Spin anisotropy due to spin-orbit coupling in optimally hole-doped Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$

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We use polarized inelastic neutron scattering to study the temperature and energy dependence of spin space anisotropies in the optimally hole-doped iron pnictide Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ ($T_c = 38$ K). In the superconducting state, while the high-energy part of the magnetic spectrum is nearly isotropic, the low-energy part displays a pronounced anisotropy, manifested by a $c$-axis polarized resonance. We also observe that the spin anisotropy in superconducting Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ extends to higher energies compared to electron-doped BaFe$_{2-x}$TM$_x$As$_2$ ($TM = Co$, Ni) and isovalent-doped BaFe$_{2-x}$As$_2$ which can be quantitatively determined by neutron polarization analysis.

I. INTRODUCTION

The parent compounds of iron pnictide superconductors, such as LaFeAsO and BaFe$_2$As$_2$, form stripe antiferromagnetic (AF) order at $T_N$ below a tetragonal-to-orthorhombic structural transition temperature $T_S$ [inset in Fig. 1(b)]. Superconductivity can be induced by partially replacing Ba by K in BaFe$_2$As$_2$ to form hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ or by partially replacing Fe by $TM$ ($TM = Co$, Ni) to form electron-doped BaFe$_{2-x}$TM$_x$As$_2$. Importantly, the resulting phase diagrams exhibit significant asymmetry between electron- and hole-doping [Figs. 1(a) and 1(b)]. For instance, while near optimal doping the stripe AF order becomes incommensurate for electron-doped BaFe$_{2-x}$TM$_x$As$_2$[10,11][see arrow in Fig. 1(b)], a double-$Q$ tetragonal magnetic structure with ordered moments along the $c$-axis is observed in hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$[28,29][see region of the phase diagram near the arrow in Fig. 1(a)]

Nevertheless, upon entering the superconducting state, a magnetic resonance mode appears in the magnetic spectrum in both cases at the AF wave vector ($Q_{AF}$[13,14]). Furthermore, by measuring the splitting of the electronic states at high-symmetry points in reciprocal space[15] angle-resolved photoemission spectroscopy (ARPES) measurements find that spin-orbit coupling (SOC) is present in both electron- and hole-doped iron pnictides with a similar energy scale $\sim 10$ meV[19]. Also common to both optimally electron-doped BaFe$_{2-x}$TM$_x$As$_2$[28,29] and hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$[22,23] is the presence of electronic nematic fluctuations, as revealed by the elastoresistance – i.e. the rate of change of the resistivity anisotropy with respect to applied in-plane uniaxial strain [Fig. 1(c)][24]. The elastoresistance diverges with a Curie-Weiss form for both classes of materials as well as for isovalent-doped BaFe$_{2-x}$As$_2$[25]. Deviation from the Curie-Weiss behavior is seen in both optimally electron- and hole-doped BaFe$_{2-x}$As$_2$ at low temperatures, while no deviation is seen in BaFe$_{2-x}$As$_2$ down to $T_c$ [Fig. 1(d)][25].

In addition to its impact on the electronic spectrum[19,20], SOC also converts crystalline anisotropies into anisotropies in spin space, as seen from nuclear magnetic resonance studies[27]. The spin anisotropy resulting from SOC plays an essential role for the double-$Q$ magnetic phase[30], in which the ordered moments align along the $c$-axis[28]. If SOC was absent, the spin excitations in the paramagnetic tetragonal state of the iron pnictides would be isotropic in spin space [Fig. 1(e)]. However, due to the presence of a sizable SOC, an anisotropy is developed in the spin excitations, which can be quantitatively determined by neutron polarization analysis[22]. In the antiferromagnetically ordered phases of the parent compounds BaFe$_2$As$_2$ and NaFeAs[30,31], where the ordered moments point parallel to the orthorhombic $a$-axis [inset in Fig. 1(b)], spin waves exhibit significant anisotropy, with $c$-axis polarized spin waves occurring at lower energy compared to $b$-axis polarized spin waves[30,31]. To elucidate the relevance of SOC to superconductivity, it is instructive to compare the behavior of the spin anisotropy in hole-doped and
electron-doped BaFe$_2$As$_2$, since the maximum values of $T_c$ are quite different in these two cases – $T_c \approx 38$ K for optimally hole-doped Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ and $T_c \approx 25$ K for optimally electron-doped BaFe$_{1.86}$Co$_{0.14}$As$_2$. Previous analysis of the electron-doped case revealed that the spin anisotropy persists in the paramagnetic tetragonal phase for doping levels up to or slightly beyond optimal doping$^{38-40}$, but vanishes in the well-overdoped regime$^{39,41}$.

In this paper, we present polarized neutron scattering studies of spin excitations in optimally hole-doped Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$.$^{17,19}$ In the normal state, we find that the spin anisotropy of Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ persists to $\sim 100$ K for $E = 3$ meV, similarly to the case of near-optimally electron-doped BaFe$_{2-x}$TM$_x$As$_2$, where spin anisotropy at $Q_{AF} = (1,0,1)$ was found below $E \approx 7$ meV and up to $\sim 70$ K$^{23,24}$. We associate the onset of normal state spin anisotropy with the nematic susceptibility deviating from Curie-Weiss behavior measured via elasto-resistance [see vertical arrows in Fig. 1(d)]$^{23}$, indicating an important role of spin excitations in transport properties of iron pnictides.

Upon entering the superconducting state, we find that while at high energies ($E \geq 14$ meV) the spectrum is nearly isotropic as found in previous work$^{23}$, at low energies the resonance mode is strongly anisotropic, being dominated by a $c$-axis polarized component. We attribute this behavior to the fact that the superconducting state is close to the double-$Q$ magnetic phase, in which the magnetic moments point out-of-plane.$^{23}$ Indeed, by adding a spin-anisotropic term that favors $c$-axis spin orientation in a simple two-band theoretical model, we find that the resonance mode in the $c$-axis polarized channel has in general a lower energy than in other channels, and that this energy difference increases as the magnetically ordered state is approached. Our analysis also reveals an interesting correlation between the energy scale of the spin anisotropy in the superconducting state and $T_{SC}$,$^{39,40,42}$ suggesting that SOC is an integral part of the superconductivity of iron pnictides.

II. EXPERIMENTAL RESULTS

Polarized inelastic neutron scattering measurements were carried out using the IN22 triple-axis spectrometer at Institut Laue-Langevin, Grenoble, France. We studied Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ single crystals ($a = b \approx 5.56$ Å, $c = 13.29$ Å) co-aligned in the $[H,0,L]$ scattering plane used in previous work$^{35,36}$. We use the orthorhombic notation suitable for AF ordered iron pnictides even though Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ has a tetragonal structure and is paramagnetic at all temperatures.$^{37,38}$ Thus, the momentum transfer is $Q = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$, with $\mathbf{a}^* = \frac{2\pi}{a}\mathbf{a}$, $\mathbf{b}^* = \frac{2\pi}{b}\mathbf{b}$ and $\mathbf{c}^* = \frac{2\pi}{c}\mathbf{c}$, where $H, K$ and $L$ are Miller indices. In this notation, magnetic order in BaFe$_2$As$_2$ occurs at $Q_{AF} = (1,0,L)$ with $L = 1,3,5 \ldots$ [Fig. 1(e)].

Three neutron spin-flip (SF) cross sections $\sigma_x^SF$, $\sigma_y^SF$ and $\sigma_z^SF$ were measured, with the usual convention $x \parallel Q$, $y \perp Q$ and in the scattering plane, and $z$ perpendicular to the scattering plane. Magnetic neutron scattering directly measures the magnetic scattering function $S_{x}^{\alpha\beta}(Q,E)$, which is proportional to the imaginary part of the dynamic susceptibility through the Bose factor, $S_{x}^{\alpha\beta}(Q,E) \propto [1 - \exp(-\frac{E - E_0}{k_BT})]^{-1}\Im \chi_{\alpha\beta}(Q,E)$. Follow-
To gain further insight into the spin anisotropy of Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$, we carried out temperature scans at $Q_{AF} = (1,0,1)$ for $E = 3$ meV and $E = 9$ meV, as shown in Figs. 3(a) and 3(b). At $E = 3$ meV, the spin anisotropy with $\sigma_{z}^{SF} > \sigma_{y}^{SF}$ persists up to $\sim 100$ K. Although below $T_c$ the magnetic signal is suppressed in all
three SF cross sections, the normal-state anisotropy persists [Fig. 3(a)]. At $E = 9$ meV, the spin anisotropy disappears above $T_c$, suggesting that the main contribution to the spin anisotropy in the superconducting state arises from the anisotropic resonance modes. In Figs. 3(c) and 3(d), $\sigma_x^{SF} - \sigma_y^{SF} \propto M_y$ and $\sigma_x^{SF} - \sigma_z^{SF} \propto M_z$ are shown. At $E = 3$ meV, $M_y > M_z$ for $T \lesssim 100$ K and both are suppressed below $T_c$. At $E = 9$ meV, while a clear resonance mode with an order-parameter-like temperature dependence is seen in $M_y$, $M_z$ remains constant across $T_c$. The temperature onset of spin anisotropy is more clearly seen in Fig. 3(c) and (f), which plots $\sigma_x^{SF} - \sigma_y^{SF} \propto M_y - M_z$ for $E = 3$ meV and $E = 9$ meV, respectively.

To obtain the temperature dependence of $M_a$, $M_b$, and $M_c$, we measured $\sigma_y^{SF}$, $\sigma_x^{SF}$, and $\sigma_z^{SF}$ at $Q_{AF} = (1, 0, 3)$ for $E = 3$ meV and $E = 9$ meV. Combining the temperature dependence for $L = 1$ and $L = 3$, $M_a$, $M_b$ and $M_c$ are obtained for odd $L$ as shown in Fig. 4(a) and 4(b). At $E = 3$ meV, $M_a \approx M_c > M_b$ within the probed temperature range, and all three channels decrease in intensity below $T_c$. At $E = 9$ meV, $M_a$ and $M_b$ display a weak temperature dependence while $M_c$ is sharply affected by $T_c$. To corroborate our conclusion, we binned data points in Fig. 4(a) and 4(b) that are well below $T_c$ ($T \leq 25$ K) and above $T_c$ ($T \geq 40$ K), as shown in the insets of Figs. 4(a) and 4(b). While magnetic excitations at $E = 3$ meV are suppressed upon entering the superconducting state, the polarization of these magnetic excitations seems to remain the same, persisting up to $T \approx 100$ K. On the other hand, at $E = 9$ meV, magnetic excitations are nearly isotropic above $T_c$, while $M_c > M_a \approx M_b$ well below $T_c$. Therefore, the $c$-axis polarized anisotropic resonance is directly coupled to superconductivity with an order-parameter-like temperature dependence.

III. DISCUSSION AND CONCLUSION

To understand the origin of this $c$-axis polarized spin resonance, we consider a simple two band model in which the resonance mode arises due to the sign change of the gap function between a hole pocket and an electron pocket displaced from each other by the AF ordering. Without SOC, the energy of the resonance mode is the same for all polarizations, being close to $2\Delta$ far from the putative magnetic quantum phase transition inside the superconducting dome [$U \ll U^*_c$ in Fig. 4(c)], but vanishing as the transition is approached [$U \to U^*_c$ in Fig. 4(c)]. SOC, however, promotes a spin anisotropy term that makes the magnetic moments point along the $c$-axis for hole-doped compound. As a result, the energy of the resonance mode polarized along the $c$-axis is suppressed much faster as the magnetic transition is approached, yielding $\omega_c < \omega_ab$ [Fig. 4(c)]. This behavior is in qualitative agreement with our experimental results, with the resonance seen in $M_c$ indeed at lower energies. It should also be noted that our model does not capture the broadening of the resonance, which is rather pronounced in the experimental data. Our simple model has two additional consequences: first, as the system is overdoped and moves farther from the magnetically ordered state, the resonance mode should become more isotropic. While spin anisotropy persists in slightly overdoped BaFe$_{1.85}$Ni$_{0.15}$As$_2$ ($T_c = 36$ K) how it evolves in K-well-overdoped samples remains to be seen. Furthermore, because in electron-doped compounds the moments point along the $a$ direction, the resonance is expected to be polarized along the $a$-axis. Although this is the case in electron-doped NaFeO$_{0.994}$Co$_{0.006}$As$_2$ the sample studied had long-range AF order. For electron-doped Ba(Fe$_{0.94}$Co$_{0.06}$)$_2$As$_2$ the anisotropic resonance was argued to be also polarized along $c$-axis, based on the assumption $M_a = M_b$ and the observation $M_y > M_z = 0$ for the anisotropic resonance. As we have shown here and in previous work even in the tetragonal state $M_a$ and
M₂ are not necessarily the same and it is unclear whether there is also significant resonance spectral weight polarized along the c-axis in previous work. Spin anisotropy of spin excitations has also been detected in the superconducting states of LiFeAs \cite{22} and FeSe₀.₅Te₀.₅ \cite{23} consistent with significant spin-orbit coupling detected by ARPES\cite{19,20} in these systems.

The normal state spin anisotropy at low energies persists to a temperature significantly higher than Tₜ [\sim 70 K in BaFe₁₀₀₋₉₀₄Nᵢ₀₋₉₀₋₉₆As \cite{24} and \sim 100 K in Ba₀₋₆₇K₀₋₃₃Fe₂As₂]. Fig. 3(e)] for both electron- and hole-doped BaFe₂As₂ near optimal doping. The temperature at which spin anisotropy onsets is similar to the temperature at which the nematic susceptibility deviates from Curie-Weiss behavior\cite{25} suggesting a common origin for both phenomena [Fig. 1(d)]. For optimally-doped BaFe₂As₁₋₄P₀.₆₆, whose nematic susceptibility shows no deviation from the Curie-Weiss form \cite{26} no spin anisotropy is observed right above Tₜ \cite{27}. While disorder is likely to play an important role in explaining this deviation from Curie-Weiss behavior in the elasto-resistance\cite{28}, our results suggest that the spin anisotropy may also be important. Indeed, previous INS experiments revealed the intimate relationship between nematicity and magnetic fluctuations.\cite{29,30} Theoretically, the nematic susceptibility increases with increasing magnetic fluctuations in all polarization channels.\cite{31} However, once a spin anisotropy sets in, fluctuations related to the spin components perpendicular to the easy axis increase more slowly with decreasing temperature. As a result, the nematic susceptibility should also increase more slowly, which may contribute to the deviation from Curie-Weiss behavior observed experimentally.

Finally, the maximum energies at which spin anisotropy is observed in the superconducting states of several doped BaFe₂As₂ compounds are plotted as function of Tₜ in Fig. 4(d). Note that the spin anisotropy of the resonance in the superconducting state is also present in BaFe₂As₁₋₄P₀.₆₆, despite the absence of spin anisotropy in the normal state\cite{32}. We note a clear positive correlation between the energy scale of the spin anisotropy and Tₜ, suggesting SOC to be an important ingredient for understanding superconductivity in iron pnictides.

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Supplementary Information:

Determination of $M_a$, $M_b$ and $M_c$

Using the method described in the supplementary of Ref. [1] and data for $\mathbf{Q} = (1,0,L)$ with $L = 1$ and 3 ($L = 0$ and 2), we extracted $M_a$, $M_b$ and $M_c$ for the magnetic zone center (zone boundary along $L$) with odd (even) $L$. The data with $L = 3$ used in combination with the $L = 1$ data [Fig. 3(a)-(b)] to obtain $M_a$, $M_b$ and $M_c$ in Fig. 4(a)-(b) are shown here in Supplementary Fig. 1. In the analysis we ignored the differences of sample illumination volume and convolution with instrumental resolution between $L = 1$ and 3, and set the scale factor that accounts for these differences to be $r = 1$ [1]. In previous works [1–4] $r$ is found to be close to unity in all cases and our results are qualitatively robust when $r$ deviates slightly from 1.

$$\mathbf{Q} = (1,0,3)$$

Supplementary Figure 1: Temperature scans of $\sigma_{x}^{SF}$, $\sigma_{y}^{SF}$ and $\sigma_{z}^{SF}$ at $\mathbf{Q} = (1,0,3)$ with (a) $E = 3$ meV and (b) $E = 9$ meV.

Measurement of resistivity anisotropy under uniaxial pressure

Previous measurements of resistivity anisotropy in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ demonstrated that compared to their electron-doped counterparts, the hole-doped compounds have much smaller and reversed resistivity anisotropy [5, 6]. Near optimal doping resistivity anisotropy was found to disappear in Ba$_{0.66}$K$_{0.34}$Fe$_2$As$_2$ [6], however recent measurements revealed significant elastoresistance in Ba$_{0.4}$K$_{0.6}$Fe$_2$As$_2$ [7]. To resolve this puzzling difference, we measured resistivity anisotropy on single crystals of Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$ and BaFe$_{1.904}$Ni$_{0.096}$As$_2$ using the Montgomery method with a mechanical clamp that can vary the applied pressure in-situ as described in Ref. [8]. Prefactors due to samples not being perfect squares were corrected for at 200 K [8], where no/negligible resistivity anisotropy is present. Several nominal pressures were applied and the actual zero pressure is determined by performing a linear fit of the change in resistivity anisotropy as a function of nominal pressure. Measured resistivity anisotropy is then scaled to $P = 15$ MPa for both samples to allow for direct comparison under the same applied pressure. Resistivity anisotropy is linearly proportional to the applied pressure in the pressure range we studied, as shown in Supplementary Figure 2 for Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$, similar to BaFe$_2$As$_2$ in which linear response persists up to 90 MPa in the paramagnetic state [8].

Our results are shown in Fig. 1(c), resistivity anisotropy with reversed sign is observed in optimally electron- and hole-doped BaFe$_2$As$_2$ samples. Significant anisotropy is seen in optimal-doped Ba$_{0.67}$K$_{0.33}$Fe$_2$As$_2$, in agreement with elastoresistance measurements. The much smaller resistivity anisotropy in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ and small applied pressure are likely causes of why it was not observed in previous work [6].

Anisotropy of the spin resonance with spin-orbit coupling

The spin excitations in the pnictides become anisotropic due to the spin-orbit coupling (SOC). At low energies, the excitations are peaked at the ordering vectors $\mathbf{Q}_1 = (\pi,0)$ and $\mathbf{Q}_2 = (0,\pi)$. Therefore, we introduce two vector
order parameters, $M_1$ and $M_2$. Following the theoretical results of Ref. [9], at sufficient hole-doping concentrations the $c$-axis becomes the easy axis, in agreement with experiments in Na-doped Ba122 [10]. Thus, the quadratic part of the free energy acquires the anisotropic contribution:

$$F(M_1, M_2) = \alpha_a (M_{1a}^2 + M_{1b}^2) + \alpha_b (M_{2a}^2 + M_{2b}^2) + \alpha_c (M_{1c}^2 + M_{2c}^2)$$

(S1)

To simplify our analysis, hereafter we consider $\alpha_a = \alpha_b \equiv \alpha_{ab}$. According to Ref. [9], $\alpha_c < \alpha_{ab}$. In addition to the reorientation of the moments in the magnetic phase, the anisotropy of the spin excitations also affects the energy of the resonance mode in the $s^+$ superconducting (SC) phase. The spin resonance mode emerges as a spin-1 collective mode protected by the SC gap.

A full calculation of the spin resonance mode in the presence of spin-orbit coupling is beyond the scope of this paper (see Ref. [11]); here, we illustrate the effect of the spin anisotropy on the resonance mode using a semi-phenomenological approach. We consider a simple two band model containing one hole and one electron pocket. For simplicity, we assume perfect nesting – deviations from perfect nesting may shift the wave-vector of the resonance mode in the $s^+$ superconducting phase. The spin resonance mode vanishes.

Here, $\Lambda$ is the high energy cutoff and $N_f$ is the density of states at the Fermi level. For small $U$, $\omega \lesssim 2\Delta$. As $U$ increases, a magnetically ordered state appears inside the SC dome at the critical value $U^* = [2N_f \ln (2\Lambda/\Delta)]^{-1}$, and the resonance mode vanishes.

Phenomenologically, the main effect of the spin anisotropy, as shown by Eq. (S1), is to shift the different components of the non-interacting spin susceptibility according to

$$\chi^{-1}(Q, \omega) \rightarrow \chi_{ii}^{-1}(Q, \omega) = \chi^{-1}(Q, \omega) + \alpha_i \quad i = ab, c$$

(S3)

As a result, the RPA condition $\chi_{ii}^{-1}(Q, \omega) = U/2$ will give different resonance energies $\omega_i$ for different channels. Effectively, the critical value of the interaction $U^*$ depends on the polarization channel. Since $\alpha_c < \alpha_{ab}$, we have $U_c^* < U_{ab}^*$. Physically, this means that for the same value of $U$, $M_c$ is closer to its instability than $M_a$ or $M_b$. Evaluation of $\omega_i$ then gives (assuming $\alpha_i$ small):

$$\frac{\omega_i}{\sqrt{(2\Delta)^2 - \omega_i^2}} \tan^{-1} \frac{\omega_i}{\sqrt{(2\Delta)^2 - \omega_i^2}} = \frac{1}{2N_f \left( \frac{1}{U} - \frac{1}{U_i^*} \right)},$$

(S4)
Since $U^*_c < U^*_{ab}$, $\omega_c < \omega_{ab}$ in general, as shown in Fig. 4(c). The difference increases as $U^*_c$ is approached, since at that point $\omega_c$ vanishes but $\omega_{ab}$ remains finite. For that figure, we used the parameters $U^*_{ab}N_f = 4/19$ and $U^*_cN_f = 0.2$. 

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