Angular Dependence of the c-axis Normal State Magnetoresistance in Single Crystal $Tl_2Ba_2CuO_6$

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We report measurements of the normal state magnetoresistance (MR) from 30 to 340 K in fields B up to 13 T for single crystals of overdoped $Tl_2Ba_2CuO_6$ ($T_c \leq 25$ K). For out-of-plane current flow, the transverse MR $\Delta\rho_c/\rho_c$ is large and positive. On rotating $\mathbf{B}$ within the a-b plane, $\Delta\rho_c/\rho_c$ exhibits a striking anisotropy with fourfold symmetry. The amplitude of this effect increases as B^4 and the maximum MR occurs for B along the [110] crystallographic directions, i.e., at 45° to the Cu-O-Cu bonds. This the first direct evidence for anisotropy of the in-plane mean free path in the cuprates.

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Understanding the unusual normal state properties of high-T_c cuprates, especially the strong variation of the Hall coefficient R_H with temperature and hole concentration p, is an important step towards the correct microscopic theory of high-temperature superconductivity [1–6]. The systematic behavior of $R_H(T,p)$ is well established experimentally, but there are at least three different theoretical approaches. The spinon-holon model [2] involves two distinct relaxation times (τ_H and t_{tr}) for momentum changes parallel and perpendicular to the Fermi surface (FS). In more conventional pictures [3,7,8], τ varies strongly around the FS, e.g., because of electron-spin fluctuation scattering [8–10]. For all these models, a key quantity is the inverse Hall angle (ρ_{xx}/ρ_{xy}) or $\cot(\theta_H)$; experimentally this varies approximately as T^2 [11] and only weakly with p [10,12]. In alternative approaches [4–6], R_H itself has been considered as the primary quantity, its unusual behavior reflecting the presence of a small energy scale [4] or a change in the effective number of carriers with T [5,6].

Normal state magnetoresistance (MR) studies on single crystals should help distinguish between these different points of view. The in-plane MR $(\Delta\rho_{ab}/\rho_{ab})$ with $B \parallel c$ is small and positive and shows large deviations from Kohler's rule [13,14] in contrast to the behavior of most metals, even those with a complicated FS. The out-of-plane MR, on the other hand, is negative and has been ascribed to the B dependence of the normal state pseudogap [15]. However, MR studies of the out-of-plane resistivity $(\Delta\rho_c/\rho_c)$ have been confined to compounds showing nonmetallic behavior $(d\rho_c/dT < 0)$ [15], and this makes comparison with the in-plane properties difficult. A further complication is that ρ_c is often large and may well arise from incoherent interlayer hopping rather than bandlike electron motion [16,17].

Here we report the first study of in- and out-of-plane MR for overdoped crystals of the single layer $Tl_2Ba_2CuO_6$ compound for which $d\rho_c/dT$ is positive. For current $\mathbf{I} \parallel c$ and $\mathbf{B} \parallel ab$, the MR is positive and surprisingly large, as found for some organic conductors [18]. Within a band picture, the physical reason for this is that (a) the Lorentz force is still large (being the product of the field and the *in-plane* velocity $v_\parallel$) and (b) because of the strong anisotropy, there is no cancellation between the Lorentz force and the Hall field. Moreover, we find that, as $\mathbf{B}$ is rotated in the a-b plane, $\Delta\rho_c/\rho_c$ shows a striking anisotropy with a fourfold symmetry that varies as B^4. On analyzing our results using Boltzmann transport theory, several interesting features emerge. There is evidence for a T-dependent anisotropy in the in-plane mean free path. However, the T dependence of the anisotropy is insufficient to account fully for $R_H(T)$. The c-axis MR obeys Kohler's rule up to about 200 K while there are large deviations for the a-b plane MR. Finally, there is a close relation between the c-axis MR and the in-plane inverse Hall angle up to 340 K.

Small ($0.3 \times 0.3 \times 0.02$ mm^3) single crystals were grown using a self-flux method in alumina crucibles sealed with gold foil [19]. They were annealed in flowing oxygen at 720 K for three days before making electrical contacts to 25 μm gold wires with Dupont 6838 silver paste fired on in oxygen for 10 min at 740 K. This gave T_c values of 15–25 K. In all, MR measurements were made on 10 crystals (seven for ρ_c and three for ρ_{ab}). Crystals from all three preparations were only 10–20 μm thick, but $\rho_c(T)$ is reliable because of the large anisotropy and because similar values were found for a-b plane dimensions between 150×180 and 300×300 μm^2.

The lower inset to Fig. 1 shows $\rho_c(T)$ and $\rho_{ab}(T)$ in zero field, with ρ_c/ρ_{ab} rising from 1000 at 300 K to 2500 at 30 K. The residual resistivity ratio for ρ_c is invariably 2–3 times smaller than for ρ_{ab}, implying that standard Bloch-Boltzmann theory with a single τ cannot be an exact description. The top inset to Fig. 1 shows that for $\mathbf{I} \parallel c$ and $\mathbf{B} \parallel ab$ there is a large positive MR at low temperatures (≈14% at 35 K and 11 T). The MR

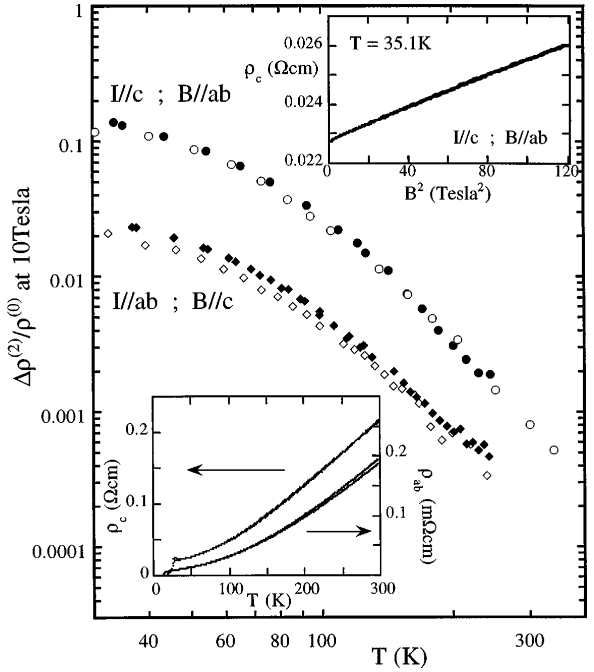

FIG. 1. T dependences of the B^2 terms $\Delta\rho^{(2)}/\rho^{(0)}$ at 10 T for c-axis MR (circles) and a-b plane MR (diamonds) in overdoped $\mathrm{Tl_2Ba_2CuO_6}$. Data for two crystals are shown in each case. Bottom inset: Zero-field $\rho_c(T)$ and $\rho_{ab}(T)$ for the crystals shown in the main figure. Top inset: MR field sweep at 35.1 K for $\mathbf{I} \parallel c$, $\mathbf{B} \parallel ab$.

varies as $\rho = \rho^{(0)} + \Delta\rho^{(2)} - \Delta\rho^{(4)}$, where $\rho^{(0)}$ is the zero-field resistivity and $\Delta\rho^{(2)}$ and $\Delta\rho^{(4)}$ are positive B^2 and B^4 terms. Using Chambers' formula [20] for the MR of a slightly warped cylindrical FS, we can show that this $-B^4$ term is associated with the crossover from B^2 at low fields to nonsaturating $|B|$ behavior in the high-field limit. The T dependences of the quadratic terms $\Delta\rho^{(2)}/\rho^{(0)}$ for $\mathbf{I} \parallel c$ and $\mathbf{I} \parallel ab$ are similar (Fig. 1), but the effect is 6–7 times larger for $\mathbf{I} \parallel c$.

For the two c-axis crystals shown in Fig. 1, the angular dependence of the MR was studied as **B** was rotated in the a-b plane; significant MR anisotropy with predominantly fourfold symmetry was observed (Fig. 2). Other smaller terms exist, but for rotations of 360° or more, the data could be unambiguously fitted by $R - R_0 = R_D\phi + R_1\cos(\phi - \phi_1) + R_2\cos2(\phi - \phi_2) + R_4\cos4(\phi - \phi_4)$. R_D represents a linear temperature drift term, and R_1 is a T-dependent Hall contribution (linear in B). R_2 was found to be essentially T independent and is associated with out-of-phase voltages induced by small movements of the sample or wires. R_4 dominates at low T, scales precisely as B^4, and falls rapidly with increasing T. In making these fits, the phases ϕ_1, ϕ_2, and ϕ_4 were kept equal to their low T values. The crystals were oriented using a Weissenberg x-ray camera, and despite the differences in contact geometries and sample shapes, the maxima in resistance occur when **B** is aligned along the (110) or equivalent directions, i.e., at 45° to the Cu-O-Cu bonds. Thus the angular dependence is an intrinsic property of the $\mathrm{CuO_2}$ planes, and we believe that this is the first direct observation of such anisotropy in the transport properties of the high-T_c cuprates.

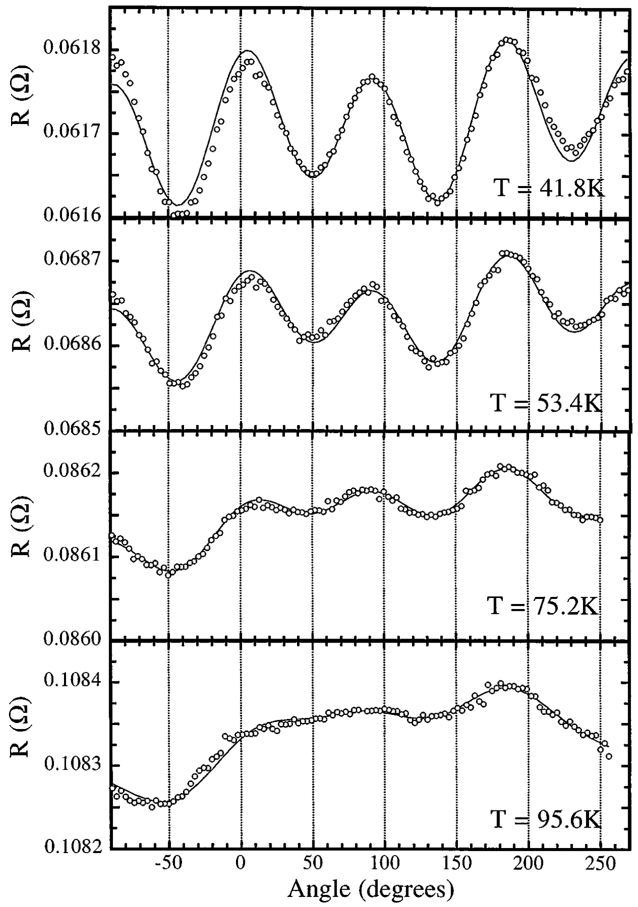

FIG. 2. Angular dependence of the c-axis transverse MR at various T for a $\mathrm{Tl_2Ba_2CuO_6}$ crystal with $T_c = 25$ K. Similar results were also obtained for the second c-axis crystal with a similar T_c.

As a first step in interpreting these results, we calculated all components of the conductivity tensor σ_{ij} for an open FS with small dispersion along the c axis ($\epsilon_\perp = -2t_\perp \cos k_\perp c$, where $c \approx 11.5$ Å is the interplanar distance and $t_\perp$ is the interplane overlap integral) using the relaxation time approximation and the Jones-Zener expansion to order B^4 [21]. For a FS with circular cross section, **B** in the a-b plane and a constant value of τ [22], this gives for the B^2 and B^4 terms, $\sigma^{(2)}_{cc}/\sigma^{(0)}_{cc} = -\Omega^2\tau^2$ and $\sigma^{(4)}_{(cc)}/\sigma^{(0)}_{cc} = 3/2(\Omega^2\tau^2)^2$, where the cyclotron frequency $\Omega = (e/\hbar c^*)v_\parallel Bc/\sqrt{2}$, c is the c-axis lattice parameter, and c^* is the velocity of light in cgs units. Inverting σ_{ij} to obtain the corresponding c-axis resistivity components gives

$$\Delta\rho^{(2)}_c/\rho^{(0)}_c = \Omega^2\tau^2, \tag{1}$$

$$\Delta\rho^{(4)}_c/\rho^{(0)}_c = (\sigma^{(2)}_{cc}/\sigma^{(0)}_{cc})^2 - \sigma^{(4)}_{cc}/\sigma^{(0)}_{cc} = -1/2(\Omega^2\tau^2)^2. \tag{2}$$

From $\Delta\rho^{(2)}_c/\rho^{(0)}_c$, we obtain $\Omega\tau$ and hence a direct estimate of the in-plane mean free path $l_\parallel = v_\parallel\tau$. At 100 K, $\Delta\rho^{(2)}_c/\rho^{(0)}_c \approx 0.025$ at 10 T and thus $l_\parallel \approx 128 \pm$

4 Å. Using the formula $\sigma_{ab} = (e^2/\hbar)k_F l/c$ for a cylindrical FS and taking $\rho_{ab}(100\ \text{K}) = 30\ \mu\Omega\ \text{cm} \pm 7\%$, we obtain $k_F \approx 0.77 \pm 0.06\ \text{Å}^{-1}$. This is in good agreement with the value determined from R_H at low T [23] and corresponds to a large FS $\approx (70 \pm 10)\%$ of the area of the Brillouin zone. Within the same model, the anisotropy ρ_c/ρ_{ab} is simply $l_{\|}^2/2\langle l_c^2\rangle$, so the value of ρ_c corresponds to a small value of $\langle l_c\rangle \approx c/3$ at 100 K. Despite this fact, which seems to indicate diffusive rather than band propagation in the c direction, many of the features discussed below are consistent with expectations for a band of fermion quasiparticles [24].

There is no anisotropy in $\Delta\rho^{(2)}$ or $\Delta\rho^{(4)}$ within the above cylindrical FS model. For tetragonal $Tl_2Ba_2CuO_6$, the structure of the CuO_2 planes will introduce fourfold anisotropy in v_F, k_F, and τ. Smoothly varying anisotropies can be represented by [21] $\tau^{-1}(\theta) = \tau_0^{-1}(1 + \varepsilon\cos^2 2\theta)$, $k_F(\theta) = k_F(1 + \alpha\sin^2 2\theta)$, and $v_F(\theta) = v_F(1 + \beta\sin^2 2\theta)$, where θ is the in-plane angle between **k** and the **a** axis. Using these formulas and the Jones-Zener method to calculate the B^2 and B^4 terms in $\rho_c(B)$ leads to a fourfold angular dependence in $\Delta\rho_c^{(4)}$ (proportional to B^4) but no anisotropy in $\Delta\rho_c^{(2)}$ [21]. Using typical values of $\alpha = 0.2$ and $\beta = 0.1$ compatible with band structure calculations [25], we calculated various quantities as a function of ε, the anisotropy of τ in the a-b plane, and compared them with the experimental results. Several, but crucially not all, of the observed features agree well with this simple model. Firstly, the maximum MR occurs for **B** $\|$ (110) is $\varepsilon > 0$, and so the experimental data imply that τ is shorter along the Cu-O-Cu bond direction. As can be seen in Fig. 3, $(\Delta\rho_c^{(2)}/\rho_c^{(0)})^2$ and $\Delta\rho_c^{(4)}/\rho_c^{(0)}$ have the same T dependence with a ratio of 0.6 ± 0.1 (top inset). For the cylindrical case ($\alpha = \beta = 0$), this ratio is 0.5 [from Eqs. (1) and (2)], but it rises slowly as α and β are increased and for $\alpha = 0.2$ and $\beta = 0.1$, it agrees with experiment for a large range of ε, $0 \le \varepsilon \le 0.7$. If ε is T dependent, the ratio of the angular anisotropy $A/\rho_c^{(0)}$ (where A is the amplitude of the fourfold term in Fig. 2) to the B^4 term $\Delta\rho_c^{(4)}/\rho_c^{(0)}$ should be constant. However, the lower inset to Fig. 3 shows that their ratio falls from 0.15 at 30 K to 0.06 at 125 K. This corresponds to an increase in ε from 0.25 to 0.40 between 30 and 125 K and shows explicitly that the anisotropy in τ is T dependent. This must affect the in-plane transport properties, namely, it will cause R_H to vary with T [3] and lead to deviations from Kohler's rule in the a-b plane MR [13]. However, the detailed calculations [21] show that larger changes in ε are required to account for $R_H(T)$ and the deviations from Kohler's rule shown in Fig. 4(a) (ε needs to increase from 0.50 to 1.25 from 30 to 125 K) than those which account for the T dependence of the MR anisotropy (ε increasing from 0.25 to 0.4). In addition, further analysis reveals that the magnitude of the a-b plane MR, $\Delta\rho_c^{(4)}/\rho_c^{(0)}$, is in fact anomalously large. The expected theoretical value for $\sigma_{ab}^{(2)}/\sigma_{ab}^{(0)}$ is $2/(k_F c)^2$ (i.e., $\approx 1/28$) of $\sigma_c^{(2)}/\sigma_c^{(0)}$. However, $\Delta\rho_{ab}^{(4)}/\rho_{ab}^{(0)}$ should be even smaller than $\sigma_{ab}^{(2)}/\sigma_{ab}^{(0)}$ because of the cancellation between the Hall field and the Lorentz force in the a-b plane. Thus the measured value, $\Delta\rho_{ab}^{(4)}/\rho_{ab}^{(0)} \approx \frac{1}{6}(\Delta\rho_c^{(2)}/\rho_c^{(0)})$ is at least 4 and probably 10 times larger than expected from the simple band treatment.

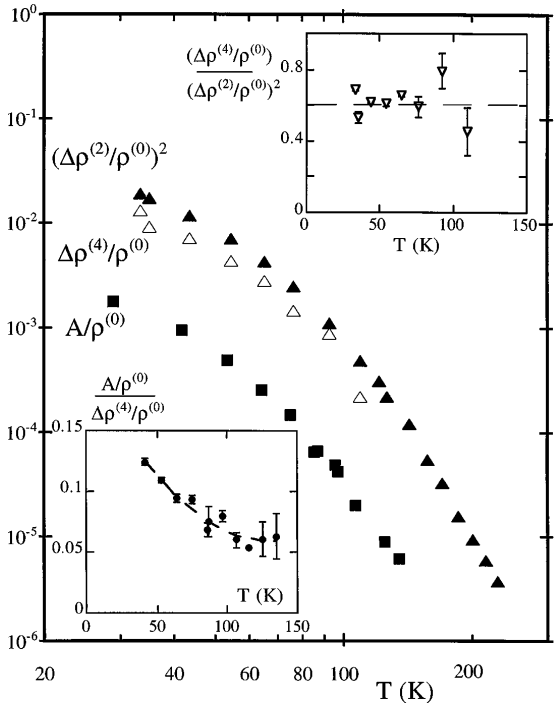

FIG. 3. T dependences of $(\Delta\rho^{(2)}/\rho^{(0)})^2$, $\Delta\rho^{(4)}/\rho^{(0)}$, and $A/\rho^{(0)}$ at 10 T for the same c-axis crystal as in Fig. 2. $\Delta\rho^{(4)}/\rho^{(0)}$ corresponds to the B^4 term at $\cos 4\phi = 0$ and A is the zero-to-peak amplitude of the fourfold symmetry term. Top inset: T dependence of $(\Delta\rho^{(4)}/\rho^{(0)})/(\Delta\rho^{(2)}/\rho^{(0)})^2$ for the data in the main figure. Bottom inset: T dependence of $A/\rho^{(0)}/(\Delta\rho^{(4)}/\rho^{(0)})$. This plot was obtained by dividing $A/\rho^{(0)}$ by $(\Delta\rho^{(2)}/\rho^{(0)})^2$ and then scaling by 1/0.6. Similar results were obtained for the second crystal.

A new ingredient may be required for a consistent explanation of all the unusual in-plane behavior. One possibility [13] is the two-lifetime model, with a transport equation of the form $g_{\boldsymbol{k}} = \tau_{\text{tr}} e\boldsymbol{E}\boldsymbol{v}_{\boldsymbol{k}}(-\partial f_0/\epsilon) - \tau_H e\boldsymbol{v}_{\boldsymbol{k}} \times \boldsymbol{B}(\partial g_{\boldsymbol{k}}/\partial\boldsymbol{k})$, where $\tau_H^{-1}(T)$ follows $\cot\theta_H(T)\,(= A + BT^2)$ and $\tau_{\text{tr}}^{-1}(T)$ is simply proportional to $\rho_{ab}(T)$. Using the Jones-Zener expansion for $g_{\boldsymbol{k}}$ [13], one finds $\sigma_{ab}^{(2)} \propto \tau_{\text{tr}}\tau_H^2$ and thus $\Delta\rho_{ab}^{(2)}/\rho_{ab}^{(0)} \propto \tau_H^2$. Hence within such a model, the Kohler plot of $\Delta\rho_{ab}^{(2)}/\rho_{ab}^{(0)}$ [Fig. 4(a)] actually represents $\tau_H^2/\tau_{\text{tr}}^2$, and its variation with temperature reflects the different behavior of the two lifetimes. However, Kohler's rule is obeyed for the c-axis MR up to 200 K [see Fig. 4(b)], implying that there is only one lifetime involved in c-axis transport. It is not clear how to apply this to the underdoped cuprates where $\rho_c(T)$ is generally nonmetallic. Finally, as shown

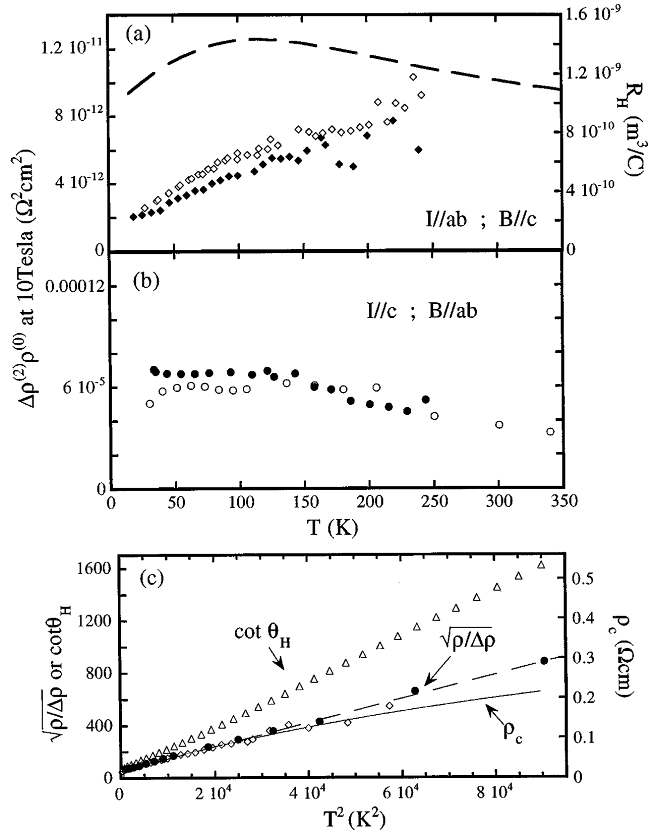

FIG. 4. (a) Kohler analysis showing $\Delta\rho_{ab}^{(2)}/\rho_{ab}^{(0)}$ at 10 T for both *a-b* plane crystals shown in Fig. 1. The dashed line is $R_H(T)$ for the same crystal whose Kohler plot is shown by (◇). (b) $\Delta\rho_c^{(2)}/\rho_c^{(0)}$ at 10 T for both *c*-axis crystals. (c) Typical T dependences of cotθ_H(△), $\sqrt{\rho_c^{(0)}/\Delta\rho_c^{(2)}}$ (●), $\sqrt{\rho_{ab}^{(0)}/\Delta\rho_{ab}^{(2)}}$ (◇), and ρ_c (solid line). $\sqrt{\rho_c^{(0)}/\Delta\rho_c^{(2)}}$ has been multiplied by 2.5. The dashed line is provided as a guide only.

in Fig. 4(c), another new result is that the relaxation rate determined from the *c*-axis MR, i.e., $(\sqrt{\rho_c^{(0)}/\Delta\rho_2^{(2)}})$ has a good T^2 dependence up to 340 K, similar to that of cot$\theta_H(T)$.

In summary, we have reported several new normal state MR effects, including an unusual angular dependence, for overdoped $Tl_2Ba_2CuO_6$ crystals. It will be interesting to explore these systematically as a function of hole doping and in other compounds, although preliminary studies indicate smaller effects in $YBa_2Cu_3O_7$ and $La_{2-x}Sr_xCuO_4$. However, the B^4 dependence of the MR anisotropy should allow these to be observed using high magnetic field facilities. We have shown that some features, including the *c*-axis MR, are consistent with single-fermion quasiparticle band theory with a large FS and a smoothly varying anisotropy in τ, but detailed agreement is lacking for the in-plane properties; namely, the T dependence of the anisotropy in τ derived from the angular measurements does not account for the observation variation of $R_H(T)$. This observation is particularly significant for an overdoped cuprate, where $R_H(T)$ is much less T dependent than in optimally doped crystals. A two lifetime model can resolve some of these discrepancies, but we have provided direct evidence for in-plane anisotropy and single lifetime effects in the *c*-axis properties which still need to be incorporated into such an approach. Other modifications of the band picture should also be considered, for example, models in which the anisotropy varies *sharply* around the FS, and models in which the normal state pseudogap, or in the overdoped case a small energy scale, is $\boldsymbol{k}$ dependent [4,26].

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