Infrared Hall Effect in High-T_c Superconductors: Evidence for Non-Fermi-Liquid Hall Scattering

J. Černe,[1,*] M. Grayson,[1] D. C. Schmadel,[1] G. S. Jenkins,[1] H. D. Drew,[1] R. Hughes,[2] A. Dabkowski,[2] J. S. Preston,[2] and P.-J. Kung[3]

[1]*Center for Superconductivity Research and Department of Physics, University of Maryland, College Park, Maryland 20741*
[2]*Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada, L8S 4M1*
[3]*Advanced Fuel Research, Inc., East Hartford, Connecticut 06108*

(Received 3 August 1999)

Infrared (20–120 and 900–1100 cm^{-1}) Faraday rotation and circular dichroism are measured in high-T_c superconductors using sensitive polarization modulation techniques. Optimally doped $YBa_2Cu_3O_7$ thin films are studied at temperatures in the range ($15 < T < 300$ K) and magnetic fields up to 8 T. At 1000 cm^{-1} the Hall conductivity σ_{xy} varies strongly with temperature in contrast to the longitudinal conductivity σ_{xx} which is nearly independent of temperature. The Hall scattering rate γ_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_c superconductors.

PACS numbers: 74.20.Mn, 74.25.Nf, 74.72.Bk, 74.76.Bz

The "normal" state of high-T_c superconductors (HTSC) exhibits many physical properties that are anomalous in comparison with conventional metals [1]. The dc resistivity ρ for HTSC 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]. Yet the cotangent of the dc Hall angle θ_H, which in simple metals has the *same* temperature dependence as ρ, *varies as* T^2 in HTSC: the so-called anomalous Hall effect [3].

Frequency dependent measurements likewise reveal anomalous Hall behavior. Normal state far infrared (far-IR) measurements just above T_c have determined that the scattering rate (γ_H) associated with θ_H is 3–4 times smaller than the scattering rate (γ_{xx}) associated with the longitudinal conductivity σ_{xx} [4]. Recent angularly resolved photoemission spectroscopy (ARPES) work reports that the quasiparticle scattering rate γ is minimum along the (π, π) direction of the Brillouin zone and varies in that direction as $\gamma \approx \max(\omega, \pi T)$ [5]. With the Fermi velocity maximum in this direction these quasiparticles should dominate *both* σ_{xx} and σ_{xy} [5] in a conventional Fermi-liquid (FL) model. Experimental evidence therefore suggests that the different behaviors underlying σ_{xx} and σ_{xy} cannot be easily reconciled in a simple FL model.

There have been many different proposed explanations for the anomalous Hall effect in HTSC [6–12]. These explanations can be classed into two basic theoretical approaches. The first approach argues that the system is still a Fermi liquid 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 [6–9]. The other theoretical approaches argue that the system has non-FL properties, with excitations composed of more exotic entities. In Anderson's theory [11] based on Luttinger liquid ideas, spinons and holons have different relaxation mechanisms. Coleman *et al.* [12] 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 temperature dependent Hall measurements into the IR, where experiments may allow a critical test of the proposed models.

In this Letter we report the first measurements of the magneto-optical response of optimally doped $YBa_2Cu_3O_7$ thin films from 900–1100 cm^{-1} in the mid-IR and compare them for the first time with temperature dependent measurements in the far-IR (20–140 cm^{-1}) [13]. We use novel polarization modulation techniques to measure the real and imaginary parts of the Faraday angle [14]. These measurements together with the zero field optical conductivity yield the full magnetoconductivity tensor as well as the complex Hall angle θ_H.

The samples are optimally doped $YBa_2Cu_3O_7$ thin films grown on Si or $LaSrGaO_4$ substrates. The primary sample used for the mid-IR measurements consists of an 150 nm thick film grown by pulsed laser deposition on $LaSrGaO_4$, with a T_c and ΔT_c of 88.2 and 0.6 K, respectively, and a dc resistivity of 60 $\mu\Omega$ cm at T_c. The infrared conductivity of this film obtained from measurements of its reflectance and transmittance is in good agreement with published data on twinned single crystals [15]. Since interference (etalon) effects can have a strong influence on IR Hall measurements, the substrates were either coated with a NiCr broadband antireflection coating [16] or wedged 1° to remove multiply reflected beams.

The measured quantity in the magneto-optical experiments is the complex Faraday angle θ_F defined as $\tan\theta_F = t_{xy}/t_{xx}$, where t_{xy} and t_{xx} are the complex transmission amplitudes. After determining σ_{xx} from zero magnetic field transmittance and reflectance measurements, Maxwell's equations can be used to transform θ_F into the

 0031-9007/00/84(15)/3418(4)$15.00

more interesting transport quantities, σ_{xy}, the off-diagonal component of the complex magnetoconductivity tensor, θ_H, the complex Hall angle defined as $\tan\theta_H = \sigma_{xy}/\sigma_{xx}$ [14], and the complex weak field Hall coefficient $R_H = \sigma_{xy}/\sigma_{xx}^2$. The $\tan\theta_H$ has the analytic properties of a response function, obeying Kramers-Kronig relations and a sum rule [17]. 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 $\cot\theta_H$ to finite frequency ω is [14,18]

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

where ω_H is the Hall frequency and γ_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 ω_c and γ 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 momentum independent scattering time, and it is the form obtained in several proposed models of the normal state transport in HTSC [7,8,11]. Furthermore, Eq. (1) represents the first two terms in a $1/\omega$ expansion of $\cot\theta_H$ [14].

In the far-IR we measure the Faraday rotation with a rotating linear polarizer. The data fit well to the empirically observed Lorentzian behavior of the Hall angle [4] [see Eq. (1)]. The Hall frequency, ω_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 $\omega_H \ll (\gamma_H, \omega)$ for this experiment so that $\tan\theta_H \cong \theta_H$. Therefore, a sensitive technique is required for θ_H measurements in this frequency range [14]. A CO_2 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 measure both the real and imaginary parts of θ_F (i.e., the rotation and ellipticity of the transmitted polarization), the transmitted radiation is analyzed using a ZnSe photoelastic modulator (PEM) with a mercury-cadmium-telluride detector and three lock-in amplifiers to 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 θ_F with a sensitivity of better than 1 part in 10^4 and 4×10^3, respectively [14].

Figure 1 shows the complex Hall conductivity σ_{xy} as a function of temperature at 8 T. The Hall conductivity is the most directly accessible magnetotransport response function determined from the experiment. It is also interesting because it is expected to be least affected by the Cu-O chains in $YBa_2Cu_3O_7$ [19]. The $\mathrm{Re}(\sigma_{xy})$ in Fig. 1(a) shows strong temperature dependence whereas the $\mathrm{Im}(\sigma_{xy})$ in Fig. 1(b) shows little temperature dependence. The factor of 4 increase in $\mathrm{Re}(\sigma_{xy})$ at low temperature is striking since in contrast both the real and imaginary parts of σ_{xx} at 1000 cm^{-1} vary by less than 20% over the same temperature range [15]. σ_{xy} is not consistent with a Drude model containing a temperature dependent carrier scattering rate. Also, the weak frequency dependence of $\mathrm{Re}(\sigma_{xy})$ and the fact that $\mathrm{Im}(\sigma_{xy})$ is not small compared with $\mathrm{Re}(\sigma_{xy})$ indicate that by 1000 cm^{-1} the system is not yet in the asymptotic ($\omega \gg \gamma$) regime where $\sigma_{xy} \propto (-1/\omega^2 + 2i\gamma/\omega^3)$, where γ is a momentum space average of the scattering rate [14,18].

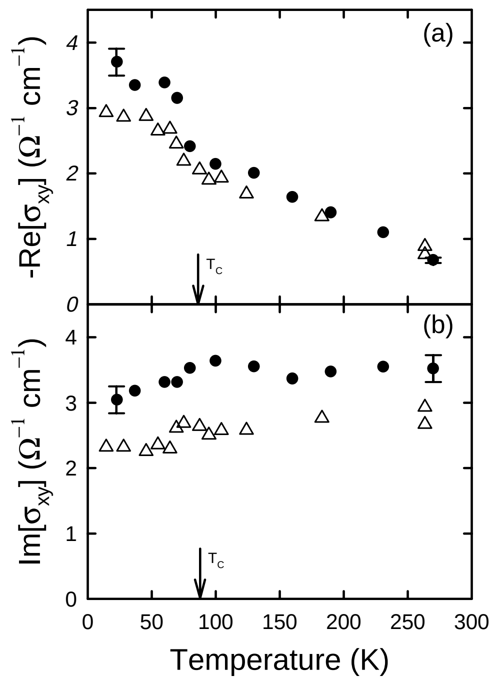

FIG. 1. The real (a) and imaginary (b) parts of the Hall conductivity σ_{xy} at 8 T as a function of temperature at 949 cm^{-1} (•) and 1079 cm^{-1} (△).

We also have examined the complex Hall angle and the Hall coefficient which, unlike σ_{xy}, are complicated by the Cu-O chains in $YBa_2Cu_3O_7$. Because of the chains, the longitudinal conductivity σ_{xx} is anisotropic in single domain samples [20,21]. Therefore, the σ_{xx} measured for the twinned thin films used in this experiment is an average of the conductivities of the chains and the Cu-O planes. It is most interesting to examine the transport quantities of the planes since the chains do not contribute significantly to the superconductivity. Reported results for σ_{xx} on single domain samples [20,21] can be used to characterize the chain conductivity in $YBa_2Cu_3O_7$. The chain conductivity is sample dependent so that we can only estimate their effects on our twinned films. Moreover, we expect the chain contribution to be smaller than observed in single domain samples since our polycrystalline films are likely to be more disordered which can easily upset the one dimensional chain conductivity. This is confirmed by our measurements of the mid-IR σ_{xx}, whose temperature and frequency behavior are consistent with a conductivity that is dominated by the planes. 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 σ_{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. Also, we note that since the chain conductivity varies more weakly with frequency compared with the plane response, the effects of the chains are less important in the far-IR [20,21].

Because of the form of θ_H given by many of the theoretical models [see Eq. (1)] it is most interesting to examine the complex inverse Hall angle θ_H^{-1}. Figure 2 shows the temperature dependence of the mid-IR θ_H^{-1} at 8 T. The $\mathrm{Re}(\theta_H^{-1})$ in Fig. 2(a) shows strong temperature dependence and no frequency dependence consistent with a temperature dependent but frequency independent γ_H. Whereas the $\mathrm{Im}(\theta_H^{-1})$ is frequency dependent but nearly temperature independent in the normal state in Fig. 2(b), which is consistent with an ω_H that is nearly temperature and frequency independent [see Eq. (1)]. The solid line in Fig. 2(a) shows the measured dc θ_H^{-1} for the same film which is seen to agree well with the mid-IR $\mathrm{Re}(\theta_H^{-1})$.

If we assume the Lorentzian form for θ_H given by Eq. (1) we can extract the normal state Hall frequency ω_H and Hall scattering rate γ_H which are shown in Fig. 3 as a function of temperature at 8 T. ω_H shows little temperature dependence in Fig. 3(a), while strong temperature dependence is observed for the mid-IR γ_H in Fig. 3(b). Both ω_H and γ_H appear to be frequency independent. Also, both the mid-IR ω_H and γ_H are in good agreement with our far-IR measurements as well as those of Ref. [4]. The agreement of the far-IR and mid-IR ω_H improves when the estimated Cu-O chain contribution to σ_{xx} is removed, which causes an increase of up to 30% in the mid-IR ω_H. This correction only weakly affects the mid-IR γ_H, which is reduced by less than 10%.

These results also can be used to determine the mid-IR complex Hall coefficient R_H ($R_H B = \theta_H^2/\sigma_{xy}$). For a Drude metal R_H is real and independent of temperature and frequency. The measured mid-IR $\mathrm{Re}(R_H)$ has a weak temperature dependence in contrast to the approximately T^{-1} behavior generally found for the dc R_H in the cuprates. The dc values of R_H are more than a factor of 5 larger than the mid-IR value at 100 K, but approach the mid-IR results at higher temperatures. The $\mathrm{Im}(R_H)$ shows strong temperature dependence and, at low temperatures, R_H is mostly imaginary showing again that, at 1000 cm^{-1}, the experiment is not near the high frequency limit.

A comparison of the mid-IR and the far-IR in Fig. 3 yields striking evidence for non-FL behavior. The agreement of the mid-IR and far-IR results in Fig. 3 shows that the frequency dependence of ω_H and γ_H is very weak. The conductivity relaxation rate γ for a Fermi liquid has the form ($\hbar = k_B = 1$):

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

where $p = 2$ is the calculated result [22] for the optical response and $p = 1$ is observed for heavy fermion systems

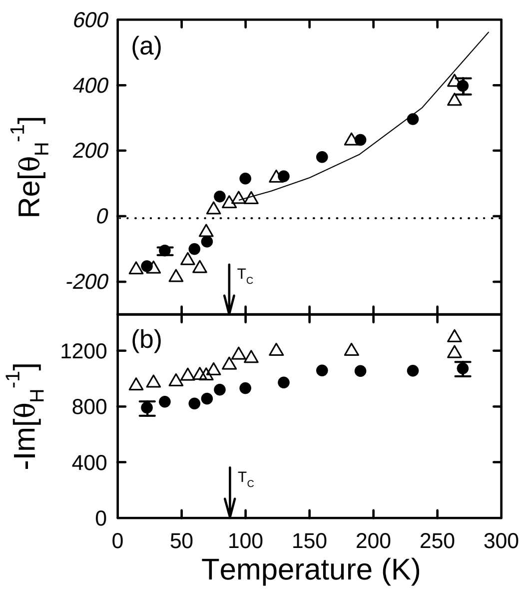

FIG. 2. The real (a) and imaginary (b) parts of the inverse Hall angle θ_H^{-1} at 8 T as a function of temperature at 949 cm^{-1} (•) and 1079 cm^{-1} (△). The thin solid line in (a) shows the values for θ_H^{-1} obtained using dc Hall measurements at 8 T.

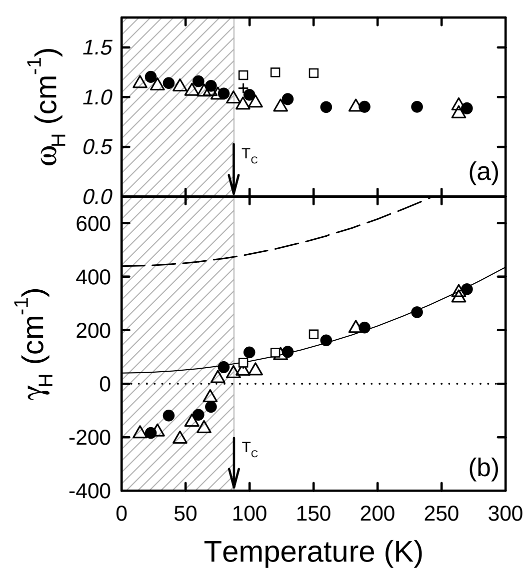

FIG. 3. (a) The Hall frequency, ω_H, and (b) the Hall scattering rate, γ_H, at 8 T as a function of temperature for far-IR (□ = 20–150 cm^{-1}; + is from Ref. [17]) and mid-IR (• = 949 cm^{-1}, and △ = 1079 cm^{-1}) frequencies. The solid line in (b) shows a T^2 fit to the mid-IR data, with the dashed 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.

[23]. W represents a characteristic energy scale for the electron system. We find $W \approx 120$ K from the far-IR data [4,13] and the mid-IR data also fit a T^2 dependence [thin solid line in Fig. 3(b)] with a W of approximately 160 K. The predicted γ_H at 1000 cm^{-1} based on Fermi-liquid theory and the observed T^2 dependence of γ_H is shown as a dashed line in Fig. 3(b) which clearly disagrees with the mid-IR results. This result demonstrates the weak frequency dependence and non-Fermi-liquid behavior of the Hall scattering.

These IR Hall results can be compared with theoretical models of the Hall conductivity. The Ong-Anderson model [11] assumes that σ_{xx} and θ_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 this case the lack of frequency dependence of γ_H is puzzling. In the two scattering time (τ) models of Refs. [8] and [12], the two τ's do not separately control σ_{xx} and θ_H so that the experimentally observed behavior is not readily accounted for. Simple assumptions about the temperature and frequency dependence of the two τ's do not lead to agreement with the IR data. The skew scattering model [10] fails because it predicts the wrong temperature and frequency dependence of the far-IR magnetoconductivity [8]. The Ioffe-Millis model [7] involves only one τ. Although this model is consistent with the mid-IR data, it appears to contradict the two τ behavior observed in the far-IR [24]. A more detailed analysis of data on several different HTSC materials may allow definitive tests of these models.

It also is interesting to compare these mid-IR Hall effect results on YBCO with those on Au and Cu films [14,18]. For Au and Cu both γ_{xx} and γ_H are temperature dependent and frequency independent as expected from electron-phonon scattering when the measurement frequency is higher than the Debye frequency. For YBCO in the mid-IR, however, while γ_H has a strong temperature dependence and no frequency dependence, γ_{xx} is temperature independent but frequency dependent (as found in IR optics and ARPES). The behavior of γ_{xx} precludes phonons or magnons as the dominant scatterers and the lack of frequency dependence of γ_H is in contrast to the predicted and observed behavior of a Fermi liquid or inelastic scattering in general. Therefore the frequency and temperature dependence of γ_H that are reported in this Letter are highly unusual and indicate a non-Fermi-liquid behavior of the normal state of $YBa_2Cu_3O_7$. We note that Ioffe and Millis [7] have recently proposed such a relaxation rate behavior based on quasielastic scattering from superconducting fluctuations in the normal state of high-T_c materials. Fluctuation effects have also been observed in the normal state of underdoped $Bi_2Sr_2CaCu_2O_8$ in measurements of the longitudinal conductivity by THz spectroscopy [25].

We acknowledge useful discussions with A. J. Millis, N. P. Ong, V. Yakovenko, and P. D. Johnson. This work was supported in part by the NSF Grant No. DMR-9705129, by the NSA, and the NSERC.

*Present address: Department of Physics, State University of New York at Buffalo, Buffalo, NY 14260.

[1] P. W. Anderson, *The Theory of Superconductivity in the High-T_c Cuprates* (Princeton University Press, Princeton, 1997).
[2] K. Kamaras *et al.*, Phys. Rev. Lett. **64**, 84 (1990); L. Forro *et al.*, Phys. Rev. Lett. **65**, 1941 (1990).
[3] T. R. Chien *et al.*, Phys. Rev. B **43**, 6242 (1991); T. R. Chien *et al.*, Phys. Rev. Lett. **67**, 2088 (1991); J. M. Harris *et al.*, Phys. Rev. B **46**, 14 293 (1992).
[4] S. G. Kaplan *et al.*, Phys. Rev. Lett. **76**, 696 (1996).
[5] T. Valla *et al.*, Science **285**, 2110 (1999); A. Kaminski *et al.*, cond-mat/9904390.
[6] A. Carrington *et al.*, Phys. Rev. Lett. **69**, 2855 (1992).
[7] L. B. Ioffe and A. J. Millis, Phys. Rev. B **58**, 11 631 (1998).
[8] A. T. Zheleznyak *et al.*, Phys. Rev. B **57**, 3089 (1998).
[9] B. P. Stojkovic and D. Pines, Phys. Rev. B **55**, 8576 (1997).
[10] G. Kotliar *et al.*, Phys. Rev. B **53**, 3573 (1996).
[11] P. W. Anderson, Phys. Rev. Lett. **67**, 2092 (1991).
[12] P. Coleman *et al.*, Phys. Rev. Lett. **76**, 1324 (1996).
[13] M. Grayson *et al.*, J. Low Temp. Phys. **117**, 1055 (1999).
[14] J. Cerne *et al.*, Phys. Rev. B **61**, 8133 (2000).
[15] Z. Schlesinger *et al.*, Phys. Rev. B **41**, 11 237 (1990).
[16] S. W. McKnight *et al.*, Infrared Phys. **27**, 327 (1987).
[17] H. D. Drew and P. Coleman, Phys. Rev. Lett. **78**, 1572 (1997).
[18] H. D. Drew *et al.*, J. Phys. Condens. Matter **8**, 10 037 (1996).
[19] For σ_{xy} in Fermi-liquid theory the mean free path is averaged over the Fermi surface with a weighting that is proportional to the local curvature. The contributions of the chain bands are expected to be small because their Fermi surface is relatively flat.
[20] D. N. Basov *et al.*, Phys. Rev. Lett. **74**, 598 (1995); S. L. Cooper *et al.*, Phys. Rev. B **47**, 8233 (1993).
[21] J. Schützmann *et al.*, Phys. Rev. B **46**, 512 (1992).
[22] R. N. Gurzhi *et al.*, Zh. Eksp. Teor. Fiz. **35**, 965 (1958) [Sov. Phys. JETP **8**, 673 (1959)].
[23] P. E. Sulewski *et al.*, Phys. Rev. B **38**, 5338 (1988).
[24] A. T. Zheleznyak *et al.*, Phys. Rev. B **59**, 207 (1999).
[25] J. Corson *et al.*, Nature (London) **398**, 22 (1999).