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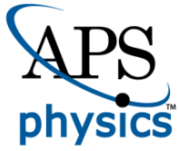
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## Spin excitation anisotropy in the paramagnetic tetragonal phase of $\text{BaFe}_{2}\text{As}_{2}$

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# Spin excitation anisotropy in the paramagnetic tetragonal phase of BaFe<sub>2</sub>As<sub>2</sub>

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We use neutron polarization analysis to study temperature dependence of the spin excitation anisotropy in BaFe<sub>2</sub>As<sub>2</sub>, which has a tetragonal-to-orthorhombic structural distortion at  $T_s$  and antiferromagnetic (AF) phase transition at  $T_N$  with ordered moments along the orthorhombic  $a$ -axis below  $T_s \approx T_N \approx 136$  K. In the paramagnetic tetragonal state at 160 K, spin excitations are isotropic in spin space with  $M_a = M_b = M_c$ , where  $M_a$ ,  $M_b$ , and  $M_c$  are spin excitations polarized along the  $a$ ,  $b$ , and  $c$ -axis directions of the orthorhombic lattice, respectively. On cooling towards  $T_N$ , significant spin excitation anisotropy with  $M_a > M_b \approx M_c$  develops below 3 meV with a diverging  $M_a$  at  $T_N$ . The in-plane spin excitation anisotropy in the tetragonal phase of BaFe<sub>2</sub>As<sub>2</sub> is similar to those seen in the tetragonal phase of its electron and hole-doped superconductors, suggesting that spin excitation anisotropy is a direct probe of doping dependence of spin-orbit coupling and its connection to superconductivity in iron pnictides.

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The iron pnictide superconductors have a rich phase diagram including an orthorhombic lattice distortion associated with ferro-orbital order and nematic phase, antiferromagnetic (AF) order, and superconductivity [1–6]. In the undoped state, a parent compound of iron pnictide superconductors BaFe<sub>2</sub>As<sub>2</sub> forms stripe AF order at  $T_N$  near a tetragonal-to-orthorhombic structural transition temperature  $T_s$  [Fig. 1(a)] [7–9]. Superconductivity can be induced by partially replacing Ba by K in BaFe<sub>2</sub>As<sub>2</sub> to form hole-doped Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> or by partially replacing Fe by  $TM$  ( $TM = \text{Co, Ni}$ ) to form electron-doped BaFe<sub>2-x</sub>TM<sub>x</sub>As<sub>2</sub> [1–3]. Although much attention has been focused on understanding the interplay between magnetism and superconductivity in these materials [1–3], a more subtle and much less explored facet involves the effect of spin-orbit coupling (SOC) [10], which translates anisotropies in real space into anisotropies in spin space and determines the easy axis of the magnetic ordered moment [Fig. 1(b)], and its connection with the electronic nematic phase and superconductivity [5, 11]. Since a nematic quantum critical point is believed to occur near optimal superconductivity in electron and hole-doped iron pnictides [12], it is important to determine the temperature and electron/hole doping evolution of SOC and its association with the nematic phase and superconductivity.

One way to achieve this in iron pnictides is to study the energy, wave vector, temperature, and doping dependence of the spin excitation anisotropy using neutron polarization analysis. Compared with angle resolved photoemission experiments [10], polarized neutron scattering experiments typically have much better energy and momentum resolution [3]. In previous work on electron-doped BaFe<sub>2-x</sub>TM<sub>x</sub>As<sub>2</sub> [13–16] and hole-doped Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> iron pnictides [17–19], there are clear ev-

idence for spin excitation anisotropy in the paramagnetic tetragonal phase with  $M_a \approx M_c > M_b$ , where  $M_a$ ,  $M_b$ , and  $M_c$  are spin excitations polarized along the  $a$ ,  $b$ , and  $c$ -axis directions of the AF orthorhombic lattice, respectively, at temperatures well above  $T_N$  and  $T_s$  [15, 19]. Although low-energy spin waves in the parent compound BaFe<sub>2</sub>As<sub>2</sub> are also anisotropic in the orthorhombic AF ordered state with  $M_c > M_b > M_a$  [20, 21], temperature dependence of the inelastic magnetic scattering at the AF ordering wave vector  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1, 0, 1)$  [Figs. 1(b) and 1(c)] and an energy transfer of  $E = 10$  meV changes from isotropic to anisotropic on cooling below  $T_N$  [20]. However, the energy scale of isotropic paramagnetic scattering at  $E = 10$  meV in BaFe<sub>2</sub>As<sub>2</sub> is considerably larger than that of the anisotropic paramagnetic spin excitation in doped superconductors ( $E < 6$  meV) [15–19]. Since the SOC-induced spin space anisotropy is present in the paramagnetic tetragonal phase of doped iron pnictide superconductors and is also expected to be present in undoped BaFe<sub>2</sub>As<sub>2</sub>, it is possible that paramagnetic spin excitations in BaFe<sub>2</sub>As<sub>2</sub> are also anisotropic, but with an energy scale smaller than  $E = 10$  meV.

To test if this is indeed the case, we carried out polarized neutron scattering experiments on BaFe<sub>2</sub>As<sub>2</sub> with  $T_N \approx T_s \approx 136$  K to study the temperature dependence of the spin excitation anisotropy [Fig. 1(d)]. In the AF ordered state at  $T = 135$  K, we find  $M_c > M_b > M_a$  at  $\mathbf{Q}_{AF} = (1, 0, 1)$  [Figs. 2(a), 2(b), and 3(a)], confirming the earlier results at 10 K [20, 21]. On warming to  $T = 138$  K ( $> T_N, T_s$ ) in the paramagnetic tetragonal state, spin excitations at  $\mathbf{Q}_{AF} = (1, 0, 1)$  are still anisotropic below  $E = 4$  meV but with  $M_a > M_b \approx M_c$  [Figs. 2(c), 2(d), and 3(b)]. For comparison, spin excitations at the AF zone boundary (ZB)  $\mathbf{Q}_{ZB} = (1, 0, 0)$  are isotropic for energies above  $E = 2$  meV [Fig. 3(d)]. Upon

further warming to  $T = 160$  K, paramagnetic scattering becomes isotropic at all energies probed ( $8 \geq E \geq 2$  meV) [Fig. 3(c)]. While temperature dependence of the spin excitations at  $E = 8$  meV and  $\mathbf{Q}_{AF} = (1, 0, 1)$  transforms from isotropic to anisotropic below  $T_N$  with no evidence of critical scattering consistent with earlier measurements at  $E = 10$  meV [20], paramagnetic scattering at  $E = 2$  meV starts to develop spin space anisotropy below about 160 K with enhanced  $M_a$  ( $> M_b \approx M_c$ ) on approaching  $T_N$  due to condensation of the longitudinal component of the magnetic critical scattering into  $a$ -axis aligned AF Bragg peak below  $T_N$  [Fig. 4(a)-4(f)] [9]. On the other hand, paramagnetic scattering at  $E = 2$  meV and  $\mathbf{Q}_{ZB} = (1, 0, 0)$  is isotropic at all temperatures above  $T_N$  [Fig. 4(g)-4(h)]. By comparing these results with spin excitation anisotropy in the paramagnetic tetragonal phase of electron/hole doped iron pnictide superconductors [15–19], we conclude that electron/hole doping in  $\text{BaFe}_2\text{As}_2$  necessary to induce superconductivity also enhances the  $c$ -axis polarized spin excitations associated with superconductivity. These results are also in line with the tetragonal  $C4$  magnetic phase with spins aligned along the  $c$ -axis in near optimally hole doped superconducting  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  [22–25].

Our polarized neutron scattering experiments were carried out using the IN22 triple-axis spectrometers at the Institut Laue-Langevin, Grenoble, France. Polarized neutrons were produced using a focusing Heusler monochromator and analyzed with a focusing Heusler analyzer with a final wave vector of  $k_f = 2.662 \text{ \AA}^{-1}$ . About 12-g single crystals of  $\text{BaFe}_2\text{As}_2$  used in previous work [26] are used in the present experiment. Figure 1(a) shows the collinear AF structure of  $\text{BaFe}_2\text{As}_2$  with ordered moments along the  $a$ -axis [7–9]. The orthorhombic lattice parameters of the AF unit cell are  $a \approx b \approx 5.549 \text{ \AA}$ , and  $c = 12.622 \text{ \AA}$ . The wave vector transfer  $\mathbf{Q}$  in three-dimensional reciprocal space in  $\text{\AA}^{-1}$  is defined as  $\mathbf{Q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$ , with  $\mathbf{a}^* = \frac{2\pi}{a}\hat{\mathbf{a}}$ ,  $\mathbf{b}^* = \frac{2\pi}{b}\hat{\mathbf{b}}$  and  $\mathbf{c}^* = \frac{2\pi}{c}\hat{\mathbf{c}}$ , where  $H$ ,  $K$  and  $L$  are Miller indices. The samples were co-aligned in the  $[H, 0, L]$  scattering plane [Figs. 1(b) and 1(c)]. In this notation, the AF Bragg peaks occur at  $[1, 0, L]$  with  $L = 1, 3, \dots$ , while the AF zone boundaries along the  $c$ -axis occur at  $L = 0, 2, \dots$ . The magnetic responses at a particular  $\mathbf{Q}$  along the  $a$ -,  $b$ -, and  $c$ -axis directions are marked as  $M_a$ ,  $M_b$ , and  $M_c$ , respectively as shown in Fig. 1(b). In the paramagnetic tetragonal state, these correspond to magnetic excitations polarized along the in-plane longitudinal, in-plane transverse, and out-of-plane directions, respectively. The neutron polarization directions  $x$ ,  $y$ , and  $z$  are defined as along  $\mathbf{Q}$ , perpendicular to  $\mathbf{Q}$  but in the scattering plane, and perpendicular to both  $\mathbf{Q}$  and the scattering plane, respectively [Fig. 1(c)]. From the observed neutron spin-flip (SF) scattering cross sections  $\sigma_x^{SF}$ ,  $\sigma_y^{SF}$ , and  $\sigma_z^{SF}$ , we can calculate the components  $M_a$ ,  $M_b$ , and  $M_c$  via  $\sigma_x^{SF} = \frac{R}{R+1}(\sin^2\theta M_a + \cos^2\theta M_c) + \frac{R}{R+1}M_b + B$ ,

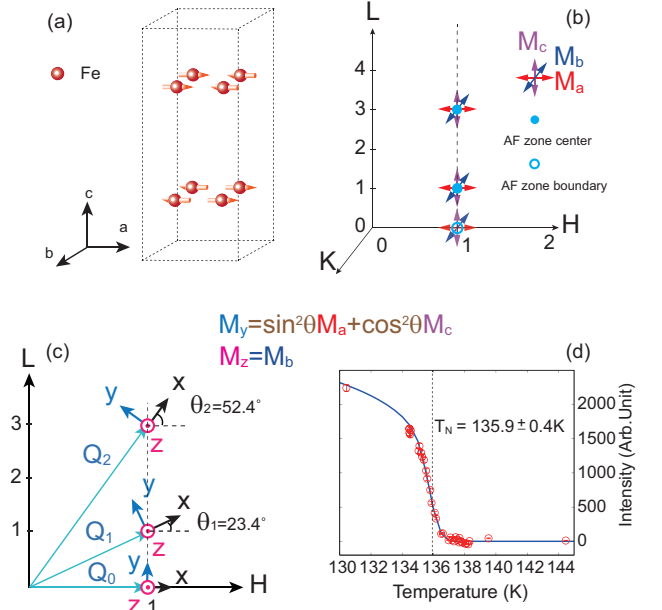


FIG. 1: (Color online) (a) The orthorhombic unit cell of  $\text{BaFe}_2\text{As}_2$  (enclosed by dashed line) with only the magnetic Fe ions shown as red spheres. The arrows indicate the ordered moment direction along the longer  $a$ -axis. Along the  $c$ -axis the nearest neighboring spins are antiparallel. (b) The positions of reciprocal space probed in the present experiment. Magnetic fluctuations polarized along the  $a$ ,  $b$ , and  $c$  directions are marked as  $M_a$ ,  $M_b$ , and  $M_c$ , respectively. (c) Schematic of the  $[H, 0, L]$  scattering plane, where wave vectors  $\mathbf{Q}_0$ ,  $\mathbf{Q}_1$ , and  $\mathbf{Q}_2$  are probed. The neutron polarization directions are along the  $x$ ,  $y$ , and  $z$ . The angle between the  $x$  direction and  $H$ -axis is denoted as  $\theta$ . (d) The temperature dependence of magnetic order parameter measured at  $\mathbf{Q}_1 = (1, 0, 1)$ . The solid line is a Gaussian convolved power law fit with  $T_N = 135.9 \pm 0.4$  K.

$\sigma_y^{SF} = \frac{1}{R+1}(\sin^2\theta M_a + \cos^2\theta M_c) + \frac{R}{R+1}M_b + B$ , and  $\sigma_z^{SF} = \frac{R}{R+1}(\sin^2\theta M_a + \cos^2\theta M_c) + \frac{1}{R+1}M_b + B$ , where  $R$  is the flipping ratio ( $R = \sigma_{Bragg}^{NSF}/\sigma_{Bragg}^{SF} \approx 13$ ) and  $B$  is the background scattering. By measuring  $\sigma_{x,y,z}^{SF}$  at two equivalent AF zone center wave vectors  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1, 0, 1)$  and  $\mathbf{Q}_2 = (1, 0, 3)$ , one can determine all three components of the magnetic response  $M_a$ ,  $M_b$ , and  $M_c$  [15, 16, 19]. For the zone boundary position at  $\mathbf{Q}_0 = (1, 0, 0)$  with  $\theta = 0$ , one can determine  $M_b$  and  $M_c$  using  $\sigma_{x,y,z}^{SF}$  at this position.

To determine the magnetic ordering temperature of  $\text{BaFe}_2\text{As}_2$ , we show in Fig. 1(d) background subtracted elastic SF cross section  $\sigma_x^{SF}$  measured at  $\mathbf{Q}_1 = (1, 0, 1)$ . The solid line is a fit of the magnetic order parameter with Gaussian convolved power-law  $M(T)^2 = B^2 \int (1 - \frac{T}{T_N})^{2\beta} e^{-(T-T_N)^2/2\sigma^2}$  [27]. Although this formula is used to account for sample inhomogeneities and a distribution of Néel temperatures in Co-doped  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  [27], we use it for pure  $\text{BaFe}_2\text{As}_2$ , where disorder is not expected to be important, to compare with  $\beta$  and  $\sigma$  in

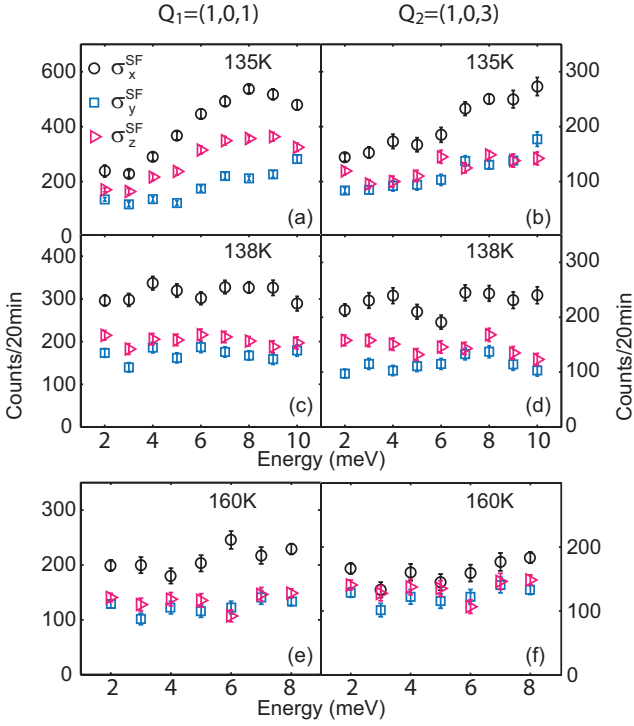


FIG. 2: (Color online) Constant- $\mathbf{Q}$  scans at the two AF wave vectors  $\mathbf{Q}_1 = (1, 0, 1)$  and  $\mathbf{Q}_2 = (1, 0, 3)$  at  $T = 135, 138,$  and  $160$  K. All three spin-flip (SF) channels  $\sigma_x^{SF}$ ,  $\sigma_y^{SF}$ ,  $\sigma_z^{SF}$  are measured at these wave vectors as defined in Fig.1 (c).

lightly Co-doped samples. We find  $T_N = 135.9 \pm 0.4$  K,  $\sigma = 0.51 \pm 0.07$ , and  $\beta = 0.1 \pm 0.02$  for  $\text{BaFe}_2\text{As}_2$ . While the value of  $\sigma$  in  $\text{BaFe}_2\text{As}_2$  is very similar to that of  $x = 0.021$  suggesting a small distribution of  $T_N$  [27], the  $\beta$  value is considerably smaller than the Co-doped samples but similar to previous value of  $\beta = 0.103$  for pure  $\text{BaFe}_2\text{As}_2$  [30]. Figure 2 shows energy scans at the AF wave vectors  $\mathbf{Q}_1 = (1, 0, 1)$  and  $\mathbf{Q}_2 = (1, 0, 3)$  at temperatures below and above  $T_N$ . In an isotropic paramagnet with negligible background scattering and  $R \rightarrow \infty$ , we would expect  $\sigma_x^{SF}/2 \approx \sigma_z^{SF} \approx \sigma_y^{SF}$ . At  $T = 135$  K below  $T_N$ , magnetic scattering at  $\mathbf{Q}_1 = (1, 0, 1)$  shows strong anisotropy with  $\sigma_z^{SF} > \sigma_y^{SF}$  [Fig. 2(a)]. Figure 2(b) plots similar scan at  $\mathbf{Q}_2 = (1, 0, 3)$  with  $\sigma_z^{SF} \approx \sigma_y^{SF}$ . Since  $\mathbf{Q}_1 = (1, 0, 1)$  and  $\mathbf{Q}_2 = (1, 0, 3)$  correspond to angles of  $\theta_1 = 23.4^\circ$  and  $\theta_1 = 52.4^\circ$ , respectively [Fig. 1(c)], we can use  $\sigma_{x,y,z}^{SF}$  at these two wave vectors to completely determine  $M_a$ ,  $M_b$ , and  $M_c$  [28, 29]. Figure 3(a) shows our calculated  $M_c$ ,  $M_b$ , and  $M_a$  ( $M_c > M_b > M_a$ ), and the outcome is similar to spin excitations of  $\text{BaFe}_2\text{As}_2$  [21] and  $\text{BaFe}_{1.91}\text{Co}_{0.09}\text{As}_2$  [16] in the low-temperature AF ordered phase.

In previous work, it was found that paramagnetic spin excitations of  $\text{BaFe}_2\text{As}_2$  above  $T_N$  and  $T_s$  are isotropic at  $E = 10$  meV and  $\mathbf{Q}_1 = (1, 0, 1)$  [20]. To see if spin excitation anisotropy is present at  $T = 138$  K ( $> T_N, T_s$ ) in the paramagnetic tetragonal state, we carried out

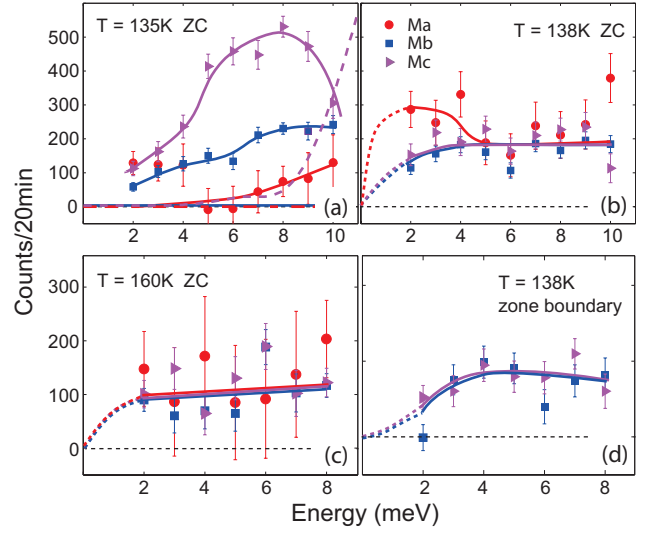


FIG. 3: (Color online) (a-c) The magnetic components  $M_a$ ,  $M_b$ , and  $M_c$  at  $\mathbf{Q}_{AF}$  and 135 K, 138 K, and 160 K obtained from data in Fig. 2. The dashed lines in (a) are results for spin waves of  $\text{BaFe}_2\text{As}_2$  at 2 K [21]. (d) The magnetic components at the zone boundary (ZB) at 138 K. Only  $M_b$  and  $M_c$  can be determined from measurements at  $\mathbf{Q} = (1, 0, 0)$ .

constant- $\mathbf{Q}$  measurements at  $\mathbf{Q}_1$  [Fig. 2(c)] and  $\mathbf{Q}_2$  [Fig. 2(d)]. Inspection of the figures finds clear difference in spin excitations ( $\sigma_z^{SF} > \sigma_y^{SF}$ ) below about  $E \approx 6$  meV at  $\mathbf{Q}_2$ . Figure 3(b) shows the energy dependence of  $M_a$ ,  $M_b$ , and  $M_c$  obtained by using the data in Figs. 2(c) and 2(d), revealing  $M_a > M_b \approx M_c$  for energies below 6 meV. Upon further warming the system to 160 K ( $> T_N, T_s$ ), magnetic signal at  $\mathbf{Q}_1$  [Figs. 2(e)] and  $\mathbf{Q}_2$  [Figs. 2(f)] becomes purely paramagnetic isotropic scattering in the energy region probed satisfying  $(\sigma_x^{SF} - B)/2 \approx (\sigma_y^{SF} - B) \approx (\sigma_z^{SF} - B)$ . The energy dependence of  $M_a$ ,  $M_b$ , and  $M_c$  shown in Fig. 3(c) confirm the isotropic paramagnetic nature of the scattering. Figure 3(d) shows the energy dependence of  $M_b$  and  $M_c$  as obtained from constant- $\mathbf{Q}$  scan at the zone boundary  $\mathbf{Q}_0 = (1, 0, 0)$ , indicating isotropic paramagnetic scattering at energies probed.

Figures 3(a)-3(c) summarize temperature evolution of the estimated  $M_a$ ,  $M_b$ , and  $M_c$  at the AF zone center  $\mathbf{Q}_{AF}$ , obtained by using data in Fig. 2 after taking into account the magnetic form factor differences at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  and other effects as shown in Ref. [29]. In the AF ordered state at  $T = 135$  K ( $\approx T_N - 1$  K), the  $M_c$  component dominates the spin excitation spectrum below 10 meV, followed by  $M_b$  and  $M_a$  [Fig. 3(a)]. For comparison, the  $M_a$  component of the spin waves is completely gapped out below  $\sim 10$  meV at 2 K [dashed line in Fig. 3(a)]. When warming the system to  $T = 160$  K ( $\approx T_N + 24$  K), paramagnetic scattering is isotropic in spin space at all probed energies with  $M_a = M_b = M_c$ . At a temperature  $T = 138$  K ( $\approx T_N + 2$  K) slightly above

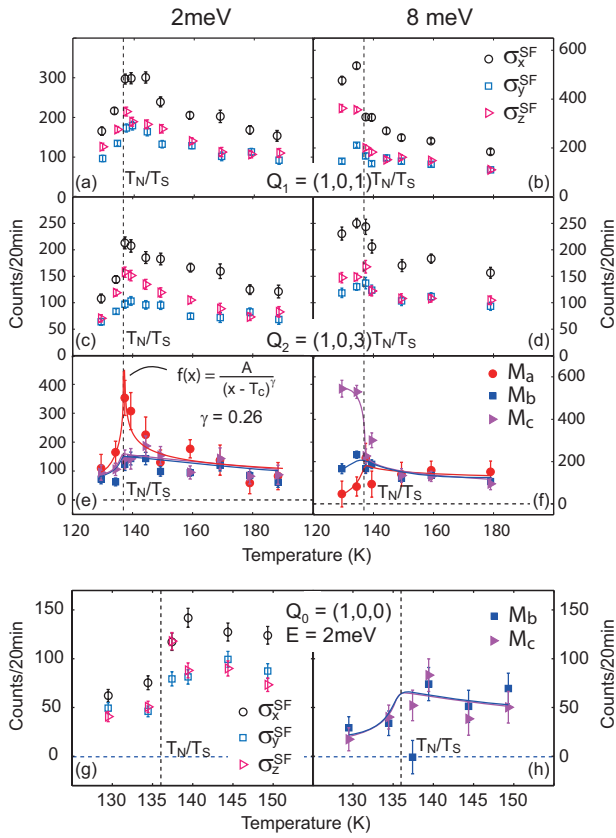


FIG. 4: (Color online) Temperature dependence of  $\sigma_x^{SF}$ ,  $\sigma_y^{SF}$ , and  $\sigma_z^{SF}$  at  $E = 2$  meV with (a)  $\mathbf{Q}_1 = (1, 0, 1)$  and (c)  $\mathbf{Q}_2 = (1, 0, 3)$ . Similar data at  $E = 8$  meV with (b)  $\mathbf{Q}_1 = (1, 0, 1)$  and (d)  $\mathbf{Q}_2 = (1, 0, 3)$ . (e) Temperature dependence of the components  $M_a$ ,  $M_b$ , and  $M_c$  at  $E = 2$  meV. The solid curve is the fitted line with function  $f(T) = \frac{A}{(T - T_c)^\gamma}$ . (f) Temperature dependence of  $M_a$ ,  $M_b$ , and  $M_c$  at  $E = 8$  meV. (g) The three SF scattering channels measured at  $\mathbf{Q}_0 = (1, 0, 0)$  and  $E = 2$  meV. (h) Temperature dependence of  $M_b$  and  $M_c$  determined from (g). The solid lines are guides to the eye. The vertical dashed lines mark  $T_N/T_s$ .

$T_N$ , paramagnetic spin excitations are anisotropic below  $\sim 5$  meV with  $M_a > M_b \approx M_c$ .

In previous unpolarized neutron scattering experiments on  $\text{BaFe}_2\text{As}_2$  [9], two-dimensional (2D) magnetic critical scattering has been observed at temperatures far above  $T_N$ . Upon cooling, the longitudinal component of the critical scattering above  $T_N$  ( $M_a$ ) is expected to increase with decreasing temperature and condense into the 3D AF Bragg positions at the 2D-3D dimensional crossover temperature  $T_{3D}$  near  $T_N$  [30]. The transverse components of spin excitations ( $M_b$  and  $M_c$ ) are the spin wave contributions not expected to diverge at  $T_N$  [9]. To test if this is indeed the case, we measured temperature dependence of  $\sigma_{x,y,z}^{SF}$  at  $E = 2$  meV and 8 meV at the AF zone center  $\mathbf{Q}_1$  [Figs. 4(a) and 4(b)] and  $\mathbf{Q}_2$  [Figs. 4(c) and 4(d)]. With decreasing temperature,  $\sigma_{x,y,z}^{SF}$  increases in intensity with the differences between  $\sigma_z^{SF}$  and

$\sigma_y^{SF}$  most obvious near  $T_N$  at  $\mathbf{Q}_2$  [Fig. 4(c)]. Using data in Fig. 4(a) and 4(c), we estimate the temperature dependence of  $M_a$ ,  $M_b$ , and  $M_c$  in Fig. 4(e). Consistent with the expectations from the magnetic critical scattering measurements [9], we see a diverging longitudinal spin excitations  $M_a$  at  $E = 2$  meV while transverse spin excitations show no critical scattering around  $T_N$ . On cooling below  $T_N$ , all three polarizations of spin excitations are suppressed due to the formation of spin gaps [20]. Similar measurements at  $E = 8$  meV show isotropic paramagnetic scattering behavior ( $M_a \approx M_b \approx M_c$ ) down to  $T_N$  before splitting into  $M_c > M_b > M_a$  seen in the AF ordered state [Fig. 4(f)]. Figure 4(g) shows temperature dependence of the spin excitations  $\sigma_{x,y,z}^{SF}$  at  $E = 2$  meV and zone boundary position  $\mathbf{Q}_0$ . We see that magnetic scattering is isotropic at all measured temperatures with no evidence of spin anisotropy.

The diverging  $M_a$  near  $T_N$  in  $\text{BaFe}_2\text{As}_2$  may arise from the longitudinally polarized spin excitation in the critical scattering regime of a Heisenberg antiferromagnet with Ising spin anisotropy [Fig. 4] [33]. This means that the effect of critical scattering in  $\text{BaFe}_2\text{As}_2$  can force the fluctuating moment along the longitudinal ( $a$ -axis) direction in the paramagnetic critical regime without the need for orthorhombic lattice distortion and associated ferro-orbital (nematic) ordering. Although this scenario is interesting, we note that temperature dependence of spin excitation anisotropy in the paramagnetic state of AF ordered  $\text{NaFeAs}$  [34] and electron underdoped  $\text{BaFe}_{1.904}\text{Ni}_{0.096}\text{As}_2$  [15] behave differently. In previous polarized neutron scattering experiments on  $\text{NaFeAs}$ , which has a collinear AF order at  $T_N = 45$  K and an orthorhombic-to-tetragonal lattice distortion at  $T_s \approx 58$  K [31, 32],  $M_a \approx M_c$  is larger than  $M_b$  in the paramagnetic orthorhombic phase below  $T_s$  and the in-plane anisotropy  $M_a - M_b$  enhances on approaching  $T_N$  from  $T_s$  [34]. When warming up to above  $T_s$ , the statistics of the data in  $\text{NaFeAs}$  is insufficient to establish possible spin anisotropy [34]. Since one of the key differences between  $\text{BaFe}_2\text{As}_2$  and  $\text{NaFeAs}$  is the coupled structural and magnetic phase transitions in  $\text{BaFe}_2\text{As}_2$ , our data suggest that the orthorhombic lattice distortion lifting the degeneracy of the Fe  $d_{xz}$  and  $d_{yz}$  orbitals also induces the  $M_c$  and  $M_b$  anisotropy. This is consistent with the observation that  $M_c$  has the lowest energy in spin waves of the AF ordered  $\text{BaFe}_2\text{As}_2$  [20, 21] and  $\text{NaFeAs}$  [34], suggesting that it costs less energy for the  $a$ -axis ordered moment to rotate out of the plane than to rotate within the plane.

For electron doped  $\text{BaFe}_{1.904}\text{Ni}_{0.096}\text{As}_2$  superconductor with  $T_c = 19.8$  K and  $T_N \approx T_s = 33 \pm 2$  K, spin excitation anisotropy at  $E = 3$  meV and zone center  $\mathbf{Q}_{AF}$  with  $M_a \approx M_c > M_b$  first appears below  $\sim 70$  K and shows no anomaly across  $T_s/T_N$  before changing dramatically below  $T_c$  [15]. For hole-doped  $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$  superconductor with  $T_c = 38$  K and no structural/magnetic

order, spin excitation anisotropy at  $E = 3$  meV and  $\mathbf{Q}_{AF}$  with  $M_a \approx M_c > M_b$  appears below  $\sim 100$  K, and also decreases abruptly  $T_c$  [19]. The similarities of these results to those of NaFeAs in the nematic temperature regime ( $T_s > T > T_N$ ) suggest that the ferro-orbital order or fluctuations [35–40] in electron and hole-doped BaFe<sub>2</sub>As<sub>2</sub> first appear in the paramagnetic tetragonal phase at temperatures well above  $T_s$  [15, 19]. Since SOC in iron pnictides is a single iron effect not expected to change dramatically as a function of electron and hole doping [41, 42], the weak/absence of  $M_c$  and  $M_b$  spin excitation anisotropy in the tetragonal phase of BaFe<sub>2</sub>As<sub>2</sub> is difficult to understand. One possibility is that the nearly coupled structural and magnetic phase transitions in BaFe<sub>2</sub>As<sub>2</sub> [8] suppress the role of the SOC induced ferro-orbital fluctuations above  $T_s$ . Although hole-doped Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> also has coupled structural and magnetic phase transitions in the underdoped regime [22], it changes to a double- $\mathbf{Q}$  tetragonal magnetic structure with ordered moments along the  $c$ -axis near optimal superconductivity [23–25]. When hole and electron doping in BaFe<sub>2</sub>As<sub>2</sub> reduces the structural and magnetic ordering temperatures, the SOC induced ferro-orbital fluctuations start to appear at temperatures above  $T_s$ . In this picture, the spin excitation anisotropy in the superconducting iron pnictides originates from similar anisotropy already present in their parent compounds below  $T_s$ . The dramatic change in spin excitation anisotropy across  $T_c$  seen in electron and hole-doped BaFe<sub>2</sub>As<sub>2</sub> suggests a direct coupling of the SOC to superconductivity. The systematic polarized neutron scattering measurements present here and in previous work on doped BaFe<sub>2</sub>As<sub>2</sub> family of materials [13–19] call for quantitative calculations on how SOC is associated with spin excitation anisotropy in iron pnictides.

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