Magnetic anisotropy in hole-doped superconducting Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ probed by polarized inelastic neutron scattering

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We use polarized inelastic neutron scattering (INS) to study spin excitations of optimally hole-doped superconductor Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ ($T_c = 38$ K). In the normal state, the imaginary part of the dynamic susceptibility, $\chi''(Q, \omega)$, shows magnetic anisotropy for energies below $\sim 7$ meV with $c$-axis polarized spin excitations larger than that of the in-plane component. Upon entering into the superconducting state, previous unpolarized INS experiments have shown that spin gaps at $\sim 5$ and 0.75 meV open at wave vectors $Q = (0.5, 0.5, 0)$, respectively, with a broad neutron spin resonance at $E_r = 15$ meV. Our neutron polarization analysis reveals that the large difference in spin gaps is purely due to different spin gaps in the $c$-axis and in-plane polarized spin excitations, resulting resonance with different energy widths for the $c$-axis and in-plane spin excitations. The observation of spin anisotropy in both optimally electron and hole-doped BaFe$_2$As$_2$ is due to their proximity to the AF ordered BaFe$_2$As$_2$ where spin anisotropy exists below $T_N$.

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Neutron polarization analysis has played an important role in determining the magnetic structure and excitations of solids. For high-transition temperature (High-$T_c$) copper oxide superconductors derived from hole or electron-doping from their antiferromagnetic (AF) parent compounds, neutron polarization analysis have conclusively shown that the collective magnetic excitation coupled to superconductivity at the AF wave vector of the parent compounds, termed neutron spin resonance, has a magnetic origin. Furthermore, by carrying out neutron polarization analysis with a spin-polarized incident neutron beam along the scattering wave vector $Q = k_i - k_f$ (where $k_i$ and $k_f$ are the incident and final wave vectors of the neutron, respectively), $\hat{x} \parallel Q$, perpendicular to $Q$ but in the scattering plane, $\hat{y} \perp Q$, and perpendicular to $Q$ and the scattering plane, $\hat{z} \perp Q$, one can use neutron spin flip (SF) scattering cross sections $\sigma_{x^2}^{SF}$, $\sigma_{y^2}^{SF}$, and $\sigma_{z^2}^{SF}$ to determine the spatial anisotropy of spin excitations. If the resonance is an isotropic triplet excitation of the singlet superconducting ground state, one expects that the degenerate triplet should be isotropic in space as pure paramagnetic scattering. Upon entering into the superconducting state, a quasi-isotropic spin resonance occurs at $E_r = 40$ meV to within the precision of the measurements and a spin anisotropy develops in the lower energy $10 \leq E \leq 30$ meV, resulting in a clear spin gap below 22 meV for the $c$-axis polarized dynamic susceptibility $\chi''$ and in-plane $\chi''_{a/b}$ for $E \geq 10$ meV. The low-energy spin anisotropy is likely due to spin-orbit coupling in the system. For optimally electron-doped copper oxide superconductor Pr$_{0.88}$LaCe$_{0.12}$CuO$_{4-\delta}$, spin excitations are isotropic both above and below $T_N$. Therefore, the spin anisotropy in the superconducting state of hole-doped YBa$_2$Cu$_3$O$_{6.9}$ is unrelated to the normal state paramagnetic scattering.

Like copper oxide superconductors, superconductivity in iron pnictides also arises when electrons or holes are doped into their AF parent compounds. Furthermore, unpolarized neutron scattering experiments have shown that both hole and electron-doped iron pnictides exhibits a neutron spin resonance similar to copper oxide superconductors. In the initial polarized neutron scattering experiment on optimally electron-doped superconductor BaFe$_{1.5}$Sn$_{0.5}$As$_2$ ($T_c = 20$ K), $\chi''$ was found to be much larger than $\chi''_{a/b}$ for energies $2 \leq E \leq 6$ meV below $T_c$, while the resonance at $E_r = 7$ meV...
is only weakly anisotropic. In a subsequent polarized neutron scattering measurement on undoped AF parent compound \( \text{BaFe}_2\text{As}_2 \), isotropic paramagnetic scattering at low-energy \( (E = 10 \text{ meV}) \) were found to become anisotropic spin waves below the Néel temperature \( T_N \) with a much larger in-plane \( (\chi_{\parallel}') \) spin gap than that of the out-of-plane gap \( (\chi_{\perp}') \). These results indicate a strong single-ion anisotropy and spin-orbit coupling, suggesting that more energy is needed to rotate a spin within the orthorhombic \( a-b \) plane than rotating it to the \( c \)-axis. However, similar polarized neutron experiments on electron-overdoped \( \text{BaFe}_{1.85}\text{Ni}_{0.15}\text{As}_2 \) \( (T_c = 14 \text{ K}) \), which is far away from the AF ordered phase, reveal isotropic paramagnetic scattering both above and below \( T_c \). Very recently, Steffens et al. report evidence for two resonance-like excitations in the superconducting state of optimally electron-doped \( \text{BaFe}_{1.88}\text{Co}_{0.12}\text{As}_2 \) \( (T_c = 24 \text{ K}) \). In addition to an isotropic resonance at \( E = 8 \text{ meV} \) with weak dispersion along the \( c \)-axis, there is a resonance at \( E = 4 \text{ meV} \) polarized only along the \( c \)-axis with strong intensity variation along the \( c \)-axis. In the normal state, there are isotropic paramagnetic scattering at AF wave vectors with \( L = 0 \) and weak anisotropic scattering with a larger \( c \)-axis polarized intensity at \( L = \pm 1 \).

If the observed anisotropic magnetic scattering in the superconducting state of optimally electron-doped \( \text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2 \) and \( \text{BaFe}_{1.88}\text{Co}_{0.12}\text{As}_2 \) are indeed associated with the anisotropic spin waves in \( \text{BaFe}_2\text{As}_2 \), one would expect similar anisotropic spin excitations in hole-doped materials not too far away from the parent compound. In this paper, we report neutron polarization analysis on spin excitations of the optimally hole-doped superconductor \( \text{Ba}_0.67\text{K}_{0.33}\text{Fe}_2\text{As}_2 \). From the previous unpolarized INS work on the same sample, we know that spin excitations in the superconducting state have a resonance at \( E_r = 15 \text{ meV} \), a small spin gap \( (E_g \approx 0.75 \text{ meV}) \) at \( Q = (0.5, 0.5, 0) \) and a large gap \( (E_g = 5 \text{ meV}) \) at \( (0.5, 0.5, 1) \). In the normal state, spin excitations at both wave vectors are gapless and increase linearly with increasing energy. Our polarized INS experiments reveal that the persistent low-energy spin excitations at the AF wave vector \((0.5, 0.5, 1) \) below \( T_c \) are entirely \( c \)-axis polarized. Although there is also superconductivity-induced spin anisotropy similar to optimally electron-doped \( \text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2 \) and \( \text{BaFe}_{1.88}\text{Co}_{0.12}\text{As}_2 \), the low-energy \( c \)-axis polarized spin excitations do not change across \( T_c \) and therefore cannot have the same microscopic origin as the spin isotropic resonance at \( E_r = 15 \text{ meV} \). We suggest that the persistent \( c \)-axis polarized spin excitations in the superconducting state of optimally hole and electron-doped iron pnictide superconductors is due to their proximity to the AF ordered parent compound. Their coupling to superconductivity may arise from different contributions of Fe \( 3d_{x^2−y^2} \) and \( 3d_{\chi^Z/Y^Z} \) orbitals to superconductivity.

Single crystals of \( \text{Ba}_0.67\text{K}_{0.33}\text{Fe}_2\text{As}_2 \) are grown by a self-flux method. About 10 grams of single crystals are coaligned in the \([H, H, L]\) scattering plane (with mosaicity \( 3^\circ \) at full width half maximum) with a tetragonal unit cell for which \( a = b = 3.93 \text{ Å} \), and \( c = 13.29 \text{ Å} \). In this notation, the vector \( Q \) in three-dimensional reciprocal space in \( \text{Å}^{-1} \) is defined as \( Q = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^* \), where \( H, K, \) and \( L \) are Miller indices and \( \mathbf{a}^* = \frac{2\pi}{a}, \mathbf{b}^* = \frac{2\pi}{b}, \mathbf{c}^* = \frac{2\pi}{c} \) are reciprocal lattice vectors. Our polarized INS experiments were carried out on the IN22 triple-axis spectrometer with Cryopad capability at the Institut Laue-Langevin in Grenoble, France. The fixed neutron wave vectors were set at \( k_f = 2.66 \text{ Å}^{-1} \) and \( k_f = 3.84 \text{ Å}^{-1} \) in order to close the scattering triangles. To compare with previous polarized INS results on iron pnictides, we converted the measured neutron SF scattering cross sections \( \sigma_{\chi^Z/Y^Z}, \sigma_{\chi^0/Y^0}, \) and \( \sigma_{\chi^Z} \) into \( c \)-axis \( (M_{001}) \) and in-plane \( (M_{110}) \) components of the magnetic

FIG. 1: (Color online) Neutron polarization analysis determined \( c \)-axis \( (\chi_{\parallel}' \propto M_{001}) \) and in-plane \( (\chi_{\parallel}'/b \propto M_{110}) \) components of spin excitations in \( \text{Ba}_0.67\text{K}_{0.33}\text{Fe}_2\text{As}_2 \) from raw SF constant-\( Q \) scans at \( Q = (0.5, 0.5, 0) \) and \( (0.5, 0.5, 2) \). To extract \( M_{001} \) and \( M_{110} \), we use methods described in Ref. and assume \( M_{110} = M_{110} \) in the tetragonal crystal. (a) Energy dependence of \( M_{001} \) and \( M_{110} \) at \( T = 45 \text{ K} \). (b) Identical scans at \( T = 2 \text{ K} \). (c) The solid and open circles show the temperature difference \((2 \text{ K}−45 \text{ K}) \) for \( M_{001} \) and \( M_{110} \), respectively. (d) The sum of \( \sigma_{\chi^Z}^{SF} + \sigma_{\chi^0}^{SF} + \sigma_{\chi^0}^{SF} \) at 45 and 2 K. Since background scattering is not expected to change between these temperatures, such a procedure will increase statistics of magnetic scattering. The black data points are collected at \( Q = (0.5, 0.5, 0) \) with \( k_f = 2.66 \text{ Å}^{-1} \), while the red data points are at \( Q = (0.5, 0.5, 1) \) with \( k_f = 3.84 \text{ Å}^{-1} \). The solid and dashed lines are guided to the eyes.
Figure 1 shows energy scans above and below $T_c$ at wave vectors $\mathbf{Q} = (0.5, 0.5, 0)$ and $(0.5, 0.5, 2)$. We chose these two equivalent wave vectors with different fixed final neutron energies to satisfy the kinematic condition for the large covered energy range. Since the iron magnetic form factors, geometrical factors, and instrumental resolutions are different at these two wave vectors, we use left and right scales for $\mathbf{Q} = (0.5, 0.5, 0)$ and $(0.5, 0.5, 2)$, respectively. In the normal state (45 K), spin anisotropy for energies below $E \approx 7$ meV is clear with $M_{001} (\chi_2^{mp})$ larger than $M_{110} (\chi_2^{mp})$ [Fig. 1(a)]. For $E > 7$ meV, spin excitations are nearly isotropic. This is different from electron-doped BaFe$_{1.85}$Cu$_{0.12}$As$_2$, where paramagnetic scattering at $\mathbf{Q} = (0.5, 0.5, 0)$ is isotropic above $T_c$.

In the superconducting state (2 K), $M_{001}$ and $M_{110}$ in Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ vanish below 5 meV, consistent with opening of a superconductivity-induced spin gap [Fig. 1(b)]$^{48}$. From $E = 5$ meV to the resonance energy at $E_r = 15$ meV, both $M_{001}$ and $M_{110}$ increase with increasing energy, but with different slope resulting significant spin anisotropy ($M_{001} > M_{110}$) appearing near $E \approx 8$ meV [Fig. 1(b)]. This is similar to the spin anisotropy in BaFe$_{1.85}$Cu$_{0.12}$As$_2$. Figure 1(c) shows the temperature difference of magnetic scattering, revealing net intensity gains for $M_{001}$ and $M_{110}$ only above $\sim 7$ and 10 meV, respectively. Figure 1(d) shows the sum of the SF magnetic scattering intensities for three different neutron polarizations, which improve the statistics, above and below $T_c$. Consistent with Fig. 1(c), the superconductivity-induced net magnetic intensity gain appears only above $\sim 7$ meV, forming a resonance at $E_r = 15$ meV.

Figure 2 summarizes the identical scans as that of Fig. 1 at the AF wave vector $\mathbf{Q} = (0.5, 0.5, 1)$ above and below $T_c$. At $T = 45$ K, we see clear spin anisotropy below $E \approx 7$ meV with $M_{001} > M_{110}$ similar to the spin excitations at $\mathbf{Q} = (0.5, 0.5, 0)$ [Fig. 2(a)]. Upon cooling to 2 K, a large spin gap opens below $E \approx 7$ meV in $M_{110}$, but there is still magnetic scattering in $M_{001}$ extending to at least 2 meV. Therefore, the low-energy signal above $\sim 1$ meV at $\mathbf{Q} = (0.5, 0.5, 1)$ reported in the earlier unpolarized neutron measurements$^{48}$ are entirely $c$-axis polarized magnetic scattering. The neutron spin resonance at $E_r = 15$ is isotropic. The temperature difference plots
between 2 and 45 K a broad and narrow peak for $M_{001}$ and $M_{110}$, respectively [Fig. 2(c)]. Fig. 2(d) shows the sum of SF magnetic scattering below and above $T_c$. Consistent with unpolarized work, we see net intensity gain of the resonance in the superconducting state for energies above $E \approx 7$ meV, different from that of BaFe$_{1.88}$Co$_{0.12}$As$_2$ where the magnetic intensity starts to gain above $E = 4$ meV in the superconducting state [Fig. 4(b)]

To further illustrate the effect of spin anisotropy, we plot in Figs. 3(a)-3(d) the differences of $(M_{001} - M_{110})$ above and below $T_c$ at wave vectors $Q = (0.5, 0.5, 0)$ and $(0.5, 0.5, 1)$. In the normal state, we see clear magnetic anisotropy with $M_{001} > M_{110}$ for energies below $\sim 7$ meV [Figs. 3(a) and 3(c)]. In the superconducting state, the $(M_{001} - M_{110})$ differences reveal similar intensity peaks centered around $\sim 7$ meV at $Q = (0.5, 0.5, 0)$ and $(0.5, 0.5, 1)$, but with a much broader width for $Q = (0.5, 0.5, 1)$ [Figs. 3(b) and 3(d)]. Since there are essentially no intensity gain in $M_{001}$ across $T_c$ near $\sim 7$ meV [Figs. 1(c) and 2(c)], the apparent peaks in $(M_{001} - M_{110})$ arise from different responses of $M_{001}$ and $M_{110}$ across $T_c$. While the intensity of $M_{001}$ across $T_c$ is suppressed below $\sim 7$ meV and enhanced above it, similar cross over energy occurs around 10 meV in $M_{110}$, thus resulting peaks near 7 meV in $(M_{001} - M_{110})$ at 2 K [Figs. 3(b) and 3(d)].

Therefore, the differences in superconductivity-induced spin gaps in $M_{001}$ and $M_{110}$ at $Q = (0.5, 0.5, 0)$ and $(0.5, 0.5, 1)$ are causing peaks in $(M_{001} - M_{110})$.

Finally, to confirm the low-energy spin anisotropy discussed in Figs. 1-3, we show in Figs. 4(a)-4(c) constant-energy scans with three different neutron polarizations at $E = 4$ meV along the $[H, H, 0]$ and $[H, H, 1]$ directions. In the normal state, $\sigma_{zz}^{SF}$ shows clear peaks at $Q = (0.5, 0.5, 0)$ and $(0.5, 0.5, 1)$ [Figs. 4(a) and 4(c)]. In both cases, we also find $\sigma_{xx}^{SF} > \sigma_{yy}^{SF}$, thus confirming the anisotropic nature of the magnetic scattering with $M_{001} > M_{110}$. In the superconducting state, while $\sigma_{xx}^{SF}$ and $\sigma_{yy}^{SF}$ are peaked at $(0.5, 0.5, 1)$, $\sigma_{zz}^{SF}$ is featureless. These results again confirm the presence of a larger superconductivity-induced spin gap in $M_{110}$ than that in $M_{001}$ [Fig. 2(b)].

From Figs. 1-4, we see anisotropic spin susceptibility in both the normal and superconducting state of Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$, different from optimally electron-doped BaFe$_{1.88}$Co$_{0.12}$As$_2$ where the anisotropy is believed to emerge only with the opening of the superconducting gap. Furthermore, our data reveal that large differences in the superconductivity-induced spin gaps at $Q = (0.5, 0.5, 0)$ and $(0.5, 0.5, 1)$ arise from the differences in spin gaps of $c$-axis polarized spin excitations. These results are similar to the previous work on electron-doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ and BaFe$_{1.88}$Co$_{0.12}$As$_2$, suggesting that the influence of a strong spin anisotropy in undoped parent compound BaFe$_2$As$_2$ extends to both optimally electron and hole-doped iron pnictide superconductors. For comparison, we note that spin excitations in superconducting iron chalcogenides are different, having slightly anisotropic resonance with isotropic spin excitations below the resonance.

In Refs. 26 and 27, it was suggested that the observed spin anisotropy in BaFe$_{1.88}$Co$_{0.12}$As$_2$ can be understood as a $c$-axis polarized resonance whose intensity strongly varies with the $c$-axis wave vector. This is not the case in Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ since we find much weaker $c$-axis modulation of the magnetic intensity. Therefore, the spin anisotropy seen in optimally electron and hole-doped superconductors is a consequence of these materials being close to the AF ordered parent compound BaFe$_2$As$_2$, where spin-orbit coupling is expected to be strong in these materials, and is not fundamental to superconductivity of these materials. To understand how spin anisotropy in optimally hole and electron-doped iron pnictide superconductors might be coupled to superconductivity via spin-orbit interaction, we note that hole and electron-doped iron pnictides are multiband superconductors with different superconducting gaps for different orbitals. If $c$-axis and in-plane spin excitations arise from quasiparticle excitations of different orbitals between hole and electron Fermi pockets, the large differences in superconducting gaps for Fermi surfaces of different orbital characters might induce the observed large spin anisotropy.

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