Inverse isotope effect in Iron based superconductor

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Abstract:

We have found that (Ba,K)Fe₂As₂ superconductor (a transition temperature, $T_c \sim 38$ K) shows an inverse Iron isotope effect ($\alpha_{Fe} = -0.18 \pm 0.03$, where $T_c \sim M^{-\alpha_{Fe}}$ and M is the iron mass), i.e. the sample containing the larger iron mass depicts higher T_c . Systematic studies using three types of Fe-isotopes (⁵⁴Fe, natural Fe and ⁵⁷Fe) reveal a clear inverse shift on T_c by measurements of temperature dependent magnetization and resistivity. The inverse isotope effect that is the first case in high- T_c superconductors strongly suggests that superconducting mechanism of the iron-based system is not explained by conventional BCS theory mediated by phonons.

KEYWORDS: isotope effect, high- T_c superconductivity, iron-based superconductor, (Ba,K)Fe₂As₂, high-pressure synthesis

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1. Introduction

Refreshment of the research in the superconductivity field was accelerated by the discovery of the superconductivity with a transition temperature (T_c) of 26 K in the Fe-based oxypnictide $\text{LaFeAsO}_{1-x}\text{F}_x$ and which explored the new high- T_c superconductors, T_c as high as 56 K [1-2]. The mechanism of superconductivity in a newly discovered iron based superconductor either conventional or unconventional is not settled yet [3-5], due to the pairing mechanism is currently in dispute. One way to settle this problem is to study the isotope effect, which deals directly with the fundamental of the pairing mechanism. In conventional superconductors, a strong effect of changing ion mass M on the transition temperature T_c implies that lattice vibrations (phonons) play an important role in the microscopic mechanism of superconductivity [6-7]. The isotope exponent α is defined by $T_c \sim M^{-\alpha}$, where M is the isotopic mass. In the classical form of BCS theory[8], α is equal to 0.5. For simple metals like Hg, Pb, Sn, and Zn, the isotope coefficient is found experimentally to be close to 0.5. A nearly zero oxygen-isotope effect ($\alpha_O \approx 0.03$) was earlier observed in optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ ($T_c \sim 92$ K) superconductor [9,10], indicating non-phonon mediated. On the other hand, large oxygen-isotope shifts were later observed in several underdoped cuprate superconductors [11-17]. The oxygen isotope effect in $\text{Ba}_{0.625}\text{K}_{0.375}\text{BiO}_3$ has a large isotope exponent ($\alpha_O \approx 0.41$), indicating that the pairing mechanism is conventional (phonon-mediated)[18]. Boron isotope effect($\alpha_B \approx 0.26$) in superconducting MgB_2 which is consistent with phonon-mediated coupling within the framework of the BCS model [19].

Here we show the iron isotope effect on T_c in $(\text{Ba,K})\text{Fe}_2\text{As}_2$ superconductor ($T_c \approx 38$ K)[20]. Negative isotope effect coefficient α_{Fe} ($= -0.18 \pm 0.03$) has been obtained between the samples synthesized simultaneously under high pressure.

2. Experimental:

We used three sorts of Fe, ^{57}Fe and ^{54}Fe enriched powder (ISO FLEX USA, purity of Fe 99.99%), and natural Fe powder ($^{\text{n}}\text{Fe}$) (Showa Chemical Industry, 99.9%). The mass of the ^{57}Fe , $^{\text{n}}\text{Fe}$ and ^{54}Fe powders are 57.00, 55.85 and 54.01, respectively. In order to evaluate the isotopic Fe powders, we measured Raman spectroscopy of Fe_2O_3 that are synthesized by oxidizing the Fe powders as shown in Fig. 1. We confirmed that the observed Raman shift is consistent with calculated one [21].

Polycrystalline samples of $(\text{Ba,K})\text{Fe}_2\text{As}_2$ were prepared by high-pressure synthesis method by using a cubic-anvil-type apparatus and details are reported elsewhere [22]. The versatility of high-pressure synthesis technique for preparation of iron-based superconductors has been already reported [23]. Variation in the T_c values due to different synthesis condition has been ruled out by synthesizing a set of two different isotope-containing samples in a BN crucible. The sample assembly for the high-pressure synthesis method is shown in the inset of Fig. 2. Three combinations of isotopic Fe, $^{\text{n}}\text{Fe}$ and ^{57}Fe , ^{57}Fe and ^{54}Fe , ^{54}Fe and $^{\text{n}}\text{Fe}$ were tested for sample preparation to ensure systematic behavior of T_c . We obtained 7 sets of samples (S1-S7) synthesized in the same conditions except for the iron mass.

Powder X-ray diffraction (XRD) patterns of the samples were measured using CuK_α radiation. The dc magnetic susceptibility was measured using a SQUID magnetometer (Quantum Design MPMS) in the temperature range 5 K to 40 K under a magnetic field of 50 Oe. The resistivity was measured by a four-probe method (Quantum Design physical property measurement system (PPMS)).

3. Results and discussion:

The XRD patterns of the samples synthesized using ^{56}Fe and ^{57}Fe isotopic powders, S1(^{56}Fe) and S1(^{57}Fe) are represented in Fig. 2 as a typical example. No impurity peaks appearance in the XRD pattern indicates the high quality samples. Most of the peaks were indexed as the $(\text{Ba,K})\text{Fe}_2\text{As}_2$ phase with tetragonal space group symmetry $I4/mmm$. The lattice parameters of the samples S1(^{57}Fe) and S1(^{56}Fe) are $a=3.919\text{\AA}$, $c=13.283\text{\AA}$ and $a=3.919\text{\AA}$, $c=13.274\text{\AA}$, respectively. The deviation in lattice parameters between the samples synthesized simultaneously is small (negligible). Judging from the c -axis parameter, K concentration, x , in $(\text{Ba}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$ is about 0.4 which is located at the optimal doping state [24], where T_c is almost constant against x so that it is suitable for verification of the isotope effect. We also confirmed the K concentration ($x \sim 0.4$) by SEM-EDX method.

Figure 3 represent the temperature (T) dependence of normalized susceptibility (χ) for the combinations of the samples, S1(^{56}Fe) and S1(^{57}Fe). Sharp superconducting transitions with a width of about 2 K, indicates good homogeneity of the samples. Surprisingly, the sample including the larger atomic weight of Fe depicts higher T_c , indicates the inverse isotope effect. The isotope exponent is estimated to be $\alpha_{\text{Fe}} = -0.16$.

In Fig. 4 the temperature (T) dependent normalized resistivity of the samples S1(^{57}Fe) and S1(^{56}Fe) are shown. This result also clearly shows that sample containing ^{57}Fe has higher T_c than ^{56}Fe , is consistent with inverse isotope effect evidenced in the magnetic susceptibility measurements. The isotope exponent ($\alpha_{\text{Fe}} = -0.15$) is slightly different than that measured by magnetic susceptibility method, but comparable. Isotope shifts in T_c (ΔT_c) determined by susceptibility measurements and α_{Fe} of the sample sets S1-S7 are listed in Table 1. The average α_{Fe} of the samples sets S1-S7 is -0.18 ± 0.03 . Our result is in contradictory with the earlier report ($\alpha_{\text{Fe}} = 0.37$) on

(Ba,K)Fe₂As₂ [25]. The contradiction may result from the difference in K concentration of the samples. It will be necessary to measure the α_{Fe} as a function of K concentration.

In conclusion, a significant inverse Iron isotope effect ($\alpha_{Fe} = -0.18 \pm 0.03$) was observed in (Ba,K)Fe₂As₂ superconductor that is the first case in high- T_c superconductors [26]. Results presented here strongly demonstrate that phonon plays insignificant role and implies the exotic coupling mechanism in this superconductivity. Understanding of this inverse isotope effect surely deals with the development of the field of conventional and unconventional superconductivity.

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Figure captions

Fig. 1. Raman spectra of Fe_2O_3 that are synthesized by oxidizing the Fe powders (^{54}Fe , ^nFe and ^{57}Fe). Main peak positions are 229.2, 225.4 and 223.2 for $^{57}\text{Fe}_2\text{O}_3$, $^n\text{Fe}_2\text{O}_3$ and $^{54}\text{Fe}_2\text{O}_3$, respectively.

Fig. 2 X-ray diffraction patterns of the sample set S1(^{57}Fe) and S1(^nFe) synthesized simultaneously in a BN crucible by high pressure synthesis technique. The inset shows the sample assembly for the high-pressure synthesis method.

Fig. 3 Temperature (T) dependence of susceptibility (χ) of the sample set S1 using combinations of the isotopic Fe, ^nFe and ^{57}Fe . The χ is normalized at the values of ZFC curve at 5K. T_c is determined from the intersection as shown in the inset. Fe isotope exponent is estimated to be $\alpha_{Fe} = -0.16$.

Fig. 4 Temperature (T) dependence of resistivity (ρ) of the sample (^{57}Fe) and (^nFe). T_c is determined from the intersection of the two extrapolated lines as shown in the inset. T_c is determined from the intersection as shown in the inset. Fe isotope exponent is estimated to be $\alpha_{Fe} = -0.15$.

Table 1: Isotope shift in T_c (ΔT_c) and α_{Fe} of the sample sets S1-S7. T_c are determined from the magnetic susceptibility measurements of (Ba,K)Fe₂As₂ samples containing ⁵⁴Fe, ⁵⁶Fe and ⁵⁷Fe isotopes. The average α_{Fe} is -0.18 ± 0.03 .

	T_c (K)			ΔT_c	α_{Fe}
	⁵⁴ Fe	⁵⁶ Fe	⁵⁷ Fe		
S1		37.79	37.91	-0.12	-0.16
S2	37.56		37.82	-0.26	-0.13
S3	37.51	37.76		-0.25	-0.20
S4	37.54	37.79		-0.25	-0.20
S5	37.32		37.75	-0.43	-0.21
S6	37.42		37.77	-0.35	-0.17
S7	37.39		37.76	-0.37	-0.18

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Shirage-PHYSC-Figure 1

Shirage-PHYSC-Figure 2

Shirage-PHYSC-Figure 3

Shirage-PHYSC-Figure 4