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Anisotropic inplane spin correlation in the parent and Co-doped $BaFe_2As_2$: A neutron scattering study

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ABSTRACT

Antiferromagnetic spin fluctuations were investigated in the normal states of the parent ($x = 0$), underdoped ($x = 0.04$) and optimally-doped ($x = 0.06$) $Ba(Fe_{1-x}Co_x)_2As_2$ single crystals using inelastic neutron scattering technique. For all the doping levels, quasi-two-dimensional antiferromagnetic fluctuations were observed as a broad peak localized at $\mathbf{Q} = (1/2, 1/2, l)$. At lower energies, the peak shows an apparent anisotropy in the $hk0$ plane; longitudinal peak widths are considerably smaller than transverse widths. The anisotropy is larger for the higher doping level. These results are consistent with the random phase approximation (RPA) calculations taking account of the orbital character of the electronic bands, confirming that the anisotropic nature of the spin fluctuations in the normal states is mostly dominated by the nesting of Fermi surfaces. On the other hand, the quasi-two-dimensional spin correlations grow much rapidly for decreasing temperature in the $x = 0$ parent compound, compared to that expected for nearly antiferromagnetic metals. This may be another sign of the unconventional nature of the antiferromagnetic transition in $BaFe_2As_2$.

1. Introduction

For iron-based superconductors [1], the conventional theory of phonon-mediated superconductivity has difficulty in explaining the high superconducting transition temperatures [2–4]. Accordingly, various other candidates for the superconducting pairing mechanism have been proposed to date, such as spin-fluctuation mediated s_{+-} model [3,5–7], as well as orbital-fluctuation mediated s_{++} model [8–11]. To determine which pairing mechanism is indeed appropriate, it is crucial to know the details of the spin and orbital fluctuations in the normal paramagnetic state. Since direct observation of the orbital fluctuations is difficult, experimental efforts have been focused on observation of the spin fluctuations using neutron inelastic scattering technique. Among a number of Fe-based superconductor compounds, AFe_2As_2 (A = Ca, Sr, Ba and K) 122-type compounds have been most intensively studied due to the availability of large single crystals with various doping levels. Both in the parent and doped compounds, rod-like low energy spin excitation with weak spin correlation along l was observed around the zone boundary $\mathbf{Q} = (1/2, 1/2, l)$ in the tetragonal paramagnetic state, for example, in $Ba(Fe_{1-x}Co_x)_2As_2$ ($0 \leqslant x \leqslant 0.08$) [12–18]. The $\mathbf{Q}$ vector connects hole Fermi surface sheets at the antiferromagnetic zone centre to electron sheets at the zone corner, and satisfies the nesting condition. Moreover, in the heavily-overdoped $Ba(Fe_{1-x}Co_x)_2As_2$ ($x = 0.24$), the inelastic excitation disappears [18] and the angle-resolved photoemission spectroscopy observed that the hole pockets disappear in $0.15 < x < 0.3$ [19,20]. These results suggest that the low-energy spin excitation originates from the Fermi surface nesting between the hole and electron sheets.

Recent studies [21–34] have detected clear inplane anisotropy in the spin correlation lengths. At low energies, the rod-like peak appears with elliptical cross section in the two-dimensional $hk0$ plane, having longer axis pointing to the transverse direction in the parent and electron doped compounds, whereas pointing to the longitudinal direction in the hole doped compounds. At high energies ($\hbar\omega > 80$ meV), the elliptical peak enlarges and splits, with no clear change on entering in the orthorhombic phase. As Park et al. explained [24], the anisotropy preserves C_4 symmetry with the symmetry axis $(0,0,l)$, and is different from rotational

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symmetry breaking. They suggest [24,35] that such inplane anisotropy, at least in the low-energy range, can be consistently reproduced by a simple random phase approximation (RPA) calculation taking account of orbital characters. It has been an issue if such a anisotropic spin correlations may be naturally attributed to the multiband nature of the Ba-122 compounds, or much intriguing idea has to be introduced, such as the frustrated $J_1 - J_2$ model [21,23], quasi propagating mode with different velocity [23], and interplay between anisotropies of the correlation length and of Landau damping [30,36]. Above controversy may be due to the lack of consistent dataset in one compound family; Park et al. [24] compared the low energy anisotropy in $Ba(Fe_{1-x}Co_x)_2As_2$ ($x = 0.075$) to that in $CaFe_2As_2$ [21], whereas another comparison was made with the hole doped KFe_2As_2 [25]. Hence, it is obvious that direct comparison between the parent compound and electron doped compound, such as $BaFe_2As_2$ and $Ba(Fe,Co)_2As_2$, under the same condition is essential. Luo et al. elaborately studied the anisotropy both in the antiferromagnetic and paramagnetic phase in $Ba(Fe_{1-x}Ni_x)_2As_2$ crystals ($0.015 \leqslant x \leqslant 0.09$) [29], however it is hard to see the doping dependence of the anisotropy in the paramagnetic phase under the same energy and temperature. Therefore, in this work, we performed electron-doping dependence study of the inplane anisotropy of low-energy spin fluctuations in $Ba(Fe_{1-x}Co_x)_2As_2$ crystals ($x = 0$, 0.04 and 0.06) by inelastic neutron scattering, focusing on the paramagnetic phase. We observed clear anisotropic inplane spin correlations for all the doping levels. The anisotropy in $BaFe_2As_2$ is smaller than the electron doped $Ba(Fe,Co)_2As_2$. This result is consistent with the Fermi surface nesting picture and indicates that the anisotropic nature of the spin fluctuations in the low energy regime are dominated by Fermi surface nesting. Concerning the temperature dependence of the peak width, the doped compounds show consistent behavior expected for nearly antiferromagnetic metals, whereas the peak in the parent compound sharpens much pronouncedly. This suggests that the quasi-two-dimensional spin correlations grow much rapidly for decreasing temperature in the $x = 0$ parent compound. This may be another sign of the unconventional nature of the antiferromagnetic transition in $BaFe_2As_2$.

2. Experimental details

Single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ ($x = 0$, 0.04(#1), 0.04(#2), and 0.06) were synthesized in a FeAs self-flux using the Bridgman method. First, FeAs precursor was prepared from 99.9% Fe and 99.9999% As powders. The starting elements were mixed, put into an alumina crucible, and sealed in a quartz tube under an argon gas atmosphere. Then the starting elements were slowly heated up to 1073 K, and kept there for 5 days.

Next, 99.9% Ba chips, 99.9% Co powder, 99.9999% As powder, and the prepared FeAs powder, weighed with a molar ratio of Ba: (Fe, Co):As as 10:45:45, were mixed, put in a carbon crucible, sealed in a tantalum crucible, and were further sealed in a quartz tube. All the procedures were performed in an argon-filled glove box with O_2 concentration being about 1 ppm to avoid oxidation. The sealed starting elements were then set in the vertical Bridgman furnace to obtain large single crystals; details of the Bridgman technique used in this study are given in [37]. We performed the Bridgman growth four times for different Co compositions, and each batch obtained was found to contain several small pieces of single crystals. The mass of the grown pieces of single crystals was between 0.3 and 1.2 g.

Co compositions of the obtained crystals were determined by energy dispersive X-ray analysis using a scanning electron microscopy JEOM JSM-5600 and Oxford Link ISIS. The resulting sample compositions are $x_{EDX} = 0.04(1)$, 0.04(1) and 0.061(4) for $x = 0.04$(#1), 0.04(#2) and 0.06 samples. The onset temperatures of the superconducting transition were confirmed by dc magnetic susceptibility measurements using a superconducting quantum interference device magnetometer Quantum Design MPMS-XL in an applied magnetic field of 10 Oe perpendicular to the c axis. Fig. 1 shows the obtained magnetic susceptibility in the low temperature region. As seen from the susceptibility data, the superconducting transition temperatures are 13, 16 and 24 K, for the doped three samples $x = 0.04$(#1), 0.04(#2) and 0.06, respectively. Antiferromagnetic transition temperatures were also found to be $T_{AF} \sim 140$, 70 and 70 K for $x = 0$, 0.04(#1) and 0.04(#2) crystals, respectively. Co compositions determined by T_c with the help of the previous report [38] are consistent with those determined by energy dispersive X-ray analysis; $x_{T_c} \sim 0.040$—0.035, 0.045 and 0.06 for $x = 0.04$(#1), 0.04(#2) and 0.06, respectively. In this study, we regard the two $x = 0.04$ samples (#1 and #2) as one composition. Although slight difference in the compositions for the two samples makes considerable change in the superconducting transition temperatures, we believe that such slight composition difference does not give rise to any significant difference in the inelastic response in the paramagnetic phase. This treatment will be accepted by the weak dependence of F and Γ parameters on the composition, as we see below.

Using the four crystals, we performed inelastic neutron scattering experiments. We used two thermal neutron triple-axes spectrometers, ISSP-GPTAS installed at JRR-3, Tokai, Japan and HB3 installed at HFIR at Oak Ridge National Laboratory, TN, USA. The parent compound ($x = 0$) and underdoped compound ($x = 0.04$(#1)) were measured at GPTAS. The same parent compound, the other underdoped compound ($x = 0.04$(#2)) and the optimally-doped compound ($x = 0.06$) were measured at HB3. Pyrolytic graphite 002 reflections were used both for the monochromator and analyzer to select an energy of neutrons. Final neutron energy was set to $E_f = 14.7$ meV. To detect small differences in peak width, collimations of 40′–80′–40′–80′ at GPTAS and 48′–80′–40′–90′ at HB3 were employed for most of the measurements. The selection of the similar collimations at the two spectrometers enables us to compare the ratio of the anisotropy reliably throughout the investigated compositions. At large energy transfers, such as $\hbar\omega = 28$ meV, where signal becomes weaker, horizontal focusing monochromator with 40′–3 blades Radial Collimator (3RC)-80′–80′ was employed at GPTAS. Higher harmonic neutrons were eliminated by using pyrolytic graphite filters.

To obtain sufficient intensity, two or three pieces of single crystals were co-aligned; the total mass of the samples was about 1 g

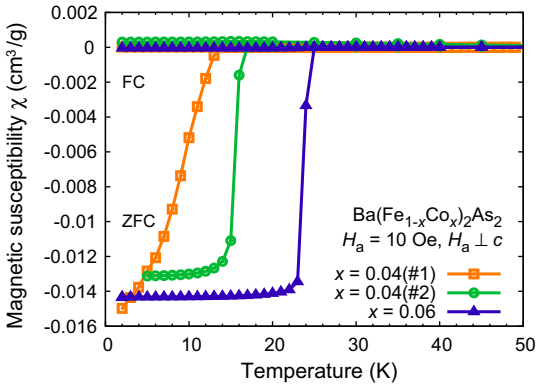

Fig. 1. Temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) dc magnetic susceptibility of $Ba(Fe_{1-x}Co_x)_2As_2$. The (orange) squares stand for $x = 0.04$(#1), (green) circles for 0.04(#2) and (blue) triangles for 0.06. The solid lines are guide to the eyes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)