model we have chosen to compare our data to is the one from Bok and Bouvier,²⁹ based on the Van-Hove singularity (VHS). We will refer to it hereafter as the BB-VHS model. In this model, the coexistence of electronlike and holelike orbits at energies near the Fermi energy, when it lies close to the VHS, is emphasized. In the BB-VHS model, R_H is given by

$$R_{H} = \frac{1}{e} \frac{n_{h}(\mu_{h})^{2} - n_{e}(\mu_{e})^{2}}{n_{h}\mu_{h} + n_{e}\mu_{e}},$$

where n_e , n_h are the density of carriers and μ_e , μ_h their mobility ($\mu_i = e \tau/m_i^*$), i = e, h. They have shown that including second-neighbor interactions in their tight-binding calculation, the FS possesses certain regions with positive and others with negative curvature. Therefore, positive and negative contributions to R_H may produce an apparent variable carrier density. The predicted sign change in R_H would be shifted to the overdoped side due to this correction.³⁰

III. EXPERIMENT

A. Sample preparation

High-quality *c*-axis oriented thin films of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$, with different *x* and δ , were prepared by dc off-axis sputtering by a method described elsewhere.³¹ Deposition time was 1.5 h, producing films with an average thickness $t \approx 1700$ Å and an uncertainty of ~13%. They were deposited on square LaAlO₃ (100) substrates of size 5×5 mm² or 10×10 mm². The doping level was adjusted by changing the Ca content *x*, and/or the O content by annealing. Four indium contacts were fixed to the corners for electrical measurements. The main source of uncertainties in



FIG. 2. Schematic configuration of electrical transport measurements with the Van der Pauw method. Left, longitudinal resistivity ρ_{xx} . Right: transversal resistivity, or Hall constant, $R_H = \rho_{xy}/B$, where *B* is the magnetic induction.

our measurements comes from the estimation of films thickness from the deposition time (according to previouslycalibrated films). This uncertainty dominates over other possible error sources, such as surface roughness (typically a few percent).

B. Measuring technique

In order to preserve the square geometry of our samples (needed for later measurements of penetration length with a microwave resonator), we used the Van der Pauw method. A current I is sent through one diagonal and voltage is measured along the second diagonal, then we exchange the connections for I with those for V and measure again. This method also requires the inversion of current sense and magnetic field direction in order to compute the average. The magnetic field was applied perpendicular to the film's surface, i.e., along the c axis, and scanned from -1 to +1 T. A linear fit to the data at different fields gives the slope dV_H/dB , where B is the magnetic induction, at every temperature point. The Hall density of carriers is then computed as $n_H = [4I/et](dV_H/dB)^{-1}$,⁵⁹ where *e* is the electronic charge and V_H is the Hall voltage. In the following, the room temperature value of n_H will be used as an indicator of the doping level. We used the lock-in amplifier technique with an ac current of 10 mA (rms) at a frequency of 1 Hz. Resistivity is measured by sending the current I through the contacts on one side of the sample and measuring voltage along the opposite side. A second measurement along the remaining pair of sides is necessary if the sample is not isotropic. Our samples are twinned with the *a* and *b* axis randomly distributed on the plane between the two crystallographic axes of the substrate, therefore they are isotropic.

Figure 2 shows schematically the electrical connections and measurement procedure. The resistivity is then calculated as $\rho = [\pi/\ln(2)]\langle V_R \rangle t/I$, where $\langle V_R \rangle$ is the voltage averaged on the different configurations. Uncertainties on measurements of ρ and R_H are dominated by t, getting close to 15%. The sample is introduced in an evacuated dewar, which is immersed in a liquid nitrogen bath. Temperatures below 77 K down to 60 K were obtained by pumping above the cryogenic liquid. A temperature controller was used in order to stabilize the sample temperature. We have observed that a small cooling rate is very crucial in order to obtain reliable



FIG. 3. Measured (longitudinal) resistivity vs temperature for samples with different doping. Inset: normalized resistivity emphasizing the deviation from the high-temperature linear fit $\rho(T) = \rho_o + bT$. ρ deviates upwards in overdoped samples and downwards in underdoped ones.

data, in particular regarding Hall measurements versus temperature. A cooling rate of less than 2 K/min was found to be slow enough.

IV. RESULTS

A. Resistivity

Resistivity measurements of samples with different doping are presented in Fig. 3. Resistivity values go down continuously with doping levels. At the highest doping level achieved, resistivity reaches 30 $\mu\Omega$ cm at 100 K. At room temperature, the conductivity varies linearly with n_H . In order to find the temperature at which ρ departs from linearity, we have fitted each curve to the equation $\rho = \rho_o + bT$, at high temperatures. The normalized curve $\left[\rho(T) - \rho_o\right]/bT$ enhances the deviation from linearity, allowing us to better determine the temperatures T^* and T_f . The inset in Fig. 3 presents normalized resistivity curves for some selected samples showing the departure from linearity. Resistivity of underdoped samples deviates downwards, while in overdoped samples it deviates upwards, below T^* and T_f , respectively. Resulting values of ρ_0 are positive in underdoped samples, decrease with doping becoming zero at optimal doping, then become negative in overdoped samples, increasing in absolute value with doping. The slope b decreases rapidly with doping in the underdoped region and remains almost constant in the overdoped regime. Figure 4 illustrates the doping dependence of ρ_0 and b. Figure 5 presents the data for T* and T_f versus n_H obtained in the way described above. We observe an almost linear drop in T^* versus n_H , similar to other reports,¹⁵ and in agreement with the phase diagram of Fig. 1. The line T_{f} , for overdoped samples, presents the opposite behavior, i.e., increasing with doping.

B. Hall effect

1. Hall constant

Figure 6 presents Hall effect versus temperature measurements for different doping levels. The data have been fitted



FIG. 4. Fit parameters for the high-temperature linear resistivity $\rho(T) = \rho_o + bT$. Note the almost constant slope *b* in the overdoped region, and ρ_o becoming zero at optimal doping.

to the equation $R_H(T) = R_H^{\infty}[1 + T_H/T]$, where T_H is a characteristic temperature above which there is a weak *T* dependence. Figure 7 presents normalized curves $R_H(T)/R_H^{\infty}$ versus T_H/T , showing a universal temperature dependence for all doping levels. The fit parameter T_H as a function of doping is shown in Fig. 8. Figure 9 shows the results from Figs. 5 and 7, together with data for the onset critical temperature T_c^{on} , showing that T_H and T_f coincide. A clear conclusion is that a canonical FL cannot exist in the overdoped region below the T_H line, because below that temperature R_H is strongly temperature-dependent.

2. Hall angle

An important parameter we can calculate from our data is the Hall cotangent, $\cot(\theta_H) = \rho_{xx}/\rho_{xy} = V_R/V_H$. Due to the cancellation of the parameters related to the sample's geometry, as emphasized in the latest expression, computed values of $\cot(\theta_H)$ possess smaller relative errors, <4%. Figure 10 presents our data fitted to the quadratic law $\cot(\theta_H) = A + BT^2$. A closer examination of the data reveals that this fit is not



FIG. 5. Characteristic temperatures for departure of $\rho(T)$ from linearity, measured from $\rho(T)$ curves in Fig. 3. The line $T^*(p)$ in the underdoped region determines the pseudogap region, and $T_f(p)$ in the overdoped region determines the presumed FL phase indicated in Fig. 1.



FIG. 6. Measured Hall constant R_H vs temperature for samples with different doping, as indicated by the Hall number n_H . Note the strong *T* dependence even at the highest doping level. Curves are fits to the equation $R_H(T) = R_H^{\infty}(1 + T_H/T)$.

equally good for different doping levels. Fitting our data to the more general power law $\operatorname{Cot}(\theta_H) = A + BT^{\gamma}$, where γ is now a free parameter, we obtain the dependence of γ on doping shown in Fig. 11.

V. DISCUSSION

A. Resistivity measurements

A constant high-temperature $(T > T_f)$ slope $d\rho/dT=b$ versus doping, as we observe in the overdoped region (Fig. 4), contradicts the FL relation $\rho = m/ne^2\tau$, which implies that $d\rho/dT$ varies as n^{-1} . Similar behavior has been reported in Ref. 24 for YBCO, and for other HTSC in Refs. 6 and 23. This result reflects a unique property of the state above the T_f line (MFL). It fits with the existence of a QCP,⁵ since the behavior at critical doping is recovered above the line $T_f(p)$ for $p > p_{op}$. Additional argumentation in favor of a QCP based on resistivity measurements is presented in Ref. 15. The high doping region below the T_f line (Fig. 1) has been



FIG. 7. Measured Hall constant normalized by the fitted parameters from the equation $R_H(T) = R_H^{\infty}(1 + T_H/T)$. We see that all the data obey this law quite well. T_H is a threshold temperature above which the *T* dependency weakens. Therefore, below the T_H line the system cannot be a canonical FL.



FIG. 8. Fitted parameter T_H vs n_H . The nonmonotonic V shape of this curve is reported here. This seems to be a particular behavior of the YBCO-123 HTSC system.

studied by ARPES.⁸ The authors found the electronic excitations to be coherent in this region, in contrast to the noncoherent excitations in the MFL region. Such a coherence in the former region indicates a metallic (FL) behavior, as pointed out by the authors. But the introductory discussion and our data on resistivity, Hall measurements and other transport properties show an anomalous behavior not compatible with a canonical FL. Therefore, we propose to call this new phase anomalous Fermi Liquid (AFL) (Fig. 9).

The superlinear behavior of $\rho(T)$ at $T < T_f$, sometimes ascribed to FL behavior, may have a more trivial explanation. Let us ignore for the moment the superconductive transition. Since ρ_0 is negative in overdoped samples, as we reduce the temperature within the linear range $(T > T_f)$, $\rho(T)$ targets a negative value. Since resistivity cannot be negative, at a certain temperature, which happens to be $T_f(p)$, $\rho(T)$ starts deviating upwards in order to reach a positive value at T=0. Given the dependence of ρ_0 on doping, $T_f(p)$ must also increase with doping, as observed. In this scenario, $T_f(p)$ does not indicate a phase transition, nor a crossover. Hence, resis-



FIG. 9. Phase diagram for our Ca-doped YBCO-123 samples. We have superposed the results from Figs. 5 and 8 together with measured values of the onset critical temperature T_c^{on} . The line T_H determines the boundary between a possible canonical FL (above) and an anomalous Fermi liquid (AFL) (below).



FIG. 10. Hall cotangent vs temperature for different doping, indicated by n_H . Curves are fittings to $\text{Cot}(\theta_H) = A + BT^2$.

tivity measurements are not conclusive regarding the FL phase. Hall effect measurements shall provide us with additional criteria in order to elucidate the nature of this region.

B. Hall effect measurements

In Fig. 6, we observe that all curves show a strong temperature dependence, even for the most overdoped samples. The fitting parameter T_H in Fig. 8 presents an unexpected V shape similar to that shown in Fig. 5. Above T_H , the system approaches the canonical FL behavior with a temperature-independent R_H . However, this conclusion is in contradiction with our discussion of resistivity measurements. As already noted, below T_H the strong temperature dependence of R_H precludes a canonical FL behavior.

The usage of an inappropriate scale in reports of data like those of Fig. 6, particularly when they include very underdoped samples which possess a large peak in $R_H(T)$, may have created the misleading impression of a gradually reducing *T* dependence with doping. This impression has probably led several researchers to conclude that, in general, over-



FIG. 11. Parameter γ vs n_H , after fitting the data of Fig. 10 to the equation $\text{Cot}(\theta_H) = A + BT^{\gamma}$. The exponent γ approaches its maximum of 1.85 only near optimal doping.

doped HTSC are canonical FL, as pictured in the general phase diagram of Fig. 1. In order to show that a strong Tdependence still remains in different overdoped HTSC systems, let us make a comparison with data published by other authors. To this effect, we have computed the ratio between the peak and room-temperature values, r_H , for different published data. For instance, in TBCO-2201 the maximum Tdependence is observed in underdoped samples where r_H \approx 1.36, and reduces gradually with doping to $r_H \approx$ 1.10 in strongly overdoped samples.^{6,33} This is in fact the HTSC system which presents the weakest T dependence in overdoped samples, which may indicate a smooth transition from MFL towards a canonical FL. The behavior of BSCO-2201 and Bi₂Sr₂Ca₁Cu₂O (BSCCO-2212) (Ref. 19) is similar to that of TBCO with the maximum T dependence in underdoped samples, $r_H \approx 1.30$ for a hole doping $p \sim 0.05$. Then it reduces slowly to $r_H \approx 1.20$ in overdoped samples with p \sim 0.23. Therefore, a substantial T dependence still remains at high doping. A contrasting behavior is found in LASCO.^{6,18} At zero doping, the nonsuperconductive phase presents a very strong T dependence with $r_H \approx 10$, which weakens by increasing doping until it almost disappears ($r_H \approx 1.04$), just below the appearance of superconductivity at $p \approx 0.04$. A further increase of doping produces a reappearance of the Tdependence, which grows proportionally to p. At optimal doping, $p \approx 0.15$, $r_H \approx 1.44$, and at high doping, $p \approx 0.25$, r_H \approx 1.70. In overdoped samples, the T dependence is more apparent at temperatures below ~ 100 K. Simultaneously, the peak shifts to lower temperatures following the reduction of T_c by doping. Above 100 K, $R_H(T)$ looks quite flat. The case of YBCO-123 is very special. From experimental data²⁵ we find an already high T dependence, $r_H \approx 2.4$, at low doping, corresponding to the oxygen content x=6.60. The T dependence is nonmonotonic. There is a minimum near x=6.85, with $r_H \approx 2.14$, and an increase to $r_H \approx 2.5$ at maximum doping, $x \approx 7.0$. This behavior, similar to our data (Figs. 6 and 8), disagrees with the expected trend from the phase diagram of Fig. 1.

Regarding the Hall angle, a fit to a variable exponent γ works better in fact than the quadratic law. The fitted exponent γ versus n_H , presented in Fig. 8, attains its maximum value ~1.85 at optimal doping. It decreases faster in the underdoped side, down to 1.55 ($n_H=0.4 \times 10^{22}$ cm⁻³), and drops more slowly in the overdoped side, down to 1.73 ($n_H=1.7 \times 10^{22}$ cm³). This behavior cannot be explained in the FL frame.

Now let us discuss our results in light of the two FL models introduced earlier. Let us start with the SP-NAFL model, which provides explicit expressions that allow us to evaluate the temperature and doping dependence of the transport coefficients. The expression for $R_H(T)$ is a polynomial [Eq. (34) in Ref. 13], which under the assumption $T > T_0$, where T_0 is a parameter in this model, can be expressed as $R_H(T) = R_H^{\infty}[1+6.5T_0/T]$. This is the same law as we have found, with the scaling $T_H = 6.5T_0$. T_0 is predicted to grow linearly with doping in underdoped samples. However, experimentally we find such a linear growth only in overdoped samples. On the contrary, in underdoped samples T_H decreases with doping, as shown in Fig. 8. The V shape of



FIG. 12. Hot and cold scattering rates computed from our transport data, according to the SP-NAFL model: cold, $1/\omega_c\tau_c = \cot(\theta_H)$; hot, $1/\omega_h\tau_h = n^2 e^2 \rho_{XX} R_H/B$. Lines are guides to the eye. Error bars for cold scattering rates are smaller than the symbols. The difference between both scattering rates persist at temperatures above T_H (Fig. 8). This renders invalid the assumption that the *T* dependence of R_H is due to τ 's anisotropy (in OD samples).

Fig. 8 cannot be explained in this model. The two scattering rates predicted in this model were computed for our samples and presented in Fig. 12. The magnitudes in the left scale show clearly that we are in the low field approximation, $(\omega \tau)^{-1} \ge 1$. The curves follow roughly the expected temperature dependence, $(\omega_h \tau_h)^{-1} \sim T$ and $(\omega_c \tau_c)^{-1} \sim T^2$, with $\tau_h < \tau_c$. If τ 's anisotropy were the origin for the temperature dependence of R_H , then the anisotropy should disappear at temperatures as low as 140 K for optimal doped samples (Fig. 8). The results of Figs. 8 and 9 are clearly not compatible with this assumption. We conclude that τ anisotropy alone cannot explain the behavior of $R_H(T)$. A difficult point in this model is the predicted FL behavior at high temperatures. While R_H satisfies that prediction for $T > T_H$, ρ instead keeps its non-FL linear on-temperature dependence up to high temperatures.

Let us now turn to the BB-VHS model. Computed curves of $R_H(T)$, at first glance, look qualitatively similar to experiment, except for the peak, which in their calculations appears at too low temperatures, below T_c . This may be due to the fact that their model does not include superconductivity. However, a careful examination of their curves shows a rise of R_H with reducing temperature that is much larger than experimentally observed. Computing the peak to roomtemperature ratio $r_H \equiv R_H^{\rm pk}/R_H(300 \text{ K})$ in their plots gives $r_H \approx 1.7$ in overdoped samples, $r_H \approx 3.5$ at optimal doping, and $r_H \approx 25$ for underdoped samples. These values are above the range between 1.1 and 2.5 found for different HTSC, as reviewed above. Another difference is that their normalized curves $R_H(T)/R_H(T^*)$ versus T/T^* , where T^* is defined as in Fig. 5, decrease continuously, apparently to zero, at high enough temperatures. Instead, in our fit, and also as found by other authors,^{16,22} it converges to a constant value R_{H}^{∞} , attained already near room temperature. No results are presented for the overdoped region. A crucial point in this model is the predicted change of sign in R_H , related to the crossing of E_f by the VHS, which should occur near p_{op} .³² Although experiments have confirmed a change of sign in R_H at heavy doping, $p \approx 0.3$ in LASCO,³² it has not been observed in our samples, nor in any other HTSC by other authors. This model predicts the correct T^* versus p linear decrease. An interesting question is what would be the predicted behavior for T_f or T_H in the overdoped regime. The same difficulty emphasized above regarding the high-temperature FL limit cannot be solved in this model. In fact, we believe it will be hard in any regular FL model to harmonize the observed high-temperature behaviors of R_H and ρ .

VI. CONCLUSIONS

Evolution of normal transport properties with doping in YBCO-123 is not monotonic, contrary to what is usually believed. Furthermore, this material, as well as other HTSC systems, does not present a clear trend towards a canonical FL at high doping. This appears to be the rule, except probably in TBCO-221. For YBCO-123 in particular, we find a minimum in the *T* dependence of R_H at p_{op} . Our measurements of the Hall effect show that $R_H(T)$ approaches a constant value, R_H^{∞} , for any doping level at temperatures above the line $T_H(p)$. Additionally, we find no range of temperature and doping where both ρ and R_H have simultaneously the canonical FL behavior. At optimal doping R_H becomes rap-

idly T-independent at $T > T_c$, but $\rho(T)$ is linear (MFL). In the overdoped region, at low temperatures, $R_H(T)$ has an anomalous behavior while $\rho(T)$ might be though of as having the canonical FL behavior, $\sim T^2$ (AFL). These results contradict the assumption of a canonical FL region in the overdoped region as shown in the phase diagram of Fig. 1. They cannot be explained by the smooth reduction of the relaxation rate anisotropy with doping predicted by the NAFL model of Stojkovic and Pines,¹³ nor by the VHS-based model of Bock and Bouvier.²⁹ In fact, these models, as well as others, do not address directly the problem of the overdoped regime but rather assume that it is a canonical FL. A correct model of the normal state of the HTSC should take into account the anomalous T-dependence behavior of ρ , R_H , and $\cot(\theta_H)$ in the overdoped region. We encourage theoreticians to undertake the challenge of discussing these points and to extend their models to the overdoped region, which up to now has been almost neglected.^{34–55}

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- ⁵⁹What we call here the "Hall carrier density" is computed as $n_H = 1/eR_H$, where R_H is measured at room temperature. We must remember that this number gives the density of carriers only for a parabolic band structure (Ref. 58). From n_H we could try to estimate p by computing the density of carriers per unit cell by using the "bond-valence sums" method (Ref. 57). However, this calculation is complicated in samples doped with Ca. An alternative method for computing p by comparing data for $T_c(n_H)$ with the universal parabola $T_c(p)$ is not straightforward because the relationship between n_H and p (or oxygen content x) is not linear. A more reliable way of computing p is by means of the universal curve for thermopower (Ref. 56), but such measurements were not made here, therefore we keep n_H as a representative parameter, which anyway does not affect our conclusions. Optimal doping $p_{\rm op}$ in our samples corresponds to $n_H \approx 0.9 \times 10^{22} \text{ cm}^{-3}$.