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Possibility of magnetic-field-induced reconstruction of the Fermi surface in underdoped cuprates: Constraints from infrared magneto-optics

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We present an analysis of infrared optical and magneto-optical conductivity data for a range of underdoped cuprate superconductors including YBa₂Cu₃O_y and La_{2-x}Sr_xCuO₄. In light of recent magneto-oscillation experiments which have been interpreted in terms of Fermi surface reconstruction in magnetic field, we search for far-infrared signatures of field-induced pockets of coherent quasiparticles. Analysis of the conductivity spectra in magnetic field reveals no sign of field-induced Drude-like response, as well as no evidence for modification of the pseudogap. By considering changes of the low-frequency spectral weight, we are able to place limits on the oscillator strength of coherent modes deriving from proposed field-induced pockets in the Fermi surface. In underdoped La_{2-x}Sr_xCuO₄ we observe a complete suppression of the superfluid density but no evidence for a coherent contribution to the conductivity. Other limits are established for YBa₂Cu₃O_y. We further discuss these results in the context of cuprate systems in which stripe order can account for the observed nodal effects but does not lead to the development of antinodal pockets with long-lived quasiparticles.

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I. INTRODUCTION

The high-temperature copper-oxygen superconductors are reasonably well-understood on the edges of their phase diagram: an antiferromagnetic Mott insulator parent compound becomes, with doping, a metal. The ground state of the intermediate region, however, has thus far been more mysterious, especially in the underdoped region. In this part of the phase diagram, where stripe-ordered and pseudogap phases compete or cooperate with superconductivity, it was long uncertain whether a well-defined Fermi surface existed at all. In fact, most studies using angle-resolved photoemission spectroscopy (ARPES) have reported disconnected Fermi arcs,^{1,2} rather than closed pockets.³ In-plane optical measurements (E polarized parallel to the CuO₂ planes) have revealed a nodal metal state with behavior resembling a Fermi liquid.⁴ Further complicating the situation is a partial suppression of the density of states, or pseudogap, in the antinodal region of reciprocal space.⁵⁻⁷ The pseudogap is seen in ARPES measurements and also in the interplane (c-axis) transport⁸ since the interlayer dynamics are dominated by antinodal states.9

In the last several years breakthrough experiments^{10–13} have identified quantum oscillations in the transport and magnetization properties in underdoped YBa₂Cu₃O_y (Y123) and YBa₂Cu₄O₈ (Y124), as well as in the electron-doped material Nd_{2-x}Ce_xCuO₄,¹⁴ providing firm evidence of quasiparticles with closed Fermi-surface orbits. The data from these high-field experiments, along with probes of the Nernst and Hall effects,¹⁵ have been interpreted as a density wave-driven reconstruction of the Fermi surface from large hole sheets to small electron and hole pockets.^{16–20} Antinodal electron pockets are claimed to play a dominant role in the

transport probed along the CuO_2 planes. These observations may move us closer to understanding the underdoped region of the cuprate phase diagram, but they also open new questions regarding the nature of the reconstruction, the groundstate properties of underdoped cuprates in the absence of the magnetic field, and the mapping of the Fermi surface in *k* space.

Most theoretical attempts at explaining the magnetooscillation data postulate the formation of electron pockets in the antinodal regions (see Refs. 16-20, for example). Recently, however, some of us presented an argument against the notion of coherent antinodal states, at least in moderate fields.²¹ In particular, we presented the counter example of a stripe-ordered sample in which electronlike thermopower occurs in a region of temperature where ARPES shows a substantial antinodal gap but a gapless nodal arc.^{22,23} We showed that since the electronlike behavior cannot be explained in terms of normal-state carriers, some other explanation, such as superconducting fluctuations, is necessary. In this paper we further address this important issue by discussing the infrared magneto-optical properties of two relevant classes of materials: $La_{2-r}Sr_rCuO_4$ (LSCO) near x=1/8, and underdoped Y123. In the former, reconstruction of the Fermi surface is likely and stripelike magnetic order is known to be induced by weak magnetic fields;²⁴ the latter is the system in which the vast majority of quantum oscillation observations have been made. We critically consider whether the c-axis conductivity data are compatible with Fermi surface reconstruction scenarios involving small pockets of light quasiparticles in the antinodal direction.

The essential capability of infrared spectroscopy is the determination of the dynamic optical constants, including the optical conductivity $\hat{\sigma}(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$. This comprehen-

sive probe of the response of an electronic system yields an intuitive and quantitative description of its excitation spectrum. The presence of a Drude component (Lorentzian mode centered at zero frequency) in $\sigma_1(\omega)$ is a signature of coherent quasiparticles. Since the interplane conductivity is dominated by states in the antinodal region of k space, 9,25-28 any reconstruction of the Fermi surface which produces coherent antinodal quasiparticles is expected to introduce a Drude signature into the interlayer optical conductivity, visible as an upturn at low frequencies. Another expectation of the reconstruction is a modification of the pseudogap. A series of ear-lier in-plane^{4,29–31} and c-axis^{5–7,31–33} studies have outlined the temperature evolution of the infrared electrodynamics. At high temperatures cuprate superconductors exhibit incoherent transport, while at low temperatures they generally form a nodal metal state accompanied by an antinodal pseudogap. The optical signature of the pseudogap is a low-frequency reduction in $\sigma_{1,c}(\omega)$. If the magnetic field were to compete with the pseudogap it would lower the gap energy and/or fill in low-frequency spectral weight. As we will show below, we do not observe either modification of the pseudogap or appearance of a coherent Drude component. Both the real and imaginary parts of the optical conductivity attest to this conclusion.

Through considerations of the low-frequency optical response we are able to place limits on the spectral weight that can be attributed to hypothetical antinodal pockets of the reconstructed Fermi surface. In underdoped samples of LSCO at least 93% of the low-temperature superfluid density is suppressed by a magnetic field of 8 T. However, this spectral weight is transferred out of the low-frequency region rather than into a coherent free-carrier mode; no new mode is directly observed in our data. Similar limits are established for underdoped Y123 in magnetic field. We will also discuss these magneto-optical data in the context of the interlayer $La_{2-x-y}Sr_x(Nd, Eu)_yCuO_4$,³⁴ electrodynamics of and La_{2-r}Ba_rCuO₄. In these systems stripe order can account for the observed nodal effects but does not lead to the development of antinodal pockets of coherent quasiparticles and is therefore is in accord with the *c*-axis data.

II. EXPERIMENTAL DETAILS AND RESULTS

Interplane reflectance measurements in magnetic field were performed for two prototypical high- T_c cuprate families, Y123 and LSCO. Sample growth and transport characterization are described in Refs. 35 and 36 and details of the magneto-optical experiment can be found in Refs. 37–39. Infrared reflectance was measured over a wide frequency range (18–40 000 cm⁻¹) in magnetic fields H=0-8 T applied parallel to the *c* axis. In all measurements the electric field of the incident light was also polarized parallel to the *c* axis to isolate the interplane response.

A. Raw reflectance

Infrared reflectance $R(\omega)$ is plotted in Fig. 1 for underdoped crystals $La_{1.90}Sr_{0.10}CuO_4$ [LSCO 0.10, Fig. 1(a)], $La_{1.875}Sr_{0.125}CuO_4$ [LSCO 0.125, Fig. 1(b)], and



FIG. 1. (Color online) Infrared reflectance spectra for (a) $La_{1.90}Sr_{0.10}CuO_4$, (b) $La_{1.875}Sr_{0.125}CuO_4$, and (c) $YBa_2Cu_3O_{6.67}$ single crystals at $T=T_c$ and at T=8 K for magnetic fields up to 8 T. Inset to (b): experimental diagram. Oscillating electric field and static magnetic field are both aligned parallel to the *c* axis of the layered cuprate structure.

YBa₂Cu₃O_{6.67} [Y123 6.67, Fig. 1(c)]. The latter sample represents a hole doping value not far from those where quantum oscillations have been observed⁴⁰ in the transport properties of Y123 (y=6.67 corresponds to a hole doping value of approximately 0.12). All crystals exhibit "insulating" behavior at temperatures $T \gtrsim T_c$, with low, mostly flat $R(\omega)$. Thus raw $R(\omega)$ data are in stark conflict with the notion of a coherent quasiparticle contribution in the antinodal region in the normal state. We will further elaborate on this point when analyzing the complex conductivity plotted in Fig. 2. Below T_c the interplane coherence of the superconducting phase allows Cooper pairs to oscillate freely between layers, producing the sharp reflectance edge of the Josephson plasma resonance (JPR). As magnetic field H is applied perpendicular to the planes, phase coherence is diminished. The JPR is suppressed, broadening and moving to lower frequencies. For the LSCO crystals, the response returns to that of the normal state, suggesting that the Josephson coupling of CuO₂ planes has been arrested. For Y123, field magnitudes of 8 T cause the spectra at 8 K to resemble that of T =45 K and zero field.

B. Signatures of condensate formation and field-induced pairbreaking in the optical conductivity

As pointed out above, the most straightforward approach for detecting coherent charge dynamics is the direct observa-



FIG. 2. (Color online) Interlayer optical conductivity $\sigma_1(\omega, H)$ of (a) La_{1.90}Sr_{0.10}CuO₄, (b) La_{1.875}Sr_{0.125}CuO₄, and (c) YBa₂Cu₃O_{6.67} single crystals for several temperatures and magnetic fields $H \parallel c$ up to 8 T. [(d)–(f)] Low-frequency spectral weight Σ_{LF} , as determined from $\sigma_2(\omega)$.

tion of a Drude feature in the optical conductivity $\hat{\sigma}(\omega)$. Such a contribution takes the form

$$4\pi\hat{\sigma}(\omega) = \frac{\omega_p^2 \tau}{1 - i\omega\tau}.$$
 (1)

Here ω_p^2 is the oscillator strength and $1/\tau$ is the free-carrier scattering rate. With this objective in mind we plot in Figs. 2(a)–2(c) the real part of the conductivity $\sigma_1(\omega)$. These curves were calculated from the reflectance spectra in Fig. 1 via the Kramers-Kronig (KK) relations. At room temperature $\sigma_1(\omega)$ consists of several strong phonons superimposed upon a nearly frequency-independent electronic background. At temperatures below $T=T^* > T_c$ we observe the dramatic suppression of low-frequency conductivity due to the pseudogap (PG).^{5,7,31,33} The PG is only a partial gap, with finite residual conductivity remaining below the gap frequency. The spectral weight missing from the gap is transferred to higher energies, above the high-energy cutoff of the data.

Upon entering the superconducting state the LSCO crystals experience a monotonic decrease in conductivity at low frequencies as the spectral weight is transferred to the superconducting $\delta(\omega)$ peak at zero frequency. This diminished electronic background in the conductivity helps to reveal the sharpening of the plasmon in the reflectance, which is associated with the superfluid response. For Y123 the presence of the bilayer plasmon feature near 400 cm⁻¹, which is coupled

to several *c*-axis phonons, produces a more complicated spectrum.⁴¹⁻⁴³ However, a similar condensate formation is observed.

Empirically, the application of magnetic field $H \parallel c$ serves to undo the effect of lowering temperature, driving the conductivity toward the normal state spectra. The most important commonality in the magneto-optical response of the three representative data sets displayed in Figs. 1 and 2 is that no new, narrow, Drude-like absorption feature is observed. The conductivity of the LSCO 0.125 and Y123 6.67 crystals are flat at lowest frequencies. The LSCO 0.10 sample does exhibit an upturn of conductivity toward low frequencies, but it is clear in this case that we are witnessing a return of the conductivity to its normal state value and not the emergence of a new coherence peak indicative of new pockets in the Fermi surface. The reversion to the normal state response is especially clear in the raw reflectance data in Fig. 1.

C. Searching for a coherence mode in magneto-optics data: Spectral weight analysis

If an emergent coherent mode had narrow width relative to the low-frequency experimental cutoff (20 cm^{-1}), then its contribution would be difficult to resolve in $\sigma_1(\omega)$, even if the lower-frequency response exhibited a marked upturn; nevertheless, such a feature would leave a tell-tale signature in the imaginary part of the optical conductivity. For example, the contribution of the superconducting condensate to the real part of the conductivity, $\sigma_1^{SC}(\omega) = \frac{\rho_s}{8} \delta(\omega)$, is matched with an imaginary contribution of $\sigma_2^{SC}(\omega) = \frac{\rho_s}{4\pi\omega}$. This consequence of the KK relations is often exploited to measure the superfluid density ρ_s in superconducting materials.⁴⁴ A narrow, but finite-width, Lorentzian resonance appearing below the low- ω cutoff will also produce an (approximate) $1/\omega$ behavior in the imaginary part of the conductivity. In the raw reflectance data both types of coherent modes $\delta(\omega)$ and Lorentzian] yield a sharp plasma edge form of $R(\omega)$. Therefore $\sigma_2(\omega)$ spectra along with raw reflectance data allow us to comment on the possibility of a coherent mode in the *c*-axis response.

Since $\sigma_2(\omega)$ registers effects due to both Cooper pair and quasiparticle coherence, the coefficient of the $1/\omega$ contribution is the sum of the spectral weights of the superfluid and the coherent quasiparticle peak. We define this quantity more generally as $\Sigma_{LF} = \rho_s + \Sigma_{QP}$. As shown in Fig. 3 for LSCO 0.10, the low-frequency limit of $\pi \omega \sigma_2(\omega)$ indicates the value $\Sigma_{LF}(H)$. The values of $\Sigma_{LF}(H)$ are displayed for each sample in the right-hand column of Fig. 2. Consistent with the behavior of the JPR in the raw reflectance data, Σ_{LF} is completely suppressed (within detection limits of 7%) by 8 T in both LSCO crystals. This unambiguously shows that all spectral weight from the *c*-axis condensate is transferred out of the low-frequency part of the spectrum, leaving no room for the formation of a coherent quasiparticle peak.

For the Y123 crystal it is possible to place bounds on the expected infrared response based on the results of the magneto-oscillation experiments. The Drude linewidth is equal to the scattering rate $1/\tau$, which is dependent upon



FIG. 3. (Color online) Magnetic field dependence of low-frequency spectral weight Σ_{LF} as determined by $1/\omega$ contribution to σ_2 . Data for LSCO 0.10 sample were collected at T=8 K, with infrared electric field and static magnetic field both aligned parallel to the interplane *c* axis.

disorder and temperature. $1/\tau$ for Y123 may be deduced from the magneto-oscillation experiments by considering the fundamental condition for the realization of quantum oscillations: $1/\tau < \omega_c = \frac{eH}{cm^*}$, where ω_c is the cyclotron frequency and *e* and m^* are the electron charge and effective mass, respectively. The smallest field at which oscillations have been reported is 30 T; inputting the effective mass m^* =1.76 determined in the same study¹² yields an upper bound on $1/\tau$ of ω_c =15 cm⁻¹. As discussed above, no feature consistent with this estimate of scattering rate is observed in our data. It is clear that for scattering rates compatible with the cyclotron resonance conditions, the interlayer conductivity contains no infrared signature of coherent antinodal quasiparticles.

Considering the low-frequency spectral weight for Y123 6.67, it is shown in Fig. 2(f) that the application of an 8 T magnetic field reduces $\Sigma_{LF}(H)$ by 50%. Since independent data are not available for the c-axis superfluid density in magnetic field, we are unable to discern between the two possible contributions to $\Sigma_{IF}(H)$. Instead we may place limits upon the allowed quasiparticle weight $\Sigma_{OP} \leq \Sigma_{LF}$. If new pockets of coherent quasiparticles were being populated through pair-breaking processes, then the transfer of spectral weight from the superconducting condensate to the narrow Drude-like mode would occur at very low energies, and Σ_{LF} would have no magnetic field dependence. Instead, we observe in our data a transfer of spectral weight out of the low-energy region. The linear suppression of Σ_{LF} is in agreement with a fairly straightforward model involving the misalignment of pinned vortices.^{45,46} Since the rate of suppression is in fact slightly faster than predicted for reasonable material parameters for Y123, the transfer of spectral weight to a narrow Drude-like mode seems unlikely.

The lack of emerging coherent interplane response is corroborated by an insensitivity of the pseudogap to magnetic field. The *c*-axis response of the underdoped cuprates is incoherent in general in the normal state, but the pseudogap serves to further reduce coherence.⁴⁷ This is accomplished by eliminating states in the antinodal region that might otherwise be associated with quasiparticles. Therefore, any formation of interplane quasiparticle coherence would require diminishing of the pseudogap. However, our data exhibit no sign of pseudogap modification: low-frequency conductivity levels are not augmented by the field. Behavior of the pseudogap in very high fields remains an open question.^{48,49} In tunneling transport measurements, even pulsed magnetic fields of the order of those used in magneto-oscillation experiments do not suppress the pseudogap.⁵⁰

III. DISCUSSION

The above analysis has revealed common trends in the magneto-optical response of a collection of underdoped cuprate superconductors. In each case the effect of magnetic field is to reduce the *c*-axis superconducting phase coherence without introducing a coherence peak or modifying the signatures of the pseudogap. This behavior is best understood within the picture of stripelike order, a scenario that also has been hypothesized to explain the quantum oscillations data.^{39,51} Therefore it may be instructive to examine the *c*-axis electrodynamics of other stripe-ordered materials.

Important examples of density-wave states competing and coexisting with superconductivity are realized in $La_{2-r-\nu}Sr_r(Nd, Eu)_{\nu}CuO_4$ and $La_{1.875}Ba_{0.125}CuO_4$. Optical studies of rare earth-doped LSCO revealed dramatic suppression of the JPR frequency, signaling the decoupling of CuO₂ planes.³⁴ This interplane decoherence coincides with the onset of stripe order and a structural transition to a lowtemperature tetragonal configuration. These combined observations vield insight into the underlying physics: when lattice distortions provide a sufficient pinning potential for collective stripe pinning, stripe order is stabilized at the expense of interlayer phase coherence. As the doping corresponding to the strongest stripe order is approached, the JPR minimum is broadened by incoherent processes⁵² and eventually vanishes. The severely diminished oscillator strength of the c-axis plasmon places a strict limit on the available spectral weight for coherent transport.

A similar situation is encountered at the $x=\frac{1}{8}$ doping level of La_{2-x}Ba_xCuO₄, a material so dominated by stripe order that its bulk T_c is suppressed to <5 K. Optical measurements show the *c*-axis response of this system to be very insulating, with low-frequency conductivity levels below 1 Ω^{-1} cm⁻¹ and no Drude-like upturn or Josephson plasmon, again placing an upper bound on the size of a coherent peak.⁵³ Recent ARPES and transport experiments have yielded further insight into the k dependence of the quasiparticle dynamics; correlations between the opening of the *d*-wavelike gap at the Fermi surface and the abrupt cessation of negative thermopower indicate that the available quasiparticle states in that system are located in the near-nodal region and not at the antinodes.²¹⁻²³ Also, reports of insulating *c*-axis resistivity in high magnetic field⁵⁴ agree with the nonmetallic interplane response observed by optical techniques.53,55

The above examples are systems which possess stripe order, yet clearly do not exhibit coherent transport along the *c* axis. Even though Fermi surface reconstruction is likely to occur in these materials, it does not lead to the creation of quasiparticle pockets in the $(\pi, 0)$ region of the Fermi surface. Again, this conclusion stems from the fact that the *c*-axis transport is highly sensitive to states in the antinodal region. This behavior is consistent with our results for Y123 and LSCO: magnetic field-induced density wave order may be driving a Fermi surface reconstruction that does not involve the formation of antinodal pockets.

There are, of course, several discrepancies in experimental conditions between the optical measurements described here and the observations of magneto-oscillations. For example, the transport experiments were conducted in extreme regions of the temperature-magnetic field parameter space which are presently inaccessible to magneto-optical techniques: the magnetic field scale accessed in the optics data (8 T) is only a fraction of that for which oscillations were discovered (50 T). However, these differences should not be significant enough to completely obscure the signatures of quasiparticle coherence in the infrared conductivity. It is important to note that one common signature of samples exhibiting quantum oscillations, electronlike transport, is observable in Y123 6.67 at 33 K for fields greater than 6 $T.^{40}$ Therefore, the relevant vortex core state at H=8 T and T =8 K is likely the same as the state reached at much higher fields and lower temperatures, where QOs are detected. Also, the 8 T field produces dramatic modifications to the interplane response. For underdoped Y123 crystals the halving of the superfluid is accompanied by a modification of the condensate formation scheme over a wide energy scale, 37,38,56 as well as by the enhancement of static incommensurate magnetic order.57

An additional difference lies in the precise chemical composition of crystals studied: quantum oscillations have only been observed for extremely clean crystals with dopings near p=0.10 in YBa₂Cu₃O_y and at p=0.125 in YBa₂Cu₄O₈, and never in La_{2-x}Sr_xCuO₄. However, the doping of YBa₂Cu₃O_y presented here (p=0.12) lies in the doping interval bounded by the above results. And although LSCO crystals are generally too disordered to permit observation of magnetooscillations, the conditions for observing coherence are more easily satisfied in IR optics than in other techniques. For instance, IR measurements can easily detect a Drude response in dirty metals which would never exhibit quantum oscillations.

It should be noted that incoherent interlayer transport is not a generic feature of anisotropic, strongly correlated electronic systems. For example, the layered ruthenate Sr_2RuO_4 is anisotropic by approximately three orders of magnitude, yet displays coherence along all three crystal axes. This is evidenced by T^2 temperature dependence of both *ab* plane and *c*-axis resistivity at low temperatures,⁵⁸ as well as by the presence of a low-frequency plasma edge in the infrared reflectance.⁵⁹ These observations are indicative of Fermiliquid-like electron-electron scattering, and Drude-like contribution to the conductivity, respectively, in stark contrast with the incoherent behavior of the cuprates discussed here.

In conclusion, we have demonstrated that optical conductivity data in magnetic field are not consistent with the existence of coherent antinodal quasiparticles. Expected consequences of this type of Fermi surface reconstruction, such as the suppression of the pseudogap and emergence of a Drudelike free-carrier response, are not observed in infrared measurements. The limits placed on the possible reconstructed Fermi pockets should be useful for guiding future theoretical efforts, especially in conjunction with ARPES and transport results.

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