High-frequency Hall effect in the normal state of $YBa_2Cu_3O_7$

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The ac Hall effect of $YBa_2Cu_3O_7$ was measured in the normal state using time-domain spectroscopy. In contrast to some theories that explain the anomalous dc Hall effect by postulating a much slower quasiparticle scattering rate for Hall conduction than for normal conduction, we report a Hall scattering rate that is consistent with the normal scattering rate. [S0163-1829(97)02926-3]

The normal-state Hall effect of the cuprate superconductors has been heavily studied because of its anomalous temperature dependence. This has been particularly true since Chien *et al.*[1] showed that the temperature dependence could be fit neatly by the expression $\cot\theta_H \equiv \sigma_{xx}/\sigma_{xy} = \alpha T^2 + C$. Anderson explained this observation in terms of spinon scattering in a ''tomographic Luttinger liquid.''[2] Since then, there have been several other explanations for the temperature dependence of the dc Hall effect, two of which make predictions for the ac behavior, as well.[3,4] These theories can be tested by measurements of the ac Hall effect.

The frequency range of importance to measurements of the ac Hall effect is 100–3000 GHz, which spans the range of Hall relaxation rates predicted by the above-mentioned theories. This ''millimeter-wave'' regime severely stretches the low-frequency limit of Fourier-transform infrared spectroscopy and the high-frequency limit of microwave techniques. Measurements of the complex conductivity in this regime can be difficult, even when only the diagonal part of the conductivity $\sigma_{xx}(\omega)$, and not the much smaller Hall conductivity $\sigma_{xy}(\omega)$, is sought. Recently Kaplan *et al.* performed an experiment using far-infrared Fourier-transform techniques, supplemented by measurements with a far-infrared laser.[5] They found that the Hall scattering rate at 95 K was half to one-quarter of the transport scattering rate. However, they were not able to measure the temperature dependence of the Hall effect because the Hall angle decreases approximately as $1/T$.

The experiments described in this paper complement those of Kaplan *et al.*. We use coherent time-domain spectroscopy to probe the real and imaginary response function without the need for Kramers-Kronig analysis. In time-domain terahertz spectroscopy, nearly single-cycle picosecond electrical pulses are guided quasioptically and transmitted through the sample.[6] The transmitted pulse is detected coherently. Fourier transformation of the transmitted pulse leads directly to the real and imaginary parts of the conductivity through the equation

$$E_{\mathrm{trans}}(\omega) = t(\omega) E_{\mathrm{in}}(\omega) \tag{1}$$

with

$$t(\omega) = \frac{4ne^{-i\phi}}{(n+1)[n+1+Z_0\sigma(\omega)d]}, \tag{2}$$

where n is the refractive index of the substrate,[7] ϕ is the phase change on passing through the substrate, $Z_0 = 377\ \Omega$ is the impedance of free space, $\sigma(\omega)$ is the complex conductivity of the film, and d is the thickness of the film.

In the presence of a magnetic field, Eq. (2) is modified by making the replacements $t_\pm$ for t and $\sigma_\pm$ for σ, where $\pm$ refers to the handedness of circularly polarized light. While the equations are simplest for circularly-polarized light, our broadband source dictates the use of linear polarizers. We placed them in the positions shown in Fig. 1 and measured the transmission through both parallel and crossed polarizers. The ratio of these two quantities is approximately equal to the cotangent of the Hall angle, $\cot\theta_H$:

$$\frac{t_{xy}}{t_{xx}} = \frac{-Z_0 d\sigma_{xy}}{n+1+Z_0 d\sigma_{xx}} \approx \frac{-\sigma_{xy}}{\sigma_{xx}}. \tag{3}$$

Since the ratio $Z_0 d\sigma_{xx}(100\ \mathrm{K})/(n+1) \simeq 7$ for this sample, the approximation is good, but not perfect. In order to calculate the Hall angle exactly, we need the magnetoconductivity, $\sigma_{xx}(H)$ at temperatures above T_c. In experiments on this sample and other similar samples, we have found that within

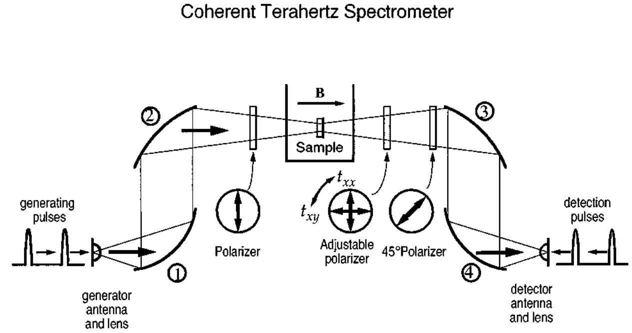

FIG. 1. Time-domain spectrometer for measurement of the complex transmission tensor in the presence of a magnetic field. Diagonal t_{xx} and off-diagonal t_{xy} components are selected by rotating the analyzing polarizer through 90°. The effect of linear birefringence is removed by reversing the field direction.

the accuracy of our measurements, $\sigma_{xx}(H)=\sigma_{xx}(H=0)$. We have also observed that within our frequency and temperature range, the conductivity is nicely fit by a Drude model: $\sigma(\omega)=ne^2/m(\Gamma-i\omega)$, with the scattering rate Γ proportional to T. We use this approximation to generate the small correction term $1+(n+1)/Z_0 d\sigma_{xx}$, which when multiplied by t_{xy}/t_{xx} yields the desired quantity $\sigma_{xy}/\sigma_{xx}\equiv\tan\theta_H$.

The data presented here were taken on a $YBa_2Cu_3O_7$ sample that was nominally 70 nm thick, with $\rho_{dc}(100\ K)=120\ \mu\Omega$ cm and $\Gamma(100\ K)=3400$ GHz. It was grown by off-axis sputtering on a sapphire substrate with a CeO_2 buffer layer and had $T_c=$ 85 K.[8] Additional data were also collected from two other films: a 40 nm film grown by off-axis sputtering on $LaAlO_3$ with $T_c=$ 88 K and a 70 nm film grown by laser ablation on $LaAlO_3$ with $T_c=$ 88 K. The data on these two other films were too noisy to show any clear temperature dependence and therefore are not presented here; however, the values of the real and imaginary parts of the Hall angle at 100 K were consistent with those reported here.

The data were all obtained at a field of magnitude 6 T. In order to eliminate signals due to leakage through the polarizers, the experiments were performed at both positive and negative fields, and the differences between these data were analyzed: $t_{xy}/t_{xx}=\frac{1}{2}[t_{xy}(6T)/t_{xx}(6T)-t_{xy}(-6T)/t_{xx}(-6T)]$. This procedure, which is described in more detail in Ref. 6, subtracts out any effects that do not depend on the sign of the magnetic field, while preserving the Hall effect, which is odd in magnetic field. Any Faraday rotation due to the substrate is also preserved, but this was measured to be negligible ($<2\times10^{-4}$ rad).

In Fig. 2 we plot $\cot\theta_H\equiv\sigma_{xx}/\sigma_{xy}$ versus frequency at temperatures from 100 to 200 K. Note that the real part of the ratio is much larger than the imaginary part and is consistent with a frequency-independent fit. These frequency-independent values are plotted against T^2 in Fig. 3. We observe that $\cot\theta_H\propto T^2$, just as is seen in dc measurements. The slope and y intercept of the data are consistent with that seen for pure samples, slightly oxygen-deficient samples ($YBa_2Cu_3O_{6.9}$), or those doped with zinc or cobalt at the 1% level.[1,9,10]

The imaginary part of $\cot\theta_H$ is much smaller than the real part, and it would be difficult to determine a functional form for it from the data. This is particularly true at higher temperatures, since t_{xy} decreases roughly as $1/T$, decreasing the signal-to-noise ratio at higher temperatures. However, it is possible to put an upper limit on the ratio of the imaginary part to the real part, and this upper limit is important in the theoretical models, as is discussed below.

In the Drude model, the cotangent of the Hall angle is

$$\frac{\sigma_{xx}}{\sigma_{xy}}=\frac{\Gamma-i\omega}{\omega_c}, \tag{4}$$

where Γ is the scattering rate and ω_c is the cyclotron frequency, $\omega_c=eB/m^*c$. If we insert into this equation the temperature dependence of Γ obtained from zero-field conductance and infrared reflectance measurements, $\Gamma\propto T$, then we find $\sigma_{xx}/\sigma_{xy}\propto T$. This is clearly not observed experimentally, so more complex models must be considered.

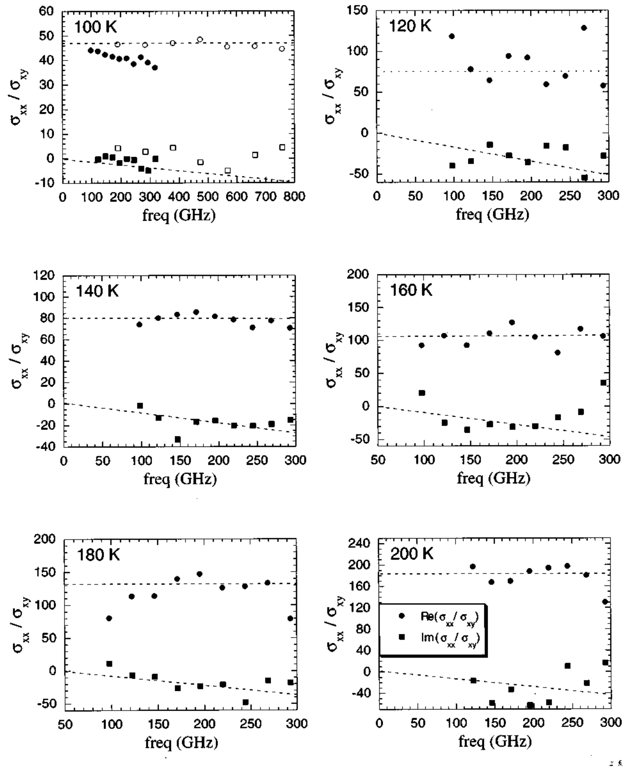

FIG. 2. Real (circles) and imaginary (squares) parts of $\sigma_{xx}/\sigma_{xy}=\cot\theta_H$ plotted versus frequency at several temperatures in a 6 T magnetic field. Notice that the frequency range is higher for the 100 K data because data were also collected with a set of higher-frequency antennas (open symbols). The dashed lines are approximate fits to the data, with an attempt to choose the highest reasonable slope for the imaginary part of the data in order to give the lowest reasonable lower bound to the Hall scattering rate, as is discussed in the text.

Anderson[2] explained the dc results by showing that within his model, a different scattering rate appears in the Hall conductivity: $\sigma_{xx}\propto\tau_{tr}$ and $\sigma_{xy}\propto\omega_c\tau_H\tau_{tr}$ with $\tau_{tr}\propto T^{-1}$ and $\tau_H\propto T^{-2}$. However, he did not extend the model to higher frequencies. Romero[11] proposed a high-frequency extension of Anderson's model. Within this model, the frequency depen-

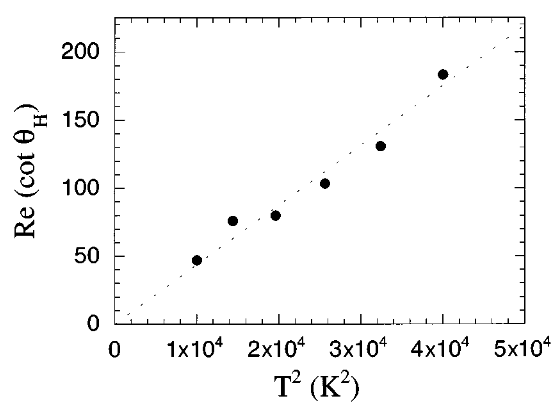

FIG. 3. The real part of $\sigma_{xx}/\sigma_{xy}=\cot\theta_H$ from Fig. 2 plotted versus T^2 at 6 T. The linear fit is the same as is seen by other groups in dc measurements.

dence of the Hall angle would be governed by the transport lifetime $\tau_{\rm tr}$. The other lifetime τ_H appears only in the combination $\omega_c \tau_H$, and thus it is not possible to use ac Hall-effect measurements to distinguish between a heavy effective mass and a long lifetime.

Coleman *et al.*[3] postulated that processes that preserve charge conjugation symmetry have one scattering rate, $\Gamma_f \propto T$, while those that do not have a slower rate, $\Gamma_s \propto T^2$. This leads to conductivities

$$\sigma_{xx} = \frac{\omega_p^2/4\pi}{(\Gamma_f + \Gamma_s)/2 - i\omega}, \tag{5}$$

$$\sigma_{xy} = \frac{\omega_c \omega_p^2/4\pi}{(\Gamma_f - i\omega)(\Gamma_s - i\omega)}. \tag{6}$$

These predictions are consistent with the dc results in the limit $\Gamma_f \gg \Gamma_s$, where

$$\cot\theta_H = \sigma_{xx}/\sigma_{xy} \cong 2\frac{\Gamma_s - i\omega}{\omega_c}. \tag{7}$$

This expression predicts that the characteristic frequency seen in ac measurements of the Hall effect (Γ_s) will be much smaller than that seen in measurements of σ_{xx}.

Kotliar *et al.*'s model[4] differs from those of Anderson and Coleman *et al.* in that the temperature dependence is caused by skew scattering. This model predicts that the ac Hall effect will show the same frequency dependence as σ_{xx}. The anomalous temperature dependence of the Hall angle appears as a temperature-dependent cyclotron frequency.

Clearly, the best way to distinguish between these models would be to measure the temperature dependence of the imaginary part of the Hall angle. Our upper bound on $\mathrm{Im}(\sigma_{xx}/\sigma_{xy})$ places constraints on the theories. Taking our cue from the form of the cotangent of the Hall angle in the simple transport model of Eq. (4), we examine the quantity

$$-\omega \frac{\mathrm{Re}(\sigma_{xx}/\sigma_{xy})}{\mathrm{Im}(\sigma_{xx}/\sigma_{xy})}, \tag{8}$$

which is equal to the scattering rate within the models of Kotliar *et al.* and Coleman *et al.* The data in Fig. 2(a) lead to a lower bound of 2500 GHz for this quantity. This is comparable to the scattering rate of 3400 GHz inferred from measurements of σ_{xx} at 100 K on the same sample.

The lower limit on the scattering rate is consistent with those models in which the frequency dependence of the Hall effect is governed by the transport scattering rate. The model of Coleman *et al.*, however, is difficult to resolve with the 100 K data. In order to yield the required T^2 temperature dependence of the Hall effect, their model requires that Γ_s is much smaller than Γ_f. If the Hall effect is to be proportional to T^2 up to 300 K, then it is necessary to have $\Gamma_s(100\ \mathrm{K}) < 500$ GHz. With the observed $\Gamma_s(100\ \mathrm{K}) > 2500$ GHz, the Hall effect would appear to be linear in T^2 only up to about 60 K.

The data presented in this paper are reasonably consistent with the measurements of Kaplan *et al.* They found that in two samples with transport scattering rates of 127 cm^{-1} (3810 GHz) and 190 cm^{-1} (5700 GHz) at 95 K, the Hall scattering rates were 68 and 52 cm^{-1}, respectively. Both their data and our data support the idea that theories requiring a much smaller Hall scattering rate than the transport scattering rate cannot be applicable. Finally, we note that our data are consistent with theories predicting that the two scattering rates are exactly the same.

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