Quantum oscillation signatures of nodal spin-orbit coupling in underdoped bilayer high $T_c$ cuprates

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(Dated: July 10, 2014)

PACS numbers: 71.45.Lr, 71.20.Ps, 71.18.+y

An inextricable link exists between the crystal structure and Fermi surface in metals, and understanding this link is crucial for arriving at a more complete understanding of the metallic state at low temperatures [1–4]. In the case of underdoped high $T_c$ superconductors, several observations made by way of quantum oscillation experiments [6] now contribute to our understanding of their metallic state. Broken translational symmetry, for example, has been shown to reconstruct the large hole Fermi surface at low temperatures [11]. The bilayer structure of YBa$_2$Cu$_3$O$_{6+x}$ has further been shown to introduce a splitting of the Fermi surface cross-section [12], leading to multiple quantum oscillation frequencies [13]. More recently, the form of the interlayer dispersion has been found to locate the Fermi surface pockets at the corners of a reconstructed body-centered orthorhombic Brillouin zone [14].

One experimental signature that remains unexplained in underdoped YBa$_2$Cu$_3$O$_{6+x}$ is the large difference in the Zeeman splitting of the Landau levels (or orbitally-averaged effective $g$-factor $g_{\text{eff}}$) for different cyclotron orbits originating from the CuO$_2$ planes [14, 17]. While the amplitude of the dominant quantum oscillation frequency $F_0 \approx 530$ T exhibits two spin zeroes (angles at which $\sin\theta = 0$) [14, 18], the amplitude of the oscillations of the frequencies $F_0 - \Delta F \approx 440$ T and $F_0 + \Delta F \approx 620$ T exhibits only a single spin zero over the same angular range (see blue curve in Fig. 1b). Since spin zeroes occur whenever the spin damped factor

$$R_s = \cos \left( \frac{\pi g_{\text{eff}} \mu_B B}{\hbar \omega_c} \right) = \cos \left( \frac{\pi m^* g_{\text{eff}}}{2m_e \cos\theta} \right)$$

(1)

renormalizing the quantum oscillation amplitude vanishes [14], which occurs when the product $(m^*/m_e \cos\theta) g_{\text{eff}}$ is equal to an odd integer, the number and location of spin zeroes can be used to infer values of $g_{\text{eff}}$. Here, $g_{\text{eff}} \mu_B B$ is the Landau level Zeeman splitting, $\hbar \omega_c$ is the cyclotron energy, $m^*/m_e \cos\theta$ is the quasiparticle effective mass in a quasi-two-dimensional metal and $m_e$ is the free electron mass. Two spin zeroes for $F_0$ imply an effective $g$ factor of $g_{\text{eff}} \approx 2$ similar to that of free electrons [14]. One spin zero for $F_0 - \Delta F$ and $F_0 + \Delta F$, by contrast, suggests a significantly reduced...

![FIG. 1: a, Simulated $\theta$-dependence of experimental quantum oscillation amplitudes at $B \cos \theta = 25$ T from Ref. 13 (having divided out the Fermi surface warping factor $R_w$)](image)
Diagonalization then yields

\[ \varepsilon^B = \varepsilon_k - \sqrt{(\alpha^2 k^2 + h^2 + t_{\perp,0}^2 \cos^2 2\phi) \pm 2h \sqrt{t_{\perp,0}^2 \cos^4 2\phi + \alpha^2 k^4 \sin^2 \theta \sin^2 (\phi - \phi_0)}} \]

\[ \varepsilon^{AB} = \varepsilon_k + \sqrt{(\alpha^2 k^2 + h^2 + t_{\perp,0}^2 \cos^4 2\phi) \mp 2h \sqrt{t_{\perp,0}^2 \cos^4 2\phi + \alpha^2 k^4 \sin^2 \theta \sin^2 (\phi - \phi_0)}} \]

in which

\[ t_{\perp,0} \approx t_{\perp,0} \cos^2 2\phi \]

-describes the variation of the bilayer hopping around the Fermi surface and

\[ H_{\text{layer}}^\pm = \varepsilon_k \sigma_0 \pm \alpha (\sigma \times k) \cdot \hat{z} + \frac{1}{2} g \mu_B (B \cdot \sigma) \]

represents each layer. Rashba-type spin-orbit interactions \[21\] of opposite sign (i.e. \( \pm \alpha \)) for each layer result from the opposing directions of the polar vectors describing the non-centro-symmetric environment in each layer. Meanwhile, \( k = (k \cos \phi, k \sin \phi, k_z) \) is the momentum vector, \( g \) is the electron \( g \)-factor (assumed to be equal to 2), \( B \) is the magnetic field, \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) is the Pauli matrix vector and \( \sigma_0 \) is the 2 \( \times \) 2 identity matrix.

An important consideration here is that the Fermi surface pocket is located at a high symmetry point in the Brillouin zone, such as at the S-point in a simple orthogonal Brillouin zone \[12\] or the T-point in a reconstructed body-centered orthorhombic Brillouin zone \[14\], causing \( t_{\perp,0} \) to vanish four times around the circumference of the pocket. We neglect the in-plane and inter-plane geometry of the Fermi surface, which are unimportant for spin-orbit interactions, by considering an unperturbed single layer electronic dispersion of the form \( \varepsilon_k = h^2 k^2 / 2m^* - \mu \) (where and \( \mu \) is the chemical potential) which produces an unperturbed circular Fermi surface of radius \( k_0 = \sqrt{2m^* \mu / h} \).

Upon expanding Equation (2) and making the substitution \( g \mu_B B = h \sin \theta \cos \phi_0, \sin \theta \sin \phi_0, \cos \theta \) (in which \( \phi_0 \) is the azimuthal angle of the in-plane magnetic field component), we obtain
for the bonding (B) and antibonding bands (AB), respectively.

A vanishing bilayer splitting at the nodes (illustrated at \( \phi = 45^\circ \) in Fig. 2) causes the spin-down bonding (\( \varepsilon_+^B \) depicted in green) and spin-up antibonding (\( \varepsilon_+^{AB} \) depicted in blue) bands to intersect in a magnetic field. A finite \( \alpha \) then causes \( \varepsilon_+^B \) and \( \varepsilon_+^{AB} \) to become mixed, leading to the opening of gaps \( \Delta_{SO} \) (shown in Fig. 2) that are offset in \( \phi \) relative to the nodes. For \( \theta = 0 \), Equation (4) reduces to the simple forms \( \varepsilon_+^B = \varepsilon_k - \sqrt{(t_{\perp,k} \pm h)^2 + \alpha^2 k^2} \) and \( \varepsilon_+^{AB} = \varepsilon_k - \sqrt{(t_{\perp,k} \pm h)^2 + \alpha^2 k^2} \). More generally, an in-plane component of the magnetic field (i.e. \( \theta \neq 0 \)) leads to the opening of additional \( \phi \)-dependent gaps \( \Delta_{\perp} \) between \( \varepsilon_+^B \) and \( \varepsilon_+^{B} \) and between \( \varepsilon_+^{AB} \) and \( \varepsilon_+^{AB} \) (shown schematically for large \( \theta \) in Fig. 2).

The presence of gaps in momentum-space transforms the Fermi surface into a network of coupled orbits [19] (shown schematically in Fig. 3). At each junction (labeled A\(_0\), B\(_\perp\), C\(_\perp\) in Fig. 3) in which the separation between neighboring orbits is small, quasiparticles undergoing cyclotron motion can either ‘tunnel,’ whereupon they switch paths to a neighboring orbit, or ‘reflect,’ whereupon they remain on the same orbit. While the reflection and tunneling amplitudes are generally complex, the phase within the complex plane contributes only a constant shift in Onsager phase of the quantum oscillations. We therefore consider a simple case in which the tunneling amplitudes (\( ip_{SO} \) and \( ip_{\perp} \)) are pure imaginary and the reflection amplitudes (\( q_{SO} \) and \( q_{\perp} \)) are real. To meet the orthogonality requirement, these are subject to the constants \( p_{SO}^2 + q_{SO}^2 = 1 \) and \( p_{\perp}^2 + q_{\perp}^2 = 1 \). Examples of possible orbits with different combinations of \( p_{SO}, p_{\perp}, q_{SO} \) and \( q_{\perp} \) are shown in Fig. 3.

While magnetic breakdown enhances \( p_{SO} \) and \( p_{\perp} \) at the expense of \( q_{SO} \) and \( q_{\perp} \) as the magnetic field is increased [19], spin-momentum locking can cause the nature of the tunneling and reflection at \( A_{SO} \) to exhibit qualitative differences from that occurring at band gaps near the Brillouin zone boundary in regular metals [22]. For the purposes of estimating the Zeeman splitting of the Landau levels, we concern ourselves here with the local effective \( g \) factor \( g_k = (2/\mu_B) \partial \varepsilon_+^{B,AB} / \partial B \) in which \( B \) is the magnitude of the magnetic field. In general, \( g_k \) depends on \( h, \theta, \phi \) and \( \phi_0 \) in the vicinity of \( A_{SO} \) in Fig. 3, and is therefore not directly related to the direction of the spin. On moving away from \( A_{SO} \) or by taking the limit \( \alpha \to 0 \), however, \( g_k \) saturates at \( \pm 2 \) whereby it then coincides with the projection of the spin along the direction of the applied magnetic field. Quasiparticles therefore experience a net change in the sign of \( g_k \) on being reflected at \( A_{SO} \), indicating that their spins are flipped. On taking the orbital average

\[
    g_{eff} = \frac{1}{2\pi} \int_{0}^{2\pi} g_k \, d\phi \tag{5}
\]

multiple spin flips cause the overall Zeeman splitting and

\[
   \Delta k_{SO} \text{ and } \Delta k_{\perp} \text{ indicate gaps in momentum space that are subject to tunneling, the latter vanishing at } \theta = 0. \quad \text{b, Schematic Fermi surface for } \alpha = 0 \text{ and } B = 0 \text{ with a finite residual nodal gap } \Delta k_{SO} \text{, indicating corresponding quantum oscillation frequencies for the bonding and antibonding Fermi surface cross-sections. c-h, Examples of magnetic breakdown orbits (dotted lines, with the corresponding quantum oscillation frequencies indicated in cyan), with the absolute transmission amplitudes } p_{\perp}, q_{\perp}, p_{SO} \text{ and } q_{SO} \text{ also shown. Dashed lines delineate segments of the orbit with opposite signs of } g_k.\]
therefore the effective $g$-factor to be strongly reduced relative to the free electron value for cyclotron orbits that include reflections at $A_{SO}$. Conversely, no net spin flip occurs for quasiparticles that tunnel between orbits at $A_{SO}$; nor does a net spin flip occur for quasiparticles that either tunnel or reflect at $\beta_{\perp}$ and $C_{\perp}$ in Fig. 3. Orbits without spin flips (e.g. $F_3$ and $F'_4$ in Fig. 3) therefore have an orbitally-averaged effective $g$-factor close to the free electron value.

Examples of orbits and their corresponding frequencies and orbitally-averaged effective $g$-factors are shown in Figs. 3-g and in Table I. In spite of the presence of multiple junctions around each orbit at which quasiparticles can tunnel or reflect, the bilayer Fermi surface obtained on projecting back to $B = 0$ in Fig. 3 is topologically identical to that considered in Refs. 13, 14. The minimum and maximum Fermi surface cross-sections, respectively yielding frequencies $F_0 - 2\Delta F$ and $F_0 + 2\Delta F$, and also the series of magnetic breakdown orbits, yielding frequencies $F_0 = \Delta F$, $F_0$ and $F_0 + \Delta F$ are therefore the same as those considered in Refs. 13, 14.

| Orbit $F_j$ | $N_j$ | $R_{MB}$ | $g_{eff}$ |
|-------------|-------|----------|-----------|
| $F_1 = F_0 - 2\Delta F$ | 1 | $p^2 q_{SO}^2$ | 0.77 |
| $F_2 = F_0 - \Delta F$ | 4 | $-p^2 q_{SO}^2$ | 0.55 |
| $F_3 = F_0$ | 2 | $p^2 q_{SO}^2$ | 2.00 |
| $F_4 = F_0$ | 4 | $-p^2 q_{SO}^2$ | 0.00 |
| $F_5 = F_0 + \Delta F$ | 4 | $-p^2 q_{SO}^2$ | 0.55 |
| $F_6 = F_0 + 2\Delta F$ | 1 | $p^2 q_{SO}^2$ | 0.77 |
| $F'_1 = F_0 - 2\Delta F$ | 4 | $p^2 q_{SO}^2$ | 1.31 |
| $F'_2 = F_0 - \Delta F$ | 4 | $p^2 q_{SO}^2$ | 2.00 |
| $F'_3 = F_0 + \Delta F$ | 4 | $p^2 q_{SO}^2$ | 1.31 |

TABLE I: Magnetic breakdown probabilities and effective $g$-factors. Only $F_1, F_2, F_3, F_4, F_5$ and $F_6$ are relevant at $\theta = 0$. Owing to the vanishing of $\Delta'_F$ at $\theta = 0$, the nodal tunneling and reflection amplitudes become $v_p = i$ and $q_L = 0$, respectively. $F'_1, F'_2, F'_3$ and $F'_4$ are examples of orbits that become finite in amplitude only when $\theta \neq 0$. Here, $g_{eff}$ is estimated from Equation 55 using $g = 2$, $B = 85$ T, and $t_{\perp,0} = 15$ meV in the limit $\alpha \to 0$.

In Fig. 4, we calculate the $\theta$-dependent quantum oscillation amplitude

$$A = \sum_j N_j R_{MB} R_s$$

(6)

according to the spin-orbit coupled bilayer Fermi surface model for the frequencies $F_0 \pm \Delta F$ and $F_0$ seen in quantum oscillation experiments (neglecting damping factors associated with Fermi surface warping and scattering). Here, $N_j$ is the number of instances an equivalent orbit is repeated (at different orientations) within the Brillouin zone, while $R_{MB} = (i\nu^\perp L (i\nu)^{SO} q_{SO}^L \nu_{SO}^L$ is the magnetic breakdown amplitude reduction factor (in which $\nu_{SO}$ enumerate tunneling events encountered $en route$ around the orbit and $\mu_L$ and $\mu_{SO}$ enumerate reflection events). The tunneling and reflection amplitudes depend on $\phi$ and must therefore be calculated at each junction around the orbit. The calculations are made for $\alpha k_0 = 1.5$ meV and $\phi_0 = -45^\circ$ using the approximate expressions $v^2 \approx -\Delta_0^2/\hbar \omega_{cF}$ and $p_{B_{eff}}^2 \approx -\Delta_0^2/\hbar \omega_{cF}$ [19]. Such forms for the tunnel probability are generally accurate in the limits $\Delta_0 \gg \hbar \omega_{cF}$ and $\Delta_0^2 \gg \hbar \omega_{cF}$, but continue also to provide a useful extrapolating formula in the opposite limits where $\Delta_0$ and $\Delta_{\perp}$ vanish. We also neglect the anisotropy of $g$ [14] (its neglect giving rise to an additional spin zero in the amplitude of $F_3$ near $\theta = 5^\circ$ in Fig. 4).

There are three ways in which bilayer spin-orbit system produces angle-dependent Zeeman splitting effects that are consistent with quantum oscillation experimental observations in Fig. 1. First, orbits $F_2$ and $F_5$, which are responsible for the $F_0 - \Delta F$ and $F_0 + \Delta F$ frequencies (assumed to be of the same amplitude), contain spin flips (i.e. reflections at $A_{SO}$ or equivalent junctions in Fig. 3) and therefore have a small orbitally-averaged effective $g$-factor ($g_{eff} < 1$ in Table I). The small effective $g$-factor produces only a single spin zero on rotating $\theta$ form 0 to 70° in Fig. 4, which is then consistent with the experimental observation of a single spin zero for these frequencies in Fig. 4. Second, the $F_3$ orbit, whose amplitude is primarily responsible for the $F_0$ frequency, involves no spin flips (i.e. no reflections at $A_{SO}$ or its equivalent junctions in Fig. 3) and therefore has an orbitally-averaged effective $g$-factor close to 2 in Table I. The two spin zeroes produced at high angles $\theta \geq 50^\circ$ in Fig. 4, are therefore consistent with those experimentally seen for the $F_0$ frequency in Fig. 4. Thirdly, orbit $F_4$, which produces a secondary weaker contribution to the $F_0$ frequency, has an orbitally-averaged effective $g$-factor of zero. A secondary contribution to $F_0$ with small effective $g$-factor was indeed found to be necessary for producing accurate simulations of the $\theta$-dependent total quantum oscillation wave form in Ref. 13.

If we assume spin-orbit interactions to control the residual splitting of the states in the nodal region, then the characteristic magnetic breakdown field $B_0 = \Delta_0^2/\hbar \varepsilon_F/\omega \approx 2.7$ T obtained from recent simulations of the experimental data as a function of $B, \theta$ and $\phi_0$ [14] can be used to estimate $\Delta_{SO} = \sqrt{\hbar \mu_B B_0/\mu} \sim 3$ meV for the magnitude of the spin-orbit interaction strength at $B = 0$. The corresponding zero field momentum space nodal gap $\Delta_{SO} = \Delta_{SO}/\hbar \varepsilon_F \sim 0.005$ Å$^{-1}$ (where $\varepsilon_F = \sqrt{2\hbar e F_0/\mu}$ is the orbitally-averaged Fermi velocity) is comparable in magnitude to the residual nodal bilayer gap of $\approx 0.01$ Å$^{-1}$ inferred from angle-resolved photoemission measurements of $Bi_2Sr_2CaCu_2O_{8+\delta}$ [22] and further consistent with its reported vanishing (below resolution limits) at hole dopings $< 15$ % in underdoped $YBa_2Cu_3O_{6.5}$ [24]. Our findings provide motivation for
performing high resolution spin-polarized photoemission experiments on Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ and YBa$_2$Cu$_3$O$_{6.5}$, and further searching for analogous multiple-layer spin-orbit interactions in bilayer ruthenates [26, 27] and heavy fermion superlattices [28].

While we have considered a scenario in which spin-orbit interactions result from polar vectors pointing perpendicular to the CuO$_2$ planes (giving rise to a Rashba-type spin-orbit interaction [21]), identical results are obtained for other types of non-centrosymmetric environment that are equal and opposite in each CuO$_2$ plane (such as those giving rise to Dresselhaus spin-orbit interactions [29]). These would require additional symmetry breaking. A global breaking of the centro-symmetric symmetry can be ruled out as a likely factor in under-doped YBa$_2$Cu$_3$O$_{6+x}$, however, as this would produce a significant spin-orbit splitting over the entire Fermi surface [30] rather than only in the vicinity of the nodes.

In summary, we have shown that the large variation of the effective g-factor for different orbits can be understood by a situation in which spin-orbit interactions control the residual bilayer splitting at the nodes [12]. We show that a scenario in which spin-orbit interactions are equal and opposite in each of the CuO$_2$ planes can qualitatively explain the observed angle-dependence of the Zeeman splitting. A significantly reduced $g_{\text{eff}}$ for some orbits is therefore likely to be the consequence of the quasiparticles undergoing a spin flip while traversing the nodal region in momentum space. Our findings provide motivation for searching for the possible consequences of nodal spin-orbit interactions for superconductivity and competing ordered phase in multilayered cuprates and other strongly correlated materials.

This work is supported by the US Department of Energy BES “Science at 100 T” grant no. LANLF100, the National Science Foundation and the State of Florida.

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