Influence of orbital nematic order on spin responses in Fe-based superconductors

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Electronic nematicity is ubiquitous in Fe-based superconductors, but what the primary nematic order parameter is and how the various nematic phenomena correlate with each other are still elusive. In this manuscript, we study the physical consequence of the orbital nematic order on spin correlations. We find that the orbital nematic order can drive a significant spin nematicity and can enhance the integrated intensity of the spin fluctuations. Our study shows that there is a cooperative interplay between the orbital nematic order and the spin correlations, and the orbital nematic order can not be taken as an unimportant secondary effect of the nematic state in Fe-based superconductors.

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I. INTRODUCTION

The electronic nematic state is a novel state in nature which breaks rotational symmetry spontaneously but is translational invariant. In the newly discovered Fe-based superconductors (FeSCs), the rotation symmetry breaking has been observed ubiquitously, in such as charge resistivity, ARPES, neutron spectra, optical conductivity, nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR), magnetic torque measurement, STM/STS, X-ray, etc. Some reviews are given in References. The rotational symmetry breaking leads to a nematic phase transition at critical temperature \( T_c \). This nematic phase transition shows close correlation to the structural and magnetic phase transitions. The origin of the nematic phase transition and its roles in superconductivity are now active topics in the field.

One complexity to study the electronic nematicity comes from the fact that the rotational symmetry breaking manifests itself in different channels simultaneously. Table I shows the various nematic parameters in experiments. From Landau’s principle of symmetry breaking, the multiplicity of the nematic parameters can be easily understood. Consider the symmetry breaking of Fe-site symmetry group from \( D_{2d} \) to \( D_2 \) in Fe-1111 and Fe-122 families. Any function \( O_\eta \) which is invariant under the operations of \( D_2 \) but not that of \( D_{2d} \) can be defined as a nematic parameter. From symmetry group theory, it can be defined as

\[
O_\eta = \sum_\gamma C_\gamma \Psi_\gamma |O_\eta\rangle = O_\eta, \forall \eta \in D_2,
\]

where \( \Psi_\gamma \) are basis of one irreducible non-identity representation of \( D_{2d} \).

The various nematic parameters show diverse manifestations of the electronic nematicity in FeSCs. In ARPES, the nematicity shows itself as orbital-relevant band shift. In inelastic neutron scattering (INS) spectra, it manifests in anisotropic spin-wave excitations (dispersion and damping) and nematic dynamic spin fluctuations, the latter of which is also shown in \( 1/T_1T \). In charge transport resistivity and conductivity, the nematicity shows as a combined effect of the anisotropic Fermi velocity and anisotropic microscopic scattering processes. The anisotropic scattering is proven as a dominant factor for the nematicity in optical conductivity shows itself in a very large energy range from zero frequency to high energy of about 2eV.

The diverse experiment manifestations of the nematicity leads to hot debates on its primary driving mechanism in FeSCs. Among the various nematic parameters, the Ising spin nematic order and the orbital nematic order are the most popular candidates as the primary one. However, it is still elusive which one is the primary driving force and how these two nematic orders correlate with each other.

In this manuscript, we will study how the orbital nematic order makes influence on the nematic spin correlations. From symmetry analysis, recently we proposed a general form of the orbital nematic order \( O = \sum_{ia,jb} F_{ia,jb} \langle d_{ia}^\dagger d_{jb}\rangle \), which involves a local form factor \( F_{ia,jb} \). The local form factor \( F_{ia,jb} \) can be on-site s-wave, nearest-neighbor d-wave and nearest-neighbor extended s’-wave, etc. These orbital nematic orders are

\[
\begin{array}{ll}
\text{Experiment technique} & \text{nematic parameter } O_\eta \\
\text{resistivity/conductivity} & \langle j_x j_x - j_y j_y\rangle \\
\text{ARPES} & \Delta(k) \langle d_{k,x}^\dagger d_{k,x} \pm d_{k,y}^\dagger d_{k,y}\rangle \\
\text{INS} & \langle S_i \cdot S_{i+\pm} - S_i \cdot S_{i+\mp}\rangle \\
\text{Raman} & \langle \rho^a \rho^b\rangle, \rho^a \text{ Raman charge density} \\
\text{STM/STS} & \langle M_{Q_1} - M_{Q_2}\rangle \\
\text{torque} & \langle S_i^x S_j^y - S_i^y S_j^x\rangle
\end{array}
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\end{array}
\]
defined as below,

\[ O_s = \sum_k \left( d_{k,xz}^\dagger d_{k,xz} - d_{k,yz}^\dagger d_{k,yz} \right), \]

\[ O_d = \sum_k \Delta_d(k) \left( d_{k,xz}^\dagger d_{k,xz} + d_{k,yz}^\dagger d_{k,yz} \right), \] \( \sum \Delta_s'(k) \left( d_{k,xz}^\dagger d_{k,xz} - d_{k,yz}^\dagger d_{k,yz} \right), \]

where \( \Delta_d(k) = (\cos k_x - \cos k_y)/2 \) and \( \Delta_s'(k) = (\cos k_x + \cos k_y)/2 \).

The on-site orbital order \( O_s \) has been taken extensively as a nematic order parameter to account for band shift in ARPES. However, a detailed investigation on the ARPES shows that the band shift of \( d_{xz} \) and \( d_{yz} \) orbitals is strong momentum-dependent. In BaFe\(_2\)As\(_2\) families, the shift is much larger at momentum near \( Q_1 = (\pi, 0) \) and \( Q_2 = (0, \pi) \) than that near \( \Gamma \) point \( k = (0, 0) \). This strong momentum-dependent band shift has been confirmed explicitly by new ARPES data. Obviously, it can not be accounted for with an on-site orbital order \( O_s \). This special band shift can be interpreted naturally with an bond order wave orbital nematic order \( O_d \), which is largest at momentum \( Q_{1,2} \) but is zero at \( \Gamma \) point. These bond orbital orders are very similar to the proposed bond charge-density-wave order parameter for cuprate superconductors. Compared to the on-site order, the bond order can avoid the energy enhancement from strong on-site Hubbard interaction, thus makes the system in a more stable and lower energy state.

In this manuscript, we further study the physical consequences of the orbital nematic order. Recently inelastic neutron scattering shows strong nematic spin correlations in FeSCs, with spin fluctuation at antiferromagnetic (AFM) momentum \( Q_1 \) much larger than that at another AFM momentum \( Q_2 \). Moreover, the integrated strength of the AFM spin fluctuations is enhanced sharply across the nematic phase transition. In this manuscript, we show that a finite orbital nematic order can drive a significant spin response nematicity and can enhance the integrated intensity of spin fluctuations. As our previous study shows that the orbital nematic order can enhance the condensation energy of magnetic ordering state, all our results show that there is strong cooperative interplay between the orbital nematic order and the spin correlations in FeSCs, and the orbital order can not be simply taken as a secondary effect caused by the symmetry breaking from spin nematicity.

Our manuscript is arranged as following. In Sec. II we specify the model Hamiltonian for FeSCs. In Sec. III we study the spin correlations with finite orbital nematic order. Sec. IV shows our discussion and conclusion.

**II. MODEL HAMILTONIAN**

The Hamiltonian for FeSCs in our study is defined as

\[ H = H_0 + H_I, \]

where \( H_0 \) is the non-interacting part and \( H_I \) describes the multi-orbital Hubbard interaction. There are two subparts in \( H_0 \). \( H_0 = H_s + H_O \). \( H_s \) describes the kinetic energy of 5-orbital 3d electrons and use the notations of Kuroki et al:\( ^{37} \)

\[ H_I = \sum_{ia \sigma} t_{ia \sigma} d_{ia \sigma}^\dagger d_{ia \sigma}, \]

where \( d_{ia \sigma} \) and \( d_{ia \sigma}^\dagger \) are the annihilation and creation operators respectively for electron at site \( i \) in orbital \( a \) with spin \( \sigma \). \( H_O \) describes the physics of orbital-relevant nematic order at mean-field,

\[ H_O = -\Delta \ O_\eta, \]

where \( O_\eta \) is defined in Eq. (2). The temperature-dependence of the orbital order parameter is given by \( \Delta = \Delta_0 (1 - T/T_c)^{1/2} \). In our study, We set 100meV as energy unit, and set \( \Delta_0 = 0.33 \) (~ 33meV) following ARPES data of Yi et al and \( T_c = 0.13 \) (~ 140K). \( k_B = \hbar = e = a = 1, \) where \( a \) is in-plane lattice constant in tetragonal structure.

The multi-orbital Hubbard Hamiltonian \( H_I \) is defined as usual,

\[ H_I = U \sum_{ia} n_{ia} n_{ia \uparrow} + \left( U' - J \right) \sum_{ia < ib} n_{ia} \Delta n_{ib} \]

where \( \Delta n_{ia} = (\cos k_x - \cos k_y)/2 \) and \( \Delta n_{ib} = (\cos k_x + \cos k_y)/2 \).

\[ -2J \sum_{ia < ib} (S_{ia} \cdot S_{ib} + J' \sum_{ia < ib} (d_{ia \uparrow}^\dagger d_{ib \uparrow}^\dagger d_{ib \uparrow} d_{ia \uparrow}^\dagger + h.c.)). \]

**III. SPIN SUSCEPTIBILITY**

In this section, we will study the influence of the orbital order on the spin susceptibility. The orbital order is defined in Eq. (2). Since the inelastic neutron spin scattering cross section is determined by the dynamical spin structure factor, our task is to calculate the spin correlation function with a finite orbital order.

Introduce a generalized multi-orbital spin operator

\[ S_{a_1 a_2} = \frac{1}{\sqrt{N}} \sum_{k \mu \mu_2} d_{k+q a_1 \mu_1}^\dagger (\sigma/2)_{\mu_1 \mu_2} d_{k a_2 \mu_2}, \]

with \( \sigma \) being the Pauli matrix, the spin (transverse) susceptibility is defined as

\[ \chi^{(+ -)}_{a_1 a_2, a_3 a_4}(q, \tau) = \langle T_{\tau} \chi^{+} \chi^{(-)} \rangle \]

The bare spin susceptibility \( \chi^{(+ -)}(q) \) with \( q = (q, i\nu_n) \) can be obtained as

\[ \chi^{(+ -)}(q) = \frac{1}{N} \sum_{k \mu_1 \mu_2} C_{\mu_1 \mu_2}^n(k, q) F_{\mu_1 \mu_2}^n(i\nu_n, k, q), \]

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The bare spin susceptibility \( \chi^{(+ -)}(q) \) with \( q = (q, i\nu_n) \) can be obtained as

\[ \chi^{(+ -)}(q) = \frac{1}{N} \sum_{k \mu_1 \mu_2} C_{\mu_1 \mu_2}^n(k, q) F_{\mu_1 \mu_2}^n(i\nu_n, k, q), \]
where \( C_{\mu\nu}^{\Lambda}(k, q) \) describes the vertex part of the spin correlation function and is determined by the orbital character of the electronic state as

\[
C_{\mu\nu}^{\Lambda}(k, q) = U_{\alpha\mu}^\dagger(k_{j1}) \frac{\sigma_{\mu\nu}^{+}}{2} U_{\alpha\mu}(k + q, \mu_2)
\]

\[
U_{\alpha\mu}(k + q, \mu_2) = \frac{\sigma_{\mu\nu}^{+}}{2} U_{\alpha\mu}(k_{j1}). \tag{8}
\]

Function \( F_{\mu_1\mu_2}^{\Lambda}(iv_n, k, q) \) in Eq. (4) is determined by the electronic band structure near Fermi energy as

\[
F_{\mu_1\mu_2}^{\Lambda}(iv_n, k, q) = \frac{f(E_{k+q\mu_2}) - f(E_{q\mu_1})}{iv_n - E_{k+q\mu_2} + E_{k\mu_1}}. \tag{9}
\]

Here \( E_{k\mu_2} \) is the m-th band energy with momentum \( k \), and \( U_{\alpha\mu}(k\mu) \) is the unitary transformation matrix to diagonalize Hamiltonian \( H_0 \). \( f(x) \) is the Fermi-distribution function. For simplicity, we will focus on the normal state without magnetic and superconducting long-range order. In this case, the unitary matrix \( U_{\alpha\mu}(k\mu) \) is spin-independent.

Because the interaction in the system is moderate and the nematic phase transition is near magnetic instability, we will include the random phase approximation (RPA) effect in spin susceptibility. The spin susceptibility from RPA enhancement due to the multi-orbital Hubbard interaction \( H_I \) can be expressed as

\[
\chi_{(\mathbf{q})}^{(+\omega)}(q) = \left(1 - \chi^{(+\omega)}(q)\hat{V}\right)^{-1} \chi^{(+\omega)}(q), \tag{10}
\]

where the interaction matrix \( \hat{V} \) is defined as

\[
\hat{V}_{\alpha_1\alpha_2\alpha_3\alpha_4} = \begin{cases} 
U, & \text{if } a_1 = a_2 = a_3 = a_4, \\
U', & \text{if } a_1 = a_3 \neq a_2 = a_4, \\
J, & \text{if } a_1 = a_2 = a_3 \neq a_4, \\
J', & \text{if } a_1 = a_4 \neq a_2 = a_3.
\end{cases}
\]

As the inelastic neutron scattering cross section is determined by the dynamical spin correlation function \( \langle S_{\alpha_1\alpha_2}(\mathbf{q}, t) S_{\alpha_3\alpha_4}(\mathbf{q}, 0) \rangle \) with \( \alpha = x, y, z \), the neutron spin response is determined by

\[
\chi(q) = \frac{1}{2} \sum_{\alpha_1\alpha_2} \chi_{\alpha_1\alpha_2\alpha_3\alpha_4}^{(+\omega)}(q). \tag{11}
\]

Here we have used the fact that \( \chi_{xx}(q) = \chi_{yy}(q) = \chi_{zz}(q) = \chi(q) \) in the state without magnetic long-range order. In our study, the parameters for Hubbard interaction are chosen not far away from a magnetic instability, and according to Kuroki et al., we set \( U = 0.96eV, U' = 0.66eV, J = J' = 0.12eV \). The rotation from imaginary frequency to real one is defined as \( iv_n \to \omega + i\delta \tau \) with \( \delta \tau = 0.05 \).

In Fig. 1 we plot the temperature-dependent spin susceptibility at \( Q_1 \) and \( Q_2 \) with \( d- \), \( s'- \) and \( s- \) orbital nematic order. Three cases are considered with frequency, \( \omega = 6meV, 15meV \) and \( 19meV \), respectively. Clearly, below critical temperature \( T_c \) with a finite orbital order, the spin fluctuation spectrum breaks the tetragonal symmetry. While the spin susceptibility at \( Q_1 \) increases with decreasing temperature, the susceptibility at \( Q_2 \) decreases with temperature. Thus the spin fluctuation spectrum breaks the discrete rotational symmetry and shows spin fluctuation nematicity.

Physically, a finite orbital nematic order would lift the degeneracy between \( d_{xz} \) and \( d_{yz} \) electronic states. The lifting of orbital degeneracy shows itself in spin susceptibility from two different ways, through function \( C_{\mu_1\mu_2}^{\Lambda} \) and through function \( F_{\mu_1\mu_2}^{\Lambda} \). To see how these two factors play role in the nematicity of spin susceptibility, we calculate the spin susceptibility in two further cases. In the first case, we call it for vertex effect, we neglect the effect of the orbital order on function \( F_{\mu_1\mu_2}^{\Lambda} \) and set orbital order zero in it. In this case, the orbital order plays role through the unitary matrix \( U \) which transforms the spin matrix from the orbital representation into the band one. In the other case, we call it for band effect, we set orbital order zero in function \( C_{\mu_1\mu_2}^{\Lambda} \). In this case, the role of the orbital order is to modify the nesting condition through the electronic band structure in function \( F_{\mu_1\mu_2}^{\Lambda} \).
The form factor with s'-wave orbital order has its largest value at momentum $\mathbf{Q}_{1,2}$ and that with $s'$-wave symmetry has largest value at $\Gamma$ and $(\pi, \pi)$. Because most weights of the low-energy $d_{zx}$ and $d_{yz}$ orbitals are near $\Gamma$ and $\mathbf{Q}_{1,2}$, the $d$-wave orbital order will lead to a more notable modification of Fermi surface nesting in FeSCs. It thus leads to a larger band effect than $s$- and $s'$-wave orbital order.

In Fig. 4 and Fig. 2 we also plot the real part of the spin susceptibility. The orbital order leads to a similar nematic behavior in real part of spin susceptibility. Fig. 3 shows the frequency dependence of spin fluctuations. In the range $\omega \in (0, 0.2)$, the nematicity of the imaginary part of spin susceptibility increases with frequency, while that of the real part of spin susceptibility decreases. At each frequency, the $s'$-wave orbital order has a weaker effect on spin fluctuation nematicity than $d$- and $s$-wave orbital orders.

Recently, neutron scattering measurement shows strong effect of the electronic nematicity on the spin fluctuations in twinned FeSCs. When temperature decreases below the nematic critical temperature $T_c$ and the system enters into nematic state without magnetic long-range order, the spin dynamic response at AFM momentum $\mathbf{Q}_{AF}$ shows a strong increase and exhibits a maximum at lower AFM transition temperature. It shows an enhancement effect of the electronic nematicity on the spin dynamic fluctuations.

In Fig. 4 we compare the imaginary part of the spin susceptibility with and without orbital nematic order. Since in twinned FeSCs, the neutron scattering spectra can not distinguish the AFM momenta $\mathbf{Q}_1$ and $\mathbf{Q}_2$, we...
define the AFM spin susceptibility as

\[ \chi_{RPA}(Q, \omega) = \frac{1}{2} (\chi_{RPA}(Q_1, \omega) + \chi_{RPA}(Q_2, \omega)). \]

It shows clearly that the orbital order with \(d\)- or \(s\)-wave form factor enhances the AFM spin fluctuations, which is consistent to the experiment observation. As a comparison, the \(s'\)-wave orbital order has a much weak effect on the integrated AFM spin fluctuations. This result rules out the \(s'\)-wave orbital order in LaFeAsO and Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) families.

**IV. DISCUSSION AND CONCLUSION**

In this manuscript, we study the influences of the orbital nematic order on the neutron spin responses. It shows that orbital nematic order can readily lead to nematicity of spin fluctuations and can enhance the total AFM fluctuation strength as well.

Though the orbital order has strong influence on the spin correlations, it should not be taken as proof that the orbital nematic order is the principle driving force for the electronic nematicity in FeSCs. One reason is that the spin nematic order can occur even without orbital degree of freedom involved\(^{23-31}\). Another reason is that the microscopic mechanism for the orbital order is still elusive. As the orbital order in the nematic state does not lead to finite gap in the electronic structure, it can not be protected by gap opening. Its energy-gain mechanism may involve the interaction in spin channel.

Whether or not the orbital nematic order is a primary driving force for the electronic nematicity, our study shows that the orbital order has a strong feedback on the spin fluctuations, both in its nematicity and its strength. Moreover, in our previous study, we show that the orbital nematic order can also enhance significantly the condensation energy of magnetic ordering state\(^{25}\). Therefore, our study shows that there is a cooperative interplay between the orbital nematic order and the spin correlations in FeSCs. This conclusion also indicates that the orbital nematic order may play important roles in magnetism and superconductivity in FeSCs.

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