Infrared Hall effect in high T\textsubscript{c} superconductors: Evidence for non-Fermi liquid Hall scattering

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Abstract

Infrared (20-120 cm\textsuperscript{-1} and 900-1100 cm\textsuperscript{-1}) Faraday rotation and circular dichroism are measured in high T\textsubscript{c} superconductors using sensitive polarization modulation techniques. Optimally doped YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7} thin films are studied at temperatures down to 15 K and magnetic fields up to 8 T. At 1000 cm\textsuperscript{-1} the Hall conductivity \(\sigma_{xy}\) varies strongly with temperature in contrast to \(\sigma_{xx}\) which is nearly independent of temperature. The Hall scattering rate \(\gamma_H\) has a \(T^2\) temperature dependence but, unlike a Fermi liquid, depends only weakly on frequency. The experiment puts severe constraints on theories of transport in the normal state of high T\textsubscript{c} superconductors.
The “normal” state of high Tc superconductors (HTSC) shows behaviors in many physical properties that are anomalous in comparison with conventional metals [1]. These anomalous properties occur in both the optimally doped materials that we discuss in this Letter and even more dramatically in the underdoped pseudogap materials. The DC resistivity \( \rho \) varies linearly with temperature \( T \) from \( T_c \) to the melting point. The Hall coefficient \( R_H \) while positive and decreasing with hole doping, varies with temperature as \( 1/T \) [2]. The cotangent of the DC Hall angle \( \theta_H \), which in simple metals has the same temperature dependence as \( \rho \), varies in HTSC as \( T^2 \) [3]. This anomalous behavior of the Hall effect appears to occur in the superconducting state as well, where thermal magneto-transport shows a similar \( T^2 \) behavior of the thermal Hall angle and thermal Hall scattering rate below \( T_c \) [4]. In contrast, the zero field quasiparticle scattering rate in the superconducting state has a \( T^4 \) dependence [4]. The normal state anomalies have been observed in DC measurements over a wide range of temperatures and in a wide variety of HTSC materials. Normal state far infrared (far-IR) measurements near \( T_c \) have determined that the scattering rate associated with \( \theta_H \) is 3-4 times smaller than the scattering rate associated with the longitudinal conductivity \( \sigma_{xx} \) [6]. On the other hand, recent angularly resolved photoemission spectroscopy (ARPES) work reports that the quasiparticle scattering rate \( \gamma \) is minimum along the \((\pi, \pi)\) direction of the Brillouin zone and varies as \( \gamma \approx \text{max}(\omega, \pi T) \) [7]. Also, since the Fermi velocity is maximum in this direction these quasiparticles should dominate both \( \sigma_{xx} \) and \( \sigma_{xy} \) [7]. Therefore experiments not only suggest that the longitudinal conductivity \( \sigma_{xx} \) and the transverse conductivity \( \sigma_{xy} \) differ fundamentally in HTCS but that they differ from the predictions of Fermi liquid theory.

There have been many different proposed explanations for the anomalous Hall effect in HTSC [8–13]. These explanations can be classed into two basic theoretical approaches. The first approach argues that the system is a Fermi liquid (FL), with excitations consisting of hole-like quasiparticles, but that the strong anisotropy of the Fermi surface and/or the quasiparticle scattering on the Fermi surface causes a non-Drude behavior of the transport [8–10]. The other theoretical approaches argue that the system has non-FL properties, with excitations composed of more exotic entities. In Anderson’s theory [12] based on Luttinger liquid ideas spinons and holons have different relaxation mechanisms. Coleman et al. [13] have considered the transport processes of the quasiparticles associated with charge conjugation symmetry of the system. Many of these models can account qualitatively for the DC measurements. This circumstance provides a motivation for extending measurements into the infrared (IR), where experiments may allow a critical test of the proposed models.

In this Letter we report measurements of the magneto-optical response of optimally doped YBa\(_2\)Cu\(_3\)O\(_7\) thin films in a broad range of far-IR (20-140 cm\(^{-1}\)) and mid-infrared (mid-IR, 900-1100 cm\(^{-1}\)) frequencies using novel polarization modulation techniques [14]. From these measurements and the zero field optical conductivity we extract the full magnetoconductivity as well as the complex Hall angle \( \theta_H \).

The samples are optimally doped YBa\(_2\)Cu\(_3\)O\(_7\) thin films grown on Si or LaSrGaO\(_4\) substrates. The primary sample used for the mid-IR measurements consists of a 150 nm thick film grown on LaSrGaO\(_4\), with a \( T_c \) and \( \Delta T_c \) of 88.2 K and 0.6 K, respectively. Since interference (etalon) effects can have a strong effect on IR Hall measurements, the substrates were either coated with a NiCr broadband antireflection coating [15] or wedged 1 degree to remove multiply reflected beams.
The measured quantity in the magneto-optical experiments is the complex Faraday angle \( \theta_F \) defined as \( \tan \theta_F = t_{xy}/t_{xx} \), where \( t_{xx} \) and \( t_{xy} \) are the complex transmission amplitudes. By determining \( \sigma_{xx} \) through zero magnetic field transmittance and reflectance measurements, Maxwell’s equations can be used to transform \( \theta_F \) into the more interesting transport quantities, \( \sigma_{xy} \), the off-diagonal component of the complex magneto-conductivity tensor, \( \theta_H \), the complex Hall angle defined as \( \tan \theta_H = \sigma_{xy}/\sigma_{xx} \) and the complex Hall coefficient \( R_H = \sigma_{xy}/\sigma_{xx}^2 \). For the experiments reported in this Letter, \( \theta_H \) is small, so that \( \tan \theta_F \approx \theta_F \) and \( \tan \theta_H \approx \theta_H \). \( \theta_H \) has the analytic properties of a response function, obeying Kramers-Kronig relations (KKR) and a sum rule [14]. In general \( \tan \theta_H \), the ratio of two response functions, is a complicated function which does not have a simple closed form. The simplest generalization of \( \theta_H^{-1} \) to finite frequency \( \omega \) is [17,14]:

\[
\theta_H^{-1} = \frac{\gamma_H}{\omega_H} - i \frac{\omega}{\omega_H},
\]

where \( \omega_H \) is the Hall frequency and \( \gamma_H \) is the Hall scattering frequency. Equation (1) is valid for a Drude metal in which case, \( \omega_H = \omega_c \) and \( \gamma_H = \gamma \), where \( \omega_c \) and \( \gamma \) are the conventional cyclotron frequency and isotropic Drude scattering rate, respectively. Equation (1) is also valid for a Fermi liquid for the case of a \( k \) independent scattering time, and it is the form obtained in several proposed models of the normal state transport in HTSC [10,9,12]. Furthermore, Eq. (1) represents the general high frequency limiting behavior of \( \theta_H \) [14].

In the far-IR we measure the Faraday rotation with a rotating linear polarizer. We fit the data to the empirically observed Lorentzian behavior of the Hall angle [6] (see Eq. 1). The Hall frequency, \( \omega_H \), and Hall scattering rate, \( \gamma_H = 1/\tau_H \) are determined from the fit.

In the mid-IR, the Hall angle is small (on the order of \( 10^{-3} \) Rad at 8 T) since \( \omega_H << (\gamma_H, \omega) \) for this experiment. Therefore, a sensitive technique is required for the mid-IR \( \theta_H \) measurements [14]. A CO2 laser produces linearly polarized mid-IR radiation which passes in the Faraday geometry through the sample, located at the center of an 8 T magneto-optical cryostat. In order to sensitively measure both the real and imaginary parts of \( \theta_F \) (i.e., the rotation and ellipticity of the transmitted polarization), the radiation that is transmitted by the sample is analyzed using a ZnSe photoelastic modulator (PEM). A liquid nitrogen-cooled mercury-cadmium-telluride element detects the radiation, and three lock-in amplifiers demodulate the resulting time-dependent signal. By analyzing both the even and odd harmonic signals of the PEM modulation frequency, one can simultaneously measure both the real and imaginary parts of \( \theta_F \) with a sensitivity of better than 1 part in \( 10^4 \) and \( 4 \times 10^3 \), respectively.

Figure [1] shows the complex Hall conductivity \( \sigma_{xy} \) as a function of temperature at 8 T. The Hall conductivity is the most directly accessible transport response function determined from the experiment. It is also interesting because it is expected to be least affected by the conductivity of the Cu-O chains. \( \text{Re}[\sigma_{xy}] \) in Fig. [1](a) shows strong temperature dependence whereas the \( \text{Im}[\sigma_{xy}] \) in Fig. [1](b) shows little temperature dependence. The factor of five increase in \( \text{Re}[\sigma_{xy}] \) at low temperature is striking since both the real and imaginary parts of \( \sigma_{xx} \) at 1000 cm\(^{-1} \) vary by less than 20% over the same temperature range [13,13]. Note that the \( \text{Re}[\sigma_{xy}] \) only shows frequency dependence at lower temperatures while \( \text{Im}[\sigma_{xy}] \) shows a uniform decrease with frequency across all temperatures. \( \sigma_{xy} \) is not consistent with a Drude model with a temperature dependent carrier scattering rate. Nor is it in the high frequency limit (\( \omega \gg \gamma \)) where general considerations imply that \( \sigma_{xy} \propto (-1/\omega^2 + 2i\gamma/\omega^3) \), where \( \gamma \) is
a momentum space average of the scattering rate [17,14]. The weak frequency dependence of \( \text{Re}[\sigma_{xy}] \) and the fact that \( \text{Im}[\sigma_{xy}] \) is not small compared with \( \text{Re}[\sigma_{xy}] \) indicates that by 1000 cm\(^{-1}\) the system is not yet in the asymptote \( (\omega > \gamma) \) regime.

We have also examined the complex Hall angle and the Hall coefficient. Since \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) contains Cu-O chains in addition to the Cu-O planes, the longitudinal conductivity \( \sigma_{xx} \) is anisotropic in single domain samples [20,21]. Therefore, the \( \sigma_{xx} \) measured for the twinned thin films used in this experiment is an average of the conductivities of the chains and planes. It is most interesting to examine the transport quantities of the Cu-O planes since the chains do not contribute significantly to the superconductivity. We can use the results for \( \sigma_{xx} \) on single domain samples in Ref. [21] to characterize the chain conductivity in \( \text{YBa}_2\text{Cu}_3\text{O}_7 \). The chain conductivity is sample dependent so that we can only estimate their effects on our twinned films. We expect the chain contribution to be smaller than observed in single domain samples since our polycrystalline films are more likely to have disorder which can very easily upset the one dimensional chain conductivity. This is confirmed by our measurements of the mid-IR \( \sigma_{xx} \), whose temperature and frequency behavior is consistent with a conductivity that is dominated by the planes. We note that the chain contribution in the far-IR has been reported to be negligible [20,21]. From these considerations we conclude that the magnitude of the chain corrections is less than 30 \% and 10 \% for the real and imaginary parts of the mid-IR \( \sigma_{xx} \) respectively. Because of the uncertainties, however, we have not removed the contribution from the Cu-O chains in this Letter but rather we will discuss their effects as we present the data.

Because of the form of \( \theta_H \) given by many of the theoretical models (see Eq. 1) it is most interesting to examine the complex inverse Hall angle \( \theta_H^{-1} \). Figure 2 shows the temperature dependence of the mid-IR \( \theta_H^{-1} \) at 8 T. The \( \text{Re}[\theta_H^{-1}] \) shows strong temperature dependence in Fig. 2(a) consistent with a temperature dependent \( \gamma_H \). The \( \text{Im}[\theta_H^{-1}] \) is nearly temperature independent in the normal state in Fig. 2(b), which is consistent with a nearly temperature independent \( \omega_H \) (see Eq. 1). The solid line in Fig. 2(a) shows the measured DC \( \theta_H^{-1} \) which is seen to agree well with the mid-IR \( \text{Re}[\theta_H^{-1}] \). The \( \text{Re}[\theta_H^{-1}] \) in Fig. 2(a) shows no frequency dependence, which is consistent with a frequency independent \( \gamma_H \), while the frequency dependence of \( \text{Im}[\theta_H^{-1}] \) is consistent with Eq. 1 and a frequency independent \( \omega_H \).

If we assume the Lorentzian form for \( \theta_H \) given by Eq. 1 we can extract the normal state Hall frequency \( \omega_H \) and Hall scattering rate \( \gamma_H \) which are shown in Fig. 3 as a function of temperature at 8 T. \( \omega_H \) shows little temperature dependence in Fig. 3(a), while strong temperature dependence is observed for the mid-IR \( \gamma_H \) in Fig. 3(b). Both \( \omega_H \) and \( \gamma_H \) appear to be frequency independent. Also, both the mid-IR \( \omega_H \) and \( \gamma_H \) are in good with agreement our far-IR measurements and with the 200 cm\(^{-1}\) result obtained by Ref. 13. The agreement of the far-IR and mid-IR \( \omega_H \) improves when the estimated Cu-O chain contribution to \( \sigma_{xx} \) is removed, which causes an increase of up to 30 \% in the mid-IR \( \omega_H \). This correction only weakly affects the mid-IR \( \gamma_H \), which is reduced by less than 10 \%.

The previous results are used to determine the mid-IR complex Hall coefficient \( R_H \) as a function of temperature. For a Drude metal \( R_H \) is real and independent of frequency. While the measured \( \text{Re}[R_H] \) shows weak temperature dependence the \( \text{Im}[R_H] \) is large and shows strong temperature dependence. At low temperatures, \( R_H \) is mostly imaginary showing again that the experiment is not near the high frequency limit at 1000 cm\(^{-1}\). The DC values of \( R_H \) are more than a factor of five larger than the mid-IR value at 100 K, but approach
the mid-IR results at higher temperatures. No frequency dependence is observed for the mid-IR \( \text{Re}[R_H] \), while the mid-IR \( \text{Im}[R_H] \) is large and weakly frequency dependent at lower temperatures.

The agreement of the mid-IR and far-IR results in Fig. 3 shows that the frequency dependence of \( \omega_H \) and \( \gamma_H \) is very weak. The conductivity relaxation rate \( \gamma \) for a Fermi liquid has the following form:

\[
\gamma = \frac{1}{W} \left[ \left( \frac{\omega}{p \pi} \right)^2 + T^2 \right],
\]

where \( p = 2 \) is the calculated result [22] for the optical response and \( p = 1 \) is observed for heavy fermion systems [23]. \( W \) represents a characteristic energy scale for the electron system. We find \( W \approx 120 \text{ K} \) from the far-IR data [3][4]. The mid-IR data fits a \( T^2 \) dependence (thin solid line in Fig. 3(b)) with a small offset. This results in an estimate for \( W \) that is approximately 160 K. The predicted \( \gamma_H \) based on Fermi liquid theory and the observed \( T^2 \) dependence of \( \gamma_H \) in the IR is shown as a dashed line in Fig. 3(b) which clearly disagrees with the mid-IR results, demonstrating the weak frequency dependence and a non-Fermi liquid behavior of the Hall scattering.

The weak frequency dependence of \( \gamma_H \) is internally consistent with the agreement of the far-IR and mid-IR values for \( \omega_H \). As discussed previously [17], a frequency dependent \( \gamma_H \) should produce, by the properties of response functions, a frequency dependent effective \( \omega_H \) saturating to its true sum rule value as \( \omega \to \infty \). It was pointed out in the earlier work [17] that \( \omega_H(\omega) \) for both the superconducting (\( \omega_H^s \)) and normal state (\( \omega_H^n \)) appeared to saturate by 200 cm\(^{-1} \), but they saturated to different values with \( \omega_H^n(200) \approx 2 \omega_H^s(200) \). The mid-IR data show that \( \omega_H^n(200) \) is close to the correct sum. Therefore it is expected that \( \text{Re}[\theta_H] \) in the superconducting state must be negative between 200 cm\(^{-1} \) and 1000 cm\(^{-1} \). This is just the behavior that the superconducting energy gap in \( \sigma_{xx} \) is expected to cause and is confirmed by the mid-IR measurement, which shows that \( \text{Re}[\theta_H] \) becomes negative below \( T_c \).

These IR Hall results can be compared with theoretical models of the Hall conductivity. The Ong-Anderson model [12] assumes that \( \sigma_{xx} \) and \( \theta_H \) are controlled by different scattering rates. The IR experiments can be explained within this model by choosing the frequency and temperature dependence of these rates appropriately. In the two scattering time (\( \tau \)) models of Refs. [10] and [13], the two \( \tau \)’s do not separately control \( \sigma_{xx} \) and \( \theta_H \) so that the experimentally observed behavior does not occur so naturally. The skew scattering model [11] fails because it predicts the wrong temperature and frequency dependence of the far-IR magneto-conductivity [10]. The Ioffe-Millis model [9] involves only one \( \tau \). Although this model is consistent with the mid-IR data, it appears to contradict the two \( \tau \) behavior observed in the far-IR [25]. More detailed analysis of data on several different HTSC materials may allow definitive tests of these models.

The Hall scattering rate obtained in this work has a \( T^2 \) temperature dependence but is independent of frequency. This behavior differs from the IR conductivity and the ARPES results in high \( T_c \) superconductors which show that the quasiparticle scattering rate has a linear dependence on both temperature and frequency. In general, the temperature dependence of carrier relaxation in solids, which usually arises from inelastic scattering from phonons, magnons, or the electron-electron interaction, has a frequency dependence similar
to the temperature dependence. Therefore the frequency and temperature dependence of $\gamma_H$ that are reported in this Letter are highly unusual and indicate non-Fermi liquid behavior of the normal state of $\text{YBa}_2\text{Cu}_3\text{O}_7$. However, Ioffe and Millis [9] have recently proposed such a relaxation rate behavior based on quasi-elastic scattering from superconducting fluctuations in the normal state of high $T_c$ materials. Fluctuation effects have also been observed in normal state of underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ in measurements of the longitudinal conductivity by THz spectroscopy [20].

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FIGURES

FIG. 1. The real (a) and imaginary (b) parts of the Hall conductivity $\sigma_{xy}$ at 8 T as a function of temperature at 949 cm$^{-1}$ (●) and 1079 cm$^{-1}$ (△).

FIG. 2. The real (a) and imaginary (b) parts of the inverse Hall angle $\theta_H^{-1}$ at 8 T as a function of temperature at 949 cm$^{-1}$ (●) and 1079 cm$^{-1}$ (△). The thin solid line in (a) shows the values for $\theta_H^{-1}$ obtained using DC Hall measurements at 8 T.

FIG. 3. (a) The Hall frequency, $\omega_H$, and (b) the Hall scattering rate, $\gamma_H$, at 8 T as a function of temperature for far-IR (□ = 20-150 cm$^{-1}$, and + is from Ref. [10]) and mid-IR (● = 949 cm$^{-1}$, and △ = 1079 cm$^{-1}$) frequencies. The solid line in (b) shows a $T^2$ fit to the mid-IR data, with the dashed line showing the expected mid-IR Hall scattering rate based on Fermi liquid theory (Eq. 2). Equation [1] ceases to be relevant in the hatched region below $T_c$, but the data are plotted for completeness.
