NMR study of hole-doped iron-pnictide superconductor $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \ (x = 0.27-1)$

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Abstract. We performed the $^{75}$As nuclear magnetic resonance measurements of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (BKFA) for $x = 0.27, 0.39, 0.58, 0.64, 0.69$ and $1$ to understand gap structure in this system. Temperature dependence of spin lattice relaxation rate ($1/T_1$) below superconducting (SC) transition temperature $T_c$ changes gradually from $x = 0.39$ to $1.0$. This result indicates that the SC gap structure changes smoothly from full gap state with maximum $T_c$ into nodal-line structure state for $x = 1$ ($\text{KFe}_2\text{As}_2$). BKFA does not have distinct SC symmetry change. The nodal gap structure appears continuously and develops gradually with increasing $x$.

1. Introduction
The discovery of superconductivity in F-doped LaFeAsO at 26 K by Kamihara et al. heralded a new era of superconductivity based on Iron [1]. Members of superconducting (SC) iron family were observed one after another in the oxygen-free compounds. Among them, $\text{AEFe}_2\text{As}_2$ ($\text{AE} = \text{Ba, Ca and Sr}$) (so-called 122 compounds) occupies a singular position since the high quality large single crystals are grown which is suitable for detail study of SC properties.

$\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (BKFA) is a hole-doped superconductor with the maximum SC transition temperature $T_c$ of 38 K around $x = 0.4$. One feature of this system is that the superconductivity appears widely for $x$ from 0.2 to 1 [2]. The extensive investigations have been performed for the optimum hole doped compound ($x \sim 0.4$) and the end member $\text{KFe}_2\text{As}_2$ ($x = 1$). Many physical property measurements, i.e., angle-resolved photoemission spectroscopy (ARPES) and magnetic field penetration measurements, indicate the appearance of fully opened multiple SC gap for $x = 0.4$ [3][4][5]. Temperature $T$ dependence of spin lattice relaxation rate ($1/T_1$) in optimum doped region has no coherence peak below $T_c$ and varies proportional to $T^{3-5}$. This $T$ dependence is different from the behavior expected for conventional $s$-wave superconductor and is usually recognized as an appearance of non $s$-wave superconductivity. In case of iron pnictide superconductor, this behavior could be explained by nodeless $s_\pm$ model in which SC phase has different sign in hole and electron band. On the contrary, $^{75}$As nuclear quadrupole resonance (NQR) and specific heat studies have pointed out appearance of multiple nodal SC
gaps in KFe$_2$As$_2$ [6]. The nodal gap structure for KFe$_2$As$_2$ was also supported by magnetic field penetration measurements [7].

The change of the gap structure associated with hole doping from $x = 0.4$ to 1 is important for the understanding of iron pnictide superconductors. Although the results indicates that two regions may have different gap symmetries, there are few reports about the overdoped region that is intermediate area. $T$ dependence of $1/T_1$ in SC state reflects thermal excitation of quasiparticles, which represents the SC gap structure. Recently we succeeded growing high quality single crystals of BKFA for $x = 0.4$, 0.58, 0.64 and 0.69. Here we show $^{75}$As nuclear magnetic resonance (NMR) measurements in BKFA.

2. Experimental

Single crystals of BKFA ($x = 0.27$, 0.39, 0.58, 0.64, 0.69, 1) were grown by the BaAs and KAs self-flux method [8]. X-ray diffraction showed that the crystals had a tetragonal ThCr$_2$Si$_2$ type structure with no impurity phase. The ratio of Ba and K was determined by the energy dispersive X-ray spectroscopy. We determined the $T_c$ in the external field by the change of inductance of NMR detecting coil with changing $T$.

The NMR experiment on the $^{75}$As nucleus ($I = 3/2$, $\gamma/2\pi = 7.292$ MHz/T) was carried out using phase-coherent pulsed NMR spectrometers and a SC magnet between approximately 2 and 8 T. The measurement was performed using a $^4$He cryostat. The NMR spectra were measured by sweeping the applied fields at a constant resonance frequency. Magnetic field was applied parallel and perpendicular to the crystal c-axis. We measured $T_1$ at a fixed frequency of 37.15 MHz with an external field of 5.06-5.09 T in the $T$ range of 2-300 K. The nuclear magnetization recovery curve was fitted by the following double exponential function as expected for the center line of the spectrum for the nuclear spin $I = 3/2$ of the $^{75}$As nucleus,

$$1 - \frac{m(t)}{m_0} = 0.1 \exp\left(-\frac{t}{T_1}\right) + 0.9 \exp\left(-\frac{6t}{T_1}\right),$$

where $m(t)$ and $m_0$ are nuclear magnetizations after a time $t$ from the NMR saturation pulse and thermal equilibrium magnetization.

3. Results and discussion

In Fig. 1 (a), we show $T$ dependence of $1/T_1$ for $x = 0.39$, 0.58, 0.69 and 1. Each arrow indicates $T_c$ under external field. $1/T_1$ has no coherence peak just below $T_c$ and decreases rapidly. $T$ dependence of $1/T_1$ from $T_c$ to 0.8$T_c$ is approximately proportional to $T^2$ for $x = 0.39$, $T^{3.2}$ for $x = 0.58$, $T^{2.2}$ for $x = 0.69$. The power-law behavior was also observed at lower temperature. The exponent $\alpha$ of power-law $T^\alpha$ decreases with increasing $x$. In order to understand this change of gap structure, we calculate $1/T_1$ in SC state with two gaps model. For simplicity we assume that both gaps have same symmetry such as full gapped $s_\pm$ or line node. The $1/T_1$ in SC state is expressed as,

$$\frac{1}{T_1} \propto \sum_{i=1,2} n_i^2 \int_0^\infty \left\{ N_i^2(E)^2 + M_i^2(E)^2 \right\} f(E) \{1 - f(E)\} dE,$$

where $N_i^2(E)^2$, $M_i^2(E)^2$, $f(E)$ are the density of states (DOS), the anomalous DOS arising from the coherence effect of Cooper paris, and the Fermi distribution functions, respectively. $n_i$ presents the fraction of DOS of the $i$-th gap and $n_1 + n_2 = 1$.

In Fig. 1 (b), we show experimentally obtained $1/T_1$ and calculated $1/T_1$. Each $1/T_1$ is normalized by the value at $T_c$ in order to clarify $x$ dependence of $1/T_1$. The experimental data for $x = 1$ was quoted from Ref.[6] using poly crystals, because we could not perform NMR/NQR
Figure 1. (color online) (a) $T$ dependence of $1/T_1$ with external field parallel to the $c$-axis was plotted for $x = 0.39, 0.58, 0.69$ and 1. Solid and open symbols are obtained at 37.15 MHz and 17.75 MHz, respectively. Each arrow indicates $T_c$ under external field. (b) The calculated $1/T_1$ are added to the experimental data. Each $1/T_1$ is normalized by the value at $T_c$. Black dashed lines are obtained by full gapped $s_\pm$ model and aqua solid lines by line node model. The data for $x = 1$ was quoted from Fukazawa et al. [6].

Table 1. Fitted parameters

| Gap type        | $x$ | $2\Delta_1(0)/T_c$ | $2\Delta_2(0)/T_c$ | $n_1$ | $\delta_i/\Delta_i$ |
|-----------------|-----|---------------------|---------------------|-------|--------------------|
| Full gaped $s_\pm$ | 0.39 | 9.6 | 2.5 | 0.76 | 0.1 |
| Full gaped $s_\pm$ | 0.58 | 5.8 | 0.92 | 0.75 | 0.1 |
| Full gaped $s_\pm$ | 0.69 | 4.8 | 0.62 | 0.65 | 0.1 |
| Full gaped $s_\pm$ | 1.0  | 4.0 | 0.54 | 0.51 | 0.1 |
| Line node       | 0.39 | 14.2 | 4.4 | 0.80 | - |
| Line node       | 0.58 | 8.1 | 0.91 | 0.75 | - |
| Line node       | 0.69 | 5.6 | 0.66 | 0.64 | - |
| Line node       | 1.0  | 4.2 | 0.48 | 0.50 | - |

measurements using single crystals owing to the drastic reduction of NMR signals in SC state. Black dashed lines are obtained by full gaped $s_\pm$ model and aqua solid lines by line node model. The best fitted parameters are listed in Table 1. Although we neglected the impurity effect, the result for $x = 0.39$ is consistent with the report by Matano et al. [4]. Both models can well
explain superconductivity in this system, although the fitting at low temperatures is worse than that at higher temperatures.

The $x$ dependence of $2\Delta_i/T_c$ for both $i$'s shows monotonous decrease with increasing $x$. The smaller gap $2\Delta_2/T_c$ rapidly decreases from $x = 0.39$ to 0.58. This probably originates from the poorer nesting condition which arises from the change of band structure by hole doping. The larger gap $2\Delta_1/T_c$ affects the inclination of $1/T_1$ just below $T_c$ and is directly related with the SC symmetry. Compared with the $x$ dependence of $2\Delta_2/T_c$, the $x$ dependence of $2\Delta_1/T_c$ is more gradual and the magnitude decreases continuously. Therefore, it can be concluded that there is no SC symmetry change in BKFA and that the nodal-line SC gap structure realized in $x = 1$ should be explained with the same SC symmetry as that realized in optimum doped region $x \sim 0.4$. We speculate that nodal-line structure emerges from $x = 0.6$-0.7 and develops gradually without the change of SC gap symmetry in this system.

However, the gap symmetry cannot be determined solely with NMR measurements, which can be clearly understood from the calculated results. To determine gap structure, not only NMR/NQR experiments but also other experiments such as specific heat, low temperature ARPES, are necessary as described in Ref.[6]. Such experiments are now in progress.

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