

Quantum oscillations in magnetic-field-induced antiferromagnetic phase of underdoped cuprates: Application to ortho-II $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$

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Abstract – The magnetic-field-induced antiferromagnetic phase of the underdoped cuprates is studied within the $t-t'-J$ model. A magnetic field suppresses the pairing amplitude, which, in turn, may induce antiferromagnetism. We apply our theory to interpret the recently reported quantum oscillations in high magnetic field in ortho-II $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ [1] and propose that the total hole density abstracted from the oscillation period is reduced by 50% due to doubling of the unit cell.

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Recently, Doiron-Leyraud *et al.* reported quantum oscillations in the clean underdoped cuprate ortho-II $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ in a high magnetic field at low temperatures [1] which imply the presence of small Fermi surface (FS) pockets. Their finding has shed new light on the nature of the anomalous spin gap phase of underdoped cuprates, a key to understanding the mechanism of high- T_c superconductivity. The existence and topology of a FS [2,3] is one of the most important issues for underdoped cuprates. Assuming the normal state is paramagnetic as in zero field, angle-resolved photoemission spectra on other underdoped cuprates [4] imply a FS consisting of four hole pockets centered on the Brillouin zone (BZ) diagonals.

In this letter we propose that the reported oscillations occur in an antiferromagnetic (AF) ordered state, hence the oscillation period corresponds to a Fermi pocket in the extended BZ. Our proposal is substantiated by examining the interplay between AF and SC in a $t-t'-J$ model using a renormalized mean-field theory (RMFT) [5]. A high magnetic field suppresses the pairing amplitude, which may in turn induce AF ordering. The FS property is studied using a phenomenological Green's function approach. The measured oscillation period in the presence

of AF ordering implies a total hole concentration of 0.075 instead of 0.15 as interpreted by Doiron-Leyraud *et al.* [1]. The latter substantially exceeds the value of 0.10 obtained from an empirical relation for $T_c = 59$ K of this compound [6] and a theoretical estimate of 0.04 based on band structure calculations [7]. The well-ordered nature of this compound should enhance T_c , and the empirical formula should overestimate, not underestimate, the hole concentration. We believe that our proposal gives a more reasonable hole density for this compound.

The interplay of AF and SC order in underdoped cuprates has been a central issue [8–12]. Recent nuclear magnetic-resonance data on multilayer cuprates have established that uniform AF order can coexist with SC order in the same CuO_2 plane even in the absence of a magnetic field and have led Mukuda *et al.* to propose a revised phase diagram of the cuprates with coexisting AF and SC order in the underdoped region [13]. Generally in cuprates AF and SC orderings are separated by a spin glass region presumably due to their crystalline disorder. Coexistence of uniform AF and SC order has been predicted from variational Monte Carlo (VMC) calculations on Gutzwiller projected wave functions [10,14]. We note that in the case of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ neutron

scattering data in an applied magnetic field revealed subgaps in spin excitations [15], which were interpreted as evidence for a field-induced AF ordering in the SC state [16]. Our microscopic theory is consistent with their phenomenological analyses [16].

We consider the $t - t' - J$ model on a square lattice [17,18],

$$H = - \sum_{i,j,\sigma} t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}) + J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

The hopping integrals are $t_{ij} = t$ for the nearest-neighbor (n.n.) pairs and $t_{ij} = t' = -t/3$ for the next-n.n. pairs. \mathbf{S}_i is a spin-(1/2) operator, and the spin exchange term ($J = t/3$) connects n.n. pairs. The constraint of no double occupation at any lattice site is implied. We use a variational wave function [9,10] to study the interplay of AF (or spin density wave (SDW)) and SC ordering,

$$|\Psi\rangle = P_D |\Psi_0\rangle, \quad |\Psi_0\rangle = \prod_{\mathbf{k}, s=\pm} (u_{\mathbf{k}s} + v_{\mathbf{k}s} d_{\mathbf{k}s\uparrow}^\dagger d_{-\mathbf{k}s\downarrow}^\dagger) |0\rangle, \quad (2)$$

where the product of \mathbf{k} runs over the reduced BZ, and $P_D = \prod_i (1 - n_{i\uparrow} n_{i\downarrow})$ is the Gutzwiller projection operator. $d_{\mathbf{k}s\sigma}$ is an annihilation operator of the SDW quasiparticles, with $s = \pm$ for the upper or lower SDW bands,

$$\begin{aligned} d_{\mathbf{k}+, \sigma} &= \cos(\phi_{\mathbf{k}}/2) c_{\mathbf{k}\sigma} - \sigma \sin(\phi_{\mathbf{k}}/2) c_{\mathbf{k}+\mathbf{Q}\sigma}, \\ d_{\mathbf{k}-, \sigma} &= \sin(\phi_{\mathbf{k}}/2) c_{\mathbf{k}\sigma} + \sigma \cos(\phi_{\mathbf{k}}/2) c_{\mathbf{k}+\mathbf{Q}\sigma}, \end{aligned} \quad (3)$$

where $\mathbf{Q} = (\pi, \pi)$ is the AF wave vector.

To carry out the variation procedure we apply the Gutzwiller approximation to replace the effect of the projection operator by a set of renormalization factors, which are determined by statistical counting [5,19] and later improvement [11,20]. This approximation or the renormalized mean-field theory [5] incorporates the resonating valence bond physics proposed by Anderson [17] and compares well with the VMC results [19]. Furthermore, as has been emphasized recently by Anderson *et al.*, the RMFT describes many key features of the phase diagram [19]. Let $\langle \hat{O} \rangle$ be the expectation value of the operator \hat{O} in the projected state $|\Psi\rangle$, and $\langle \hat{O} \rangle_0$ be that in the corresponding unprojected state $|\Psi_0\rangle$, then

$$\langle c_{i\sigma}^\dagger c_{j\sigma} \rangle = g_{t,\sigma}^{ij} \langle c_{i\sigma}^\dagger c_{j\sigma} \rangle_0, \quad \langle S_i^x S_j^x \rangle = g_s^x \langle S_i^x S_j^x \rangle_0, \quad (4)$$

where $\tau = x, y, z$, and g 's are the renormalization factors. In the AF state, electron populations in the two sublattices A (with net spin up) and B (with net spin down) are different. For the n.n. pair (ij) , $g_t = 2\delta(1-\delta)/(1-\delta^2+4m_0^2)$. For the next-n.n. pairs on sublattice A or B , $g_{t'\sigma}^A = g_{t'\sigma}^B = g_t(1+\delta+2\sigma m_0)/(1+\delta-2\sigma m_0)$. In the above expressions, δ is the hole density, $m_0 = \langle S_i^z (-1)^i \rangle_0$ is the staggered magnetic moment in units of μ_B in the state $|\Psi_0\rangle$. To address the interplay between SC and AF ordering, we use an improved Gutzwiller approximation of Ogata and Himeda for the spin exchange renormalization

factors, which takes the effect of intersite correlations into account [11]. In their formalism, g_s^τ are also functions of m_0 and the pairing amplitude Δ in eq. (6), and are given by $g_s^{x(y)} = g_t^2/\delta^2(1+\eta_1)^{-7}$, and $g_s^z = g_s^x(1+\eta_2)$ with η_1, η_2 both small positive numbers, given in¹. The difference between g_s^z and $g_s^{x(y)}$ is due to the asymmetry in spin space.

Within the Gutzwiller approximation, varying a projected state for H in eq. (1) reduces to varying an unprojected state $|\Psi_0\rangle$ for a renormalized H_0 ,

$$H_0 = - \sum_{i,j,\sigma} g_t^{ij} t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}) + J \sum_{\langle i,j \rangle, \tau} g_s^\tau S_i^\tau S_j^\tau. \quad (5)$$

We introduce two mean-fields, $\chi_x = \chi_y = \chi$ for hopping and $\Delta_x = -\Delta_y = \Delta$ for d -wave pairing,

$$\chi = \sum_{\sigma} \langle c_{i+x\sigma}^\dagger c_{i\sigma} \rangle_0, \quad \Delta_x = \langle c_{i+x\uparrow} c_{i\downarrow} - c_{i+x\downarrow} c_{i\uparrow} \rangle_0. \quad (6)$$

The singlet SC order parameter $\Delta_{\text{SC}} \approx \bar{g}_t \Delta$, with $\bar{g}_t = (g_{t'\uparrow}^A + g_{t'\uparrow}^B)/2$. The pairing amplitude and the SDW state described below define $|\Psi_0\rangle$. We choose a standard SDW form,

$$\cos(\phi_{\mathbf{k}}) = \epsilon_{\mathbf{k}}/\zeta_{\mathbf{k}}, \quad \zeta_{\mathbf{k}} = \sqrt{\epsilon_{\mathbf{k}}^2 + \tilde{\Delta}_{m,\mathbf{k}}^2}, \quad (7)$$

where $\epsilon_{\mathbf{k}} = -(2tg_t + 3J\bar{g}_s\chi/4)\gamma_+(\mathbf{k})$ is the kinetic energy contribution of n.n. hopping including a self-energy term of χ , with $\bar{g}_s = (g_s^z + 2g_s^{xy})/3$, and $\tilde{\Delta}_{m,\mathbf{k}} = m_v + 2t'(g_{t'\uparrow}^A - g_{t'\uparrow}^B)\theta_{\mathbf{k}}$. m_v is a variational parameter which determines m_0 , $\gamma_{\pm}(\mathbf{k}) = \cos k_x \pm \cos k_y$, and $\theta_{\mathbf{k}} = \cos k_x \cos k_y$. The d -wave pairing variational functions are chosen as

$$\begin{aligned} v_{\mathbf{k}\pm}^2 &= (1 - \xi_{\mathbf{k}\pm}/E_{\mathbf{k}\pm})/2, \\ u_{\mathbf{k}\pm} v_{\mathbf{k}\pm} &= \Delta_v(\mathbf{k})/2E_{\mathbf{k}\pm}, \\ \xi_{\mathbf{k}\pm} &= \pm\zeta_{\mathbf{k}} - 4t'\bar{g}_t\theta_{\mathbf{k}} - \mu, \\ \Delta_v(\mathbf{k}) &= (3/4)J\bar{g}_s\Delta_{v0}\gamma_-(\mathbf{k}), \end{aligned} \quad (8)$$

with $E_{\mathbf{k}\pm} = \sqrt{\xi_{\mathbf{k}\pm}^2 + \Delta_v(\mathbf{k})^2}$, and Δ_{v0} is a variational parameter. Note that because of the mean-field dependence of the g -factors here, $\Delta_{v0} \neq \Delta$ in general.

The variational energy per site $E = \langle H \rangle/N_s$ can then be expressed in terms of the mean-field parameters,

$$\begin{aligned} E &= -4g_t t \chi - 8t'\bar{g}_t \chi_1 + 4t'(g_{t'\uparrow}^A - g_{t'\uparrow}^B) \chi_2 \\ &\quad - (3\bar{g}_s/4)J(\Delta^2 + \chi^2) - 2g_z J m_0^2, \end{aligned} \quad (9)$$

where $\chi_1 = -(1/2N_s) \sum_{\mathbf{k},s} \xi_{\mathbf{k}s} \theta_{\mathbf{k}}/E_{\mathbf{k}s}$, $\chi_2 = -(1/2N_s) \times \sum_{\mathbf{k},s} s \tilde{\Delta}_{m,\mathbf{k}} \xi_{\mathbf{k}s} \theta_{\mathbf{k}}/\zeta_{\mathbf{k}} E_{\mathbf{k}s}$. The variational ground state is obtained by minimizing E with respect to m_v , Δ_{v0} , χ , μ ,

¹The small parameters η_1 and η_2 in $g_s^{x(y)}$ and g_s^z are given by $\eta_1 = 2X/(1-\delta^2+4m_0^2)^2$, $\eta_2 = 8m_0^2(2X_1+X_1^2)/(8m_0^2+X_2)$, $X = \delta^2(\Delta^2 - \chi^2) + 4m_0^2 X_2 + X_2^2/4$, $X_2 = \chi^2 + \Delta^2$, and $X_1 = 3X_2 \times (1-\delta)^2/[(1-\delta^2+4m_0^2)(1+\eta_1)^3]$.

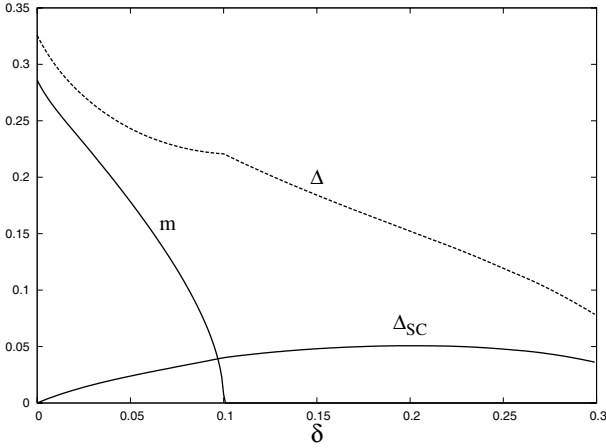


Fig. 1: Pairing mean-field Δ (dashed line), superconducting order parameter Δ_{SC} and antiferromagnetic staggered magnetization m (solid lines) as functions of hole concentration δ for the $t - t' - J$ model, with $J = t/3$, $t'/t = -1/3$.

under constraint of the constant total hole concentration δ . In fig. 1, we plot the pairing mean field Δ , the SC order parameter Δ_{SC} , and the staggered magnetization $m = \sqrt{g_s^z} m_0$ in the projected state $|\Psi\rangle$, as functions of doping δ . At $\delta < \delta_c = 0.1$, AF order coexists with SC order; at $\delta > \delta_c$, the ground state has only SC order with $m = 0$. This result is similar to the variational Monte Carlo calculation which gives $\delta_c \simeq 0.10$ for the $t - J$ model [14].

We now consider the effect of an external magnetic field on the phase diagram. Cuprates are type-II superconductors, and a magnetic field penetrates in the form of vortices. The SC order outside the vortex cores is substantially suppressed if the applied field H_{app} is comparable with the critical field H_{c2} as in the experiments in ref. [1], but quantum oscillations may still occur [21].

We use the RMFT to show that the suppression of the SC pairing amplitude due to the magnetic field may induce AF ordering. Consider the case $\delta \geq \delta_c$, so that the ground state at $H_{\text{app}} = 0$ is a pure SC state with the values in eq. (6) Δ_0 and $m_0 = 0$. We approximate the effect of H_{app} on the background SC region by a suppression of the pairing amplitude from Δ_0 to an average value $\bar{\Delta} = \alpha \Delta_0$, with $\bar{\Delta}$ estimated below. Then eq. (9) allows us to carry out the variation for a given Δ or equivalently for a fixed value of the variational parameter Δ_{v0} . We have found that at $\delta > \delta_c$, a suppression of Δ_{SC} and hence Δ will induce AF ordering $m \neq 0$. In fig. 2 we plot m as a function of α for $\delta = 1.2\delta_c = 0.12$. $\bar{\Delta}$ may be estimated as follows. Let ξ_{coh} be the SC coherence length, and d the average distance of two neighboring vortices. We have $d/\xi_{\text{coh}} \approx \sqrt{H_{\text{app}}/H_{c2}}$, and $\bar{\Delta} = \pi^{-1}(d/2)^{-2} \int_{r \leq d/2} \Delta(\mathbf{r}) d\mathbf{r}$, where $\Delta(\mathbf{r})$ is the shape of the pairing mean field around a vortex. For a rough estimate, we use for the s -wave the form $\Delta(\mathbf{r}) = \Delta_0 \tanh(r/\xi)$ [22]. The value of H_{c2} of underdoped high- T_c superconductors is about 160 T [23], so the suppression ratio α at $H_{\text{app}} = 10 \text{ T}$ ($\sim H_{c2}/16$),

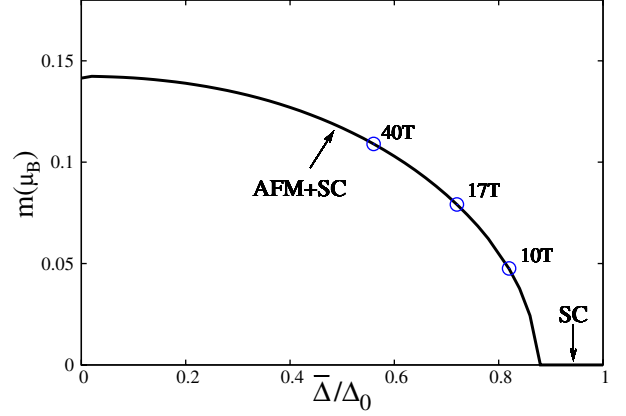


Fig. 2: Staggered magnetization m obtained from eq. (9) of RMFT as a function of the ratio of the suppressed pairing amplitude and the zero-field pairing amplitude $\alpha = \bar{\Delta}/\Delta_0$ at doping $\delta = 1.2\delta_c$ ($\delta_c = 0.10$) ($t = 0.3 \text{ eV}$, $J = 0.1 \text{ eV}$, $t' = -0.1 \text{ eV}$), where $\bar{\Delta}$ is the spatially averaged pairing mean field $\Delta(\mathbf{r})$ as discussed in context, Δ_0 is the pairing amplitude at zero field. Circles indicate the estimated corresponding applied fields (see the text).

18 T ($\sim H_{c2}/9$), and 40 T ($\sim H_{c2}/4$) are 0.82, 0.72, and 0.56, respectively. The points corresponding to these values are denoted in fig. 2 with open circles.

We turn now to the implications of our calculations for the interpretation of the recent experiments by Doiron-Leyraud *et al.* [1]. In their experiments, the Hall resistance of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$, as a function of magnetic field, shows clear oscillation in the vortex liquid phase. The FS area thus obtained is found by using the Onsager relation and corresponds to small FS pockets with a charge carrier density of 0.0375. The compound is SC without AF ordering at low temperature in the absence of magnetic field. Estimates of the hole doping from the SC transition temperature give a value $\delta = 0.1$. Since Fermi arcs have been observed in angle-resolved photoemission spectra around the nodal points in Na-CCOC at similar hole doping [4], Doiron-Leyraud *et al.* interpreted the full FS to be composed of 4 such Fermi pockets. Here we point out that while the low-temperature phase at $H_{\text{app}} = 0$ is SC, the high magnetic field may induce AF ordering as discussed above. If this is the case, the area enclosed by the full FS will be double the pocket area. Thus, the quantum oscillation may actually indicate a hole doping of only 0.075 instead of 0.15.

Since quantum oscillations in the vortex liquid phase are known to reproduce those of the underlying normal state [21], we examine the FS pockets in the non-SC state. To this end we extend the phenomenological theory we developed earlier for the pseudogap state [24] to include AF ordering. We make an ansatz for the coherent part of the Green function (GF) which represents a quasiparticle,

$$G_{\text{AF}}^s(\mathbf{k}, \omega) = \frac{z}{\omega - \xi_{\mathbf{k}s} - \Sigma_s(\mathbf{k}, \omega)}, \quad (10)$$

where z is a numerical factor for the weight of the coherent part. The self-energy takes the form

$$\Sigma_s(\mathbf{k}, \omega) = \Delta^2 \gamma_-^2(\mathbf{k}) / (\omega + s\zeta_{\mathbf{k}}), \quad (11)$$

where $\zeta_{\mathbf{k}s}$ and $\xi_{\mathbf{k}s}$ are given in eqs. (7) and (8) with the replacement of m_v by m_0 . The Luttinger sum rule [25,26] for the number of electrons can be formulated as

$$N_e = \sum_{\mathbf{k}s} \Theta[\text{Re } G_{\text{AF}}^s(\mathbf{k}, \omega = 0)], \quad (12)$$

where $\Theta(x)$ is the Heaviside step function. Note that the boundary of the reduced BZ in the SDW state coincides with the Luttinger surface of zeroes of the corresponding GF in the absence of AF ordering. The FS of G_{AF}^s depends on the staggered magnetization m_0 . In fig. 3, we show the FS for several values of m_0 at a hole doping $\delta = 0.075$. In that case, the upper SDW band is completely empty and the lower SDW band is partially filled. Δ is chosen to be the value defined as in eq. (6) that gives the lowest energy in the RMFT for a given m_0 . In such case the orbits of a quasiparticle in a magnetic field are confined to the reduced BZ and consist of two ellipses each composed of two half-ellipses centered on opposite diagonals BZ. The period of quantum oscillations corresponds to one half of the total hole density. In the absence of AF order, *i.e.* $m_0 = 0$, the phenomenological GF has now four separate pockets near the nodal points and associating the period of the quantum oscillations with the area of a pocket leads to a total hole density twice as large.

The electron GF can be obtained from the SDW GF. The resulting electron spectra weights relevant for angle-resolved photoemission spectra measurements are shown in fig. 3 by the width on the Fermi pockets. The spectral weight distribution inside the reduced zone at small m_0 is similar to our phenomenological theory for the pseudogap state [24], *i.e.* a substantial weight only on the inner edge. The spectral weight outside the reduced BZ is very small but grows as m_0 increases. With increasing m_0 , the pockets become more ellipsoidal.

In summary, we have used the renormalized mean-field theory to show that a high magnetic field may induce antiferromagnetism in underdoped superconducting state. Applying our theory to the recently observed quantum oscillations in a magnetic field in underdoped cuprates [1], we propose that the period measured in oscillations implies a hole density of 0.075, instead of 0.15 as originally proposed.

Since the original manuscript was prepared, quantum oscillations have been observed in $\text{YBa}_2\text{Cu}_4\text{O}_8$ [27]. Again assuming 2 rather than 4 pockets gives a more reasonable value of the hole density $\delta = 0.1$. Lee has drawn our attention to his proposal of a doubled unit cell due to a staggered flux phase [28] as an alternative to the AF phase considered here. We have carried out a similar analysis for this case and found that it has a slightly lower energy than the AF phase. It may well be possible to observe the

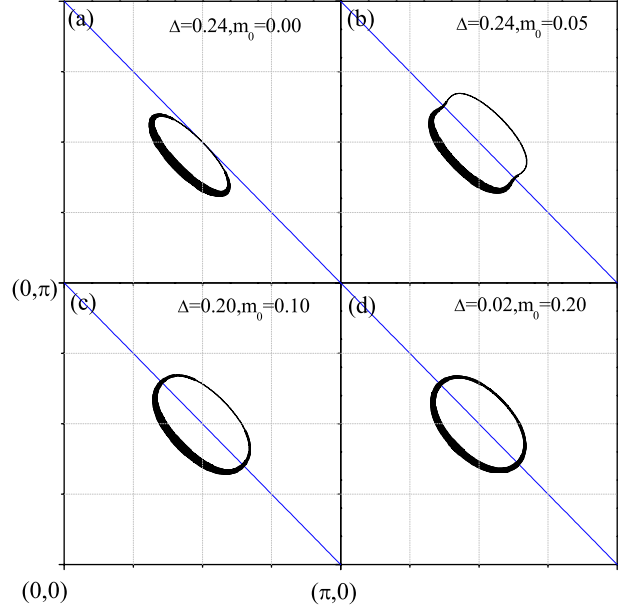


Fig. 3: Evolution of the Fermi surface in one quadrant of BZ calculated from eqs. (10)–(12) at hole doping $\delta = 0.075$. The period of quantum oscillations is given by the area of the closed Fermi pocket, which is $\delta/4$ for (a) in the absence of AF order and is $\delta/2$ for (b)–(d) in the AF ordered state. The thickness of the curves represents the spectral weight.

broken symmetry in moderate fields (*e.g.* see fig. 2) and so check for both possibilities experimentally. Renormalized mean-field theory shows that a high magnetic field may induce antiferromagnetism in underdoped superconducting state. Applying our theory to the recently observed quantum oscillations in a magnetic field in underdoped cuprates [1], we propose that the period measured in oscillations implies a hole density of 0.075, instead of 0.15 as originally proposed.

After we submitted the paper, LeBoeuf *et al.* [29] published their new data which strongly suggest that at high magnetic fields in both YBCO compounds an unpaired normal state is stabilized with a longer period AF or other superlattice rather than the simple AF or flux ordering that we consider in this paper. This implies that the overall negative sign of the Hall signal cannot be attributed to a contribution from vortex motion and is due rather to electron than to hole pockets. Millis and Norman have found that such an electron pocket may appear and dominate magnetotransport in the presence of an AF superlattice with an 8-fold period perpendicular to the chains at $1/8$ doping [30]. Further calculations extending our results are underway and will be reported elsewhere.

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