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Iron Isotope Effect on T_c in optimally-doped (Ba,K)Fe₂As₂ (T_c = 38 K) and SmFeAsO_{1-y} (T_c = 54 K) Superconductors

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Abstract:

We report the iron isotope effect on a transition temperature (T_c) in an optimally-doped (Ba,K)Fe₂As₂ (T_c = 38 K) and SmFeAsO_{1-y} (T_c = 54 K) superconductors. In order to obtain the reliable isotope shift in T_c , twin samples with different iron isotope mass are synthesized in the same conditions (simultaneously)

under high-pressure. We have found that (Ba,K)Fe₂As₂ shows an inverse iron isotope

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effect $\alpha_{Fe} = -0.18 \pm 0.03$ while SmFeAsO_{1-y} shows a small iron isotope effect $\alpha_{Fe} = -0.02 \pm 0.01$, where the isotope exponent α is defined by $T_c \sim M^{-\alpha}$ (M is the isotopic mass). The results show that α_{Fe} changes in the iron-based superconductors depending on the system. The distinct iron isotope effects imply the exotic coupling mechanism in the iron-based superconductors.

KEYWORDS: isotope effect, high- T_c superconductivity, iron-based superconductor, $(\text{Ba,K})\text{Fe}_2\text{As}_2$ and SmFeAsO_{1-y} , high-pressure synthesis

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1. Introduction

The discovery of high- T_c superconductivity in $\text{LaFeAsO}_{1-x}\text{F}_x$ ($T_c=26$ K)[1], decorated the new horizon in the field of superconductivity and the scientific community welcomed these materials as a new high- T_c family. The highest T_c so far tuned in these materials is crossing a value of 50 K. Many techniques are employed and theories are proposed to reveal the glue behind the existence of so high- T_c in these materials. One of the tools to explore the pairing mechanism in superconductivity is the study of the nuclear mass dependent T_c , which was discovered [2] well before the establishment of so-called Bardeen-Cooper-Schrieffer (BCS) classical theory. T_c varies with the isotopic mass of the sample was the first direct evidence for interaction between the electrons and the lattice (phonon). This indicates that lattice vibrations are a part of the superconducting process. This was an important clue in the process of developing the BCS theory because it suggested lattice coupling, and in the quantum treatment suggested that phonons were involved. The origin of this pairing is one of the great unsolved mysteries in the field of high- T_c superconductivity. Many elemental superconductors and alloy superconductors are well fitted with BCS-theory whose isotope exponent is close to 0.5 ($\alpha = 0.5$) can be treated as the conventional superconductors and then suggests that electron-phonon interactions are involved. On the other hand, the optimal doped cuprate superconductor's exhibits negligibly small isotope effect that indicates these materials are unconventional (electron-phonon interactions are not essential). In the recently discovered an iron based high- T_c superconductors, the issue of pairing mechanism is unclear as large isotope effect[3], inverse isotope effect[4,5] and very small isotope effect[6] are reported. Here we demonstrate the experimental and most reliable results on iron isotope effect, in which

the samples are prepared by high pressure synthesis technique, one of the effective techniques to grow the high quality samples.

2. Experimental

The usefulness of high-pressure synthesis technique was subsequently demonstrated in the iron-based superconductors [7]. Polycrystalline samples were prepared by high-pressure synthesis method using a cubic-anvil-type apparatus and the sample preparation is reported elsewhere [4-6]. Variation in the T_c values due to different synthesis condition has been ruled out by synthesizing simultaneously a set of two different isotope-containing samples. The samples synthesized simultaneously are almost single phase and the lattice parameters of the twin samples are almost identical to each other [4-6]. We used three sorts of Fe, ^{57}Fe and ^{54}Fe enriched powder (ISOFLEX USA, purity of Fe 99.99%), and natural Fe powder ($^{\text{n}}\text{Fe}$, mass = 55.85) (Showa Chemical Industry, 99.9%). In case of SmFeAsO_{1-y} samples, we used the $^{\text{n}}\text{Fe}$ powder instead of using the ^{57}Fe and ^{54}Fe metal powders directly, because the degree of oxidization of the raw (as-purchased) isotope powders turned out to be different from each other. In our process, 57% of Fe in the samples is from the $^{\text{n}}\text{Fe}$ powder so that the average isotope masses of Fe are 56.35 and 55.05 for the samples synthesized using the $^{57}\text{Fe}_2\text{O}_3$ and $^{54}\text{Fe}_2\text{O}_3$ powders, respectively.

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) under a magnetic field of 5 Oe. The resistivity of the two isotope containing samples was measured simultaneously by a four-probe method (Quantum Design PPMS).

3. Results and discussion

3.1 Inverse iron isotope effect on T_c in $(\text{Ba,K})\text{Fe}_2\text{As}_2$

We have prepared seven sets of samples and **Fig. 1** represent the temperature (T) dependence of normalized susceptibility (χ) for the samples synthesized from the natural iron powder ($^{\text{n}}\text{Fe}$) and ^{54}Fe enriched one as a typical example. The both samples show sharp superconducting transitions at ~ 38 K, which corresponds to optimally doped state [8]. Superconducting transition width of about 2 K indicates good homogeneity of the samples. T_c is determined from the intersection of two extrapolated lines (see the inset of Fig.1). A clear isotope shift in T_c ($\Delta T_c = 0.25$ K) can be seen in this combination. Surprisingly, the sample including the larger atomic weight of Fe depicts higher T_c , indicates the inverse isotope effect ($\alpha_{Fe} = -0.20$). An average isotope exponent α_{Fe} for seven sets of samples is $\alpha_{Fe} = -0.18 \pm 0.03$.

In Fig. 2 the temperature (T) dependent normalized resistivity of the same samples in Fig. 1 are shown. T_c is determined by the definition as a similar way stated in the susceptibility measurement shown in the inset of Fig. 2. This result also clearly shows that sample containing $^{\text{n}}\text{Fe}$ has higher T_c than ^{54}Fe , is consistent with negative isotope effect evidenced in the magnetic susceptibility measurements (see Fig.1). The isotope exponent is estimated to be $\alpha_{Fe} = -0.15$, which is comparable with isotope exponent determined from the magnetic susceptibility measurements. Our result is in contradictory with the earlier report on $(\text{Ba,K})\text{Fe}_2\text{As}_2$ ($\alpha_{Fe} = 0.37$) [3]. It may be due to different doping level. The inverse isotope effect in $(\text{Ba,K})\text{Fe}_2\text{As}_2$ is the first case in high- T_c superconductors[9].

3.1 A small iron isotope effect on T_c in SmFeAsO_{1-y}

Fig. 3 represents the temperature (T) dependence of field-cooled (FC) magnetic susceptibility (χ) for the samples synthesized from the $^{57}\text{Fe}_2\text{O}_3$ and $^{54}\text{Fe}_2\text{O}_3$ powders (the average iron isotope masses are 56.35 and 55.05, respectively) as a typical example. The magnitude (χ) is normalized at 5 K (-1.0) and 56 K (0.0) values. Superconducting transitions are sharp enough, allowing us to define T_c accurately. T_c is determined as stated in Fig. 1 and shown in the inset of Fig. 3. As indicated in the inset, shift of T_c is at most -0.01 K, which correspond to the iron isotope coefficient $\alpha_{Fe} = -0.01$. An average iron isotope coefficient for five sets of sample is $\alpha_{Fe} = -0.02 \pm 0.01$. This iron isotope effect is negligibly small, first case in the iron based superconductors and it is different from that of $(\text{Ba,K})\text{Fe}_2\text{As}_2$ even though both systems are in an optimally doped state.

Our result is also in apparently contradictory with a large $\alpha_{Fe} = 0.34$ on $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$ ($T_c = 41$ K) by Liu *et al.* [3]. Giving lower T_c 's of their samples in comparison to the present study, their samples are probably located at the so-called under-doped region and the apparent difference between the two experiments is likely due to the different doping level of the samples. If this is the case, this resembles the situation with the cuprate superconductors where the oxygen-isotope effect is negligibly small in the optimally doped samples, while that in the under- or over-doped one is large, almost close to 0.5 [10-18]. It should be necessary to evaluate α_{Fe} as a function of doping level in the iron-based superconductors, which is really technically challenging as the variation due to different doping level and isotope effect can not be easily distinguished.

Conclusions:

The inverse iron isotope effect in $(\text{Ba, K})\text{Fe}_2\text{As}_2$ ($T_c = 38$ K) at its optimal doping state is the first report in the family of high- T_c superconductor to show this

phenomenon. While a small isotope effect at its optimal state in SmFeAsO_{1-y} ($T_c = 54$ K) superconductor was evidenced. This anomalous isotope mass dependence on T_c will be a key to understand the superconducting mechanism in the iron based system.

Acknowledgments

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Figure captions:

Fig. 1 Temperature (T) dependence of susceptibility (χ) of (Ba,K)Fe₂As₂ samples synthesized from the natural iron powder (ⁿFe) and ⁵⁴Fe enriched one. The χ is normalized at the values of ZFC curve at 5 K. Fe isotope exponent is estimated to be $\alpha_{Fe} = -0.20$.

Fig. 2 Temperature (T) dependence of resistance (R) of the (Ba,K)Fe₂As₂ samples synthesized from the natural iron powder (ⁿFe) and ⁵⁴Fe enriched one. T_c is determined from the intersection of the two extrapolated lines as shown in the inset. Fe isotope exponent is estimated to be $\alpha_{Fe} = -0.15$.

Fig. 3 Temperature (T) dependence of the field-cooled (FC) susceptibility (χ) normalized at 5 K and 56 K for SmFeAsO_{1-y} samples with the average isotope masses of Fe are 56.35 and 55.05. The inset shows the T dependence of χ near T_c . An iron isotope coefficient is estimated to be $\alpha_{Fe} = -0.01$.

Figure 1

Figure 2

Figure 3