Orbital magnetic response and the anisotropy of magnetic susceptibility in the Iron-based superconductors

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(Dated: January 13, 2013)

We propose that the orbital angular momentum of the conduction electrons in the Iron-based superconductors is activated in their low energy physics. Using a five-band tight-binding model derived from fitting the LDA band structure, we find that the orbital magnetic susceptibility of the conduction electrons in such a multi-orbital system is several times larger than the Pauli spin susceptibility and is comparable in magnitude to the observed total magnetic susceptibility. The orbital magnetic susceptibility in the Fe-As plane($\chi^L_z$) is found to be larger than that perpendicular to the Fe-As plane($\chi^L_z$) by a factor about two and the total magnetic susceptibility in the normal state can be fitted with formula $\chi(T,\theta) \approx \chi_s(T) + \chi_L(\theta)$, where $\chi_s(T)$ is the temperature dependent isotropic part due to spin and $\chi_L(\theta)$ is the temperature independent anisotropic part due to orbital. In the superconducting state, $\chi^L_z$ is found to be significantly reduced as the pairing gap develops, while $\chi^L_z$ is almost not affected by the superconducting transition. We argue the large anisotropy observed in the bulk magnetic susceptibility and the Knight shift in the Iron-based superconductors should be attributed to the orbital magnetic response of their conduction electrons.

PACS numbers:

One special feature of the newly discovered Iron-based superconductors\textsuperscript{1–4} is their multi-orbital nature. In conventional superconductors, in which only one orbital plays an essential role around the Fermi energy, the orbital angular momentum is quenched either as a result of the s-wave character of the orbital, or by the crystal field splitting effect. However, from LDA band structure calculation, it is reported that all the five Fe 3d orbital play essential role in forming the low energy degree of freedom around the Fermi energy and the crystal field splitting is extremely small. This situation has caused great complexities in the model study. However, it also generates the interesting opportunity to explore the physics of the orbital degree of freedom in this system. The orbital character near the Fermi energy has now been extensively studied by angle-resolved photoemission spectroscopy (ARPES) and the LDA result is to a large extent confirmed\textsuperscript{5–8}.

Many novel properties of the Iron-based superconductors have been attributed to their multi-orbital nature. For example, the intimate relation between the structural and magnetic phase transition\textsuperscript{9,10} has been proposed to originate from an orbital-related mechanism\textsuperscript{11,12}. The same picture also provides a reasonable interpretation for the unusual in-plane anisotropy in resistivity\textsuperscript{13,14} and the $d_{xz}$ and $d_{yz}$ band splitting observed in the ARPES measurement\textsuperscript{8}. In these situations, the multi-orbital nature manifests itself in the form of a static structure, a more interesting possibility is that the orbital degree of freedom appearing as a dynamic mode in the low energy physics, contributing to various kinds of response and relaxation processes. The purpose of this paper is to investigate the contribution of the orbital angular momentum of the conduction electrons to the magnetic susceptibility.

The magnetic susceptibility of a metal is usually attributed to the response of its spin degree of freedom, which in the absence of the spin-orbital coupling is isotropic. The orbital magnetic susceptibility, which is in principle anisotropic, is usually small as a result of the quenching of the orbital angular momentum of the conduction electrons. The small remnant orbital magnetic response, namely the well known Van Velck paramagnetism, is controlled by the large gap separating the occupied bands from the unoccupied atomic levels that carry orbital angular momentum and is expected to show negligible temperature dependence. However, if the conduction electron itself carries orbital angular momentum, the orbital magnetic response of the system would be much larger and be sensitive to the changes of electronic state of the system.

Measurement of the magnetic susceptibility on the Iron-based superconductor has produced several intriguing results. Firstly, the magnetic susceptibility is found to show significant temperature dependence\textsuperscript{17–21}. Such a temperature dependence is unexpected for a weakly correlated system with a large band width. In the literature, this unusual temperature dependence is either attributed to the strong correlation effect, or to the proximity of the system to a semimetal phase\textsuperscript{22–24}. Secondly, the magnetic susceptibility is found to be strongly anisotropic\textsuperscript{17–21}. As shown in Fig. 1 the susceptibility in the Fe-As plane is much larger than that perpendicular to the Fe-As plane and their difference is nearly temperature independent. An understanding of such anisotropy in the magnetic susceptibility is still absent and we will show that it can be originated from the contribution of the orbital angular momentum of the conduction electrons.

In this paper, we study the orbital magnetic response of the conduction electrons in the Iron-based supercon-
The magnetic susceptibility data of Ba122 system are quoted from Ref. 21. Here the superscripts $xy/x$ and $z$ denote the direction of the applied magnetic field.

The model Hamiltonian Eq. (1) can be diagonalized in momentum space, in which it takes the form

$$H_0 = \sum_{\mathbf{k}, \alpha, \sigma} \epsilon_{\mathbf{k}, \alpha} c_{\mathbf{k}, \alpha, \sigma}^\dagger c_{\mathbf{k}, \alpha, \sigma},$$

where $\epsilon_{\mathbf{k}, \alpha}$ is the band energy for the $\alpha$-band.

To describe the superconducting state, we model the pairing potential with a sign-changing $s$-wave form $\Delta_{\alpha\beta}(\mathbf{k}, T) = \Delta(T) \delta_{\alpha\beta} \cos(k_x) \cos(k_y)$. Note that only the inband pairing is considered. Such a pairing symmetry is consistent with the weak coupling spin-fluctuation-exchange mechanism or the strong coupling superexchange mechanism for the superconductivity in the iron-based superconductors. However, as will be clear below, the detailed form of the pairing potential (except its inband pairing nature) is not essential for the uniform susceptibility. So we will take the pairing potential adopted only as a simplified way to induce a full gap on the Fermi surfaces of the system.

The orbital magnetic susceptibility is defined through the correlation function of the orbital magnetic moment in the following way

$$\chi_L^\alpha(\mathbf{q}, \tau) = -(g_L \mu_B)^2 \langle T_\tau L^\alpha(\mathbf{q}, \tau) L^\alpha(-\mathbf{q}, 0) \rangle,$$

in which $L^\alpha(\mathbf{q}, \tau)$ denotes the Fourier component of the orbital magnetic moment density in the $\alpha$-direction. The orbital magnetic moment on a given site $i$ is defined as $L^\alpha_i = \sum_{\mu, \nu, \sigma} c^\dagger_{i, \mu, \sigma} l^\alpha_{\mu, \nu, \sigma} c_{i, \nu, \sigma}$, where $l^\alpha_{\mu, \nu, \sigma}$ is the matrix element of the orbital magnetic moment in the space spanned by the five orbital $3d_{xz}, 3d_{yz}, 3d_{x^2-y^2}, 3d_{x^2}, 3d_{yz}$ respectively. $t(\Delta x, \Delta y; \mu, \nu)$ denotes the in-plane hopping integral between the $\mu$-th and $\nu$-th orbitals at the lattice distance $(\Delta x, \Delta y)$. $\varepsilon_{\mu}$ is the on-site energy of the $\mu$-th orbital. The values of these model parameters are given in Ref. 19.
and

\[
P^{\tau} = \begin{pmatrix}
0 & -i\sqrt{3} & 0 & 0 \\
0 & 0 & 0 & -i \\
i\sqrt{3} & 0 & 0 & i \\
0 & 0 & i & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}.
\]

The orbital magnetic susceptibility is readily obtained as follows

\[
\chi_{q}^{m}(T) = \lim_{q \to 0} \frac{1}{N} \sum_{k,\alpha,\beta} \left[ f(E_{k+q,\beta}) - f(E_{k,\alpha}) \right] \frac{E_{k,\alpha}E_{k+q,\beta}}{E_{k,\alpha}E_{k+q,\beta}} \\
\times \left( 1 + \frac{\xi_{k,\alpha}\xi_{k+q,\beta} + \Delta_{k,\alpha}\Delta_{k+q,\beta}}{E_{k,\alpha}E_{k+q,\beta}} \right) \times |O_{k,\alpha,\beta}^{m}|^2
\]

Here \( E_{k,\alpha} \) denotes the excitation energy of the quasi-particle in the \( \alpha \)-th band and is given by \( E_{k,\alpha} = \sqrt{\xi_{k,\alpha}^2 + \Delta_{k,\alpha}^2} \). \( \xi_{k,\alpha} = \xi_{k,\alpha} - \mu \) and \( \Delta_{k,\alpha} \) are the band energy and the pairing gap of the \( \alpha \)-th band, \( \mu \) is the chemical potential. \( O_{k,\alpha,\beta}^{m} = \sum_{\nu} U_{\nu}^{\alpha,\beta} \sum_{\nu'} U_{\nu'}^{\beta,\alpha} \) is the matrix element of \( P^{\tau} \) in the basis of the band eigenstate at momentum \( k \). \( U_{\nu,\alpha} \) is the \( \alpha \)-th eigenvector of the band Hamiltonian at momentum \( k \). In deriving Eq. (5), we have used the inversion symmetry of the system and the fact that \( P_{\nu,\nu'}^\alpha = -P_{\nu,\nu'}^\alpha \).

For comparison, we also calculate the Pauli spin susceptibility of the band electrons. The Pauli susceptibility is defined through the following spin-spin correlation function,

\[
\chi S(q, \tau) = -(g_{SB})^2 \langle T_{S^z}(q, \tau)S^z(-q, 0) \rangle, \quad (6)
\]

where \( g_{SB} = 2 \) and the spin density operator at lattice site \( i \) can be written as \( S_{i}^z = \sum_{\mu, s' s} c_{i, \mu, s}^{\dagger} \sigma_{s}^{z} c_{i, \mu, s'} \). The uniform bare spin susceptibility can be shown to be given by

\[
\chi S(T) = \lim_{q \to 0} \frac{1}{N} \sum_{k,\alpha} \left[ f(E_{k+q,\alpha}) - f(E_{k,\alpha}) \right] \frac{E_{k,\alpha}E_{k+q,\alpha}}{E_{k,\alpha}E_{k+q,\alpha}} \frac{E_{k,\alpha}}{E_{k,\alpha}} \\
\times \left( 1 + \frac{\xi_{k,\alpha}\xi_{k+q,\alpha} + \Delta_{k,\alpha}\Delta_{k+q,\alpha}}{E_{k,\alpha}E_{k+q,\alpha}} \right) \times |O_{k,\alpha,\beta}^{m}|^2
\]

The difference between Eq. (5) and Eq. (7) lies in the fact that the orbital magnetic response has contributions from both the intraband and interband processes, while the spin susceptibility has only intraband contributions, the reason for the latter is that the spin density operator is diagonal in the orbital space. Such a difference will have important consequence in the superconducting state. As the coherence factor, \( 1 - \frac{\xi_{k,\alpha}\xi_{k+q,\alpha} + \Delta_{k,\alpha}\Delta_{k+q,\alpha}}{E_{k,\alpha}E_{k+q,\alpha}} \), vanishes when \( q \to 0 \) in the superconducting state, \( \chi S(T) \) will show activation behavior if there is a full gap on the Fermi surface. On the other hand, the coherence factor for the orbital magnetic susceptibility, \( 1 - \frac{\xi_{k,\alpha}\xi_{k+q,\alpha} + \Delta_{k,\alpha}\Delta_{k+q,\alpha}}{E_{k,\alpha}E_{k+q,\alpha}} \), is in general nonzero for \( \alpha \neq \beta \) when \( q \to 0 \). As a result, \( \chi q^2(T) \) will in general be nonzero in the zero temperature limit. However, the intraband contribution to the orbital magnetic susceptibility should be suppressed in exact the same manner as the spin susceptibility in the superconducting state.

Now we present the numerical results for both the orbital and the spin magnetic susceptibilities. In our calculation, the chemical potential is self-consistently determined by solving the particle number equation at each temperature. We then use Eq. (5) and Eq. (7) to calculate the magnetic susceptibilities. The temperature dependence of the pairing gap is modeled by

\[
\Delta(T) = \Delta \sqrt{1 - \frac{T}{T_c}},
\]

in which we have set \( \Delta = 0.02eV \) and \( k_B T_c = 0.005eV \). These parameters are typical for the Iron-based superconductors. The band filling will be fixed at \( n = 6 \) at first, as the band structure calculation leading to model Hamiltonian Eq. (1) is done at such a commensurate filling.

![Figure 2: The spin and orbital magnetic susceptibility of the Iron-based superconductor calculated from the five-band model as functions of temperature.](image)

The results for the orbital and spin magnetic susceptibilities are shown in Fig. 2. Both susceptibilities are found to show little temperature dependence below \( k_B T = 0.05eV \) in the normal state. This is in accordance...
with the expectation for a typical band metal. The orbital magnetic susceptibility is found to be several time larger than the Pauli spin susceptibility and is already comparable in magnitude with the observed total susceptibility. More specifically, $\chi^T_0(T)$ is found to be about 3 times larger than the Pauli spin susceptibility $\chi_S(T)$, which is estimated to be about one-fourth of the observed total magnetic susceptibility at 150 K.21 The orbital magnetic susceptibility also show large anisotropy. The in-plane orbital susceptibility $\chi^L_0(T)$ is found to be about 1.8 times larger than the out-of-plane orbital susceptibility $\chi^z_0(T)$.

In our calculation, we have neglected the interaction correction. The interaction correction is believed to induce enhancement of the effective mass and thus the spin susceptibility. It is also believed that the interaction correction will induce temperature dependence in spin susceptibility. However, the interactions, such as the local Coulomb repulsion and the Hund’s rule coupling, are not expected to renormalize the orbital magnetic susceptibility directly. Thus, although the bare spin susceptibility calculated in this paper may not be a reliable estimation for the real spin response, the results for the orbital magnetic susceptibility should be robust.

In the superconducting state, the spin susceptibility $\chi^S(T)$ drops abruptly as the pairing gap develops on the Fermi surface and approaches zero in the zero temperature limit. The in-plane orbital magnetic susceptibility $\chi^L_0(T)$ also exhibits a significant reduction in the superconducting state, but remains nonzero in the zero temperature limit. On the other hand, the signature of the superconducting transition in $\chi^z_0(T)$ is almost unobservable. To understand why there is such a difference between $\chi^L_0(T)$ and $\chi^z_0(T)$, we note that the diagonal matrix element $O_{\tilde{k},\alpha,\alpha}$ is identical zero as the result of the tetragonal symmetry of the system, while $O_{\tilde{k},\alpha,\alpha}$ is in general nonzero. As we have mentioned above, the intra-band contribution to the orbital magnetic susceptibility also suffers from a suppression by the coherence factor in the superconducting state.

To see how our results depend on the band filling, we have carried out the calculation for several different electron concentrations. It is found that the qualitative features of both the spin and the orbital magnetic susceptibilities, for example, their temperature dependence and anisotropy, is quite robust, but their magnitudes are reduced as we increase the electron concentration of the system. The detailed dependence of the susceptibilities and their relative ratio in the normal state on the band filling are shown in Fig.3. The calculation is done at a fixed temperature $k_BT = 0.03eV$ and the rigid band approximation is assumed. It is found that both the magnitude and the anisotropy of the orbital magnetic susceptibility do not change significantly with band filling. On the other hand, the spin susceptibility show much stronger band filling dependence.

Now we turn back to the experimental data shown in Fig. [1] As a common feature of all the data shown, both the in-plane and the out-of-plane susceptibility exhibit linear temperature dependence above $T_c$ (or $T_N$) with almost the same slope. The in-plane susceptibility is seen to be much larger than the out-of-plane susceptibility in all measurements. Thus the total susceptibility can be interpreted as consisting of two contributions: an isotropic component with a linear temperature dependence and a temperature independent component that is anisotropic. It is quite natural to attribute the isotropic component to the spin magnetic response, which with interaction correction can exhibit strong temperature dependence.22-24 The remaining anisotropic component is more likely to be the contribution of the orbital magnetic moment, which is not directly renormalized by the usual local electron correlation effect and should be temperature independent in the normal state. In principle, the anisotropy in magnetic susceptibility can also be caused by the spin-orbital coupling effect.25 However, in the Iron-based superconduc-
tors, the spin-orbital coupling is negligible small. Therefore, we feel our proposal for the anisotropy is more realistic for the Iron-based superconductors.

In summary, we have shown that the orbital angular momentum of the conduction electrons in the Iron-based superconductors can play a significant role in its low energy physics. It contributes a large temperature-independent anisotropic component to the magnetic susceptibility. As a result, the total magnetic response in the normal state can be separated into a temperature-dependent isotropic part $\chi_s(T)$ and a temperature-independent anisotropic part $\chi_L(\theta)$, where $\theta$ is the angle between the magnetic field and the normal of the Fe-As plane. In other words, a fit of the experimental susceptibility to the formula $\chi(T, \theta) = \chi_s(T) + \chi_L(\theta)$ should be feasible. We note that the orbital magnetic moment can also contribute to the relaxation of the nuclear spins and other fluctuation effect at low energy. A full investigation of the dynamical orbital response will be presented in future works.

YHS is support by NSFC Grant No. 10974167 and TL is supported by NSFC Grant No. 10774187 and National Basic Research Program of China No. 2007CB925001 and No. 2010CB923004.

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