Study of magnetic excitation spectra of several Fe-pnictide systems

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Abstract. Magnetic excitation spectra $\chi''(Q, \Delta E)$ have been measured for several Fe-pnictides including Ca-Fe-Pt-As system ($T_c \sim 30$ K), one of new superconducting systems with FeAs planes. For the Ca-Fe-Pt-As system, data were taken with the neutron spectrometer 4SEASONS at J-PARC for a large crystal. The spectral weight $\chi''(Q, \Delta E)$ of this new system observed at the M point in the reciprocal space is enhanced with decreasing temperature $T$ through $T_c$ in the broad energy ($\Delta E$) region around $\sim 12.5$ meV. However, it seems not to be significant as compared with the sharp and strong enhancement in the $Q$ and $\Delta E$ spaces expected for the $S^\pm$ symmetry, although the observed shape of $\chi''(Q, \Delta E)$ is not masked by the resolution effect, indicating the result seems to be hard to explain, unless the $S^\pm$ symmetry is introduced.

1. Introduction

A naive but important question on the superconductivity of Fe-based systems is whether its pairing mechanism is new or well-known. To answer the question, it is essential to identify the symmetry of their superconducting order parameters: If the two order parameters on the disconnected Fermi surfaces around $\Gamma$ and M points in the reciprocal space have the opposite signs, ($S_\pm$ symmetry), the spin fluctuation mechanism is considered to be relevant [1, 2]. It is widely believed, because many physical characteristics of the systems are very similar to those of high-$T_c$ Cu-oxides, in which the magnetic interaction is considered to induce the pairing. However, because the Fe-based systems have multi band nature in contrast to the single band nature of Cu oxides, careful studies are necessary not to miss a new pairing mechanism related to the orbital degrees of freedom of the former.

A direct way to achieve this purpose is to see if the relative signs of the above two order parameters around the $\Gamma$ and M points, $\Delta_\Gamma$ and $\Delta_M$ are same or opposite. [We use the reciprocal space for the two

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dimensional unit cell with one Fe atom within a FeAs layer, where the M point correspond to (±0.5, 0) or (0, ±0.5) (reciprocal lattice unit). We have experimentally studied three physical quantities sensitive to the relative signs of Δ_F and Δ_M, doping effects of nonmagnetic impurities on T_c [3, 4], magnetic excitation spectra χ″(Q, ω) [5, 6] and the NMR relaxation rate 1/T_1 [3, 7]. Among these studies, results of the doping studies on LnFe_1-xM_xAsO_1-yF_y (Ln=La, and Nd; M=Co, Ni, Mn, Ru) systems have strongly indicated that Δ_F and Δ_M should have the same signs, suggesting the so-called S_++ symmetry of the order parameter. Because it is difficult that the ordinary electron-phonon superconductivity can realize the T_c value as high as ~55 K observed for NdFeAsO_1-xF_x, the result suggests the existence of a new pairing mechanism different from both of the phonon- and spin-fluctuation mediated ones. Therefore, careful studies on two other quantities stated above seem to be important to see the relative signs of Δ’s.

On the magnetic excitation spectra χ″(Q, ω) in the superconducting state, Maier and Scalapino [8] are the first who argued the relationship between χ″(Q, ω) and the symmetry, pointing out that a sharp “resonance peak” appears in the χ″(Q, ω) at Q=(0.5, 0), if Δ_F and Δ_M have the opposite signs, and that no peak appears for the same signs of these two. Experimentally, the enhancement of χ″(Q, ω) by the superconductivity have been observed at Q=(0.5, 0) in many Fe-based systems [9, 10]. However, the peak structure is not very sharp as compared with the calculated results. On this point, Onari et al. [11,12] have pointed out that even for the S_+ symmetry, the enhancement of χ″(Q, ω) appears by a mechanism different from that of ref. 8. Then, it seems to be important to distinguish which symmetry experimentally observed data of χ″(Q, ω) support. We study on this subject in this report.

2. Neutron experiments and their results

2.1. LaFeAsO_1-xF_x (x=0.11) and Ba(Fe_1-xCo_x)As_2 (x=0.1)

We have studied the magnetic excitation spectra for LaFeAsO_1-xF_x (polycrystalline sample with x~0.11; T_c~23 K) [5] and Ba(Fe_1-xCo_x)As_2 (aligned crystallites; x~0.1 and T_c~23 K ) [6] with the triple axis spectrometer 5G PONTA installed at JRR-3, and for Ca-Fe-Pt-As (a crystal of about 4.5 g and T_c~30 K) with the spectrometer 4SEASONS at the pulse neutron facility of J-PARC.

For all the samples, we observed the magnetic inelastic peak at the point corresponding to the M point [Q=(0.5, 0)=Q_0]. Here, we just mention the following on LaFeAsO_1-xF_x (x=0.11) (details are in ref. 5). Although a slight enhancement of χ″(Q, ΔE) or, its Q-integrated value, χ″(ΔE), has been observed at 3.5 K (≈T_c) in the region of the transfer energy (ΔE) above ~10 meV, the enhancement is rather small as compared with the prediction of ref. 8. We have not found significant anomalies in the T dependences of the peak- and Q-integrated-intensities with decreasing temperature through T_c, even though the resolution effects do not mask the intrinsic T dependence of the χ″(Q, ΔE) shape.

For Ba(Fe_1-xCo_x)As_2 (x=0.1), data were collected by the constant-E_F mode (E_F=14.7 meV), where the scattering vector Q=(Q_x, Q_y) was scanned along the Q_x direction through (0.5, 0) at various fixed ΔE values at 2.5 K (≈T_c), obtaining the peak of the magnetic scattering centered at (0.5, 0). (The suffices x and y indicate the two directions connecting neighboring Fe-Fe sites, respectively.) The energy position of the peak of the χ″(ΔE)-ΔE curve is ~9.0 meV, χ″(ΔE) being the Q_x-integration of the peak.

The value of χ″(ΔE) at 2.5 K (≈T_c) is estimated to be at most the twice of the value observed slightly above T_c. In the T dependence of the height and width of the observed profile, we have not found any significant anomalies at T_c. In Fig. 1, the observed value of χ″(ΔE) is plotted against ΔE together with the calculated curves for the S_x and S_y symmetries obtained after the consideration of the resolution effect [6]. In the figure, the peak heights and peak positions of the calculated lines are scaled to roughly coincide with those of the observed data. (They were deduced by using the results reported in Fig. 3 of Onari et al. [11]. The curve expected from the data of ref. 8 for the S_1 symmetry is sharper than that of the calculation. The simple comparison between the observed and calculated data indicates that a better fit of the profile width can be found for the S_y symmetry than for the S_x
symmetry. Moreover, as stated above, the observed profile width does not exhibit, with decreasing \( T \) through \( T_c \), any sign of the sharpening expected for the \( S_\perp \) symmetry, even though the resolution effects do not mask the intrinsic \( T \) dependence of the \( \chi''(Q, \Delta E) \) shape for the present case, too [6].

2.2. Ca-Fe-Pt-As system

A large single crystal (~4.5 g) of the Ca-Fe-Pt-As system was prepared and used in the present neutron studies. The superconductivity of this system was found by Nohara's group [13]. The structure analyses carried out by the powder Rietveld analysis revealed that there are two types of structures both of which have FeAs planes common to other Fe pnictide superconductors [14]. We used one of these two with the lattice parameters \( a=3.9029(3) \) Å; \( c=10.5122(6) \) Å (tetragonal; space group \( P4/nmm \). Here, \( a \) corresponds to a unit cell with two Fe atoms within a FeAs plane.). After our Rietveld analysis, the single-crystal structure-determination has been carried out on this type of system by Ni et al. [15], and the detailed superstructure have been reported. In this sense, the structure reported in ref. 14 is just the basic structure of the system. An important point we stress is that the Pt fraction in the FeAs layers is very large (at least ~10%). Even in this high level Pt doping, the superconductivity survives, making the possibility of the \( S_\perp \) symmetry very small, because the symmetry is very fragile to the impurity scattering [3, 6].

In the measurements, the crystal was set on the Al sample holder with the \( c \) axis parallel to the direction of the incident neutron beam [\( z \)-direction]. We collected data at five temperatures from 4 K to 72 K, where the incident neutron energy of 40.4 meV was mainly used. The energy resolution is \(~1.7\) meV at the neutron transfer energy \( \Delta E \sim 10 \) meV. In the map of the magnetic scattering intensity \( S(Q, \omega)\)\(=(n+1)\chi''(Q, \omega) \) collected through the \( \Delta E \) window of 11 meV\(<\Delta E(=\omega)<14 \) meV, for example, we can find the intensity peaks at \( (Q_x, Q_y)=(0.5, 0) \), corresponding to the \( M \) point, and other equivalent points as for many other Fe pnictide superconductors [9, 10]. Although, the \( Q_x \) value depends on \( \Delta E \) for each \( (Q_x, Q_y) \) point, we do not show it explicitly for this quasi two dimensional system.

We have analyzed the data in the region \( \Delta E < 20 \) meV, and found the following. (1) In the entire \( T \) region studied, we have not found significant \( T \) dependence of the width of the magnetic scattering peak along the radial \( Q \) direction [width of the peak in the top panel of Fig. 2]. (2) The peak width along the tangential \( Q \) direction [width of the peak in the bottom panel of Fig. 2] is larger than that along the radial direction. (These widths are intrinsic, because they cannot be explained by the resolution effects, unless the unrealistic mosaic spread is introduced.) (3) The \( \chi''(Q, \Delta E)\) curve or almost equivalently because the \( Q \) width of the magnetic scattering is \( T \)-independent, \( \chi''(\Delta E)\) curve does not show anomalous behaviors at \( T_c \), except that the peak value exhibits gradual increase with decreasing \( T \) through \( T_c \). At least, we do not see "a resonance-like enhancement" or a sharp peak in the \( [Q, \Delta E(=\omega)] \) space (see Fig. 3). At the energy of the \( \chi''(Q, \Delta E) \) peak, the enhancement is smaller than twice of the value above \( T_c (~30 \) K).
3. Other discussion and summary

The enhancement of $\chi''(Q_m, \Delta E)$ we have observed seems not to be significant as compared with that for the opposite signs between $\Delta T_c$ and $\Delta T_m$. Instead, they seem to be explained by the model pointed out in refs. 11 and 12. One might say that even for the $S_z$ symmetry, various origins of the peak broadening exist. For example, details of the Fermi surface shapes may affect the profile width. The anisotropy of the profile widths along the radial and tangential directions may be the evidence for the effect of the Fermi surface shapes [16]. We believe, however, that the $S_{zz}$ symmetry cannot, at least, be excluded. The nonexistence of a clear anomaly at $T_c$ in the $T$ dependences of the peak intensities strengthens the above arguments. Then, it is interesting to consider a mechanism related to the orbital degrees of freedom [17, 18].

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