Dynamical Spin Susceptibility Studied by Inelastic Neutron Scattering on LaFeAsO$_{1-x}$F$_x$

S. Shamoto$^{1,2,*}$, M. Ishikado$^{1,2}$, S. Wakimoto$^{1,2}$, K. Kodama$^{1,2}$, R. Kajimoto$^{2,3}$, M. Arai$^{2,4}$

$^1$Quantum Beam Science Directorate, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
$^2$JST-TRIP, Tokyo 102-0075, Japan
$^3$Research Center for Neutron Science and Technology, Comprehensive Research Organization for Science and Society (CROSS), Tokai, Ibaraki 319-1106, Japan
$^4$J-PARC Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan

E-mail: shamoto.shinichi@jaea.go.jp

Abstract. Low-energy spin excitations on polycrystalline LaFeAsO$_{1-x}$F$_x$ samples have been studied by inelastic neutron scattering. The $Q$-integrated dynamical spin susceptibility $\chi''(\omega)$ at 11 meV decreases with increasing fluoride content $x$ across the phase transition from antiferromagnetic to superconducting phases. For superconducting samples, a peak in $\chi''(\omega)$ develops at $\omega=E_{\text{res}}$ below $T_c$, accompanied by its reduction below $E_{\text{res}}$. Fe $3d$ orbital contribution on the dynamical spin susceptibility and the electron scattering between $\Gamma$ and $M$ Fermi surfaces are discussed.

1. Introduction

Since the discovery of high-$T_c$ superconductivity (SC) in iron-based materials [1], a number of iron-based superconductors have been discovered. These superconductors have similar cylindrical Fermi surfaces at $\Gamma$ and $M$ points as schematically shown in Fig. 1 [2]. Because of the strong nesting condition from hole Fermi surfaces at $\Gamma$ point to electron Fermi surfaces at $M$ point, the Lindhard response function calculated from the band structure has a sharp peak at antiferromagnetic (AF) wave vector $Q_{\text{AF}}=(1/2, 1/2, L)$ [3]. The non-interacting magnetic susceptibility is renormalized by the superexchange interaction $J$. The imaginary part of renormalized dynamical spin susceptibility $\chi''(Q,\omega)$ in itinerant magnetic materials can be probed by inelastic neutron scattering. The dynamical spin susceptibility in one of the optimally doped iron-based superconductors, BaFe$_{1.85}$Co$_{0.15}$As$_2$, has been described as antiferromagnetic metal within an itinerant electron picture [4]. Here, in addition to the dynamical spin susceptibility of LaFeAsO$_{1-x}$F$_x$ measured by inelastic neutron scattering [5,6,7,8], Fe $3d$ orbital contribution on the dynamical spin susceptibility and the electron scattering between $\Gamma$ and $M$ Fermi surfaces are discussed.
2. Experimental methods

High quality polycrystalline LaFeAsO$_{1-x}$F$_x$ samples with $x = 0$, 0.057, 0.082, and 0.158 have been synthesized by solid state reaction. The fluorine concentration, $x$, in the synthesized sample was determined by secondary ion microprobe mass spectrometry measurement. Based on the Meissner signal measured with a superconducting quantum interference device magnetometer, the superconducting transition temperatures ($T_c$) in LaFeAsO$_{1-x}$F$_x$ with $x = 0$, 0.057, 0.082, and 0.158 were determined to be 0, 25, 29, and 7 K, respectively, as shown in Fig. 2. The unit cell volume decreased monotonically with increasing fluorine content $x$. Details of syntheses and characterizations of all samples were reported elsewhere [5]. Inelastic neutron scattering measurements were performed at the following spectrometers, the triple-axis spectrometers TAS-1 at JRR-3 (JAERI), HB-3 at HFIR (ORNL), the chopper spectrometers 4SEASONS at J-PARC, and MERLIN at ISIS (RAL), using large amounts of polycrystalline samples with masses of 25-36 g. Observed scattering intensities were normalized among samples by using (0 0 2) nuclear Bragg peak intensity.

3. Results and Discussion

3.1. Dynamical spin susceptibility and spin resonance mode of LaFeAsO$_{1-x}$F$_x$

In the parent LaFeAsO compound, the spin excitation at $Q_{AF} = 1.2$ Å$^{-1}$ has a long tail to the higher momentum transfers. This is due to the powder averaging of a magnetic rod in reciprocal lattice space [5,9]. In the superconducting compounds, the low-energy spin excitations remain with the similar intensity. However, the peak width in $Q$ becomes broad in the superconducting compounds, due to the shorter AF spin correlation length. The broad peak is approximately fitted by a Gaussian peak.

The non-interacting susceptibility, $\chi(q, \omega)$, is proportional to the number of electron transitions from occupied state to unoccupied state, as follows.

$$\chi_0(q, \omega) = \sum_x \frac{f(\varepsilon_x) - f(\varepsilon_{x+q})}{\varepsilon_x - \varepsilon_{x+q} - \omega - i\delta}$$

(1)

The non-interacting susceptibility is renormalized by the superexchange interaction $J$, leading to the dynamical spin susceptibility as follows.

$$\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - J\chi_0(q, \omega)}$$

(2)
The \( q \)-integrated imaginary part of dynamical spin susceptibility, \( \chi'(q) \), of LaFeAsO\(_{1-x}\)F\(_x\) at 11 meV around \( Q_{\text{SF}} \) in the normal state is shown in Fig. 3. The value at \( x=0.082 \) was scaled from that below \( T_c \) in ref. 6 based on the spin resonance ratio in ref. 7. Volume shrinkage in Fig. 2 affects the \( \chi'(q) \) value. Except for the effect, the decrease of dynamical spin susceptibility with increase of \( x \) can be attributed to two possible reasons. One is the decrease of the number of electron transitions in equation (1). The other is the decrease of the superexchange interaction \( J \) in equation (2). In the case of LaFeAsO\(_{1-x}\)F\(_x\), the electron number increases with increase of doping. Consequently, the hole Fermi surface may disappear, as observed in overdoped Ba(Fe,Co)\(_2\)As\(_2\) [10]. Recent NQR measurement of the similarly overdoped LaFeAsO\(_{1-x}\)F\(_x\) superconductor exhibits a Hebel-Slichter peak in \( 1/T \) below \( T_c \) [11]. This result suggests that the symmetry of superconducting order parameter changes from \( s_\pm \) to normal \( s \) wave with electron doping. These changes correspond to the disappearance of the hole Fermi surface. It should be noted that the normal \( s \) wave SC in this iron-based compound results only in low \( T_c \). Recent inelastic neutron scattering of overdoped Ba(Fe,Co)\(_2\)As\(_2\) also shows the disappearance of dynamical spin susceptibility at nearly the same electron concentration (\( x=0.15 \)) [12] where angle resolved photoemission spectroscopy (ARPES) shows the disappearance of hole Fermi surface [10]. Therefore, the decrease of dynamical spin susceptibility is consistent with the decrease of density of states at the hole Fermi surface, leading to no electron transition at low energies from occupied state to unoccupied state at the momentum transfer \( Q_{\text{SF}} \) in equation (1).

In optimally doped LaFeAsO\(_{0.92}\)F\(_{0.08}\) an enhancement in \( \chi'(Q_{\text{SF}},q) \) has been observed below \( T_c \) [6,7]. The observed peak energy of \( E_{\text{res}} \sim 13 \) meV corresponds to \( 5.2 \) \( k_B \)\( T_c \). This intensity enhancement is attributed to spin resonance mode [3,13]. The peak appears as a result of coherence in superconducting state. Because of the time reversal symmetry of spin scattering between two Fermi surfaces at \( \Gamma \) and \( M \), dynamical spin susceptibility exhibits a peak when superconducting order parameters in two Fermi surfaces has a sign reversal as in the \( s_\pm \) pairing symmetry [14,15]. This is because the dynamical spin susceptibility is multiplied by a coherence factor in the superconducting state described as follows.

\[
\frac{1}{2} \left( 1 - \frac{E_k E_{k+q}}{E_k E_{k+q}} \right) \approx \frac{1}{2} \left( 1 - \frac{\Delta_k \Delta_{k+q}}{|\Delta_k| \Delta_{k+q}} \right)
\]

(3)

where \( E_k = \sqrt{\varepsilon_k^2 + \Delta_k^2} \) is quasi-particle dispersion relation and \( \varepsilon_k \) is band energy measured relative to Fermi energy. When the signs of order parameter on different FSs are inverse, this coherence factor remains to be one, leading to the resonance mode. In contrast, it becomes zero in the case of same signs, resulting in the suppression of dynamical spin susceptibility. Meanwhile, the superconducting gaps may not have a simple structure. One side of Fermi surface may have different gap structure from the other. The SC gap may not be uniform along \( k \). Therefore, such a superconducting gap structure can be studied from inelastic neutron scattering spectrum through the coherence factor. Low energy gap of 7 meV observed in the measurement suggests that optimally doped LaFeAsO\(_{0.92}\)F\(_{0.08}\) exhibits fairly uniform full gap structure along \( k \) on all the Fermi surfaces. In BaFe\(_{1.85}\)Co\(_{0.15}\)As\(_2\), the resonance mode has been observed independent on \( L \) [16]. On the other hand, it becomes \( L \) dependent in BaFe\(_{1.92}\)Co\(_{0.08}\)As\(_2\) [17] and BaFe\(_{1.92}\)Ni\(_{0.1}\)As\(_2\) [18]. Recent work about thermal conductivity along \( c \)-axis of the Ba(Fe,Co)\(_2\)As\(_2\) system shows nodal behavior in the wide range of Co doping level except for the optimally doped compound [19]. This proximity to the nodal gap structure in the optimally doped compound, BaFe\(_{1.85}\)Co\(_{0.15}\)As\(_2\), may correspond to the low energy gap of about 2.5 meV observed in the dynamical spin susceptibility at 4 K [4]. These results suggest that the superconducting gap may have a complicated structure corresponding to 3-dimensional band dispersion in iron-based superconductors, possibly resulting in common broad spin resonance peaks. In any case, the optimally doped high-\( T_c \) superconductor shows twice enhancement in the dynamical spin susceptibility at the peak energy, \( E_{\text{res}} \). In addition, the details of electronic band structure of an iron-based superconductor are sensitive to structural parameters, such as the pnictogen height [20]. According to a band structure calculation [21], \( \Delta^\pm \)-orbital-type Fermi surface in LaFePO with low pnictogen height appears instead.
of $xy$-orbital-type Fermi surface. Note that the orbital-type is the main orbital contribution to the Fermi surface at $\Gamma$ point. For this case, the nesting condition between two Fermi surfaces at $\Gamma$ and $M$ points may disappear. In other words, the superexchange interaction $J$ between different orbitals in the iron square lattice may become zero depending on the hopping integral between those orbitals. According to the orbital dependence of $J$, $\chi''(\omega)$ obtained by inelastic neutron scattering mainly shows identical orbital contribution in two Fermi surfaces separated by $Q_{AF}$. Theoretically, the importance of the intra-orbital pair scattering for spin fluctuation mediated SC has been proposed [3]. Based on the spin fluctuation mechanism, observed $\chi''(\omega)$ reflects important part associated with SC gaps.

Note that a recent theoretical calculation can also explain the peak structure by $s^{++}$ wave with strong dumping effect, which is consistent with impurity effects such as Co doping on the SC [22]. The SC gap opening effect seems similar to that of $s^\pm$ wave. One of the important differences in spin excitation between $s^\pm$ and $s^{++}$ waves is the peak energy of $\chi''(\omega)$ relative to the summation of two superconducting gap energies at hole and electron Fermi surfaces.

Anyway, these results may suggest that an intimate relationship between electron scattering between $\Gamma$ and $M$ Fermi surfaces and high-$T_c$ SC in iron-based superconductors.

![Figure 3. Q-integrated dynamical spin susceptibility $\chi''(\omega)$ at 11 meV in the normal state as a function of F concentration $x$ in LaFeAsO$_{1-x}$F$_x$.](image)

![Figure 4. Schematic illustration of electron scattering between $\Gamma$ and $M$ Fermi surfaces. $\Gamma$ and $M$ Fermi surfaces have anti-bonding and bonding characters, respectively. Their electron scattering between them corresponds to lattice contraction or expansion, resulting in the lattice breathing.](image)

3.2. Lattice breathing by electron round scattering between $\Gamma$ and $M$ Fermi surfaces

$\Gamma$ and $M$ Fermi surfaces in iron-based superconductors have hole and electron characters, respectively. In a band, a bottom part has bonding character whereas the top part has anti-bonding character, because of the energy dependence on the node number. Carriers in the bottom and top parts are usually called as hole and electron, respectively. In the sense, a lattice expands in the case of electron loss or hole loss, while a lattice contracts in the opposite case, i.e., electron gain or hole gain. When an electron carrier is scattered from $\Gamma$ to $M$ Fermi surfaces, $\Gamma$ Fermi surface gains a hole and $M$ Fermi surface gains an electron, resulting in lattice contraction. Alternatively, when an electron carrier is scattered from $M$ to $\Gamma$ Fermi surfaces, $\Gamma$ Fermi surface loses a hole and $M$ Fermi surface loses an electron, resulting in lattice expansion. Therefore, electron round scattering between $\Gamma$ and $M$ Fermi surfaces is lattice breathing. As observed by inelastic neutron scattering, strongly enhanced dynamical spin susceptibility at $Q_{AF}$ due to the good nesting condition between $\Gamma$ and $M$ Fermi surfaces suggests the frequent electron hopping between them. It may result in active lattice breathing. The importance of breathing mode has also been discussed in superconductors, BaPb$_x$Bi$_{1-x}$O$_3$ and Ba$_{1-x}$K$_x$BiO$_3$, in...
relation to charge disproportionation on bismuth atoms [23]. In this sense, there might be possible similarity between iron-based superconductors and these high-\(T_c\) oxide superconductors.

**4. Summary**

The dynamical spin susceptibility, \(\chi''(\omega)\), observed in LaFeAsO\(_{1-x}\)F\(_x\) samples by inelastic neutron scattering is discussed as the result of good nesting condition between \(\Gamma\) and \(M\) Fermi surfaces separated by \(Q_{AF}\). It consists mainly of the same orbital components in two Fermi surfaces. The decrease of \(\chi''(\omega)\) as a function of fluorine content \(x\) in LaFeAsO\(_{1-x}\)F\(_x\) samples is mainly attributed to the disappearance of hole Fermi surface except for the volume shrinking effect. The electron round scattering between hole and electron Fermi surfaces is introduced in possible relation to lattice breathing.

**Acknowledgements**

This work was supported by JST, Transformative Research-Project on Iron Pnictides (TRIP). Inelastic neutron scattering experiments were partially conducted under UK-Japan and US-Japan collaborations. Work at ORNL was supported by the Scientific User Facilities Division, Office of Basic Energy Sciences, US Department of Energy.

**References**

[1] Kamihara Y, Watanabe T, Hirano M, Hosono H 2008 J. Am. Chem. Soc. 130 3296

[2] For example, Singh D J and Du M H 2008 Phys. Rev. Lett. 100 237003

[3] Mazin I I, Singh D J, Johannes M D, Du M H 2008 Phys. Rev. Lett. 101 057003

[4] Inosov D S, Park J T, Bourges P, Sun D L, Sidis Y, Schneidewind A, Hradil K, Haug D, Lin C T, Keimer B, Hinkov V 2010 Nature Phys. 6 178

[5] Ishikado M, Kajimoto R, Shamato S, Arai M, Iyo A, Miyazawa K, Shirage P M, Kito H, Eisaki H, Kim S-W, Hosono H, Guidi T, Bewley R, Bennington S M 2009 J. Phys. Soc. Jpn. 78 043705

[6] Wakimoto S, Kodama K, Ishikado M, Matsuda M, Kajimoto R, Arai M, Kakurai K, Esaka F, Iyo A, Kito H, Eisaki H, Shamato S 2010 J. Phys. Soc. Jpn. 79 074715

[7] Shamato S, Ishikado M, Christianson A D, Lumsden M D, Wakimoto S, Kodama K, Iyo A, Arai M 2010 Phys. Rev. B 82 172508

[8] Shamato S, Ishikado M, Wakimoto S, Kodama K, Kajimoto R, Arai M, Fukuda T, Nakamura H, Machida M, Eisaki H 2010 Physica C 470 S284

[9] Warren B E 1941 Phys. Rev. 59 693

[10] Sekiya Y, Sato T, Nakayama K, Terashima K, Richard P, Bowen J H, Ding H, Xu Y-M, Li L J, Cao G H, Xu Z-A, Takahashi T 2009 New J. Phys. 11 025020

[11] Mukuda H, Nitta M, Yashima M, Kitaoka Y, Shirage P M, Eisaki H, Iyo A 2010 J. Phys. Soc. Jpn. 79 113701

[12] Matan K, Ibuka S, Morinaga R, Chi Songxue, Lynn J W, Christianson A D, Lumsden M D, Sato T J 2010 Phys. Rev. B 82 054515

[13] Kuroki K, Onari S, Arita R, Usui H, Tanaka Y, Kontani H, Aoki H 2008 Phys. Rev. Lett. 101 087004

[14] Maier T A, Scalapino D J 2008 Phys. Rev. B 78, 020514R

[15] Korshunov M M, Eremin I 2008 Phys. Rev. B 78 140509R

[16] Lumsden M D, Christianson A D, Parshall D, Stone M B, Nagler S E, MacDougall G J, Mook H A, Lokshin K, Egami T, Abernathy D L, Goremychkin E A, Osborn R, McGuire M A, Sefat A S, Jin R, Sales B C, Mandrus D 2009 Phys. Rev. Lett. 102 107005

[17] Christianson A D, Lumsden M D, Nagler S E, MacDougall G J, McGuire M A, Sefat A S, Jin R, Sales B C, Mandrus D 2009 Phys. Rev. Lett. 103 087002

[18] Chi S, Schneidewind A, Zhao J, Harriger L W, Li L, Luo Y, Cao G, Xu Z, Loewenhaupt M, Hu J, Dai P 2009 Phys. Rev. Lett. 102 107006

5
[19] Reid J-Ph, Tanatar M A, Luo X G, Shakeripour H, Doiron-Leyraud N, Ni N, Bud’ko S L, Canfield P C, Prozorov R, Taillefer L 2010 Phys. Rev. B 82 064501
[20] Kuroki K, Usui H, Onari S, Arita R, Aoki H 2009 Phys. Rev. B 79 224511
[21] Vildosola V, Pourovskii L, Arita R, Biermann S, Georges A 2008 Phys. Rev. B 78 064518
[22] Onari S, Kontani H, Sato M, 2010 Phys. Rev. B 81 060504R
[23] For example, Pei S, Jorgensen J D, Dabrowski B, Hinks D G, Richards D R, Mitchell A W, Newsam J M, Sinha S K, Vaknin D and Jacobson A J 1990 Phys. Rev. B 41 4126