Detecting the orbital character of the spin fluctuation in the iron-based superconductors with the resonant inelastic X-ray scattering spectroscopy

DA-WEI YAO and TAO LI

Department of Physics, Renmin University of China - Beijing 100872, PRC

received 16 August 2018; accepted in final form 8 February 2019
published online 11 March 2019

PACS 74.70.Xa – Pnictides and chalcogenides
PACS 75.25.Dk – Orbital, charge, and other orders, including coupling of these orders
PACS 78.70.Ck – X-ray scattering

Abstract – The orbital distribution of the spin fluctuation in the iron-based superconductors (IBSs) is the key information needed to understand the magnetism, superconductivity and electronic nematicity in these multi-orbital systems. In this work, we propose that the resonant inelastic X-ray scattering (RIXS) technique can be used to probe selectively the spin fluctuation on different Fe 3d orbitals. In particular, the spin fluctuation on the three t2g orbitals, namely, the 3d_{xz}, 3d_{yz} and the 3d_{xy} orbital, can be selectively probed in the σ → π′ scattering geometry by aligning the direction of the outgoing photon in the y-, x- and z-direction. Such orbital-resolved information on the spin fluctuation is invaluable for the study of the orbital-selective physics in the IBSs and can greatly advance our understanding of the relation between orbital ordering and spin nematicity in the IBSs and the orbital-selective pairing mechanism in these multi-orbital systems.

The multi-orbital nature of the iron-based superconductors (IBSs) is the most important origin for the novel properties of this new family of unconventional superconductors. Mounting evidences have been accumulated through the years for the importance of the orbital degree of freedom in the magnetism, superconductivity and the electronic nematicity of the system. For example, electrons in different Fe 3d orbitals may experience different strengths of electron correlation and a two-component picture with both itinerant electron and local moment may be necessary to understand the magnetism of the IBSs [1–5]. Indeed, recent ARPES measurements have found a great difference in the mass renormalization factor for electrons in the five Fe 3d orbitals. Such a phenomenon is generally termed as orbital-selective Mottness (OSMT). At the same time, electrons in different Fe 3d orbitals may prefer different magnetic correlation patterns [6–10]. More specifically, electrons in the 3d_{yz} and 3d_{xz} orbital prefer magnetic order with a wave vector Q = (π, 0) and Q′ = (0, π), respectively, as a result of their different nesting condition on the Fermi surface. Such a phenomenon is called orbital-selective spin fluctuation (OSSF) by Fanfarillo et al. [11–14]. The study of the origin and nature of the OSMT and OSSF phenomena is the key to understand the relation between orbital ordering and spin nematicity in the IBSs and the orbital selective pairing mechanism in these multi-orbital systems [15–21].

With such rich expectations on the orbital-selective physics in the magnetism of the IBSs, it is quite embarrassing to find that there is almost no efficient way to probe the orbital character of the spin fluctuation in the IBSs. In principle, the orbital character of the magnetic fluctuation can be inferred from the atomic form factor in the inelastic neutron scattering measurement. However, the difference in the atomic form factor of the Fe 3d orbitals is rather small, since the atomic form factor is determined solely by the electron density distribution, rather than the wave function of the electron orbital.

In this paper, we propose that the resonant inelastic X-ray scattering (RIXS) technique can be used to probe the orbital character of the spin fluctuation in the IBSs. We find that one can couple selectively to the spin fluctuation on a given Fe 3d orbital by choosing properly the polarization of the incident and outgoing photon. We have derived the explicit form for the RIXS selection rule and suggested the relevant scattering geometry for studying the OSMT and OSSF physics in the IBSs. The
The general theory of direct RIXS process is now well established [25] and has been summarized in the excellent review article [22] by Ament et al. Here we will adopt the notations of this review. Using the second-order perturbation theory, the transition amplitude induced by the photon scattering can be expressed as [22]

$$F_{f,g} = \langle f | \mathcal{D}^\dagger_{\text{out}} G(z) \mathcal{D}_{\text{in}} | g \rangle,$$

(1)

Here $|g\rangle$ and $|f\rangle$ are the initial and final state of the system. $G(z) = \sum_n \frac{|n\rangle\langle n|}{z - E_n}$ is the propagator for the intermediate state after the first photon excitation. $|n\rangle$ is the intermediate state with a core hole, $z = E_g + \hbar \omega + i \Gamma_n$. $E_g$ and $E_n$ are the energies of the ground state and the intermediate state, $\Gamma_n$ is the inverse lifetime of the intermediate state caused by other nonradiative atomic processes. $\hbar \omega$ is the energy of the incident photon. $\mathcal{D} = \frac{1}{\hbar \omega} \sum_n e^{i k \cdot r_i} \epsilon \cdot p_i$ is the transition operator corresponding to the nonmagnetic coupling between the photon and the electron (this is just the usual $\mathbf{A} \cdot \mathbf{p}$ term). Here $\epsilon$ is the polarization vector of the incident or outgoing photon. $r_i$ and $p_i$ are the position and momentum operator of the $i$-th electron.

The expression of the transition amplitude $F_{f,g}$ can be simplified by the following two approximations. Firstly, since the radius of the Fe 3d orbital is much smaller than the distance between neighboring Fe ions in the IBSs, the dipole approximation is applicable [22] and we can approximate the plane-wave factor $e^{i k \cdot r_i}$ in the transition operator $\mathcal{D}$ by a constant $e^{i k \cdot R_i}$, in which $R_i$ is the position of the atomic site in which the $i$-th electron resides. Under the dipole approximation, the RIXS transition amplitude becomes

$$F_{f,g} = \langle f | (\epsilon_{\text{out}} \cdot \mathcal{D}^\dagger_{\text{out}}) G(z) (\epsilon_{\text{in}} \cdot \mathcal{D}_{\text{in}}) | g \rangle,$$

(2)

in which $\mathcal{D}_k = \sum_i e^{i k \cdot R_i}$ is the dipole operator at the photon wave vector $k$. Here we have used the operator identity $p_i = \frac{m_i}{2 \hbar} \mathbf{r}_i + V(r_i), r_i = \frac{m_i}{2 \hbar} [H_i, r_i]$ to rewrite the matrix element of $\mathcal{D}$ as

$$\langle n | \mathcal{D} | g \rangle = \frac{E_n - E_g}{\hbar \omega} \left\langle n \left| \sum_i e^{i k \cdot R_i} \epsilon \cdot r_i \right| g \right\rangle,$$

and then used the resonance condition $\hbar \omega = E_n - E_g$. As a second approximation, we neglect the interaction effect in the intermediate state and treat the denominator of $G(z)$, namely, $z - E_n$, as a constant. This is the so-called fast-collision approximation [26,27], which is valid for the direct RIXS process studied in this paper\footnote{For indirect RIXS processes, the coupling between the valence electron and the core hole can have important consequences.}. Under the fast-collision approximation, $G(z)$ of the Fe $L_3$-edge RIXS becomes

$$G(z) \approx \frac{1}{\pi} \frac{1}{2p_\mathbf{k}} \sum_{i,m_j} \left| \frac{3}{2}, m_j \right\rangle \left\langle i, \frac{3}{2}, m_j \right|.$$

Here $|i, \frac{3}{2}, m_j\rangle$ denotes the state in which the $m_j$-th component of the Fe 2$p^\uparrow$ multiplet is created by the photon excitation at site $i$ in the IBSs with the RIXS technique.

---

**Fig. 1:** Illustration of the Fe $L_3$-edge RIXS process studied in this work. The Fe $L_3$-edge RIXS process is a direct RIXS process and is composed of the following two steps: (a) the incident photon with momentum $k_{\text{in}}$ excites a Fe 2$p^\uparrow$ core electron into the Fe 3$d$ shell; (b) the Fe 2$p^\uparrow$ core hole left behind by the photon excitation is annihilated by another electron in the Fe 3$d$ shell, accompanied by the emission of a scattered photon with momentum $k_{\text{out}}$. The RIXS process effectively creates a particle-hole pair in the Fe 3$d$ shell.
Under these two approximations, one finds that
\[ F_{j,g} \propto \left< f \right| \sum_i e^{i q R_i} T_i \left| g \right>, \] (3)
in which \( q = k_{in} - k_{out} \) is the momentum transfer from the scattered photon. \( T_i \) is a transition operator acting on the Fe 3d electrons on site \( i \) and is given by
\[ T_i = \sum_{m_j} r_{out} \left| i, \frac{3}{2}, m_j \right> \left< i, \frac{3}{2}, m_j \right| r_{in}, \] (4)
in which \( r_{in} = \epsilon_{r} \cdot \mathbf{r} \) and \( r_{out} = \epsilon_{out} \cdot \mathbf{r} \). As a result of the spin-orbital coupling in the 2p core level, \( T_i \) can induce both spin conserving and spin flip transitions in the Fe 3d shell. Completing the summation over \( m_j \), we find that \( T_i \) can be written as the following second quantized form:
\[ T_i = \sum_{\mu, \nu, \sigma} n_{\mu, \nu, \sigma} \hat{c}^\dagger_{i, \mu, \sigma} \hat{c}_{i, \nu, \sigma} + \sum_{\mu, \nu, \sigma} s_{\mu, \nu, \sigma} \hat{c}^\dagger_{i, \mu, \sigma} \hat{c}_{i, \nu, \sigma} + \sum_{\mu, \nu} s_{\mu, \nu} \hat{c}^\dagger_{i, \mu, \downarrow} \hat{c}_{i, \nu, \downarrow} + \sum_{\mu, \nu} s_{\mu, \nu} \hat{c}^\dagger_{i, \mu, \uparrow} \hat{c}_{i, \nu, \uparrow}. \] (5)

Here \( \mu, \nu = 1, \ldots, 5 \) is the index of the Fe 3d orbitals. We will use the following convention for the 3d orbitals, namely, \( |1\rangle = |xz\rangle \), \( |2\rangle = |yz\rangle \) and \( |3\rangle = |xy\rangle \) for the three orbitals in the \( t_{2g} \) subspace, \( |4\rangle = |3z^2 - r^2\rangle \) and \( |5\rangle = |x^2 - y^2\rangle \) for the two orbitals in the \( e_g \) subspace. \( \sigma = \uparrow, \downarrow \) is the spin of the valence electron. \( \hat{c}_{i, \mu, \sigma} \) is the annihilation operator for a spin-\( \sigma \) electron on the \( \mu \)-th orbital of site \( i \).

The matrix elements in eq. (5) are given by
\[ n_{\mu, \nu} = \frac{2}{3} \sum_m \langle \mu | r_{in} | m \rangle \langle m | r_{out} | \nu \rangle, \]
\[ s_{\mu, \nu}^z = \frac{1}{3} \sum_m \langle \mu | r_{in} | m \rangle \langle m | r_{out} | \nu \rangle, \]
\[ s_{\mu, \nu}^x = \frac{1}{3} \sum_m A_m \langle \mu | r_{in} | m \rangle \langle m + 1 | r_{out} | \nu \rangle, \]
\[ s_{\mu, \nu}^y = \frac{4}{3} \sum_m A_m \langle \mu | r_{in} | m \rangle \langle m + 1 | r_{out} | \nu \rangle, \]
in which \( |m\rangle \) is the eigenstate of \( l_z \) with eigenvalue \( m \), \( A_m = \sqrt{2} - m(m + 1) \).

To simplify the analysis of the selection rule in the tetragonal environment, we expand the 2p core level \( |m\rangle \) in the basis of real harmonics with the \( p_x, p_y \) and \( p_z \) character as
\[ |+1\rangle = -\frac{1}{\sqrt{2}}(|x\rangle + i|y\rangle), \]
\[ |0\rangle = |z\rangle, \]
\[ |-1\rangle = \frac{1}{\sqrt{2}}(|x\rangle - i|y\rangle). \] (7)

One then finds, that the RIXS matrix elements become
\[ n_{\mu, \nu} = \frac{2}{3} \sum_{\alpha} \langle \mu | r_{in} | \alpha \rangle \langle \alpha | r_{out} | \nu \rangle, \]
\[ s_{\mu, \nu}^z = \frac{1}{3} \sum_{\beta, \gamma} \epsilon_{\alpha \beta \gamma} \langle \mu | r_{in} | \beta \rangle \langle \gamma | r_{out} | \nu \rangle. \] (8)

Here \( \alpha = x, y, z \), \( \epsilon_{\alpha \beta \gamma} \) is the anti-symmetric tensor.

With these matrix elements in hand, we can calculate the RIXS cross-section of the Fe 3d excitation in both the density and the spin channel. In the paramagnetic phase, in which the spin is a conserved quantity, they are given by
\[ I_{\text{RIXS}}^{\text{density}}(q, \omega) = -\text{Im} \left\{ \sum_{\mu, \nu, \mu', \nu'} n_{\mu, \nu} n_{\mu', \nu'} D_{\mu \nu, \mu' \nu'}(q, \omega) \right\}, \]
\[ I_{\text{RIXS}}^{\text{spin}}(q, \omega) = -\text{Im} \left\{ \sum_{\mu, \nu, \mu', \nu'} \langle s_{\mu, \nu}^z \rangle_{\mu', \nu'} \chi_{\mu \nu, \mu' \nu'}(q, \omega) \right\}. \] (9)

Here \( D \) and \( \chi \) are the density and the spin correlation function defined as follows:
\[ D_{\mu \nu, \mu' \nu'}(q, \tau) = \langle T_{\tau} n_{\mu, \nu} n_{\mu', \nu'}(-q, 0) \rangle, \]
\[ \chi_{\mu \nu, \mu' \nu'}(q, \tau) = \langle T_{\tau} s_{\mu, \nu}^z n_{\mu', \nu'}(-q, 0) \rangle, \] (10)
in which \( n_{\mu, \nu} = \sum_{\sigma} c_{i, \mu, \sigma}^\dagger c_{i, \nu, \sigma} \) is the density operator on site \( i \), \( s_{\mu, \nu}^z = \frac{1}{2} \sum_{\gamma, \gamma'} c_{i, \mu, \gamma}^\dagger c_{i, \nu, \gamma'} \sigma_{\gamma \gamma'}^z \) is the spin density operator in the \( \alpha \)-direction on site \( i \). In the magnetic ordered phase with a collinear AF order along the \( z \)-axis, the density fluctuation is entangled with the longitudinal spin fluctuation. One should then defined separately the density operator for the up-spin and the down-spin component, whose fluctuations are entangled [24].

To see more clearly the selection rule in the RIXS process, we split \( s_{\mu, \nu}^z \) into its Hermitian and anti-Hermitian part, which correspond to the spin density and spin current excitation in the orbital space. In this study, we will focus on the spin density excitation, since the spin current excitation is in general a manifestation of the itinerant nature of the electron motion and only contribute a weak continuum in the spectrum. It is easy to show that the Hermitian part of \( s_{\mu, \nu}^z \) is asymmetric with respect to the exchange between \( r_{in} \) and \( r_{out} \) and thus proportional to \( \epsilon_{in} \times \epsilon_{out} \). As a comparison, we note that the Hermitian of \( n_{\mu, \nu} \) is symmetric with respect to the exchange between \( r_{in} \) and \( r_{out} \).

Such a difference in the symmetry property of the spin and density excitation can be used to separate them in the RIXS measurement. To detect the spin excitation, one can employ the \( \sigma \rightarrow \pi' \) scattering geometry (see fig. 2). Here \( \sigma \) denotes that the polarization of the incident photon is perpendicular to the scattering plane, \( \pi' \) denotes that the polarization of the scattered photon is within the scattering plane. Such a scattering geometry has already been used in a recent RIXS study on an electron-doped cuprate superconductor [28]. On the other hand, to detect the
Fig. 2: The RIXS scattering geometries proposed to detect the spin fluctuation on the (a) 3d\textsubscript{xz}, (b) 3d\textsubscript{yz} and the (c) 3d\textsubscript{xy} orbital and the corresponding RIXS matrix elements. Here we have adopted the $\sigma \rightarrow \pi'$ scattering geometry to maximize the matrix element of spin excitation. $k_{\text{out}}$ is aligned with the y-, x- and z-axis in (a), (b) and (c). The scattering plane has been set to be the x-y, x-y and the y-z plane in (a), (b) and (c) for clarity and can be rotated freely around $k_{\text{out}}$. $q||$ is the momentum transfer in the x-y plane and $c$ is a constant. The spin fluctuation excited in (a), (b) and (c) is along the y-, x- and z-direction in the spin space. $3z^2 - r^2 \leftrightarrow x^2 - y^2$ denotes the mixed $3z^2 - r^2$ and $x^2 - y^2$ orbital character.

Before discussing the application of these results to the study of the IBSs, we note one limitation of the Fe L\textsubscript{3}-edge RIXS. Since the largest momentum transfer at the Fe L\textsubscript{3}-edge (with a photon energy of about 710 eV) is only about $\pi/2$ (see footnote \textsuperscript{2}), the Fe L\textsubscript{3}-edge RIXS can only cover one-quarter of the first Brillouin zone in the IBSs. In particular, the most important momentum for the stripy magnetic correlation in the IBSs, namely, $Q = (\pi,0)$ or $Q' = (0,\pi)$, is out of the reach of the Fe L\textsubscript{3}-edge RIXS. In the magnetic ordered phase, Q is folded back to the $\Gamma$-point so that the spin fluctuation around Q becomes accessible to the Fe L\textsubscript{3}-edge RIXS measurement. At the same time, local moment fluctuation in the paramagnetic phase is expected to be robust even for momentum far away from the underlying magnetic ordering wave vector. The Fe L\textsubscript{3}-edge RIXS can thus be used to study the orbital character of the spin fluctuation in both the magnetic ordered phase and the paramagnetic phase.

In a previous study \cite{24}, the spin fluctuation in both the magnetic ordered phase and the paramagnetic phase of the IBSs are computed with RPA for a five-band itinerant model. The result is then used to compute the RIXS cross-section at the Fe L\textsubscript{3}-edge. It is claimed that the long-wavelength spin wave excitation in the magnetic ordered phase has dominant 3d\textsubscript{xy} character. On the other hand, it is found that no well-defined dispersive mode can be resolved around the $\Gamma$-point in the paramagnetic phase from such a calculation. However, it is well known that the RPA scheme is unsuitable for the description of the local moment physics in the paramagnetic phase. In particular, it predicts that the spin fluctuation spectrum around the $\Gamma$-point in the paramagnetic phase should be essentially unchanged by the RPA correction and be composed of particle-hole continuum. This has been found to be incorrect by recent experiment on heavily doped IBSs, in which a robust dispersive magnon mode has been observed around the $\Gamma$-point in the paramagnetic phase \cite{23}. At the same time, the power of polarization resolution of the RIXS technique has not been fully explored in this early study, in which only the polarization of the incident photon is specified. This greatly limits the possibility of resolving the orbital character of the spin fluctuation in the IBSs with the RIXS technique.

Here we first consider the spin wave excitation of the magnetic ordered phase of the IBSs. As a symmetry restoration mode in the spin space, the long-wavelength spin wave excitation is expected to inherit the orbital character of the ordered moment. In a multi-orbital system, the magnetic order parameter is in general a matrix in the orbital space, with its matrix element defined by $M_{\mu,\nu} = \frac{1}{v} \sum_{\sigma} \sigma (c_{i,\mu,\sigma} c_{i,\nu,\sigma})$. The orbital character of the ordered moment is encoded in the eigenvalues and density difference between different Fe 3d orbitals, for example, in the electronic nematic phase of the IBSs, one can employ the $\sigma \rightarrow \sigma'$ scattering geometry.

$3a \approx 4 \, \text{Å}$ is the lattice constant of the IBSs.
eigenvectors of this matrix. In the IBSs, the lattice symmetry around the Fe ions is broken to $D_3$ in the magnetic ordered phase and the symmetry allowed magnetic order parameter can only take the form of

$$M = \begin{pmatrix}
M_{11} & 0 & 0 & 0 \\
0 & M_{22} & 0 & 0 \\
0 & 0 & M_{33} & 0 \\
0 & 0 & 0 & M_{44} \\
M_{45} & M_{45} & M_{45} & M_{45}
\end{pmatrix}. \quad (11)$$

Thus, the ordered moment has either pure $t_{2g}$ or pure $e_g$ character. Previous studies indicate that the magnetic moment in the IBSs is mainly contributed by the $t_{2g}$ orbitals [4,7]. At the same time, since both the OSMT and the OSSF happen in the $t_{2g}$ subspace, it is also the most interesting place to study the orbital character of the spin fluctuation in the IBSs. We will thus focus on the spin fluctuation in the $t_{2g}$ subspace in the following discussion.

The selection rule for the Fe $L_3$-edge RIXS process becomes extremely simple in the $t_{2g}$ subspace. If we approximate the Wannier orbitals with the corresponding atomic orbitals$^3$, one finds that the spin transition matrix element within the $t_{2g}$ subspace is given by

$$s_{\mu,\nu}^\alpha = \frac{c}{4}[(e_{\mu} \cdot (\epsilon_{in} \times \epsilon_{out}))(e_{\nu} \cdot e_\alpha) + \mu \leftrightarrow \nu](1 - 2\delta_{\mu \nu}). \quad (12)$$

Here $c$ is a constant. $e_\mu$ is a unit vector and is defined as $e_{xz} = e_y$, $e_{yz} = e_x$ and $e_{xy} = e_z$. $\epsilon_{in}$ and $\epsilon_{out}$ are the polarization vectors of the incident and outgoing photon. For completeness’ sake, the spin transition matrix element within the $e_g$ subspace is given by

$$s_{\mu,\nu}^\alpha = -\frac{c}{6}[(e_\alpha \cdot (\epsilon_{in} \times \epsilon_{out}))(1 - 2n_\alpha \cdot \vec{\pi}). \quad (13)$$

Here $\vec{\pi}$ is the Pauli matrix in the $e_g$ subspace, $n_\alpha$ is a unit vector and is defined as $n_\alpha = \cos \phi_\alpha e_z + \sin \phi_\alpha e_x$, with $\phi_x = 0$, $\phi_y = -\frac{\pi}{2}$ and $\phi_\alpha = \frac{\pi}{2}$.

We now discuss how to separate the spin fluctuation in the three Fe $t_{2g}$ orbitals in the RIXS measurement. First, since the RIXS matrix element for spin excitation is proportional to $\epsilon_{in} \times \epsilon_{out}$, the $\sigma \rightarrow \sigma'$ scattering geometry is most favorable for the detection of spin excitations. Second, according to eq. (12), the orbital character of the spin fluctuation excited in the RIXS process is determined by the factor $e_{\nu} \cdot (\epsilon_{in} \times \epsilon_{out})$. However, in the $\sigma \rightarrow \sigma'$ scattering geometry, $\epsilon_{in} \times \epsilon_{out}$ is nothing but the unit vector in the direction of the outgoing photon. Thus, to measure the spin fluctuation in the $\mu$-th orbital, one should align the direction of the outgoing photon with $e_\mu$. The excited spin fluctuation is also in the $e_\mu$-direction in spin space. This is illustrated in fig. 2, in which we have listed the matrix elements for collective spin excitation in all major channels for the IBSs. Except for the uncertainty in the small spin fluctuation weight on the two $e_g$ orbitals, the orbital character of the spin fluctuation in the IBSs can thus be fully resolved by the proposed RIXS scattering geometries.

In the magnetic ordered phase of the IBSs, the ordered moment is found to lie in the FeAs plane and to be perpendicular to the ordering wave vector [29]. Assuming $Q = (\pi, 0)$, the ordered moment is then pointing into the $y$-direction. According to the selection rule listed above, one can either excite the transverse spin fluctuation in the $z$-direction in the $3d_{xy}$ orbital by aligning the direction of the outgoing photon with the $z$-axis, or excite the transverse spin fluctuation in the $x$-direction in the $3d_{yz}$ orbital by aligning the direction of the outgoing photon with the $x$-axis. The transverse spin fluctuation in the $3d_{xz}$ orbital is inaccessible with the Fe $L_3$-edge RIXS. One way to solve this problem is to apply a magnetic field in the $y$-direction so as to rotate the ordered moment to the $x$-direction or $z$-direction. In the paramagnetic phase, RIXS can be used to measure the spin fluctuation in all the three $t_{2g}$ orbitals with the same efficiency.

Such orbital-resolved information on the spin fluctuation can be used to address some important issues of the IBSs. The first issue is about the origin and nature of the OSMT in the IBSs. It has long been anticipated that electrons on different Fe $3d$ orbitals may experience different strengths of electron correlation and that a two-component picture with both itinerant electrons and local moments is needed to understand the magnetism of the IBSs [1,2]. Among the five Fe $3d$ orbitals, it is generally believed that the $3d_{xy}$ orbital is the most strongly correlated [3,4]. Unlike the magnetism of the itinerant electrons, the local moment is expected to be robust against carrier doping and should survive even in the paramagnetic phase. Indeed, recent RIXS measurement do find evidence for the existence of dispersive magnon excitation in the heavily doped IBSs [23], whose dispersion is found to be very similar to the spin wave dispersion of the magnetic ordered phase of the parent compound. A natural question to ask is then if the observed local moment fluctuation in the heavily doped IBSs has indeed a dominant $3d_{xy}$ orbital character. This problem can be answered by the RIXS measurement suggested above.

The second issue is about the origin and nature of the OSSF or the relation between orbital ordering and spin nematicity in the IBSs. Within the itinerant picture, it is straightforward to see that the electrons on the $3d_{xz}$ and the $3d_{yz}$ orbitals prefer different magnetic ordering patterns, since the electronic states around $(\pi, 0)$ and $(0, \pi)$ have dominant $3d_{yz}$ and $3d_{xz}$ character. The orbital order

$^3$The RIXS selection rule for spin excitation in the $t_{2g}$ subspace discussed in the main text is essentially unchanged even if we do not approximate the Wannier orbitals with the corresponding atomic orbitals. In fact, using the fact that the three $t_{2g}$ orbitals and the three $2p$ orbitals each form a distinct one-dimensional representation of the $D_3$ group, it is easy to obtain the following selection rules:

| Orbital | $x_\alpha$ | $y_\alpha$ | $z_\alpha$ | Spin direction | Transition probability |
|---------|------------|------------|------------|-----------------|------------------------|
| $xz$    | $x(z)$    | $x(z)$    | $y$        | $[x(z)x(z)]^2$  |
| $yz$    | $y(z)$    | $z(y)$    | $x$        | $[y(z)y(z)]^2$  |
| $xy$    | $x(y)$    | $y(x)$    | $z$        | $[x(y)x(y)]^2$  |
that breaks the symmetry between the 3d_{xz} and 3d_{yz} orbitals will thus be linearly coupled to the stripy magnetic order with wave vector Q or Q′ and be generated spontaneously in the magnetic ordered phase [8,10–14]. This scenario of OSSF can be verified by comparing the size of the ordered moment on the 3d_{xz} and the 3d_{yz} orbitals in the magnetic ordered phase, which again can be inferred from the orbital character of the long-wavelength spin wave excitation. In principle, the orbital selectivity in the spin fluctuation pattern can also occur in the local moment scenario. To decide if this is the case, one can compare the spin fluctuation spectral weight in the 3d_{xz} and the 3d_{yz} orbital around the Γ-point in the paramagnetic phase, in which case the itinerant spin fluctuation becomes very weak.

Finally, we discuss the general structure of the spin fluctuation in the orbital space of the paramagnetic phase. For this purpose, we express the generalized spin susceptibility in terms of its spectral representation as follows:

\[ \chi_{\mu\nu,\mu'\nu'}(q, \omega_n) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{R_{\mu\nu,\mu'\nu'}(q, \omega')}{i\omega_n - \omega'} , \]  

(14)

Here \( R_{\mu\nu,\mu'\nu'}(q, \omega) \) denotes the matrix elements of the spectral density matrix and is given by

\[ R_{\mu\nu,\mu'\nu'}(q, \omega) = e^{i\Omega} \sum_{m,n} \langle n|s_{\mu\nu}(q)|m\rangle \langle m|s^\dagger_{\mu'\nu'}(-q)|n\rangle \times \frac{(e^{-\beta E_n} - e^{-\beta E_m})2\pi\delta(\omega + E_n - E_m)}{E_n - E_m} , \]  

(15)

in which \( E_n \) denotes the energy of the many-body eigenstate \( |n\rangle \), \( \Omega \) is the grand potential of the system. From this expression, it is straightforward to show that \( R_{\mu\nu,\mu'\nu'}(q, \omega) \) is a positive definite Hermitian matrix with respect to its index \( \mu, \mu' \). The 25 eigenvalues \( \rho^{(n)} \) and eigenvectors \( V^{(n)}_{\mu,\nu} \) of \( R \) can then be interpreted as the spectral weight and the orbital character of the spin fluctuation at momentum \( q \) and frequency \( \omega \). In general, the eigenvector \( V^{(n)}_{\mu,\nu}(q, \omega) \) can have very complex momentum and frequency dependence and is not diagonal in the index \( \mu \) and \( \nu \). More specifically, it is composed of contributions from both the spin density and the spin current excitation in the orbital space. Such complications are manifestations of the itinerant nature of the spin fluctuation in the system, since the orbital character of local moment fluctuations is expected to be much more definite and much less sensitive to \( q \) and \( \omega \). A measurement of the orbital character of the spin fluctuation in the paramagnetic phase can thus provide invaluable information on the electron correlation effect in the IBSs. We note that RIXS can be used to detect both the spin density and the spin current fluctuation in the orbital space.

In summary, we propose that the Fe L3-edge RIXS can be used to detect the orbital character of spin fluctuations in the IBSs. In particular, we find that the spin fluctuations on the three \( t_{2g} \) orbitals, namely, the 3d_{xz}, 3d_{yz} and the 3d_{xy} orbitals, can be selectively probed in the \( \sigma \rightarrow \pi' \) scattering geometry by aligning the direction of the outgoing photon with the y-, x- and z-axis. We show that such orbital-resolved information on the spin fluctuations can be very useful in the study of the OSMT and the OSSF physics in the IBSs and deepens our understanding of the origin of the electronic nematicity and pairing mechanism in the IBSs. The proposed technique can also be used in the study of other transition metal oxides in which the \( t_{2g} \) orbital is dominating the low-energy physics.

**REFERENCES**

[1] Zhang G. M., Su Y. H., Weng Z. Y., Lee D. H. and Xiang T., *EPL*, 86 (2009) 37006.
[2] Kou S. P., Li T. and Weng Z. Y., *EPL*, 88 (2009) 17010.
[3] Yi M., Liu Z. K., Zhang Y., Yu R., Zhu J. X., Lee J., Moore R., Schmitt F., Li W., Riegs S., Chu J. H., Lv B., Hu J., Hashimoto M., Mo S. K., Hussain Z., Mao Z. Q., Chu C. W., Fisher I., Si Q. M., Shen Z. X. and Lu D. H., *Nat. Commun.*, 6 (2015) 7777.
[4] Yu R. and Si Q. M., *Phys. Rev. Lett.*, 110 (2013) 146402.
[5] Medici L., Giovannetti G. and Capone M., *Phys. Rev. Lett.*, 112 (2014) 177001.
[6] Lee C. C., Yin W. G. and Ku W., *Phys. Rev. Lett.*, 103 (2009) 267001.
[7] Daghofer M., Luo Q. L., Yu R., Yao D. X., Moreo A. and Dagotto E., *Phys. Rev. B*, 81 (2010) 180514.
[8] Nevidomskyy A. H., arXiv:1104.1747 (2011).
[9] Su Y. H., Liao H. J. and Li T., *J. Phys. C*, 27 (2015) 105706.
[10] Su Y. H., Zhang C. and Li T., *Phys. Lett. A*, 380 (2008) 2016.
[11] Fanfarillo L., Cortijo A. and Valenzuela B., *Phys. Rev. B*, 91 (2015) 214515.
[12] Fanfarillo L., Benfatto L. and Valenzuela B., *Phys. Rev. B*, 97 (2018) 121109(R).
[13] Fanfarillo L., Mansart J., Toulemonde P., Cerceilier H., Le Fèvre P., Bertran F., Valenzuela B., Benfatto L. and Brouet V., *Phys. Rev. B*, 94 (2016) 155138.
[14] Benfatto L., Valenzuela B. and Fanfarillo L., *npj Quantum Mater.*, 3 (2018) 56.
[15] Baek S. H., Efremov D. V., Ok J. M., Kim J. S., van den Brink J. and Bùchner B., *Nat. Mater.*, 14 (2015) 210.
[16] Luo H. Q., Wang M., Zhang C. L., Lu X. Y., Regnault L. P., Zhang R., Li S. L., Hu J. P. and Dai P. C., *Phys. Rev. Lett.*, 111 (2013) 107006.
Detecting the orbital character of the spin fluctuation in the IBSs with the RIXS spectroscopy

[17] Kuroki K., Onari S., Arita R., Usui H., Tanaka Y., Kontani H. and Aoki H., Phys. Rev. Lett., 101 (2008) 087004.
[18] Ran Y., Wang F., Zhai H., Vishwanath A. and Lee D. H., Phys. Rev. B, 79 (2009) 014505.
[19] Yin Z. P., Haule K. and Kotliar G., Nat. Phys., 10 (2014) 845.
[20] Kreisel A., Andersen B. M., Sprau P. O., Kostin A., Séamus Davis J. C. and Hirschfeld P. J., Phys. Rev. B, 95 (2017) 174504.
[21] Sprau P. O., Kostin A., Kreisel A., Böhmer A. E., Taufour V., Canfield P. C., Mukherjee S., Hirschfeld P. J., Andersen B. M. and Davis J. C. Séamus, Science, 357 (2017) 75.
[22] Ament L. J. P., van Veenendaal M., Devereaux T. P., Hill J. P. and van den Brink J., Rev. Mod. Phys., 83 (2011) 705.
[23] Zhou K. J., Huang Y. B., Monney C., Dai X., Strocov V. N., Wang N. L., Chen Z. G., Zhang C., Dai P., Patthey L., van den Brink J., Ding H. and Schmitt T., Nat. Commun., 4 (2013) 1470.
[24] Kaneshita E., Tsutsui K. and Tohyama T., Phys. Rev. B, 84 (2011) 020511(R).
[25] Ament L. J. P., Ghiringhelli G., Sala M. M., Bracic L. and van den Brink J., Phys. Rev. Lett., 103 (2009) 117003.
[26] Luo J., Trammell G. T. and Hannon J. P., Phys. Rev. Lett., 71 (1993) 287.
[27] van Veenendaal M., Phys. Rev. Lett., 96 (2006) 117404.
[28] da Silva Neto E. H., Minola M., Yu B., Tabis W., Bluschke M., Unruh D., Suzuki H., Li Y., Yu G., Betto D., Kummer K., Yakhou F., Brookes N. B., Letacon M., Greven M., Keimer B. and Damascelli A., Phys. Rev. B, 98 (2018) 161114.
[29] de la Cruz C., Huang Q., Lynn J. W., Li J., Ratcliffe H. W., Zarestky J. L., Mook H. A., Chen G. F., Luo J. L., Wang N. L. and Dai P. C., Nature, 453 (2008) 899.