# Reduced Hall carrier density in the overdoped strange metal regime of cuprate superconductors

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**Efforts to understand the microscopic origin of superconductivity in the cuprates are dependent on knowledge of the normal state. The Hall number in the low temperature, high field limit $n_{\mathrm{H}}(0)$ has a particular significance because within conventional transport theory it is simply related to the number of charge carriers, and so its evolution with doping gives crucial information about the nature of the charge transport. Here we report a study of the high field Hall coefficient of the single layer cuprates $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) and (Pb/La) doped $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201) which shows how $n_{\mathrm{H}}(0)$ evolves in the overdoped, so-called strange metal, regime of cuprates. We find that $n_{\mathrm{H}}(0)$ increases smoothly from $p$ to $1+p$, where $p$ is the number of holes doped into the parent insulating state, over a wide range of doping. The evolution of $n_{\mathrm{H}}$ correlates with the emergence of the anomalous linear-in-$T$ term in the low-$T$ in-plane resistivity. The results could suggest that quasiparticle decoherence extends to dopings well beyond the pseudogap regime.**

In the search for the microscopic origin of high temperature superconductivity in the cuprates, much effort has been directed to understanding their normal state properties and how these are linked to superconductivity as a function of temperature and doping. The underdoped regime exhibits pseudogap phenomena as well as tendencies toward several types of order, or incipient order, including charge and spin density waves (CDW/SDW) [1]. In the regime where CDW order has been detected, at low temperature and high fields, $n_{\mathrm{H}}$ changes sign [2] suggesting some form of Fermi

surface reconstruction [3]. In some overdoped cuprates, beyond the doping level $p^*$ where the pseudogap disappears, there do not appear to be any competing orders and so potentially these materials provide a simpler starting point from which to understand the emergence of high temperature superconductivity.

In the far overdoped regime, the normal state behaviour of Tl2201 resembles, in many aspects, that of a conventional Fermi liquid with coherent quasiparticles around the entire Fermi-surface [4–6], whose shape is found to be well described by conventional density functional theory [7, 8], albeit with a large (factor 3) renormalisation in the effective mass which derives from a narrowing of the band [5, 6]. One aspect of the overdoped regime which contrasts with that of conventional metals is the evolution of the in-plane resistivity $\rho_{xx}(T)$ with doping. $\rho_{xx}(T)$ evolves smoothly from linear, close to optimal doping, to quadratic in the far overdoped regime [9, 10] leading to this being called the 'strange metal' regime [11, 12].

The close resemblance of the cuprate phase diagram to that of other material families, such as heavy-fermions and iron-pnictides, where superconductivity occurs close to an antiferromagnetic quantum critical point (QCP) [13] has led to speculation that the linear resistivity close to optimal doping in the cuprates may be a marker for a quantum critical transition to a hidden ordered phase [14]. The idea is that quantum fluctuations of the hidden phase provide the scattering mechanism which gives rise to both the linear resistivity and also provide the pairing mechanism for high temperature superconductivity. Possible candidates for this order are the pseudogap or CDW, but the thermodynamic evidence for a true phase transition of any type at finite temperature is weak and there is little evidence that either of these have a quantum critical end-point [15, 16]. For example, there are no observable anomalies in the specific heat [17] as the material is cooled into the pseudogap or CDW regimes. On the other hand, a number of recent experiments might support the existence of a QCP close to optimal doping. First, quantum oscillations in $YBa_2Cu_3O_{6+x}$ (Y123) show that the quasiparticle mass $m^*$ increases with doping, beyond $p = 0.12$, with $1/m^*$ extrapolating to zero at $p \sim 0.18$ [18]. Second, measurements suggest that in Y123, $n_{\rm H}(0)$ undergoes a rapid increase from $p$ to $1 + p$ over a narrow doping range $0.16 < p < 0.20$ [19], revealing possible critical behaviour near the doping where the pseudogap is believed to end in this material.

Whether such features are really caused by a QCP and whether this QCP is relevant to superconductivity requires further study. Recently, a high-pressure study of $YBa_2Cu_4O_8$ (Y124) [20] showed that as the maximum $T_c$ is approached by pressure tuning, rather than by chemical doping, $m^*$ actually decreases. This suggests that although the mass increase in Y123 near optimal doping may be linked to quantum CDW fluctuations, these fluctuations may not be the primary cause of the high $T_c$.

Interpreting $n_{\rm H}(0)$ as a planar hole density may have complications in some of the systems studied to date. In Y123, the quasi-one-dimensional CuO chains layer increases the $b$-axis conductivity but does not contribute to the Hall conductivity ($\sigma_{xy}$), thus increasing the measured $n_{\rm H}(0)$ over that expected from the CuO planes alone (see SI). In $La_{2-x}Sr_xCuO_4$ (LSCO), it is found that $n_{\rm H}(0) \simeq p$ but only for $p < 0.08$ [21, 22]. At higher doping, $n_{\rm H}$ increases well above $(1 + p)$ [21, 22] probably because the Fermi-surface develops electron-like curvature for doping close to $p \simeq 0.2$ where there is believed to be a Lifshitz transition [23]. In Nd doped LSCO the rise in $n_{\rm H}$ with $p$ is sharper

than in LSCO [24] but again the interpretation may also be complicated by a change in Fermi-surface curvature [25]. A detailed discussion of these issues can be found in the Supplementary information.

Here we have studied the evolution of $n_{\rm H}(0)$ in two cuprate families, Tl2201 and Bi2201, which have simple single band Fermi-surfaces and which may be overdoped to the edge of the superconducting dome or beyond without suffering Lifshitz transitions [6, 26]. Single crystals spanning a wide range of doping from slightly underdoped to strongly overdoped were used in this study. We find that $n_{\rm H}(0)$ evolves smoothly as a function of $p$ right across the overdoped regime so that $n_{\rm H}(0)$ does not reach the value $1 + p$ until close to the edge of the superconducting regime (note that our doping scales differ from some previous works as described in the SI). The behaviour correlates well with the evolution of the linear component of $\rho_{xx}(T)$ suggesting that the two have a common origin. Moreover there does not appear to be a simple correlation between the evolution of $n_{\rm H}(0)$ with $p$ and the closing of the pseudogap as previously conjectured [19, 24].

Tl2201 has unique properties for this study. It has a quasi-two-dimensional band-structure with a single $CuO_2$ layer giving rise to one band crossing the Fermi level. Its maximum $T_c$ is 94 K and it may be sufficiently overdoped so that it becomes non-superconducting yet remains electronically sufficiently clean for quantum oscillations to be observed [4–6]. For the overdoped compositions with $T_c \lesssim 26$ K, the Fermi surface geometry, scattering rate anisotropy and temperature dependence have all been accurately determined by quantum oscillation, angle-dependent magnetoresistance and angle resolved photoemission (ARPES) measurements [4–8, 27, 28].

Bi2201 is another single-layered cuprate which has a significantly reduced maximum $T_c$ (= 34 K) and upper critical field ($H_{c2}$), allowing superconductivity to be suppressed over a wider range of field and temperature space thereby reducing uncertainty in $n_{\rm H}(0)$. While quantum oscillations have not been observed in Bi2201, probably because it more disordered than Tl2201, its electronic structure has nonetheless been well characterized via ARPES and scanning tunneling microscopy [26, 29–32].

Figure 1 shows the field and temperature dependence of the Hall coefficient $R_{\rm H}$ for five representative Tl2201 samples. The evolution of $R_{\rm H}$ as a function of field and temperature at fixed doping can be understood, to some extent, in the overdoped regime using conventional Boltzmann transport theory. We have calculated the expected field and temperature dependence of $R_{\rm H}$ for Tl2201 using the known Fermi surface geometry as well as the anisotropy and $T$-dependence of the scattering rate determined independently from $c$-axis magnetoresistance measurements [27, 33](see SI). At low field, $R_{\rm H}$ is enhanced with respect to $1/ne$ (where $n$ is the carrier density determined by the Fermi surface volume) due to anisotropy in the Fermi velocity and scattering rate. At high field this anisotropy is averaged out as the electrons complete increasing fractions of their cyclotron orbits before being scattered and consequently $R_{\rm H}$ tends toward $1/ne$ (see Fig. S7). The low field enhancement in $R_{\rm H}$ is reduced at low temperature as scattering becomes dominated by isotropic impurity scattering and this together with the smaller scattering rate means that $R_{\rm H}$ approaches $1/ne$ at lower fields. Higher impurity scattering increases the field scale at which $R_{\rm H}$ approaches its infinite field value, but it also considerably diminishes the enhancement of $R_{\rm H}$ over $1/ne$, so $R_{\rm H} \simeq 1/ne$ at relatively low fields.

$R_{\rm H}(H,T)$ for the most overdoped sample of Tl2201 in Fig. 1, with $p = 0.27$, follows well the calculated behaviour. $R_{\rm H}$ at high temperature (120 K) decreases slowly with increasing field ($\sim 10\%$ in 60 T), but as the temperature is lowered, this field dependence becomes weaker and for $T \leq 10$ K, it is essentially constant once superconductivity is suppressed ($\mu_0 H > 20$ T at $T = 4.2$ K). The limiting value of $R_{\rm H}$ at this doping is close to that expected for the Hall number $n_{\rm H} \sim 1 + p$. This limiting behaviour is made clearer in the right hand panels of Fig. 1 where $R_{\rm H}$ is replotted against $H/\rho_{xx}^0$ which is proportional to $\omega_c\tau$ which reflects the fraction of a cyclotron orbit traversed by an electron before it scatters (here $\rho_{xx}^0$ is the extrapolated zero field resistivity).

For the $p = 0.26$ sample, at elevated temperatures the $H$ dependence of $R_{\rm H}$ becomes larger but again saturates in the high $H/T$ limit at a value consistent with $n_{\rm H} \sim 1 + p$. For $p = 0.22$, the irreversibility field has increased significantly. Nevertheless, 60 T still appears to be sufficient to reach the limiting value of $R_{\rm H}$, though in this case, it is found to be significantly higher than that expected from $n_{\rm H} \sim 1 + p$. Indeed, for the optimally doped sample ($p = 0.19$) and the slightly underdoped sample ($p = 0.14$), the values of $R_{\rm H}$ at the highest field and lowest temperature correspond more closely to $n_{\rm H} \sim p$ than to $n_{\rm H} \sim 1 + p$. For these higher $T_c$ samples, the temperature dependence of $R_{\rm H}$ is stronger than calculated from the estimated anisotropic scattering, although $R_{\rm H}$ still decreases with increasing $H$ as expected.

For Bi2201, the field dependence of $R_{\rm H}$, above $H_{c2}$, is much weaker than in Tl2201 (Fig. S3) consistent with a much higher isotropic elastic scattering rate (residual resistivities are almost one order of magnitude larger), and so we would expect the high field/low temperature values of $n_{\rm H}$ to more accurately reflect the carrier density.

The $T$-dependence of $n_{\rm H}$ at fixed high fields, for the different dopings are shown in Figure 2 for both Tl2201 and Bi2201. Extrapolating $n_{\rm H}(T)$ to $T = 0$ for each composition gives an estimate of $n_{\rm H}(0)$ whose evolution with doping is plotted in the lower panels of Figure 3. $n_{\rm H}(0)$ is found to evolve smoothly as a function of $p$ for both materials. For some doping values, multiple samples were measured and these gave consistent results, giving confidence that the error bars in $n_{\rm H}$ are accurate. For underdoped Tl2201 $n_{\rm H}(0) \simeq p$ but then increases over a broad doping range until it reaches the $n_{\rm H}(0) = 1 + p$ line approximately at $p = 0.25$ ($T_c = 40$ K). In Bi2201, previous measurements have shown that $n_{\rm H}(0)$ follows a non-monotonic behaviour below optimal doping [34] possibly due to the presence of a CDW similar to that found in Y123 [2]. In the overdoped regime however, $n_{\rm H}(0)$ in Bi2201, monotonically increases with increasing $p$ suggesting there are no regions of CDW order there. By taking into account the CuO chain conductivity, we show in the SI (Fig. S9) that the evolution of the planar contribution to $n_{\rm H}(0)$ in Y123 may show a very similar evolution of $n_{\rm H}(0)$ with $p$ to that found here for Tl2201 and Bi2201, although further measurements of the resistance anisotropy are needed to confirm this.

One potential interpretation of our results is that the evolution in $n_{\rm H}(0)$ evidences a slow closing of the pseudogap in the overdoped regime in these materials. Such behaviour has been suggested by a recent phenomenological model based on heterogeneous localization [35]. One of the clearest experimental signatures of the pseudogap is a collapse of the size of the anomaly in the electronic specific heat $\gamma$ at $T_c$ ($\Delta C(T_c)$) and a decrease in $\gamma(T)$ above $T_c$ [36]. In Tl2201, $\Delta C(T_c)$ is largest at the lowest doping measured ($p \simeq 0.20$) and $\gamma(T)$ is temperature independent above $T_c$

[37] which strongly suggests that $p^* < 0.20$ and so the $p$ to $1+p$ transition in $n_{\rm H}(0)$ occurs in a regime where there is no pseudogap. This is consistent with NMR Knight shift measurements which show that for Tl2201 the susceptibility $\chi(T)$ is temperature independent above $T_c$ for $p > 0.21$ [38]. For a sample with $T_c = 85\,{\rm K}$ a small downturn in $\chi(T)$ evident which suggests that $p^* < 0.194$ [39]. For Bi2201, analysis of $\rho(T)$ of our samples (see SI), suggests that $p^* < 0.215$ implying that again, the transition in $n_{\rm H}(0)$ occurs at least partially in the region where there is no pseudogap. For Bi2201 other probes suggest a larger value of $p^*$. NMR results give $p^*$ in the range 0.23-0.25 [40], and ARPES in the range 0.23-0.24 [26, 41] (see SI for a discussion). Nevertheless, these estimates are still in the range where $n_{\rm H}(0) < (1+p)$.

It is possible, in principle, that a pseudogap energy scale may be below the zero field $T_c$ and this may cause the reduction in $n_{\rm H}(0)$ we see. Such a pseudogap would not be manifest in $\gamma(T)$ above or at $T_c$, but instead there should be an anomaly below $T_c$ [42]. Furthermore, this should be accompanied by an anomalous reduction in the growth of the superfluid density $(1/\lambda^2)$ and $H_{c2}$ as temperature is lowered, as both are known to be strongly reduced in the pseudogap regime [42]. None of these signatures are observed experimentally in Tl2201 [37, 43, 44]. Indeed, $1/\lambda^2(T=0)$ and $\Delta C(T_c)$ are both found to be maximum at the lower doping measured ($p \simeq 0.20$), which seems to rule out this scenario in Tl2201.

In the cuprates, impurity scattering may be anisotropic [45], arising, for example, from a region of the Fermi surface that lies close to a van Hove singularity (vHs). This could reduce $n_{\rm H}$ at low field, but not the high-field limit estimated here. Moreover, in both Tl2201 and Bi2201 all indications are that the vHs remains above the Fermi level at all doping levels studied ([26] and SI). With decreasing doping, the Fermi level becomes ever further removed from the vHs. Thus, anisotropic scattering is unlikely to account for the decrease in $n_{\rm H}(0)$ with decreasing doping.

The fall in $n_{\rm H}(0)$ could also be interpreted as evidence of an, as yet undetected, reconstruction of the Fermi surface which begins in the far-overdoped regime. A reconstruction of the Fermi surface by a density wave could reduce $n_{\rm H}(0)$ [46], however, this should also give rise to small Fermi pockets. As quantum oscillations (QO) from the full $(1+p)$ Fermi-surface are observed in Tl2201 for $p > 0.28$, the non-observation of such QO from small pockets is evidence that they do not exist. Although it is possible that the QO from these pockets could be damped if the density wave has poor coherence.

Previously, it was shown that $\rho_{xx}(T)$ in both Tl2201 and LSCO could be modelled as the sum of $T$ and $T^2$ components [12]. The $T^2$ (Fermi-liquid like) component remains approximately independent of doping, whereas the anomalous linear-in-$T$ component ($\rho_{\rm lin}$) rises almost linearly with $p$ as $p$ is decreased from the edge of the superconducting dome [12, 47]. Recently [48], $\rho_{\rm lin}$ has been associated with scattering at the so-called Planckian limit which is the maximum allowed rate at which energy can be dissipated. Such strong scattering may derive from quasiparticle decoherence. As shown in Figure 3, the reduction of $n_{\rm H}(0)$ appears to correlate closely with the emergence of $\rho_{\rm lin}$ in both Tl2201 and Bi2201, and so an alternative interpretation of the reduction in $n_{\rm H}$ is that it evidences a growth of quasiparticle decoherence on part of the Fermi-surface. The Hall conductivity $\sigma_{xy}$ is strongly weighted by parts of the Fermi surface with strong curvature. So if decoherence developed on the flat sections of the

Fermi-surface but the quasiparticles remained coherent on the corners, this would lead to decrease in $\sigma_{xx}$, but would leave $\sigma_{xy}$ relatively unchanged, resulting in a decrease of $n_H \sim \sigma_{xx}^2/\sigma_{xy}$ as observed here. At lower doping, once the pseudogap has developed, $n_{\rm H}(0) = p$ [19, 21, 22, 49]; a response which presumably comes solely from the remaining Fermi arcs, although the mechanism for this remains open to debate.

Intriguingly, for Tl2201, the region ($p > 0.275$) where $n_{\rm H}(0)$ merges with the $1 + p$ line is the only region where quantum oscillations from the full $(1 + p)$ Fermi-surface have been observed [5]. The quasi-classical model of angle-dependent, out-of-plane magnetoresistance, so successful in modelling the lower $T_c$ samples of Tl2201, also fails for doping less than this ($T_c \gtrsim 40\,{\rm K}$) [33]. Although there could be several reasons [6] why quantum oscillations were not observed for higher $T_c$ samples, such as increased impurity scattering, in the light of these new results it is plausible that a loss of quasiparticle coherence around the Fermi surface is preventing quantum oscillatory phenomena from being realized. The endpoint of the transition in $n_{\rm H}(0)$, where $n_{\rm H}(0) \simeq p$ appears to occur approximately at optimal doping (Figure 3) which is also where $\rho_{\rm lin}$ is maximum [12, 47], again showing the close correlation between the two properties. If this is indeed caused by decoherence, it appears that this onsets well before the pseudogap is evident in resistivity or specific heat. Understanding, exactly how decoherence affects the transport properties and superconductivity could prove to be a crucial part of the high-$T_c$ cuprate puzzle.

## DATA AVAILABILITY

Source data are available for this paper. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request

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## AUTHOR CONTRIBUTIONS

The project was conceived by AC, CP and NEH. Pulsed field measurements on Tl2201 were performed by CP and ZW at HLD-Dresden and by CP,SB,WT and JA and at LNCMI-Toulouse. JL and SL contributed to the Hall effect

measurement on Bi2201 at HMFL-Nijmegen. Samples of Tl2201 were grown by LM and JRC. Samples of Bi2201 were grown by TK and TT. AC performed the numerical simulations of $R_H(T, H)$. The manuscript was written by AC and NEH with input from all the coauthors.

## COMPETING INTERESTS

The authors declare no competing interests.

## ADDITIONAL INFORMATION

Supplementary information is available for this paper at XXXX Correspondence and requests for materials should be addressed to A.C., C.P. or N.E.H.


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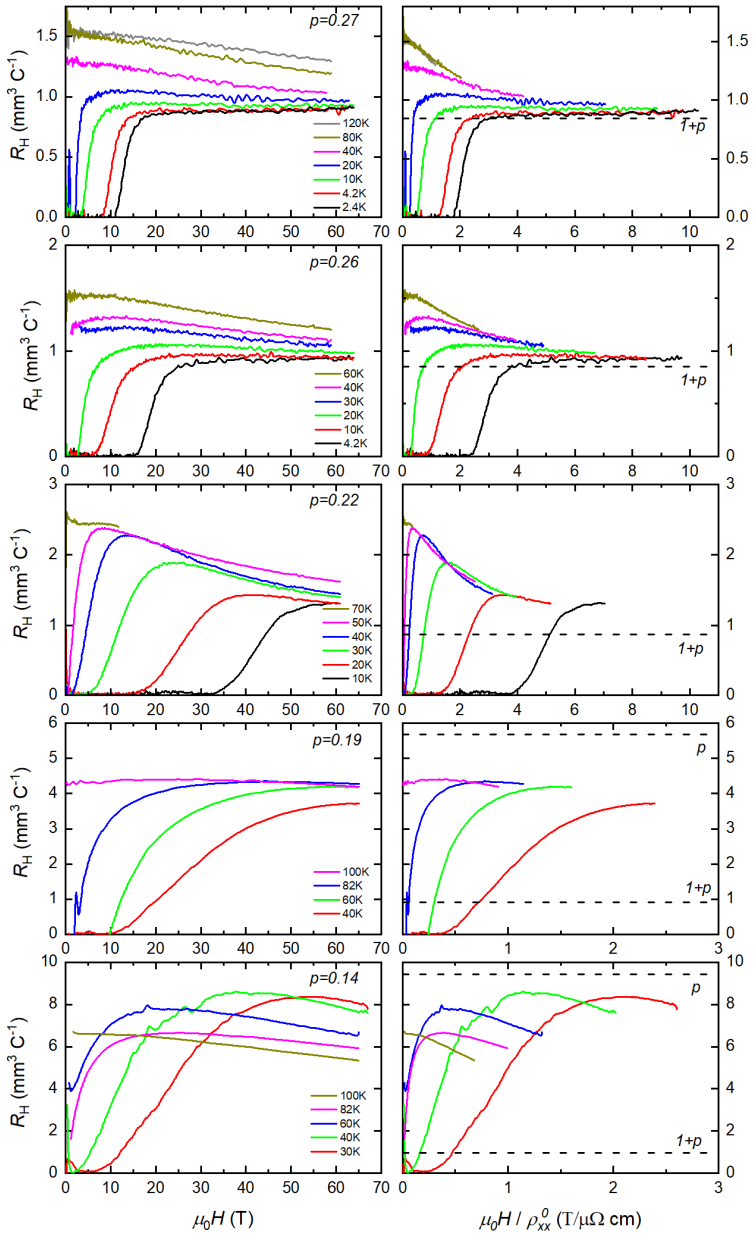


FIG. 1. Field dependence of the Hall coefficient for Tl2201. The left hand panels show the raw data at different temperatures and the right panels show the same data with the field scaled by the estimated value of the resistivity $\rho(T)$ at $H = 0$. The doping value ($p$) is indicated in each panel. The dashed lines on the left hand panels show Hall coefficient ($R_{\rm H}$) values corresponding to $1/n_{\rm H}e$ where $n_{\rm H} = 1 + p$ or $n_{\rm H} = p$.

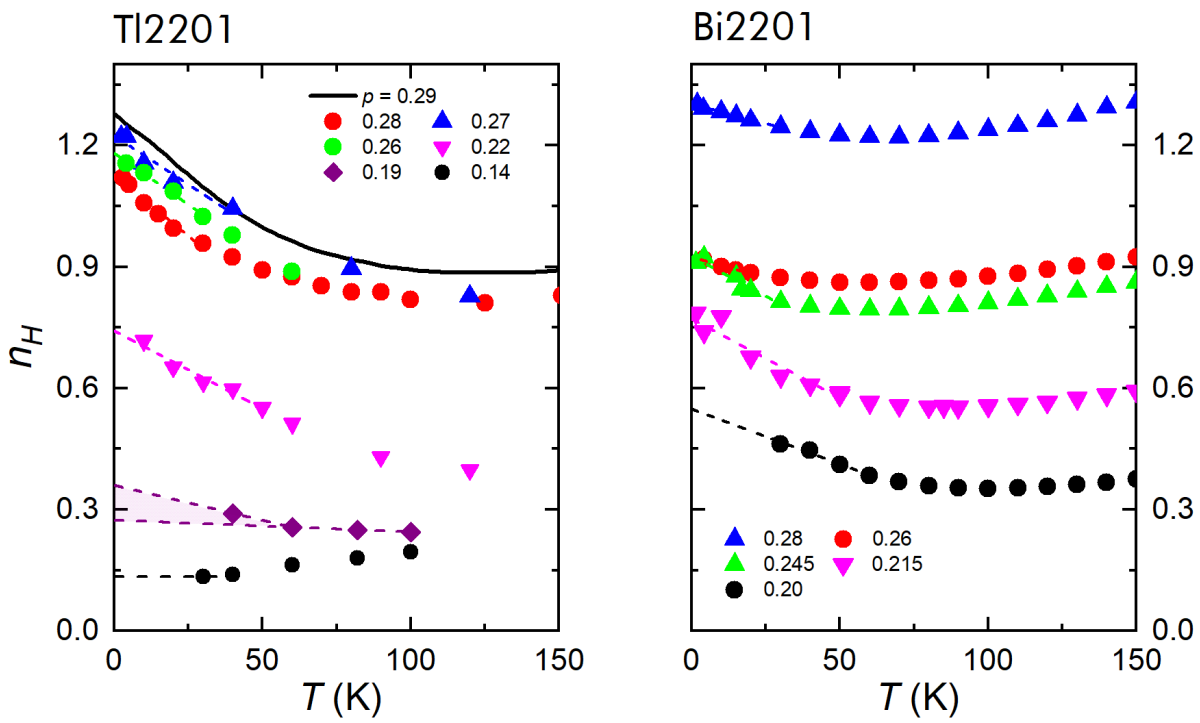


FIG. 2. Evolution of the Hall number with temperature and doping for Tl2201 and Bi2201. Hall number $n_{\rm H}$ values are taken at the highest values of field ($\mu_0 H = 60$ to 65 T) in Figure 1) expect $p = 0.28$ which was measured in 14 T. The dashed lines show the extrapolation to $T = 0$ to give $n_{\rm H}(0)$. For the sample with $p = 0.19$ we show two possible extrapolations. The solid line shows $n_{\rm H}(T)$ for a $p = 0.29$ sample of Tl2201 taken from Ref. [50], with $\mu_0 H = 16$ T.

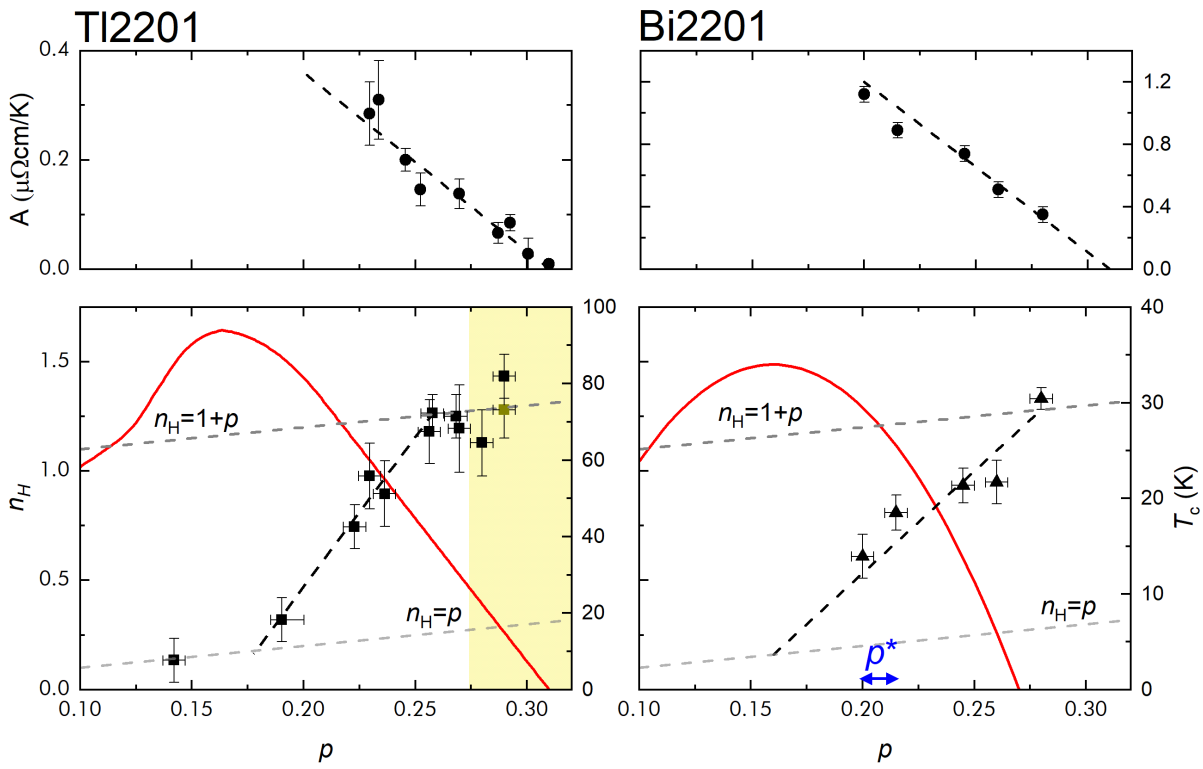


FIG. 3. Doping dependence of the low temperature Hall number and linear-in-$T$ component of resistivity for Tl2201 and Bi2201. Bottom panel: High field and low temperature values of the Hall number $n_{\rm H}(0)$ versus doped holes ($p$) for Tl2201 and Bi2201, from the extrapolations shown in Figure 2. The data point for the most overdoped sample is taken from Ref. [50]. The grey dashed lines show the behaviour expected for $n_{\rm H}(0) = p$ and $n_{\rm H}(0) = 1 + p$ and the black dashed line is a guide for the eye. The error bars in $n_{\rm H}$ reflect the geometric uncertainty, whereas those for $p$ reflect the uncertainty in $T_c$. The assumed evolution of $T_c(p)$ is shown with a solid red line for each compound (right hand scale, see SI). The shaded area on the Tl2201 panel shows the doping range where quantum oscillations have been observed. The end of the pseudogap regime ($p^*$) as found from $\rho_{xx}(T)$ measurements in our samples is indicated ($0.20 < p < 0.215$). Top Panels: Evolution of linear-in-$T$ coefficient of the zero field resistivity of Tl2201 [12] and Bi2201. The $A$ error bars reflect the geometric uncertainty.