Magneto-optical Evidence for a Gapped Fermi Surface in Underdoped $YBa_2Cu_3O_{6+x}$

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The infrared (900–1100 cm^{-1}) Faraday rotation and circular dichroism are measured in the normal state of underdoped High T_c superconductors and used to study the magnetotransport. $YBa_2Cu_3O_{6+x}$ thin films are investigated in the temperature range 10–300 K in magnetic fields up to 8 T and as a function of 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.

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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 gaps, are controversial [1]. Therefore, a key question is the route by which these systems approach the Mott state as the doping is reduced from the optimal T_c. Angle-resolved photoemission spectroscopy (ARPES) measurements on $Bi_2Sr_2CaCu_2O_8$ (BSCCO), which show a clear Fermi surface at optimal or overdoping, show a d-wave symmetric pseudogap in the underdoped materials. The Fermi surface near $(\pi, 0)$ disappears, leaving only "Fermi arcs" in the (π, π) direction [2]. One interpretation of the Fermi arcs is in terms of density wave states associated with $Q = (\pi, \pi)$ scattering of magnetic excitations which partially gap the Fermi surface. One example that has recently been advanced is the d-density wave state in which the density wave gap has d-wave symmetry, which implies broken time reversal symmetry [3,4]. The gapping of the Fermi surface leads to small Fermi surface pockets of which only one-half have appreciable ARPES spectral weight. The plasma frequency (Drude spectral weight) decreases, because of reduced effective carrier density, and vanishes at the metal insulator transition but the Hall frequency (Hall angle spectral weight) and the cyclotron frequency, $\omega_c = 2\pi\frac{eB}{\hbar c}[\oint dk_t/|\nu|]^{-1} \equiv eB/cm_c^*$, are enhanced. Charge ordering in the form of stripes has also been suggested and is observed in some nonsuperconducting cases such as $La_{1.475}Nd_{0.4}Sr_{0.125}CuO_4$ [5]. At other Sr dopings and for other underdoped cuprates the evidence is not so direct or unambiguous [6]. In the stripe phase scenario, the conduction becomes pseudo one dimensional (at least locally) and the Hall conductivity is suppressed while the longitudinal conductivity is not radically affected, leading to a large reduction in ω_H and only modest changes in the plasma frequency. Whereas, in the Mott transition scenario, at least in the case of 3D metals and dynamic mean field theory (DMFT), the Fermi surface remains intact, but spectral weight transfers from a quasiparticle band at the Fermi level to the Hubbard bands away from the Fermi energy [7,8]. Equivalently, the effective mass diverges and the Fermi velocity vanishes as the metal insulator transition is approached. This leads to a vanishing of both the plasma frequency and the Hall frequency, $\omega_H \equiv eB/m_H^*$ [8]. The nature of the Mott transition in quasi-2D is not established and is at the core of the pseudogap mystery. Therefore, as these different scenarios lead to characteristic predictions for the optical spectral weight and the Hall frequency (and cyclotron frequency) as a function of carrier density, measurement of the Hall frequency is important in addressing this important question.

In this Letter we present evidence that supports partial gapping of the Fermi surface by a measurement of the Hall frequency from high frequency magneto-optical measurements on underdoped $YBa_2Cu_3O_{6+x}$ (YBCO). By making the measurements at frequencies above the carrier relaxation rate ($\omega\tau > 1$) the Hall frequency is obtained essentially independently of the unknown k-space dependence of the carrier scattering rates that complicate the dc Hall effect. The experiment shows that the Hall frequency is significantly enhanced as the hole concentration is lowered below optimal doping in $YBa_2Cu_3O_{6+x}$.

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 σ_{xx} determined separately from zero magnetic field transmittance and reflectance measurements. Also the samples have very little magnetoresistance at 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 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 $YBa_2Cu_3O_7$, underdoped $YBa_2Cu_3O_{6.65}$, severely underdoped $YBa_2Cu_3O_{6.4}$, and optimally doped $Bi_2Sr_2CaCu_2O_8$ thin films. The $YBa_2Cu_3O_7$ 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 $Bi_2Sr_2CaCu_2O_8$ 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 $YBa_2Cu_3O_{6+x}$ thin films are complicated by the Cu-O chain contributions to the conductivity. Their nearly one-dimensional character implies not only small contributions to σ_{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 σ_{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 $YBa_2Cu_3O_{6+x}$ 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 σ_{xy} is strongly temperature dependent as σ_{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 $YBa_2Cu_3O_{6+x}$ have shown that the spectral response function of the Hall angle in the mid-IR regime follows, approximately, a conventional Lorentzian form where ω_H is the Hall frequency and γ_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:

$$\theta_H^{-1} = \frac{\gamma_H}{\omega_H} - i\frac{\omega}{\omega_H}. \tag{1}$$

Figure 2 shows the frequency dependent measurement for a $x = 0.65$ underdoped $YBa_2Cu_3O_{6+x}$ sample at 100 K and for an optimally doped $Bi_2Sr_2CaCu_2O_8$ 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 $\mathrm{Im}(\theta_H^{-1})$ for optimally doped $Bi_2Sr_2CaCu_2O_8$ 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 σ_{xx} in YBCO from simulations with estimated chain conductances. The imaginary part of the inverse Hall angle is found to

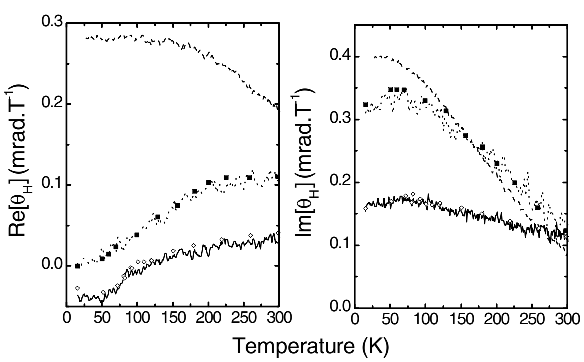

FIG. 1. Temperature dependence of the real (left panel) and imaginary (right panel) part of the Hall angle for $YBa_2Cu_3O_{6+x}$ thin films with $x = 0.93$ (solid line and open circles), $x = 0.65$ (dotted line and solid squares), and $x = 0.4$ (dashed line).

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 σ_{xx}, both $\mathrm{Re}(\theta_H^{-1})$ and $\mathrm{Im}(\theta_H^{-1})$ are raised nearly uniformly over the whole temperature range.

We note that the trend in the change of the T^α temperature dependence seen in dc measurements of $\cot(\theta_H)$ on BSCCO [12] and YBCO [13] (i.e., from $\alpha \approx 1.75$ at optimal doping ($p = 0.16$) to $\alpha \approx 2$ 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 $x = 0.65$ ($p \approx 0.10$). This behavior even persists in the $T_c = 0$ phase ($x = 0.4$, $p \approx 0.05$) where $\alpha > 2$. As in dc Hall angle measurements [14], no feature in the temperature dependence of the $\mathrm{Re}(\theta_H^{-1})$ for underdoped samples is observed which could be linked to T^*, 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]:

$$\omega_H = \frac{eB}{\hbar c} \frac{\oint dk v \times dv/dk}{\oint dk|v|} = \frac{eB}{\hbar c} \frac{\oint dv \times v}{\oint dk|v|}, \qquad (2)$$

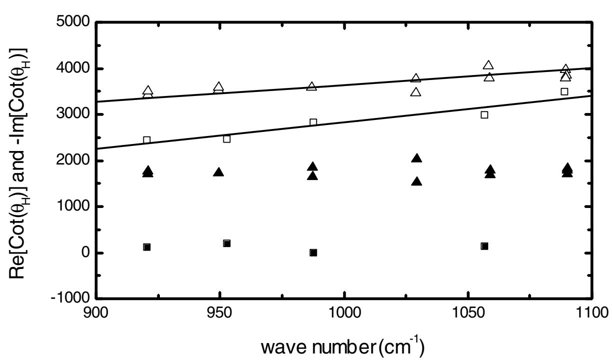

FIG. 2. Frequency dependence of the real (full symbols) and the imaginary part (open symbols) of the cotangent of the Hall angle for a $YBa_2Cu_3O_{6.65}$ thin film at 100 K (square) and for a $Bi_2Sr_2CaCu_2O_8$ thin film at 300 K (triangle). The solid lines are linear fit to the imaginary part.

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 $\omega_H = \frac{eB}{\hbar c}(v_F/k_F) \approx 0.33\ \mathrm{cm}^{-1}/\mathrm{T}$, where $k_F = 0.71\ \mathrm{\AA}^{-1}$ is the radius of the Fermi surface and $v_F = 1.8\ \mathrm{eV\,\AA}$, 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 $YBa_2Cu_3O_{6+x}$ 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 $YBa_2Cu_3O_{6+x}$. 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 $\omega_H = 0.22\ \mathrm{cm}^{-1}$ with a zone-diagonal Fermi velocity (v_F) of $1.3\ \mathrm{eV\,\AA}$ and a Fermi surface radius of $0.74\ \mathrm{\AA}^{-1}$ [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 (π, π) direction, and they show very little doping dependence of the Fermi velocity. Since k_F^* also does not change significantly with doping the Fermi mass is nearly constant or only weakly decreas-

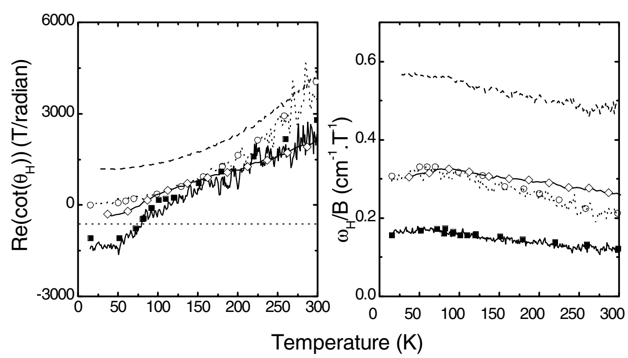

FIG. 3. Left: Temperature dependence of the real part of the cotangent of the Hall angle for $YBa_2Cu_3O_{6+x}$ thin films with $x = 0.93$ (solid line and solid squares), $x = 0.65$ (dotted line and open circles), $x = 0.4$ (dashed line), and optimally doped BSCCO (solid line and open diamonds). Right: Temperature dependence of the Hall frequency for $YBa_2Cu_3O_{6+x}$ thin films with $x = 0.93$ (solid line and solid squares), $x = 0.65$ (dotted line and open circles), $x = 0.4$ (dashed line), and optimally doped BSCCO (solid line and open diamonds).

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. ω_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 ω_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 σ_{xx} spectral weight below the DDW gap and σ_{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 underdoped cuprates for different dopings. The frequency dependence of the Hall angle is Drude-like and indicates a quasielastic relaxation process for optimal and underdoped 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 ω_H is consistent with the presence of Fermi pockets due to a partial gapping of the Fermi surface.

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