

Spectral Measurement of the Hall Angle Response in Normal State Cuprate Superconductors

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We measure the temperature and frequency dependence of the complex Hall angle for normal state YBa₂Cu₃O₇ films from dc to far-infrared frequencies (20–250 cm⁻¹) using a new modulated polarization technique. We determine that the functional dependence of the Hall angle on scattering does not fit the expected Lorentzian response. We find spectral evidence supporting models of the Hall effect where the scattering Γ_H is linear in T , suggesting that a single relaxation rate, linear in temperature, governs transport in the cuprates.

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The normal state Hall effect in cuprate superconductors exhibits an anomalous temperature dependence that cannot be explained using conventional transport theory for metals. According to the simple Drude model, the resistivity of a metal and the cotangent of its Hall angle, $\cot(\theta_H) = \frac{\sigma_{xx}}{\sigma_{xy}}$, should share the same temperature dependence, both proportional to the scattering rate of the charge carriers. However, the normal state resistance of cuprate superconductors is linear with temperature, $\rho \sim T$, while the Hall angle has a robust $\cot(\theta_H) \sim T^2$ behavior [1] over a wide range of oxygen doping [2], and with substitutional doping [3] in a variety of the cuprates [4]. This apparent duality of scattering rates characterizes the anomalous Hall transport in the cuprates. The spin-charge separation model of Anderson assumed that two scattering rates were in fact involved, with the two species of quasiparticles relaxing at the different observed rates [5]. Subsequent explanations focused either on alternative non-Fermi liquid mechanisms [6,7] or on the effects of k -space scattering anisotropies [8–11]. The common feature of all the above theories is a dominant term that is linear in the scattering rate, $\cot(\theta_H) \sim \gamma_H$. In contrast, a recent theory by Varma and Abrahams [12] treats anisotropic scattering in a marginal Fermi liquid and predicts a *square-scattering* response, $\cot(\theta_H) \sim \gamma_H^2$.

These different models can be distinguished at finite frequency. The linear- and square-scattering models correspond to Lorentzian and square-Lorentzian spectral responses, respectively, and, although Hall experiments have been performed at finite frequencies [13–15], this paper is the first to study both temperature *and* frequency dependence of the Hall response in a frequency range that discerns a line shape and extrapolates to the dc limit.

We begin by reviewing the concept of a frequency dependent Hall angle [16] using the Drude model as an example of a Lorentzian response. All parameters are implicitly spectral, i.e., $\theta_H = \theta_H(\omega)$, and in the present case of strong scattering, $\tan(\theta) \approx \theta \ll 1$. Quasiparticles circling at the cyclotron frequency $\omega_H^* = eB/mc$

traverse a fraction $\omega_H^* \tau_H^*$ of a cyclotron orbit during the time τ_H^* between scattering events. The dc longitudinal current j_x is thereby deflected into j_y by this small arc angle $\theta_H \approx \frac{j_y}{j_x} = \omega_H^* \tau_H^* = \omega_H^* / \gamma_H^*$, where γ_H^* is defined as the Hall scattering rate. For the ac response, we substitute $\gamma_H^* \rightarrow \gamma_H^* - i\omega$ yielding a Lorentzian:

$$\theta_H(\omega) = \frac{\omega_H^*}{\gamma_H^* - i\omega}. \quad (1)$$

Linear scattering models of the anomalous Hall transport predict this same Lorentzian form for the frequency response near the dc limit.

For the experiment at infrared frequencies, the Hall angle θ_H cannot be measured directly but must be deduced from transmission studies of polarized light [13–15]. The measurable quantity, the Faraday angle θ_F , is the angle of rotation of polarized light induced after passing through a thin conducting film in the presence of a normal magnetic field B . In the thin film limit ($d \ll \lambda, \delta$) with d the film thickness, λ the wavelength, and δ the penetration depth, θ_H follows from θ_F according to Maxwell's equations:

$$\theta_H = \left(\frac{1 + n}{Z_o \sigma_{xx}} + 1 \right) \theta_F. \quad (2)$$

σ_{xx} is the experimentally determined complex sheet conductivity, Z_o the free space impedance, and $n = 3.4$ the refractive index of the Si substrate. With our highly conductive films, the term in parentheses is near unity, so the functional dependence on σ_{xx} is minimal. θ_F and θ_H are both causal response functions, so their Re and Im parts obey Kramers-Kronig relations and correspond to real space rotation and ellipticity, respectively [16].

The cuprate sample investigated was a pulsed-laser deposited, twinned film of 500 Å YBa₂Cu₃O₇ on a (1 × 1) cm² × 350 μm insulating silicon substrate, with an intermediate strain relieving layer of 100 Å yttrium stabilized zirconate [13]. Partial data on a second sample confirmed the results reported here. The sample was mounted in an 8 T Oxford Spectromag cryostat with the B field

oriented along the optical axis, normal to the sample surface. Two 8 cm diameter Kapton cryostat windows on either side of the sample allowed direct optical access. Sourced by a broadband quartz Hg arc lamp, the spectra were measured using a step-scan Fourier transform interferometer with wire-grid polarized beam splitters having a density of 40 wires per mm. The novel technique consisted of a mechanically rotating optical element which modulated the polarization of light incident on the sample, and the transmitted signal was measured with a bolometer detector using standard lock-in techniques at harmonics of the rotator frequency. Measurements of cyclotron resonance in GaAs verified this technique.

Schematics for measuring $\text{Re}(\theta_F)$ and $\text{Im}(\theta_F)$ are shown in the insets of Fig. 1. To measure the Faraday angle $\text{Re}(\theta_F)$, polarized light was projected through a mechanically rotating linear polarizer with rotation angle $\phi(t) = 2\pi f_{\text{rot}}t$, ($f_{\text{rot}} = 70$ Hz). The light then passed through the sample, striking a stationary polarizer before reaching the bolometer detector. The phase shift in the second harmonic of the bolometer signal is identically the Faraday rotation, $\text{Re}(\theta_F) = \text{Re}(t_{xy}/t_{xx})$, where t_{ij} is the transmittance tensor relating the transmitted field in the j direction to the incident field in the i direction [15].

$\text{Im}(\theta_F)$ is measured analogously (inset of Fig. 1), with the light modulated into right and left elliptical polariza-

tions by a mechanically rotating wave plate and omitting the polarizer previously in front of the detector. The wave plate shifts the optical phase of the field component along its extraordinary axis by the retardance $\beta(\omega)$. In this case, the in-phase second harmonic response is

$$P = 2|E_i|^2 \sin(\beta) \text{Im}(t_{xx}^* t_{xy}). \quad (3)$$

Calibrating separately $\beta(\omega)$ and the transmitted spectral intensity, $S \approx \frac{|E_i|^2}{2}(t_{xx}^* t_{xx})$, we find

$$\text{Im}(\theta_F) = \text{Im}\left(\frac{t_{xy}}{t_{xx}}\right) = \left[\frac{P}{4S \sin(\beta)}\right]. \quad (4)$$

Figure 1 plots the directly measured $\text{Re}(\theta_F)$ and $\text{Im}(\theta_F)$ parts of the Faraday angle. The agreement of $\text{Im}(\theta_F)$ with the Kramers-Kronig (KK) transform of $\text{Re}(\theta_F)$ confirms the consistency of our measurements, and justifies the use of the lower noise, wider bandwidth KK transform to represent $\text{Im}(\theta_F)$ in the following calculations.

Combining this complex θ_F with the complex longitudinal conductivity determined from extended-Drude fits to the transmission data [17,18] (inset, Fig. 2), the Hall angle is determined over the full 20–250 cm^{-1} range using Eq. (2) (Fig. 2). We see that $\text{Re}(\theta_H^{-1})$ extrapolates to the dc transport values measured on a separate sample using the Van der Pauw geometry, and the dc points exhibit the T^2 law universally observed in the cuprates.

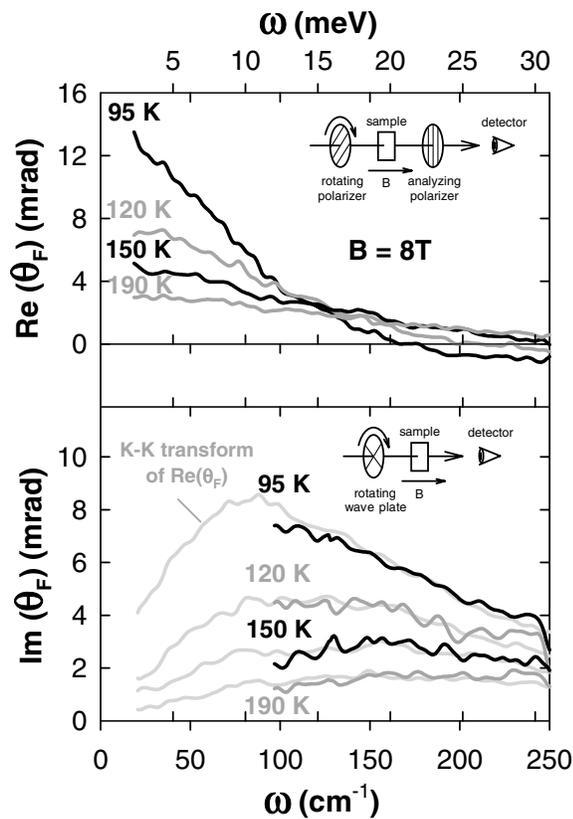


FIG. 1. The measured complex Faraday angle. Insets show separate experimental setups for measuring $\text{Re}(\theta_F)$ and $\text{Im}(\theta_F)$. The Kramers-Kronig transform of $\text{Re}(\theta_F)$ is plotted in gray.

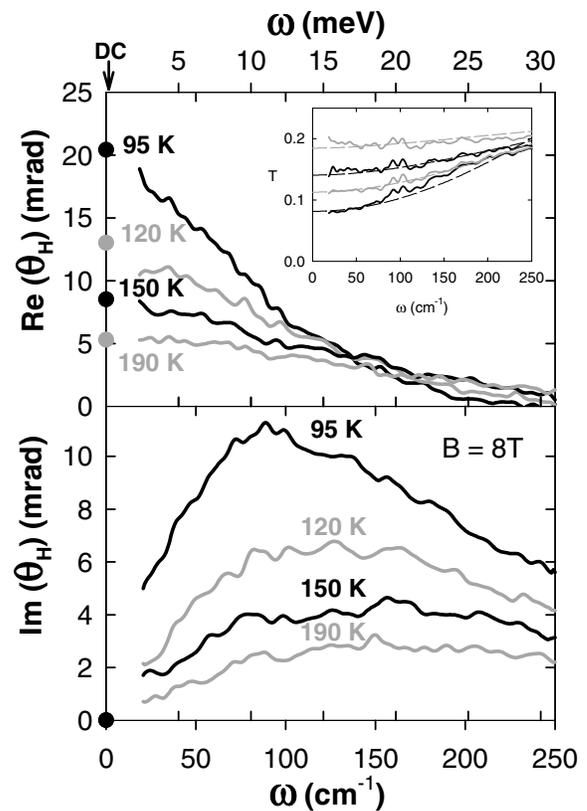


FIG. 2. The derived complex Hall angle. Inset shows transmission data with extended Drude conductivity fits for each temperature in dashed lines.

We first check consistency with the linear-scattering (Lorentzian) form of Refs. [5–11] by solving Eq. (1) for the Lorentzian parameters $\omega_H^*(\omega) = -\omega/\text{Im}[\theta_H^{-1}(\omega)]$ and $\gamma_H^*(\omega) = \omega_H^*(\omega)\text{Re}[\theta_H^{-1}(\omega)]$ in Fig. 3. This parametrization demonstrates that θ_H is *not* Lorentzian over the full frequency range since $\gamma_H(\omega)$ shows additional frequency dependence, decreasing significantly above 100 cm^{-1} particularly at low temperatures. This result is puzzling, because ac conductivity [18] and angle resolved photoemission spectroscopy (ARPES) [19] both show the generally expected *increase* in scattering at higher frequencies.

For the moment, we assume this behavior may result from other energy scales in the problem not treated in the Lorentzian models. (The π - π resonance, superconducting gap energy, and $\omega \sim T$ all occur around $300\text{--}500\text{ cm}^{-1}$.) Accordingly, we will focus on the low-frequency limit to critically evaluate the Lorentzian theories. We plot the dc limit values $\gamma_H^{*\text{dc}} = \lim_{\omega \rightarrow 0} \gamma_H^*(\omega)$ and $\omega_H^{*\text{dc}} = \lim_{\omega \rightarrow 0} \omega_H^*(\omega)$ in the inset of Fig. 3 as averaged from 20 to 80 cm^{-1} . Open squares plot consistency with Ref. [13] at 95 K. Looking at the temperature dependence, we immediately see that $\gamma_H^{*\text{dc}}(T) \sim T$ and $\omega_H^{*\text{dc}}(T) \sim 1/T$. This confirms the Lorentzian behavior proposed in one spinon-

holon model [7] and in the skew-scattering model [11]. However, this data does not support the other Lorentzian theories which posit a temperature *independent* numerator and a scattering rate quadratic in temperature [5,6,8–10]. We reemphasize, however, that *all* of the Lorentzian theories break down at moderate frequencies above 100 cm^{-1} .

As an alternative to the linear-scattering Lorentzian model, we consider a square-scattering form for the Hall angle, which corresponds to a Lorentzian-squared ac response. This form was predicted by Varma and Abrahams in their marginal Fermi liquid treatment of Hall scattering in the cuprates [12,20]. Extending their result to finite frequencies, we get the square-Lorentzian form:

$$\theta_H(\omega) = \frac{\omega_H^* \Omega_p^*}{(\Gamma_H^* - i\omega)^2}. \quad (5)$$

ω_H^* is still the cyclotron frequency, linear in B , and Ω_p^* is indicative of a new energy scale in the problem, interpreted as a Fermi surface average of the scattering derivative [12]. Solving for the square-scattering rate Γ_H^* and weighting parameter $\omega_H^* \Omega_p^*$, one sees at once the independence of $\omega_H^* \Omega_p^*$ from both temperature and frequency in Fig. 4. The existence of such a robust parameter that is constant over $20\text{--}250\text{ cm}^{-1}$ in infrared energy and $95\text{--}190\text{ K}$ in temperature is remarkable, and provides convincing evidence that the square-Lorentzian analysis may elucidate the dominant physics. In the dc limit, the scattering rate, $\Gamma_H^{*\text{dc}}$, is again linear in temperature, but with twice the value as in the Lorentzian analysis. At higher frequencies $\Gamma_H^*(\omega)$ now shows a moderate *increase*, qualitatively consistent with scattering rates observed in ac conductivity [18] and ARPES [19].

Although the square-Lorentzian functional form for the Hall angle is a valid causal response function, it cannot be correct at all frequencies since it leads to a Hall sum of zero [16]: $\omega_H = \frac{2}{\pi} \int_0^\infty \text{Re}[\tan[\theta_H(\omega)]] d\omega = 0$, inconsistent with the finite ω_H implied by ARPES measurements of the Fermi surface [19] and band theory. The positive Hall angle measured at 1000 cm^{-1} by Cerne *et al.* [15] is an indication that the functional form is already changing. The exact behavior of this crossover is therefore an interesting topic for further investigation.

It is interesting to consider not only Γ_H^* , the renormalized ac observable deduced from the square-Lorentzian analysis, but also the *bare* scattering Γ_H . At dc, the bare and renormalized values satisfy $\omega_H \Omega_p / \Gamma_H^2 = \omega_H^* \Omega_p^* / \Gamma_H^{*2}$. This bare Hall scattering Γ_H is linear in temperature just as the bare longitudinal scattering γ_o is linear in temperature as seen in the famous $\rho \sim T$ relation. The renormalizations enter differently, however, between longitudinal and Hall transport since Γ_H^* stays linear in temperature, but γ_o^* increases superlinearly due to a decreasing renormalized transport mass m_o^* [18]. This difference might be accounted for with vertex corrections in the Kubo formula which enter differently in σ_{xx} and σ_{xy} . The $\cot(\theta_H) \sim \Gamma_H^{*2}$ form obtained by

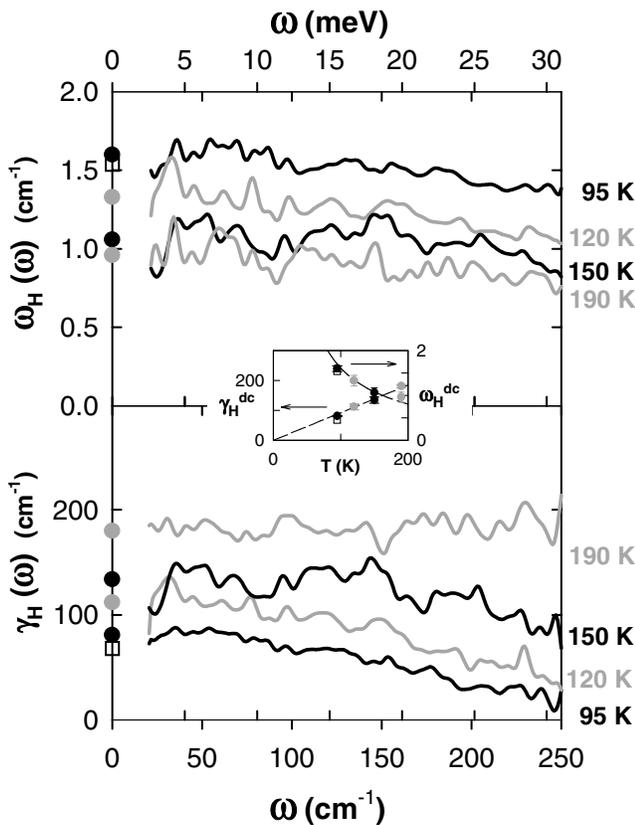


FIG. 3. Lorentzian parametrization of Hall angle. The Hall scattering γ_H develops strong downwards frequency dependence above 100 cm^{-1} , implying a deviation from simple Lorentzian behavior. Inset plots the low-frequency limit of these parameters vs T , open squares plot Ref. [13] at 95 K.

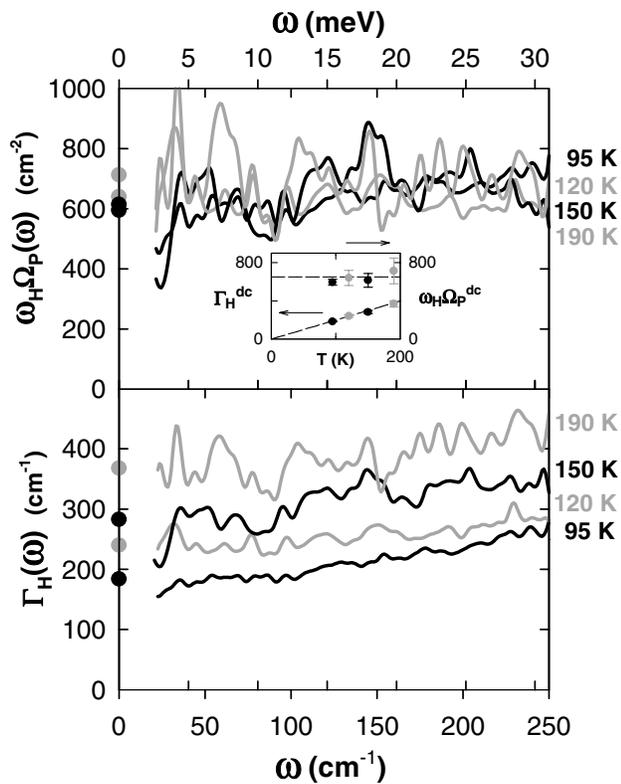


FIG. 4. Square-Lorentzian parametrization of Hall angle. The scattering parameter Γ_H shows a moderate increase with frequency and $\omega_H \Omega_p$ is notably temperature and frequency independent. Inset plots the low-frequency limit of these parameters vs T .

Ref. [12] is itself a consequence of vertex corrections in σ_{xy} . Therefore, the relation between γ_o^* and Γ_H^* presents an interesting subject for future theoretical and experimental work.

In summary, we have measured the complex Hall response at infrared frequencies. The observed $\theta_H(\omega)$ does not fit the Lorentzian line shape predicted by many models of transport in the cuprates or conventional transport theory; only at low frequencies is the data consistent with a subset of linear-scattering models [7,11] that predict the observed behavior: $\gamma_H^*(T) \sim T$, $\omega_H^*(T) \sim 1/T$. Alternately, the data show a good fit to a square-Lorentzian form, over the *entire* frequency range, with the temperature dependence of the response function coming again from a relaxation rate linear in temperature, $\Gamma_H^*(T) \sim T$. In all cases, the experiments suggest that transport in the cuprates is governed throughout by a relaxation rate that is linear in temperature.

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