The undoped cuprates are two-dimensional Mott insulators with a large antiferromagnetic exchange interaction ($J \sim 0.2$ eV). Upon hole doping the antiferromagnetism is destroyed at hole density $p \sim 0.1$ and the superconductivity is optimized at $p \sim 0.16$. Further doping leads to decreasing $T_c$ and more or less conventional Fermi-liquid behavior. The underdoped metallic region between $0.1 \leq p \leq 0.16$ has attracted much attention because anomalies in many physical properties have led to a widespread belief that therein lies the key to understanding high temperature superconductivity. Specific heat, magnetic susceptibility, transport, and optical measurements suggest a partial gapping of the Fermi surface that has been termed a pseudogap [1]. The various interpretations of this pseudogap, which include an anisotropic Mott gap or various types of density waves, are controversial [1].

Therefore, a key question is the route by which these different scenarios approach the Mott state as the doping is reduced. The underdoped metallic region between $0.1 \leq p \leq 0.16$ has attracted much attention because anomalies in many physical properties have led to a widespread belief that therein lies the key to understanding high temperature superconductivity. Specific heat, magnetic susceptibility, transport, and optical measurements suggest a partial gapping of the Fermi surface that has been termed a pseudogap [1]. The various interpretations of this pseudogap, which include an anisotropic Mott gap or various types of density waves, are controversial [1].

In this Letter we present evidence that supports partial gapping of the Fermi surface by a measurement of the Hall frequency from underdoped YBa$_2$Cu$_3$O$_{6+x}$ samples. Which shows a clear Fermi surface at optimal oxygen concentration. A dramatic increase of the Hall frequency is observed for underdoped samples, which is not consistent with the approach to a Mott transition but is consistent with a partial gapping of the Fermi surface as predicted in density wave models.
concentration is lowered below optimal doping in YBa$_2$Cu$_3$O$_{6+x}$.

The measured quantities in the magneto-optical experiments are the Faraday rotation and the circular dichroism. The $\tan\theta_H$ is determined from the magneto-optical data and $\sigma_{xx}$ determined separately from zero magnetic field transmittance and reflectance measurements. Also the samples have very little magnetoresistance as the laser frequency as the sample transmission has no measurable variation with field. In our case of low fields and strong scattering, the Faraday and Hall angles are very small (a few milliradians at most at 8 T) and nearly equal to their tangents. The measurement of such small Faraday angles requires a very sensitive technique which is accomplished by using a ZnSe photoelastic modulator to analyze the change in the polarization of IR radiation from a modulator to analyze the change in the polarization of which is accomplished by using a ZnSe photoelastic

Small Faraday angles requires a very sensitive technique which is accomplished by using a ZnSe photoelastic modulator to analyze the change in the polarization of IR radiation from a CO$_2$ laser after it is transmitted by the sample. Experimental details are described elsewhere [9].

The samples used in this study were optimally doped YBa$_2$Cu$_3$O$_7$, underdoped YBa$_2$Cu$_3$O$_{6.65}$, severely underdoped YBa$_2$Cu$_3$O$_{4.5}$, and optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_8$ thin films. The YBa$_2$Cu$_3$O$_7$ samples are 100 nm thick films grown on LaSrGaO$_4$ substrates. Deoxygenation of the underdoped samples is obtained by annealing at controlled temperature and oxygen pressure. The optimal and underdoped samples reported here have $T_c$’s of 89 K and 55–60 K, respectively. The 6.4 sample does not show any superconducting transition down to 4.2 K. The Bi$_2$Sr$_2$CaCu$_2$O$_8$ samples are approximately 200 nm thick films peeled from a bulk single crystal and placed on a 0.5 mm thick BaF substrate.

Before presenting our results, we note that the analysis of the Hall measurements on YBa$_2$Cu$_3$O$_{6+x}$ thin films are complicated by the Cu-O chain contributions to the conductivity. Their nearly one-dimensional character implies not only small contributions to $\sigma_{xy}$ but also an anisotropic longitudinal conductivity $\sigma_{xx}^{\mathrm{chain}}$ that is observed to be sample dependent and comparable to the plane conductivity at 1000 cm$^{-1}$ [10]. Consequently, the chain contributions cannot be reliably subtracted to obtain the pure in-plane $\sigma_{xx}$ for our twinned films as has been discussed elsewhere [11]. Therefore, we have not attempted to correct the Hall data presented here for the effects of chains. This leads to uncertainties in the determination of the Cu-O plane Hall response. However, the chain conductivity is nearly frequency independent and real in the mid-IR so that its effects on the Hall angle are relatively benign and easy to characterize qualitatively [11]. The comparison of the optimally doped YBa$_2$Cu$_3$O$_{6+x}$ and chainless optimally doped BSCCO shows that while the results differ in detail the important effects are clearly observable. We discuss the effects of the chains on the results as we discuss the data.

Figure 1 displays the results of the temperature dependence of the real part (left panel) and the imaginary part (right panel) of the Hall angle for the doping values of $x = 0.4$, $x = 0.65$, and $x = 0.93$ at 950 cm$^{-1}$. For $x = 0.65$ and 0.93 doping, two different samples measured with two different techniques (one based on magnetic field sweeps at fixed temperature, the other one on temperature scans at fixed field) are shown with very good agreement between each data set. In all cases, this strong temperature dependence for the Hall angle shows that $\sigma_{xy}$ is strongly temperature dependent as $\sigma_{xx}$ is only weakly temperature dependent in the mid-IR as has been reported in the literature [10] and as we have confirmed in zero field measurements on our samples.

Previous frequency dependence measurements on optimally doped YBa$_2$Cu$_3$O$_{6+x}$ have shown that the spectral response function of the Hall angle in the mid-IR regime follows, approximately, a conventional Lorentzian form where $\omega_H$ is the Hall frequency and $\gamma_H$ is the Hall scattering rate [11]. This Drude-like form can be obtained from Fermi-liquid theory for $\gamma_H \ll \omega$ and from many of the theoretical models for the magnetotransport in the normal state of cuprates mentioned in the introduction. With this form of spectral response, it is most revealing to plot the inverse Hall angle:

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

Figure 2 shows the frequency dependent measurement for a $x = 0.65$ underdoped YBa$_2$Cu$_3$O$_{6.65}$ sample at 100 K and for an optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_8$ sample at 300 K. In both cases a Drude-like behavior is observed with a frequency independent real part of the inverse Hall angle and an imaginary part linear with frequency. While $\text{Im}(\theta_H^{-1})$ for optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_8$ linearly extrapolates to zero, the extrapolation for the YBCO films gives a positive intercept. We understand this as a consequence of the chain contributions to $\sigma_{xx}$ in YBCO from simulations with estimated chain conductances. The imaginary part of the inverse Hall angle is found to

![FIG. 1. Temperature dependence of the real (left panel) and imaginary (right panel) part of the Hall angle for YBa$_2$Cu$_3$O$_{6+x}$ thin films with $x = 0.93$ (solid line and open circles), $x = 0.65$ (dotted line and solid squares), and $x = 0.4$ (dashed line).](image-url)
depend strongly on the doping of the sample and is only weakly temperature dependent while the real part is strongly temperature dependent (see Fig. 3). If an estimated chain conductivity is removed from $\sigma_{xx}$, both $\text{Re}(\theta_H^{-1})$ and $\text{Im}(\theta_H^{-1})$ are raised nearly uniformly over the whole temperature range.

We note that the trend in the change of the $T^\alpha$ temperature dependence seen in dc measurements of $\cot(\theta_H)$ on BSCCO [12] and YBCO [13] (i.e., from $\alpha = 1.75$ at optimal doping ($p = 0.16$) to $\alpha = 2$ for $p = 0.05$ underdoping) is observed in a similar but more dramatic way in our mid-IR data with $\alpha \approx 1$ at optimal doping ($x = 0.93$) and $\alpha \approx 2$ for $x = 0.65$ ($p = 0.10$). This behavior even persists in the $T_c = 0$ phase ($x = 0.4$, $p = 0.05$) where $\alpha > 2$. As in dc Hall angle measurements [14], no feature in the temperature dependence of the $\text{Re}(\theta_H^{-1})$ for underdoped samples is observed which could be linked to $T^\alpha$, the characteristic temperature of the opening of a pseudogap. From the real and imaginary parts of the inverse Hall angle the temperature and frequency dependences of the Hall scattering rate and the Hall frequency in the normal state can be directly extracted using Eq. (1).

For YBCO and BSCCO samples, little to no frequency dependence is observed for the Hall scattering rate (not shown but obtainable from Fig. 2) as reported earlier for optimally doped YBCO [11].

The right panel of Fig. 3 displays the temperature dependence of the Hall frequency for the different samples which is seen to vary only weakly with temperature but to increase substantially in underdoped YBCO. It is interesting to compare these experimental values with the values that can be estimated from the ARPES data for optimally doped cuprates. For a Fermi-liquid the Hall frequency can be expressed in terms of integrals of the Fermi velocity over the Fermi surface as [15]:

$$\omega_H = \frac{eB}{hc} \frac{\int dk \nu \times \nu dk}{\int dk |\nu|} = \frac{eB}{hc} \frac{\int dk \nu \times \nu}{\int dk |\nu|},$$  \(2\)

where tetragonal symmetry is assumed. ARPES measurements show that the Fermi surface is approximately circular for optimally doped BSCCO and the velocity varies little around the Fermi surface; therefore we find $\omega_H = \frac{eB}{hc} (v_F/k_F) = 0.33 \text{cm}^{-1}/\text{T}$, where $k_F = 0.71 \text{Å}^{-1}$ is the radius of the Fermi surface and $v_F = 1.8 \text{eV} \text{Å}^{-1}$, both in the zone-diagonal direction [2]. The ARPES measured quantities $k_F$ and $v_F$ correspond to an effective mass, $m^* = \hbar k_F/v_F = 3m_0$, which is about twice the band value and in good agreement with the mass deduced from the infrared conductivity.

In the case of YBa$_2$Cu$_3$O$_{6+x}$ there is comparatively much less ARPES data because of the generally poor quality of the cleaved surfaces [2,16]. As a consequence, we cannot make as reliable a Hall frequency comparison for YBa$_2$Cu$_3$O$_{6+x}$. However, the existing data confirm that YBCO electronic structure, except for the chains, is very similar to that of BSCCO [15,17]. From ARPES data on optimally doped YBCO, we get $\omega_H = 0.22 \text{cm}^{-1}$ with a zone-diagonal Fermi velocity ($v_F$) of $1.3 \text{eV} \text{Å}$ and a Fermi surface radius of $0.74 \text{Å}^{-1}$ [16,17]. Removing the Cu-O chain conductivity contribution estimated from the literature [10] leads to an increase of about 20% of the measured midinfrared Hall frequency which is then in good agreement with the ARPES deduced Fermi mass. Therefore, we base our general discussion of the behavior of the Hall frequency in underdoped YBCO on the behavior of the ARPES data on underdoped BSCCO which are assumed to be similar.

For underdoped BSCCO, the ARPES dispersion curves have been measured in the $(\pi, \pi)$ direction, and they show very little doping dependence of the Fermi velocity. Since $k_F^2$ also does not change significantly with doping the Fermi mass is nearly constant or only weakly decrea-
ing. Therefore, if we first assume that the Fermi surface topology has not changed significantly, as band theory indicates [18], and that the zone-diagonal values of the Fermi velocity are not wildly unrepresentative of the rest of the Fermi surface as is found in optimally doped BSCCO, the Hall frequency would not be expected to change appreciably. Therefore the observed strong increase in the Hall frequency in YBCO as the hole doping is reduced from optimal doping immediately suggests that the Fermi surface topography changes significantly in the pseudogap state and is counter to the expected behavior of the approach to a Mott transition or a stripe phase.

It is interesting to examine the behavior of the Hall frequency expected in the presence of density wave states [3]. Tewari et al., have recently reported a calculation of the Hall angle within the d-density wave (DDW) model [19]. Using the semiclassical Boltzmann theory in the weak field limit they calculate the high frequency Hall response as a function of hole doping at zero temperature. \( \omega_H \) is extracted from the theory from the imaginary part of \( \cot(\theta_H) \) using Eq. (1). Choosing representative parameters for the band structure and the DDW gap, they find a robust growth of \( \omega_H \) similar to the behavior reported here for YBCO. The enhancement of the Hall frequency means that the Drude weight (below the DDW gap) reduces more rapidly than the Hall conductivity. Experimentally the low frequency spectral weight is observed to reduce rapidly with underdoping in the cuprates [1,20]. This behavior corresponds, in part, to the pseudogap phenomenology within the enhanced Drude analysis of the optical data. We note, however, that no evidence for a DW gap has been reported from IR experiments. Within density wave scenarios this is explained with the hypothesis of fluctuating density wave order. Fluctuations should not strongly affect our interpretation since the rise in Hall frequency results primarily from the reduction of the \( \sigma_{xx} \) spectral weight below the DDW gap and \( \sigma_{xy} \) would be affected by fluctuations only near the density wave zone boundary. Because of the many approximations in the theory and the imperfect knowledge of the YBCO band structure and because of the uncertainties in the measurements, arising from the chains in YBCO, the semiquantitative agreement that is observed is the most that can be expected. However, it is clear that other pseudogap states that produce Fermi pockets would give similar qualitative results. Therefore these preliminary results beg for IR Hall measurements on a cuprate with a better characterized band structure and the absence of CuO2 chains such as BSCCO or the electron doped cuprates, which would allow more stringent comparison with theoretical models.

In conclusion, we have reported the first measurements of the mid-IR Hall effect in the normal state of under-doped cuprates for different dopings. The frequency dependence of the Hall angle is Drude-like and indicates a quasielastic relaxation process for optimal and under-doped samples. The observation of a dramatic increase of the Hall frequency in the pseudogap state is the main result of this work. This large increase is inconsistent with expectations from the band structure, ARPES data, the behavior of the Mott transition in 3D or DMFT, or the stripe phase. The rapid increase of \( \omega_H \) is consistent with the presence of Fermi pockets due to a partial gapping of the Fermi surface.

We thank A. Millis, V. Yakovenko, and S. Chakravarty for fruitful discussions and D. Romero for cleaving the BSCCO crystals. The research was partially supported by the NSF under Grant No. DMR-0303112.