Nodeless energy gaps of single-crystalline $\text{Ba}_{0.68}\text{K}_{0.32}\text{Fe}_2\text{As}_2$ as seen via $^{75}\text{As}$ NMR

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Abstract

We report $^{75}\text{As}$ nuclear magnetic resonance studies on a very clean hole-doped single-crystal $\text{Ba}_{0.68}\text{K}_{0.32}\text{Fe}_2\text{As}_2$ ($T_c = 38.5$ K). The spin-lattice relaxation rate $1/T_1$ shows an exponential decrease below $T \simeq 0.45T_c$ down to $T \simeq 0.11T_c$, which indicates a fully-opened energy gap. From the ratio $(T_1)_c/(T_1)_a$, where $a$ and $c$ denote the crystal directions, we find that the antiferromagnetic spin fluctuation is anisotropic in the spin space above $T_c$. The anisotropy decreases below $T_c$ and disappears at $T \to 0$. We argue that the anisotropy stems from spin-orbit coupling whose effect vanishes when spin-singlet electron pairs form with a nodeless gap.
The discovery of superconducting transition in electron-doped iron-arsenide LaFeAsO$_{1-x}$F$_x$ provides a new route to high temperature superconductivity [1]. Remarkably, many other $R$FeAsO$_{1-x}$F$_x$ ($R$: rare earth) were synthesized and $T_c$ was raised to 55 K in SmFeAsO$_{1-x}$F$_x$ [2], which is the highest among materials except cuprates. Soon after these works, the hole-doped BaFe$_2$As$_2$ was also found to be superconducting [3]. The large single crystals of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ are easy to obtain, which makes them a good system for studying many physical quantities.

One of the most outstanding issues for a new superconductor is the symmetry of the electron pairs which is directly related to the paring mechanism. Nuclear magnetic resonance (NMR) experiments found the electron pairs to be in the spin-singlet state [4] and indicated the existence of multiple energy gaps [4, 5]. The multiple-gap property is likely associated with the multiple electronic bands. The Fermi surfaces consist of two hole-pockets centered at the $\Gamma$ point and two electron pockets around the $M$ point [6]. However, whether there are nodes in the gap function or not is still under hot debate. Angle-resolved photoemission spectroscopy (ARPES) [7] suggested fully opened gaps, but thermal conductivity [8, 9] measurements suggested nodal gaps. The penetration depth measurements by different groups have led to opposite conclusions [10, 11].

Theoretically, the sign-reversing $s^{\pm}$-wave model has been considered as the most promising candidate [12–14], but $d$ wave or $s$ wave with zero gap, and even a conventional $s^{++}$ wave was also proposed [15–17]. It has been shown that the $s^{\pm}$-wave or a multiple-gap $d$-wave model can fit quite well the spin-lattice relaxation rate, $1/T_1$, which shows a rapid decrease below $T_c$ with a hump structure at $T \sim T_c/2$ [4, 5, 18–20]. However, an important feature that $1/T_1$ should decrease as an exponential function of $T$ expected for the $s$-wave gaps has not been observed so far, because of impurity scattering in the samples. The impurity scattering can also alter other physical properties [21]. Thus, the conclusions on the gap symmetry drawn so far are still controversial. Measurements in sufficiently clean samples are highly needed to resolve the issue.

Here we report $^{75}$As NMR study on a very clean single crystal Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ with $T_c = 38.5$ K that is the highest among reports for this family. We obtained two pieces of evidence for fully-opened gaps. First, we observe an exponential decay of $1/T_1$ below $T \sim 0.45T_c$ down to $T \sim 0.11T_c$. For the second piece of evidence, we find that the antiferromagnetic (AF) spin fluctuation (SF) is anisotropic in the spin space above $T_c$, but
The vertical axis for $H \parallel a$ is offset for clarity. (b) The $T$ dependence of the Knight shift with $H \parallel a$ axis and $H \parallel c$ axis, respectively. The arrow indicates $T_c$ for $H \parallel a$.

The anisotropy decreases below $T_c$ and disappears at $T \to 0$. We argue that the anisotropy is due to spin-orbit coupling, whose effect vanishes at $T \to 0$ because the electron pairs are in the spin-singlet state with nodeless gap.

The single crystal of Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ was grown by using the self-flux method and characterized as discussed elsewhere [22]. Both dc susceptibility measured by a superconducting quantum interference device and ac susceptibility measured by the NMR coil indicates $T_c = 38.5$ K at zero magnetic field. The $T_c$ is 37.6 K for $\mu_0 H (=7.5T) \parallel a$ axis and 36.4 K for $\mu_0 H (=7.5T) \parallel c$ axis. The $1/T_1$ was determined from an excellent fitting to $1 - M(t)/M(\infty) = 0.1 \exp(-t/T_1) + 0.9 \exp(-6t/T_1)$, where $M(t)$ is the nuclear magnetization at time $t$ after the saturation pulse [23].

Figure 1 (a) shows the $^{75}$As-NMR spectra by scanning the magnetic field at a fixed frequency, $\omega_0/2\pi = 55.1$ MHz. The nuclear quadrupole frequency $\nu_Q$ is found to be 5.1 MHz at 100 K which is smaller than that in the Sn-flux-grown sample (5.9 MHz) [20]. Since doping of K increases $\nu_Q$ [20], this suggests that the Sn-flux-grown crystal had a higher doping rate. The Knight shift $K$ was obtained from the central transition peak and determined with respect to $\omega_0/\gamma$ with the nuclear gyromagnetic ratio $\gamma = 7.2919$ MHz/T. Below $T_c$, $K$ is obtained by scanning $\omega_0$ at a fixed field to avoid the vortex pinning effect. Above $T_c$, we confirmed that the results obtained by scanning field and scanning frequency agree well.

The effect of the nuclear quadrupole interaction was taken into account in extracting $K_a$. As shown in Fig. 1 (b), both $K_a$ and $K_c$ show a sharp decrease below $T_c$, which indicates

![FIG. 1: (Color online) (a) $^{75}$As-NMR spectra at a frequency of $\omega_0/2\pi = 55.1$ MHz and $T=100$ K. The vertical axis for $H \parallel a$ is offset for clarity. (b) The $T$ dependence of the Knight shift with $H \parallel a$ axis and $H \parallel c$ axis, respectively. The arrow indicates $T_c$ for $H \parallel a$.](image-url)
FIG. 2: (Color online) The $T$ dependence of $1/T_1$. The error is within the size of the symbols. The dashed line shows the $T^3$ variation. The curves below $T_c$ are fits to a two-gap $s^\pm$ model using the same parameters for both directions. The inset shows the semilog plot of $1/T_1 T$ vs $T_c/T$, which evidences an activation type $T$ dependence of the relaxation.

spin-singlet pairing [4].

The main panel of Fig. 2 shows the $T$ dependence of $1/T_1$ that decreases rapidly below $T_c$, with the reduction over about five decades. The decrease at low $T$ is much faster than $T^3$ that is expected for a $d$-wave gap. As in other materials, $1/T_1$ shows a “knee” shape around half $T_c$, which indicates multiple gaps [4, 5]. To see the low-$T$ behavior more clearly, we plot $1/T_1 T$ as a function of inverse reduced-temperature $T_c/T$ in the inset. As can be seen there, $1/T_1 T$ shows a very good exponential behavior below 17 K. This is strong evidence for a fully opened gap.

Using the $s^\pm$-wave model and introducing the impurity scattering rate $\eta$ in the energy spectrum, $E = \omega + i \eta$ [24], but neglecting the quasiparticle damping effect for simplicity, we can fit the data quite well. For a sign-reversing two-gap model, as seen in Fig. 2, we obtain $\Delta^+_1 = 5.63 \ k_B T_c$, $\Delta^+_2 = 1.11 \ k_B T_c$, $N_1 : N_2 = 0.85 : 0.15$, where $N_i$ is the density of state (DOS) on band $i$, and $\eta = 0.044 \ k_B T_c$. For a model of three bands corresponding to ARPES [7], we obtain $\Delta^+_1 = 4.7 \ k_B T_c$, $\Delta^+_2 = 0.96 \ k_B T_c$, $\Delta^-_3 = 4.7 \ k_B T_c$, $N_1 : N_2 : N_3 = 0.85 : 0.15 : 0.044$.
FIG. 3: (Color online) $T$-dependence of the $\gamma^2(1/T_1 T)$. The arrow indicates $T_c$.

$N_3 = 0.44 : 0.12 : 0.44$, and $\eta = 0.022 k_B T_c$. The $\eta$ is much smaller than $\eta = 0.15 k_B T_c$ in LaFeAsO$_{0.92}$F$_{0.08}$ and $\eta = 0.22 k_B T_c$ in the Sn-flux grown Ba$_{0.72}$K$_{0.28}$Fe$_2$As$_2$, meaning that the present sample is much cleaner, as supported by a small resistivity of 26$\mu$Ω at $T_c$ and the much sharper spectrum-width that is only half the value for the Sn-flux grown crystal. The cleanness of the present crystal is the reason for the exponential behavior of $1/T_1$ at low $T$; impurity scattering brings about finite DOS that results in seemingly power-law $T$-dependence of $1/T_1$ at $T_c$.

Next we move to the normal state. Figure 3 shows the $T$ dependence of $1/T_1 T$ which increases with decreasing $T$ down to $T_c$, indicating strong AF SF. The $1/T_1 T$ stems from the magnetic susceptibility at all wave vectors. When there exists strong AF SF, one may assume $1/T_1 T = (1/T_1 T)^{AF} + (1/T_1 T)^0$, where $(1/T_1 T)^{AF}$ is due to the susceptibility at the AF wave vector $Q$, and $(1/T_1 T)^0$ is due to $s$-band electrons and the orbital hyperfine interaction. Note that above $T = 250$K, $1/T_1 T$ becomes a constant. By taking the averaged value of $1/T_1 T$ at $T \geq 250$K as $(1/T_1 T)^0$, $(1/T_1 T)^{AF}$ is then obtained. Figure 4 shows the ratio of the relaxation due to AF SF, $(T_1)^{AF}/(T_1)^0$, which is about 2. This result indicates that the SF is anisotropic in the spin space, as elaborated below.

Generally, $1/T_1$ is related to the transverse fluctuating internal magnetic field, $\delta H$, as follows:

$$\left( \frac{1}{T_1} \right)_z = \frac{\gamma^2}{2} \int_{-\infty}^{\infty} dt \cos(\omega_0 t) \left\langle \delta H_x(t) \delta H_x(0) + \delta H_y(t) \delta H_y(0) \right\rangle,$$  

where $\langle \cdots \rangle$ denotes the statistical average. $\delta H$ is related to the fluctuating moment $\mathbf{S}$ of
Fe as $\delta \mathbf{H} = \mathbf{A} \cdot \mathbf{S}$, where $\mathbf{A}$ is the hyperfine coupling tensor between the As nucleus and Fe spins.

For $Q = (\pi, 0)$ and $Q = (0, \pi)$ AF SF, one has \[29\]

$$
\mathbf{A}(\pi, 0) = \begin{pmatrix} 0 & 0 & A \\ 0 & 0 & 0 \\ A & 0 & 0 \end{pmatrix}, \quad \mathbf{A}(0, \pi) = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & A \\ 0 & A & 0 \end{pmatrix},
$$

(2)

respectively. One therefore obtains

$$
\left(1/T_1\right)^{AF}_{a,b} = \frac{\gamma^2}{4} A^2 \int_{-\infty}^{\infty} dt \cos(\omega_0 t) \langle S_a(t) S_a(0) + S_b(t) S_b(0) + S_c(t) S_c(0) \rangle,
$$

(3)

$$
\left(1/T_1\right)^{AF}_{c} = \frac{\gamma^2}{2} A^2 \int_{-\infty}^{\infty} dt \cos(\omega_0 t) \langle S_c(t) S_c(0) \rangle.
$$

(4)

Since $\langle S_j(t) S_j(0) \rangle$ ($j = a, b, c$) can be expressed in terms of the imaginary part of the susceptibility $\chi''_j$ through the fluctuation-dissipation theorem, \[f_{\infty} \cos(\omega_0 t) \langle S_j(t) S_j(0) \rangle = \frac{2k_B T \chi''_j(\omega_0) }{} \], the anisotropy of the relaxation can be expressed as

$$
R_{AF} = \frac{\left(1/T_1\right)^{AF}_{a,b}}{\left(1/T_1\right)^{AF}_{c}} = \frac{\chi''_a(\omega_0, Q) + \chi''_b(\omega_0, Q)}{2\chi''_c(\omega_0, Q)} + \frac{1}{2}.
$$

(5)

If $\chi''_a(\omega_0, Q) = \chi''_b(\omega_0, Q) = \chi''_c(\omega_0, Q)$, namely, if the SF is isotropic in the spin space, then

$$
R_{AF}^{iso} = 1.5.
$$

(6)

The observed $R_{AF}$ shown in Fig. 4 is much larger than 1.5, which follows from Eq. (5) that $\chi''_{a,b}(\omega_0, Q)$ is larger than $\chi''_c(\omega_0, Q)$ by about 50%.

We propose that the anisotropy in the SF stems from spin-orbit coupling (SOC) that mixes spin and orbital freedoms so that the magnetic susceptibility bears some orbital character, which is anisotropic. We use a two-band model \[30\] involving spin-orbit coupled $d_{xz}$ and $d_{yz}$ and calculate the anisotropy. Our theoretical study starts from a two-dimensional Hamiltonian:

$$
H = H_{TB} + H_{LS}.
$$

(7)

Here $H_{TB} = \sum_{k \mu \nu \sigma} \epsilon_{\mu \nu} d_{k \mu \sigma}^\dagger d_{k \nu \sigma}$ is the tight-binding Hamiltonian with orbit index $\mu, \nu = 1(xz), 2(yz)$ and spin index $\sigma = +1(\uparrow), -1(\downarrow)$. The SOC is described by $H_{LS} = 2\lambda_{LS} \sum \mathbf{L} \cdot \mathbf{S}_l = -i \lambda_{LS} \sum \kappa \sigma d_{k,1\sigma}^\dagger d_{k,2\sigma} + h.c.$.
FIG. 4: (Color online) $T$-dependence of the anisotropy of $T_1$ due to AF spin fluctuation. The dashed line marks the value for isotropic AF spin fluctuation.

The dynamical magnetic response is calculated at the random-phase-approximation level, with the on-site intra-orbit Hubbard interaction $H_U = \sum_{l \in \text{Lattice}, \mu} U n_{l\mu\uparrow} n_{l\mu\downarrow}$. The longitudinal (transverse) susceptibility $\bar{\chi}_c$ ($\bar{\chi}_t$) at $q=Q = (\pi, 0)$ or $(0, \pi)$ is

$$\bar{\chi}_{c,+}^{-} (q, i\nu_n) = \frac{1}{1 - \bar{\chi}_{0,c,+} (q, i\nu_n) \Gamma (q)} \bar{\chi}_{0,c,+} (q, i\nu_n),$$

where $\bar{\chi}$ is a $2 \times 2$ matrix in the orbital space with the matrix elements defined by $\bar{\chi}_{c,+}^{\mu\nu} (q, \tau) = \langle T_\tau S_\mu^z (-q, \tau) S_\nu^z (q, 0) \rangle$ and $\bar{\chi}_{t,+}^{\mu\nu} (q, \tau) = \frac{1}{2} \langle T_\tau S_\mu^+ (-q, \tau) S_\nu^- (q, 0) \rangle$. $\Gamma (q) = \text{diag}(U, U)$ is the Hubbard interaction vertex in the spin-spin channel. The bare susceptibility $\chi_0 (q, i\nu_n)$ can be easily obtained by diagonalizing the Hamiltonian shown in Eq. (7). We choose the nearest-neighbor hopping integral $t_1 = -0.1051 \text{ eV}$ [31].

The calculated magnetic anisotropy above $T_c$ with $\lambda=0.2|t_1| (U = 9.7|t_1|)$ and $\lambda=0.1|t_1| (U = 9.87|t_1|)$, leading to $R_{AF} \sim 2$ at $T = T_c$, is respectively shown in Fig. 5, which is in qualitative agreement with the experimental finding. Here $T_c=0.065|t_1|$ is obtained by a self-consistent calculation of a mean-field BCS model with a gap $\Delta = 0.309|t_1|$.

A particular feature we find experimentally is that the $T_1$ ratio decreases below $T_c$ and it approaches the characteristic value 1.5 for the isotropic SF. Below $T_c$, it is less trivial to subtract the contribution of $(1/T_1 T)^0$, so we simply plot the raw data as shown in Fig. 6. We emphasize, however, that this approximation does not affect our conclusion [32], since the contribution from $(1/T_1 T)^0$ to the observed $1/T_1 T$ is only 15% for $H \parallel a$ and 20% for $H \parallel c$ at $T = T_c$.

The asymptotic value 1.5 for $(T_1)_c/(T_1)_a$ implies that the SOC effect vanishes at $T \to 0$. 

\[ \text{FIG. 4: (Color online) $T$-dependence of the anisotropy of $T_1$ due to AF spin fluctuation. The dashed line marks the value for isotropic AF spin fluctuation.} \]**

\[ \text{The dynamical magnetic response is calculated at the random-phase-approximation level, with the on-site intra-orbit Hubbard interaction $H_U = \sum_{l \in \text{Lattice}, \mu} U n_{l\mu\uparrow} n_{l\mu\downarrow}$. The longitudinal (transverse) susceptibility $\bar{\chi}_c$ ($\bar{\chi}_t$) at $q=Q = (\pi, 0)$ or $(0, \pi)$ is} \]

\[ \bar{\chi}_{c,+}^{-} (q, i\nu_n) = \frac{1}{1 - \bar{\chi}_{0,c,+} (q, i\nu_n) \Gamma (q)} \bar{\chi}_{0,c,+} (q, i\nu_n), \]

\[ \text{where $\bar{\chi}$ is a $2 \times 2$ matrix in the orbital space with the matrix elements defined by $\bar{\chi}_{c,+}^{\mu\nu} (q, \tau) = \langle T_\tau S_\mu^z (-q, \tau) S_\nu^z (q, 0) \rangle$ and $\bar{\chi}_{t,+}^{\mu\nu} (q, \tau) = \frac{1}{2} \langle T_\tau S_\mu^+ (-q, \tau) S_\nu^- (q, 0) \rangle$. $\Gamma (q) = \text{diag}(U, U)$ is the Hubbard interaction vertex in the spin-spin channel. The bare susceptibility $\chi_0 (q, i\nu_n)$ can be easily obtained by diagonalizing the Hamiltonian shown in Eq. (7). We choose the nearest-neighbor hopping integral $t_1 = -0.1051 \text{ eV}$ [31].} \]

\[ \text{The calculated magnetic anisotropy above $T_c$ with $\lambda=0.2|t_1| (U = 9.7|t_1|)$ and $\lambda=0.1|t_1| (U = 9.87|t_1|)$, leading to $R_{AF} \sim 2$ at $T = T_c$, is respectively shown in Fig. 5, which is in qualitative agreement with the experimental finding. Here $T_c=0.065|t_1|$ is obtained by a self-consistent calculation of a mean-field BCS model with a gap $\Delta = 0.309|t_1|$.} \]

\[ \text{A particular feature we find experimentally is that the $T_1$ ratio decreases below $T_c$ and it approaches the characteristic value 1.5 for the isotropic SF. Below $T_c$, it is less trivial to subtract the contribution of $(1/T_1 T)^0$, so we simply plot the raw data as shown in Fig. 6. We emphasize, however, that this approximation does not affect our conclusion [32], since the contribution from $(1/T_1 T)^0$ to the observed $1/T_1 T$ is only 15% for $H \parallel a$ and 20% for $H \parallel c$ at $T = T_c$.} \]

\[ \text{The asymptotic value 1.5 for $(T_1)_c/(T_1)_a$ implies that the SOC effect vanishes at $T \to 0$.} \]
FIG. 5: (Color online) Calculated magnetic anisotropy above $T_c$ due to SOC. The parameters and $k_B T$ are in units of $|t_1|$, where $t_1$ is the nearest-neighbor hopping integral.

FIG. 6: (Color online) $T$-dependence of the $T_1$ anisotropy below $T_c$. The dashed straight line indicates the value for isotropic AF SF.

This can happen only when the gaps are fully opened in the case of spin-singlet pairing with $\Delta > \lambda_{LS}$. When there are nodes in the gap function, $1/T_1$ at low $T$ is governed by the nodal quasiparticles that are spin-orbit coupled, thereby $(T_1)_c/(T_1)_a$ should resume its value of 2.0 at $T = T_c$. Note also that $(T_1)_c/(T_1)_a$ should become $\sim 0.75$ if the AF SF completely vanishes [29]. Thus, our finding of the decrease of $(T_1)_c/(T_1)_a$ to 1.5 at $T \rightarrow 0$ is another strong evidence for nodeless gap and implies that the AF SF persists in the superconducting state.

In conclusion, from the NMR measurements on a clean single crystal Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$, we find the long-sought exponential decrease of $1/T_1$ at low $T$, which evidences a fully-opened gap. In the normal state, the AF SF is anisotropic in the spin space. However, the anisotropy diminishes below $T_c$ and vanishes at the zero-$T$ limit, which is a feature indicating
nodeless gap.

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[32] Below $T_c$, if we assume that $(1/T_1 T)_0$ follows a function as shown by the solid curve in Fig. 2 and subtract it from $1/T_1 T$, then $R_{AF}$ decreases from 2 at $T = T_c$ to 1.58 at the lowest temperature $T = 0.11 T_c$. 