Origin of the anomalous Hall effect in the overdoped *n*-type superconductor $Pr_{2-x}Ce_xCuo_4$: Current-vertex corrections due to antiferromagnetic fluctuations

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The anomalous magnetotransport properties in electron doped (*n*-type) cuprates were investigated using Hall measurements at THz frequencies. The complex Hall angle was measured in overdoped $Pr_{2-x}Ce_xCuO_4$ samples ($x=0.17$ and 0.18) as a continuous function of temperature above T_c at excitation energies 5.24 and 10.5 meV. The results, extrapolated to low temperatures, show that inelastic scattering introduces electronlike contributions to the Hall response. First-principle calculations of the Hall angle that include current-vertex corrections (CVC) induced by electron interactions mediated by magnetic fluctuations in the Hall conductivity reproduce the temperature, frequency, and doping dependence of the experimental data. These results show that Fermi-liquid CVC effects are the source of the anomalous Hall transport properties in overdoped *n*-type cuprates.

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The various anomalous properties exhibited by the cuprates above the superconducting transition temperature continues to challenge the condensed-matter community. In particular, the unusual magnetotransport has often been cited as evidence that the cuprates are not consistent with Fermi-liquid theory,[1,2] a paradigm of physics for over 50 years. Despite the large simple convex holelike Fermi surfaces (FSs) observed by angular-resolved photoemission spectroscopy (ARPES), both optimally doped *n*- and *p*-type cuprates exhibit anomalous and strongly temperature-dependent Hall coefficients. In overdoped *n*-type cuprates, R_H has zero crossings that suggest a mixed electron and hole response. Consensus on an explanation for the apparent non-Fermi-liquid behavior of the cuprates has not been achieved despite much theoretical and experimental effort.[1,2]

One proposed explanation involves current-vertex corrections (CVC) to the standard relaxation-time approximation (RTA) for the Hall conductivity.[2] In this Fermi-liquid scenario, inelastic electron interactions mediated by antiferromagnetic (AF) fluctuations are the operative mechanism. In an alternative explanation thermally induced magnetic fluctuations at finite temperatures reconstruct the FS dynamically leading to fluctuating electronlike and holelike Fermi-surface segments over an area determined by the AF correlation length and a time scale associated with the AF correlation time.[3] In both of these scenarios, Fermi-liquid behavior is recovered at $T=0$ where inelastic scattering and thermal fluctuations vanish.

In this paper we report experiments that directly address these issues by extending magnetotransport measurements into the frequency domain in the electron-doped cuprates. We exploit the fact that inelastic scattering increases with temperature and frequency while magnetic fluctuations depend only on temperature. We report temperature- and doping-dependent Hall data on overdoped $Pr_{2-x}Ce_xCuO_4$ (PCCO) at THz frequencies $\lesssim 10$ meV that strongly support the Fermi-liquid interpretation in which CVCs are included.

Thin-film PCCO *c*-axis oriented samples were grown via pulsed laser deposition onto 100-μm-thick $LaSrGaO_4$ (001) substrates. The two samples reported here have a chemical doping of $x=0.17$ (Ref. 4) and 0.18 with thicknesses 40 and 125 nm and T_c's of 13 and 9 K, respectively. Resistivity and dc Hall measurements were similar to previously reported data.[5,6] However, the low-temperature dc Hall coefficient R_H measured on these new films is more consistent with the simple large holelike Fermi surface centered at (π,π) measured by ARPES.[7] Luttinger's thereom is satisfied since the carrier number density associated with the FS volume measured by ARPES is consistent with the stoichiometric doping level.[6,8,9] As temperature is raised, the dc Hall coefficient rapidly decreases and becomes negative at a temperature which increases with *x*, and R_H eventually returns to positive values around room temperature.

Below a doping of $\sim 16\%$ where PCCO undergoes a quantum phase transformation to an antiferromagnetic state, the low-temperature dc Hall coefficient deviates sharply from the observed overdoped behavior[6] and ARPES data show so called Fermi arcs.[7,10] In this paper we address the overdoped paramagnetic phase in which the large holelike Fermi surface remains intact.

The Faraday rotation and circular dichroism were measured (expressed as the complex Faraday angle, θ_F) at discrete frequencies as a continuous function of temperature. The output of a far-infrared molecular vapor laser was polarization modulated with a rotating quartz quarter-wave plate and subsequently transmitted through the *c*-axis oriented sample at normal incidence in an applied magnetic field up to 8 T. The detector signal was harmonically analyzed to directly extract the complex Faraday angle, a technique that is detailed elsewhere.[11,12]

In the thin-film limit, the complex Hall angle is related to the Faraday angle via $\theta_H=(1+\frac{n+1}{Z_0\sigma_{xx}d})\theta_F$, where σ_{xx} is the longitudinal conductivity, n is the index of refraction of the substrate, Z_0 is the impedance of free space, and d is the

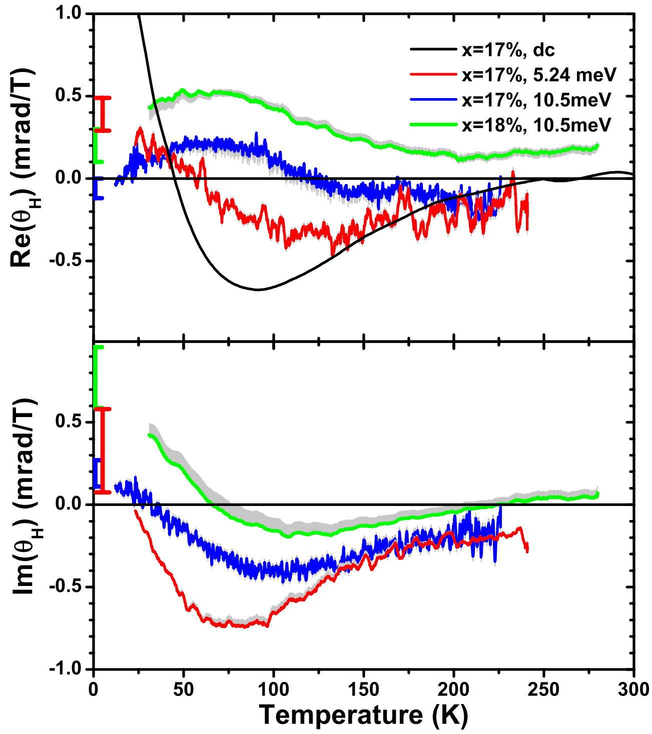

FIG. 1. (Color online) The real and imaginary parts of the Hall angle measured at dc, 5.24 and 10.5 meV. The negative value at finite temperature in both the real and imaginary parts of the Hall angle signify strong electronlike contributions which dominate the σ_{xy} response, a clear deviation from the simple holelike Fermi surface observed by ARPES. The brackets at low temperature are the zero-temperature extrapolations of the data as described in the text. Depicted in gray with each data set are error bars derived from the juxtaposition of uncertainties in thickness, σ_{xx}, and θ_F. The included dc Hall data is taken from Ref. 5.

thickness of the film.[12] Fourier transform infrared (FTIR) spectroscopic transmission measurements were performed in the spectral range from 2 to 13 meV at a set of discrete temperatures ranging from 5 to 300 K, and the complex conductivity σ_{xx} was extracted by fitting to a Drude form.

We have measured the real and imaginary parts of the Hall angle at 10.5 and 5.24 meV. The Hall signals were found to be linear in field over the measured magnetic field range of ± 8 T. The complex Hall angle as a function of temperature is plotted in Fig. 1. As in the case of the dc Hall effect in overdoped PCCO,[5,13] we observe a complex temperature dependence with zero crossings that depend on doping.

The observed Hall angle differs in important ways compared to the dc behavior (see Fig. 1). Most importantly, $\mathrm{Im}(\theta_H)$ becomes nonzero. $\mathrm{Im}(\theta_H)$ is negative at high-temperature crossing zero at a temperature that increases with doping and frequency. The low-temperature $\mathrm{Im}(\theta_H)$ values are positive as expected for a holelike Fermi surface as observed by ARPES. However the negative values at high temperatures is inconsistent with the holelike Fermi surface within Drude-type models. The differences between the $\mathrm{Re}(\theta_H)$ and the dc Hall angle increase with frequency and doping. The peak in $\mathrm{Re}(\theta_H)$ is expected in the simple Drude model for $\omega \sim \gamma_H$, the Hall scattering rate.

Motivated by the simple Fermi-liquidlike behavior of the low-temperature dc Hall coefficient, we begin our discussion by comparing the IR Hall response with a simple Drude model where the complex Hall angle in the weak-field approximation is given by

$$\theta_H = \frac{\sigma_{xy}}{\sigma_{xx}} = \frac{\omega_H}{\gamma_H - i\omega}, \quad (1)$$

where σ_{xy} is the Hall conductivity, σ_{xx} is the longitudinal conductivity, $\omega_H = qB/m_H$ is the Hall frequency, m_H is the effective Hall mass, ω is the radiation frequency, B is the applied magnetic field, and q is the effective charge of the quasiparticle. While Eq. (1) cannot be expected to accurately represent either the dc or IR Hall data of Fig. 1 at finite temperature because of the apparent combined electronlike and holelike response, it may be expected to correctly describe the low-temperature results.

To accomplish this, it is necessary to extrapolate the Hall data to $T \simeq 0$. Although the extrapolation of data to $T \simeq 0$ may at first glance appear problematic, the fundamental observation of a dramatic suppression of the Hall response at finite frequency is robust to the extrapolation method. To begin the analysis, it should be noted that generally in conducting condensed-matter systems when the energy scale associated with temperature becomes less than the energy scale set by the frequency, the temperature dependence of the conductivity weakens so that the exponent which may describe the power law of the corrections to the low-temperature finite-frequency response is expected to be larger than that of the dc response. Therefore, as a guide we use the reported dc Hall coefficient, Hall angle, and longitudinal resistivity data[5,6] for 17% and 18% PCCO. All these data were fit between 0 and 50 K and demonstrate temperature power laws with exponents ranging from 0.8 to 1.5. The dc Hall angle obeys a power law with an exponent of 1.0 (1.2) for the 17% (18%) sample. A power law of 0.8 actually overestimates the $T \simeq 0$ extrapolated ac Hall response leading to an overestimate of ω_H. A larger temperature exponent greater than that of the dc Hall angle results in a *smaller* Hall response as expected, characterized as a smaller ω_H. We choose a reasonable upper bound of 2.0. The zero-temperature extrapolations of these power law fits to the IR Hall angle data between 0 and 50 K with exponents ranging from 0.8 to 2 are shown in Fig. 1. Even with these generous overestimates of uncertainties associated with the extrapolations, the IR Hall angle is much smaller than the dc Hall angle (2.6 and 3.4 mrad/T for the 17% and 18% doped samples, respectively).[5,6]

This suppression of the finite-frequency Hall response is more quantitatively characterized by comparing the low-temperature Hall frequency ω_H in the Drude representation to that expected from ARPES measurements. As noted previously, the low-temperature dc Hall coefficient above the quantum critical doping is correctly given by the Fermi-surface properties measured by ARPES. All the magnetotransport response functions including ω_H can be derived directly from ARPES data within the RTA in which σ_{xx} and σ_{xy} are both expressed as integrals around the Fermi surface involving the Fermi velocities and scattering rates.[14–16] ARPES measurements determine the size and shape of the

TABLE I. Utilizing the extrapolated zero-temperature complex Hall angle values in Fig. 1, the finite-frequency zero-temperature Hall frequency ω_H may be extracted normalized to the Hall frequency predicted by ARPES measurements and Boltzmann transport theory, $\omega_H^0=0.052$ meV/T.

Doping, x (%)	ω (meV)	ω_H/ω_H^0
17	5.24	0.20 ± 0.13
17	10.5	0.04 ± 0.02
18	10.5	0.16 ± 0.04

Fermi surface, Fermi velocities, and momentum distribution widths. For optimal doping, the Hall frequency derived from the large holelike Fermi surface observed by ARPES (Refs. 7 and 11) yields $\omega_H^0=0.052$ meV/T, equivalent to an effective Hall mass $m_H=2.2m_e$.

The estimated $T\simeq0$ Hall frequencies normalized to the values deduced from ARPES data, given by $\omega_H/|\omega_H^0|=-(\omega/|\omega_H^0|)[\mathrm{Im}(1/\theta_H)]^{-1}$, are presented in Table I. These data replicate our main observation that the Hall response is severely suppressed at finite frequency compared to the low-temperature dc values and, equivalently, the RTA expectations deduced from ARPES. The uncertainties associated with the IR Hall angle extrapolations are small compared to this suppression. Also discernable in Table I is a rapid reduction in the Hall response with frequency similar to the rapid decrease in R_H and the dc Hall angle[5] with temperature. This reduction with increasing frequency can be seen directly in the Table I (rows 1 and 2), and both values are much smaller than the dc values deduced from ARPES measurements. It should also be noted that the reduction in the Hall response with temperature or frequency, expressed in the same units, is of a similar magnitude. From the doping dependence in Table I (rows 2 and 3), it is also seen that the Hall frequency suppression increases as the doping is reduced from 18% to 17% toward the quantum critical doping.

We can also examine the frequency dependence of R_H from these data. Again, comparing the ac and dc values is particularly interesting since the $T\simeq0$ dc R_H is consistent with the Luttinger theorem implying a Fermi-liquid analysis is valid. The low-temperature ac Hall coefficient $R_H=\theta_H/\sigma_{xx}$ is calculated using σ_{xx} from FTIR measurements. The Luttinger R_H at $T\simeq0$ and $\omega=0$ implies that the scattering rates associated with the off-diagonal and diagonal conductivities are equal ($\gamma_{xy}=\gamma_{xx}$). As frequency is increased to 10 meV ($\sim$20 K), γ_{xx} is estimated to increase by only $\sim$10%.[17] At low temperatures our data show $\mathrm{Im}(R_H)>0$ at finite frequencies which necessarily requires $\gamma_{xy}<\gamma_{xx}$ within a Drude analysis. Therefore, in the low-frequency limit where $\mathrm{Re}(R_H)\sim\gamma_{xx}^2/\gamma_{xy}^2$, the Drude expectation is a slight increase in $\mathrm{Re}(R_H)$ between dc and 10 meV. However, $\mathrm{Re}(R_H)$ at finite frequency is found to be severely suppressed. In particular, we find that for the $x=17$% sample at 10.5 meV and $T\simeq0$, $\mathrm{Re}(R_H)$ is suppressed by a factor of 4 well outside of any experimental errors. Since choosing a γ_{xy} with the constraint $\gamma_{xy}<\gamma_{xx}$ cannot account for this suppression, a Fermi-liquid Drude scenario does not capture the $T\simeq0$ frequency dependence of R_H. It should be noted that our measured $T\simeq0$ suppression of $\mathrm{Re}(R_H)$ in frequency is consistent with the $\omega=0$ suppression in temperature[17] provided $\omega/2\sim\pi T$, where ω is the optical frequency, as predicted by Fermi-liquid theory. The suppression of $\mathrm{Re}(R_H)$ is definitively weaker for lower frequencies and higher doping but errors associated with measurements of σ_{xx} as well as the $T\simeq0$ extrapolation of the Hall angle are relatively large.

The observed reduction in the Hall angle and Hall coefficient with frequency (and/or temperature) corresponds to a large suppression in σ_{xy} caused by the presence of electronlike contributions. The electronlike contributions at $T\simeq0$ and finite frequency cannot be due to AF fluctuations since thermally excited AF fluctuations vanish at zero temperature. Instead these results suggest a breakdown of the RTA except when both temperature and frequency are zero where inelastic scattering vanishes. While the experiments implicate inelastic scattering as the underlying cause of the anomalous Hall effect in overdoped PCCO, the mechanism is not obvious from the experimental findings. However, current-vertex corrections to the conductivity in the presence of antiferromagnetic fluctuations can lead to currents that are not necessarily parallel to the quasiparticle velocity which can produce negative contributions to the Hall conductivity.[2] In these nearly antiferromagnetic metals, the Hall conductivity is strongly modified by the CVCs which represents electron-electron (Umklapp) scattering processes associated with the magnetic Brillouin zone.[2] Including the CVCs has successfully reproduced the strong temperature dependence of dc Hall coefficient in both electron- and hole-doped cuprates.[2] These effects also modify the frequency-dependent response since inelastic scattering is frequency dependent and occurs even at $T\simeq0$.

To examine the role of CVCs on the IR Hall response, we have calculated the frequency- and temperature-dependent conductivity for overdoped PCCO within the fluctuation-exchange (FLEX) conserving approximation. The starting point is the Hubbard model with model parameters for the tight-binding hopping amplitudes representing the band structure (t,t',t'') and the Hubbard U.[2] In the FLEX approximation the effective interaction potential is proportional to the derived dynamical spin susceptibility which has the following functional form: $\chi_{\mathbf{q}}^s(\epsilon)\propto\xi^2[1+\xi^2(\mathbf{q}-\mathbf{Q})^2-i\epsilon/\omega_{sf}]^{-1}$, where the AF correlation length ξ is much larger than the lattice spacing in PCCO at low temperatures.[18] With this interaction, an electron with $\mathbf{k}$ on the Fermi surface is scattered to $\mathbf{k}+(-)\mathbf{Q}$ by absorbing (or emitting) a low-energy AF magnon fluctuation. This scattering process in nearly AF metals not only shortens the quasiparticle lifetime (i.e., hot spot) but also changes the direction of electron motion. In Fermi-liquid theory, the former and the latter effects are described by the self-energy and the CVC, respectively. How the electron scattering due to magnetic fluctuations leads to strong CVC effects is illustrated in Fig. 3. An excited electron at $\mathbf{k}$, after a quasiparticle lifetime $1/2\gamma_{AF}$, is scattered to $\mathbf{k}+\mathbf{Q}$ due to AF fluctuations $\mathbf{Q}\approx(\pm\pi,\pm\pi)$. Therefore, $\mathbf{J_k}\propto\mathbf{v_k}+\mathbf{v_{k+Q}}$ in the hydrodynamic regime $\omega\ll\gamma_{AF}$.

Figure 2 shows the numerical results for the ac Hall angle obtained by the FLEX+CVC approximation. The nearest-neighbor hopping integral is taken as $t=-0.36$ eV and the

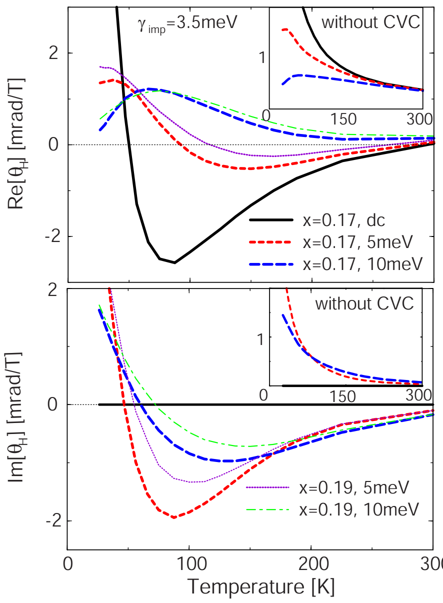

FIG. 2. (Color online) Prediction of the real and imaginary part of the Hall angle by incorporating CVCs induced by magnon scattering. The Hall angle calculated in the FLEX approximation without including CVCs is shown in the insets.

quasiparticle damping rate due to elastic scattering was set at $\gamma_{\text{imp}}=3.5$ meV corresponding to $\rho_{\text{imp}}\sim 5$ $\mu\Omega$ cm. In the absence of CVC the complex frequency-dependent θ_H is positive due to the large holelike Fermi surface corresponding to the ARPES data on electron overdoped cuprates. In Fig. 2 in which CVCs are included, both $\text{Re}(\theta_H)$ and especially $\text{Im}(\theta_H)$ take on increasingly negative values as temperature is lowered from room temperature and the AF fluctuations become stronger. Below 100 K when the damping rate due to AF fluctuations γ_{AF} becomes smaller than the elastic scattering rate, CVCs become less important. However, since γ_{AF} increases with ω, CVC effects will be more important for larger ω. As ω and T approach zero, the inelastic scattering reduces and both $\text{Re}(\theta_H)$ and $\text{Im}(\theta_H)$ become positive. In the collisionless regime where $\omega \gg \gamma_{AF}$ the electron-electron scattering becomes less important and $\mathbf{J}_\mathbf{k}\sim \mathbf{v}_\mathbf{k}$.

Also, as can be seen in Fig. 2 as T approaches zero, $\text{Re}(\theta_H)$ at 5 and 10 meV is much smaller than the zero frequency value. In addition, the figure shows the calculated results for two different doping levels which demonstrate the reduction in the CVC effects as the system is doped further from the AF phase boundary.

Although the temperature, frequency, and doping dependence of θ_H is qualitatively well described by the FLEX +CVC theory, the overall amplitude of the experimental data in Fig. 1 is smaller by a factor of 2–3 compared with the theoretical results in Fig. 2. This discrepancy may result

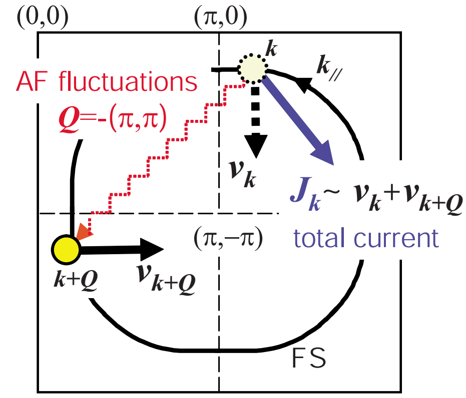

FIG. 3. (Color online) Electron scattering process $\mathbf{k}\rightarrow\mathbf{k}+\mathbf{Q}$ due to AF fluctuations $\mathbf{Q}\approx(\pm\pi,\pm\pi)$. The total current $\mathbf{J}_\mathbf{k}$ is composed of quasiparticle velocities at $\mathbf{k}$ and $\mathbf{k}+\mathbf{Q}$.

from the fact that the FLEX approximation, as a low-energy theory, can provide an accurate description of the magnetic correlation effects but cannot give a good account of the Mott correlations in the Hubbard model. In particular, the theory fails to account for the high-frequency Hubbard band optical transitions for $\omega \gtrsim U$. This implies, by the optical conductivity sum rules, that the theory overestimates the spectral weight of the low-energy Drude-type response. The suppression of the Drude-type spectral weight in the cuprates has been discussed by Millis *et al.*[16,19] Experiments suggest that the Drude oscillator strength associated with σ_{xy} is more strongly suppressed than that associated with σ_{xx}.[20,21] Therefore, since $\theta_H=\sigma_{xy}/\sigma_{xx}$, there is an overall suppression of the Hall angle Drude-type oscillator strength[22] that is not captured by the FLEX theory. The nature of these Mott correlations is an important theoretical issue that deserves and is receiving further study.[23]

In conclusion, the low-temperature Hall response of PCCO is observed to be strongly suppressed at finite frequencies. The full complex temperature, frequency, and doping dependencies of θ_H in overdoped PCCO are found to be naturally reproduced in the Fermi-liquid FLEX+CVC approximation for this strongly interacting nearly antiferromagnetic electron system. These results show that current-vertex corrections contribute significantly to the magnetotransport properties of PCCO and, by extension, support the conclusion that CVC effects are the underlying mechanism causing the anomalous Hall transport in the hole-doped cuprate systems.[2]

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