Ong construction for the reconstructed Fermi surface of underdoped cuprates

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Using the Ong construction for a two-dimensional metal, we show that the sign change in the Hall coefficient R_H of underdoped hole-doped cuprates at low temperature is consistent with the emergence of biaxial charge order recently proposed to explain the observation of low-frequency quantum oscillations. The sharp evolution of R_H with temperature, however, can only be reconciled by incorporating a highly anisotropic quasiparticle scattering rate. The magnitude and form of the scattering rate extracted from the fitting imply that those quasiparticles at the vertices of the reconstructed pocket(s) approach the boundary of incoherence at the onset of charge order.

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Understanding the nature of the pseudogap phase in underdoped (UD) cuprates is regarded as one of the key steps in unravelling the origin of high-temperature superconductivity. Prior to 2007, the prevailing view of the electronic structure of UD hole-doped cuprates was one of Fermi arcs: disconnected regions of coherent quasiparticles located near the intersection of the underlying full Fermi surface and the zone diagonals. These arcs evolve out of an incoherent high-temperature phase via the formation of the pseudogap—an anisotropic depression in the density of states [1] or spectral weight [2].

The 2007 discovery of quantum oscillations (QO) in UD $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y123) [3,4] and $\text{YBa}_2\text{Cu}_4\text{O}_8$ (Y124) [5,6] raised a fundamental question about the nature of the electronic ground state in UD cuprates, namely how could the prevailing view of Fermi arcs be reconciled with the existence of pockets? The observation of a sign change in R_H , the thermopower S , and the Nernst coefficient ν in Y123 [7–9], Y124 [7], and $\text{HgBa}_2\text{CuO}_4$ (Hg1201) [10] at low temperatures hinted that these pockets must result from Fermi surface reconstruction (FSR) induced by some form of (short-range) charge ordering that might itself be enhanced by the application of a high magnetic field, i.e., by the suppression of superconductivity. Biaxial charge order with wave vectors at $(0, +\delta)$ and $(+\delta, 0)$ (where δ is a fraction of the Brillouin zone coordinates) had already been apparent, in other cuprate families at least, in Fourier-transform scanning tunneling spectroscopy (FT-STs) [11,12] and later in resonant inelastic x-ray scattering (RIXS) [13,14]. Its observation in Y123 [15–17] ultimately connected all these phenomena together and suggested that the electron pocket inferred from QO and transport experiments originated from a reconstruction of the pseudogapped FS via wave-vector displacements linked to the extremities of the remnant Fermi arcs [18,19]. The relation between the interaction driving the FSR and that responsible for the formation of the pseudogap itself, however, has not yet been established.

In addition to this central question, there are other important details which still remain to be resolved, such as the actual number of pockets that appear following FSR and whether the FSR itself is a purely field-induced phenomenon or can also be present in zero field. In order to address such questions, it is instructive to compute the transport and thermodynamic properties for the different scenarios and compare with experimental data. One interesting feature of the “nodal” electron pocket inferred from the biaxial charge order picture

is its negative curvature, which endows it with both electron- and holelike character. According to Ong [20], negative FS curvature in a quasi-two-dimensional metal leads to different contributions to the Hall conductivity σ_{xy} of opposite sign as the mean free path l is integrated over the FS. Harrison and Sebastian (HS) claimed recently that such a diamond-shaped pocket, together with an isotropic transport lifetime τ , can account for a number of key features of the (field-induced) normal state of UD cuprates, including the sign reversal in R_H and QO appearing in the Hall conductivity [21]. HS evaluated $R_H(T, B)$ using both the Ong construction and the Shockley-Chambers tube integral form of the Boltzmann equation. With the latter, the sign of R_H is determined by the product of the orthogonal components of the Fermi velocity $v_x(t)v_y(t + dt')$ which in turn is determined by the FS curvature and $\omega_c\tau$, the product of the cyclotron frequency and the quasiparticle lifetime. With the former, HS introduced anisotropy in v_F (through the opening of the hybridization gap Δ), v_F being smaller at the vertices of the pocket than along the original arcs. It is this anisotropy in v_F , coupled with the negative curvature, that induces a sign change in R_H . However, since the anisotropy in v_F does not vary with temperature and τ is isotropic, R_H as calculated using the Ong construction has no implicit T dependence. Within the HS model, therefore, T dependence in R_H can only arise from changes in $\omega_c\tau$, implying that the sign reversal itself is a purely field-induced effect.

Significantly, in Y123 at a hole doping $p = 0.12$, R_H becomes negative at a field-independent temperature $T_0 = 70$ K, i.e., above the zero-field superconducting transition temperature $T_c = 65$ K—see Fig. 5 of Ref. [22]. This implies that a large magnetic field is not necessary to induce the sign reversal in $R_H(T)$ and in turn, that the HS model is not generally applicable to the UD Y-based cuprates. As the Ong formalism is in essence a low-field construct, here we adopt the Ong construction to calculate $R_H(T)$ first for a single electron pocket, taking as our starting point the FSR stemming from this proposed biaxial charge order [18,19], then to explore the case of one electron and two smaller hole pockets, as advocated in Ref. [19] and suggested by recent experiments [23]. We show that if the sign change in $R_H(T)$ can be induced in arbitrarily low fields, then for a single electron pocket, τ itself must be anisotropic. Otherwise one needs to invoke the existence of additional hole pocket(s), in contrast to the conclusions drawn by HS [21]. Even with multiple pockets, however, the fitting

is much more satisfactory with an anisotropic τ . An intriguing relation between the strength of the scattering and the FSR is then revealed.

According to the Ong construction [20],

$$R_H = 2\pi^2 c A_\ell / e (\bar{I} S)^2, \quad (1)$$

where c is the c -axis lattice spacing, $A_\ell = \frac{\mathbf{H}}{H} \cdot \int d\mathbf{l}(k) \times \mathbf{l}(k)$ is the so-called Stokes area swept out by the mean-free-path vector $\mathbf{l}(k) [= \mathbf{v}(k)\tau(k)]$ as k moves around the FS, and S is the FS perimeter. The local curvature of the FS gives rise to different “circulation” of the $\mathbf{l}$ vector and thus to contributions to σ_{xy} of opposite sign. To simplify our parametrization and to make comparison with the HS model more explicit, we use the FS area inferred from QO experiments for the electron pocket (for $p = 0.12$) and assume the same form for v_F introduced by HS [24]. The curvature of the pocket is set by the unreconstructed (bonding) FS derived from band structure calculations for ortho-II Y123 [25]. (Similar results were obtained for the antibonding FS, as described in the Supplemental Material [26].) Panel (a) of Fig. 1 shows the resultant nodal pocket superimposed on the unreconstructed FS [25]. As in Ref. [21], α is the angle subtended by the extrema of the pocket to the corner (e.g., X point) of the Brillouin zone [27]. However, here we constrain α and the geometry of the electron pocket using band structure and the magnitude of the QO frequency.

The only unknown in our model is the scattering rate $\Gamma(T, \phi)$. At low T , the in-plane resistivity $\rho_{ab}(T)$ of UD Y123 and Y124 is found to obey a T^2 dependence [22,28,29], characteristic of dominant electron-electron scattering. We therefore introduce a scattering rate of the form:

$$\Gamma(T, \phi) = \Gamma_0(\phi) + \Gamma_2 \cos^2(2\phi) T^2, \quad (2)$$

where Γ_2 is the strength of the anisotropic T^2 term, and ϕ is the angle inside the electron pocket defined with respect to the O-Cu-O bond direction [30]. It is this component that generates the strong T dependence (and sign change) in R_H . Γ_0 , the impurity scattering rate, may also have k dependence, e.g., due to a predominance of small-angle scattering off impurities located away from the CuO₂ planes [31], that can also influence

$R_H(T)$ [32]. Here, however, we invoke the isotropic-I approximation and assume that $\Gamma_0(\phi)$ has the same angle dependence as v_F , so that the product v_F/Γ_0 is independent of ϕ .

As T is raised, it is unphysical for $\Gamma(T)$ to vary quadratically with temperature without bound. Indeed, it is well known that in UD cuprates, $\rho_{ab}(T)$ becomes T linear above a characteristic temperature T^* related to the magnitude of the pseudogap [33]. One simple and well-explored procedure to account for this deviation from a pure T^2 dependence at intermediate temperatures is to introduce a maximum (saturation) scattering rate $\Gamma_{\max}$ in parallel with $\Gamma(T)$ [34]. The magnitude of $\Gamma_{\max}$ is set to v_F/a —commonly referred to as the Mott-Ioffe-Regel (MIR) limit—corresponding to a minimum mean-free-path $\mathbf{l} \sim a$, the (in-plane) lattice constant. The introduction of $\Gamma_{\max}$ also ensures that the effective scattering rate becomes isotropic at elevated temperatures, as expected.

In panels (b) and (c) of Fig. 1, simulations of $R_H(T)$ and $\rho_{ab}(T)$ are plotted for the electron pocket depicted in panel (a). The values of Γ_0 and Γ_2 for each curve are given in the figure caption. The red horizontal dashed line in panel (b) is obtained for a purely isotropic T^2 scattering rate. As discussed in the introduction, within the Ong formalism, $R_H(T)$ for such a pocket can only have intrinsic T dependence if Γ has an anisotropy that changes with increasing T , as illustrated by the red solid curve in panel (b). In that simulation, $R_H(T)$ changes sign around 65 K, i.e., at or around T_c , consistent with the behavior observed for $p = 0.12$ [7]. This contrasts with the conclusions of HS who argued that the sign change can occur even for isotropic scattering. We stress that this is only possible if the change of sign is a high field phenomenon.

The anisotropy required to induce a sign change in R_H (for the given FS configuration) is large, as indicated in Fig. 2 where the variation of $\mathbf{l}(\gamma)$ (the so-called $\mathbf{l}$ curve) is plotted for the two different regimes (low- T , R_H negative and high- T , R_H positive). Here, γ is the angle subtended by the vector normal to the Fermi surface (i.e., by v_F) relative to the k_x axis. In each plot, the two closed loops of contrasting circulation give rise to contributions to σ_{xy} and R_H that are of opposite sign. The ratio of the respective areas of the two loops ultimately determines the sign of the Hall coefficient [20]. Note that the

FIG. 1. (Color online) (a) Reconstructed diamond pocket (red) formed from translation of the remnant arc of the original FS of ortho-II Y123 at a hole doping $p = 0.12$ (black). T dependence of the Hall coefficient (b) and resistivity (c) calculated for the single pocket assuming anisotropic (solid lines) and isotropic (dashed line) scattering. [Note that in (c), the two lines are indistinguishable on this scale of plot]. For these simulations, $\Gamma_0 = 6.5$ meV and $\Gamma_2 = 0.052$ meV/K² [see Eq. (2)].

FIG. 2. (Color online) I curve for the nodal pocket shown in Fig. 1 at two representative temperatures where ρ_{xy} has opposite sign. The tangential arrows indicate the circulation of each loop, and the $-/+$ signs indicate the corresponding sign of ρ_{xy} . The resultant ρ_{xy} is determined by the difference in the areas of the two counter-rotating loops [20]. For clarity, the values of I_x and I_y at each T have been scaled by the average $|I|$ ($= 17.0$ nm and 7.0 nm at $T = 25$ K and 125 K, respectively).

inner loop (which determines the magnitude of the negative component in R_H) is almost circular. This is because the region with negative curvature is confined to a very small angular (ϕ) window around the vertex where $|I|$ is essentially constant.

For completeness, we introduce in Fig. 3 a second scenario in which the electron pocket coexists with a pair of smaller hole pockets [panel (a)]. This picture is motivated by theoretical modeling of bidirectional charge order in UD cuprates [18] and the recent observation of a second frequency in QO experiments on UD Y123 [23]. In contrast to the electron pocket, the hole pockets have no region of negative curvature. In order to model the transport data, we use the QO frequency reported in Ref. [23] to define the area of the hole pockets and assume

that v_F in the smaller hole pockets is isotropic with a value one half that of the electron pocket. Thus, $\Gamma(T)$ is once again the only unknown. To simplify further, we assume the same form of scattering rate for both the electron and hole pockets.

Panels (b) and (c) of Fig. 3 show simulations of $R_H(T)$ and $\rho_{ab}(T)$ for the multipocket FSR scenario depicted in panel (a). The solid blue line in panel (b) is obtained assuming anisotropic scattering of the form shown in Eq. (2). The set of parameters for each curve is again listed in the figure caption. The overall temperature dependence is very similar to that obtained in Fig. 1 for the single electron pocket. The blue dashed line is the $R_H(T)$ curve obtained for a purely isotropic T^2 scattering rate [i.e., $\Gamma(T) = \Gamma_0 + \Gamma_2 T^2$] using the standard two-carrier formalism. Inspection of panel (b) in Figs. 1 and 3 illustrates that the only way to induce a sign change with purely isotropic scattering is to invoke the existence of additional hole pocket(s), in contrast to the conclusions drawn by HS [21]. In the case of isotropic scattering, however, the T dependence of $R_H(T)$ is much more gradual in the simulation than is seen in the experimental data [7]. Indeed, in order to drive $R_H(T)$ positive with increasing temperature, one needs to introduce a significantly larger (T^2) scattering rate on the electron pocket, but since the area (carrier density) of this pocket is three times that of the two hole pockets, it is the dominant contribution to the electrical resistivity, and thus its overall strength must be constrained in order to be consistent with the slope of $\rho_{ab}(T)$.

Momentum dependence of the scattering rate has long been recognized as a necessary ingredient in the transport properties and spectroscopic response of both underdoped [35,36] and overdoped [37,38] cuprates. In overdoped cuprates, the total scattering rate consists of two T -dependent terms, one that is linear-in- T with a d -wave angle dependence and one almost isotropic term that varies as T^2 [37,39]. In the UD regime, the pseudogap develops preferentially at the zone boundaries, thereby gapping out those FS sections where the T -linear term is dominant, leaving Fermi arcs in which only the T^2 scattering term prevails at sufficiently low doping and temperatures. The additional characteristic uncovered here is that below T^* , additional and strong anisotropy also develops in the T^2 channel due to the emergent charge-ordering instability.

FIG. 3. (Color online) (a) FSR scenario with one electron and two hole pockets for ortho-II Y123 at $p = 0.12$ (black). The area of the hole pockets have been scaled to match that deduced from QO data [23]. (b) $R_H(T)$ and (c) $\rho_{ab}(T)$ simulations for the combined pockets. The solid lines are for $\Gamma_0 = 6.5$ meV and $\Gamma_2 = 0.052$ meV/K² for the electron pockets and $\Gamma_0 = 13.0$ meV and $\Gamma_2 = 0.052$ meV/K² for the two hole pockets [see Eq. (1)]. The dashed lines represent simulations based on the two-carrier model with isotropic scattering.

In both the single and multiple pocket scenarios, the strength of the anisotropic scattering term required to induce the sign change around $T_0 \sim 70$ K turns out to be the same. From the magnitudes of Γ_2 and α [which sets the range of ϕ over which Eq. (2) is valid], we see that $\Gamma[T, \phi = (\pi - 2\alpha)/4]$ exceeds $\Gamma_{\max}$ at a temperature around 120 K, i.e., at $T \sim T_{CO}$. This implies that the quasiparticles at the vertices of the arcs (above T_{CO}) are on the boundary of coherence at the onset of charge ordering. According to Hartnoll [40], there are two universal bounds on the transport processes that occur in strongly correlated metals. One is the MIR bound described above, beyond which the quasiparticles are destroyed—resulting in a loss of (optical) spectral weight at low frequencies [41]. The other is a diffusivity bound (also known as the “Planckian” bound [42]) beyond which transport is driven by diffusion of charge and energy rather than by momentum relaxation. The bound itself is characterized by a rate of dissipation $\hbar/\tau = \eta k_B T$, where η is a numerical factor. In cuprates, this bound (with $\eta = 2\pi$) has been argued to be responsible for the loss of quasiparticle coherence at the Brillouin zone boundary (i.e., where the pseudogap first opens) [39].

It would appear then that both bounds are at play in UD cuprates, though a key ingredient missing from current models is the k -space anisotropy that creates a coexistence of coherent and incoherent states whose ratio evolves with temperature. Above T_{CO} , where coherent quasiparticles reside only along the Fermi arcs, R_H is positive due to the curvature of the original FS. States located at the tips of the arcs, however, are on the boundary of coherence, lying closest in momentum to those (pseudogapped) states with zero or diminished spectral weight. Since the ordering vector connects these tips, one also expects scattering off (charge) fluctuations there to be most intense. For $T < T_{CO}$, the mean free path of these quasiparticles, while now satisfying the MIR criterion, is still shorter than the charge order correlation length. Thus, despite the short-range nature of the correlations, the electronic structure begins to undergo zone folding. However, since the vertices of the emergent pockets are where scattering is most intense, $R_H(T)$ remains positive. As T is reduced further, Γ becomes increasingly isotropic, and the more pronounced “curvature” of the pocket near the vertices ensures that the electronlike contribution to $R_H(T)$ eventually wins out.

At this stage, it is difficult to ascertain which of the scenarios (single versus multiple pockets) is the correct description of the reconstructed state in UD Y123, though recent reports of additional frequencies in QO studies [23] or of the small A coefficient in $\rho_{ab}(T)$ [29] suggest the latter. Nevertheless,

the proposed FSR driven by bidirectional charge order does appear to produce a FS of the appropriate size and geometry. In addition to Y123, CDW modulations have also been reported for Hg1201 [43], $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$ (Bi2201) [13] and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) [44], suggesting that they are universal to hole-doped cuprates. Only in UD Hg1201, however, does $R_H(T)$ become negative at low T and high B [10]. In Eu-doped LSCO, both spin and charge (stripe) order is stabilized (in zero field), and R_H does again change sign at low T [45]. Clearly, whether charge correlations within the CuO_2 planes are short- or long-range depends very sensitively on the orbital configuration of the spacer layer. The mobility of the charge carriers appear to be less of a determining factor since it is well known that in all members of the LSCO family, the mobility of doped holes is much lower than in Y123 and Hg1201. At low T , where quasiparticle scattering is dominated by impurities, one expects the scattering rate to have only weak anisotropy and thus the sign of R_H to be determined by the topology of any reconstructed pockets. Thus the absence of a sign change in $R_H(T)$ in Bi2201 and LSCO suggests strongly that there the electron pockets never form. The reason for this intriguing difference in behavior is not yet known.

In summary, we have explored the T dependence of the in-plane Hall coefficient in UD Y123 using Boltzmann transport analysis applied to the FS geometry induced by bidirectional charge order. The overall T dependence is found to be in good agreement with the experimental data. Our analysis has also uncovered two important and previously overlooked characteristics of the quasiparticle states in the reconstructed phase of clean underdoped cuprates. Firstly, the sign change in $R_H(T)$ can only be reconciled with the existence of a single electron pocket provided that the electron-electron scattering rate is highly anisotropic, in contrast to the conclusions of Ref. [21]. Otherwise, it is necessary to invoke the presence of a second set of hole pockets, as inferred from recent transport studies. Secondly, we find that the quasiparticle states at the vertices of the reconstructed pocket(s) are on the boundary of coherence (i.e., their scattering rate is maximal as permitted by the Mott-Ioffe-Regel limit) at the charge-ordering temperature. It would be extremely informative to look for direct evidence of both these intriguing properties, e.g., in future angle-resolved photoemission experiments.

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